FACTORS DETERMINING THE OCCURRENCE AND CHARACTERISTICS OF NEW PARTICLE FORMATION EVENTS – A STUDY OVER A CONTINENT

by

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Abstract

New Particle Formation (NPF) events, an important source of ultrafine particles in the atmosphere, were studied in multiple sites across Europe. Apart from the importance of meteorological conditions and atmospheric composition variables and compounds, such as the solar radiation intensity, the relative humidity or the condensation sink in the occurrence and development of the events, the importance of the origin of the incoming air masses and the specific characteristics that come with them is underlined. The increased formation rate of particles of 10 nm diameter calculated in the present study (being at a greater size than that of the initial particle formation occurring at about 1.5 nm and thus affected as a metric by the growth of the particles) and growth rates observed within the urban environment are associated with the increased presence of condensable species found in such environments, regardless of the geographical location within the European continent.

The NPF process is also studied according to the different aspects that define it, the frequency of the events, the formation and growth rates of the particles, and the specific role of each one of the atmospheric variables was calculated throughout Europe using the large dataset available, providing an insight of the effect of these variables on the NPF mechanisms. Finally, the range of the effect of these events on the ultrafine particle composition in each area was studied and the importance of the events in the air quality of a given area is displayed.

To my father,
my example, my inspiration
and my biggest supporter.

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1. **Dimitrios Bousiotis**, Manuel Dall'Osto, David C.S. Beddows, Francis D. Pope, and Roy M. Harrison**:** Analysis of new particle formation (NPF) events at nearby rural, urban background and urban roadside sites**,** Atmos. Chem. Phys., 19, 5679–5694, https://doi.org/10.5194/acp-19-5679-2019, 2019
2. **Dimitrios Bousiotis**, Francis D. Pope, Manuel Dall'Osto, Andreas Massling, Jacob Klenø Nøjgaard, Claus Nørdstrom, Jarrko V. Niemi, Harri Portin, Tuuka Petäjä, Noemi Perez, Andrés Alastuey, Xavier Querol, Giorgos Kouvarakis, Stergios Vratolis, Konstantinos Eleftheriadis, Alfred Wiedensohler, Kay Weinhold, Maik Merkel, Thomas Tuch and Roy M. Harrison: A Phenomenology of New Particle Formation (NPF) at Thirteen European Sites

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- **Dimitrios Bousiotis**, Manuel Dall'Osto, David C.S. Beedows, Francis D. Pope, Andreas Massling, Roy M. Harrison, (2019), "Analysing the relation of New Particle Formation (NPF) events to local environmental factors in northern Europe", presented as an oral presentation to the European Geosciences Union (EGU) General Assembly, Vienna, Austria
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- **Dimitrios Bousiotis**, Manuel Dall'Osto, David C.S. Beedows, Francis D. Pope, Roy M. Harrison (2018), "Analysis of New Particle Formation (NPF) Events at a Nearby Rural, Urban Background and Urban Roadside Sites, presented as a poster and a short oral presentation to the Aerosol Society Conference, Birmingham, UK

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1. Introduction

1.1 Air pollution

Air pollution is the presence of excessive quantities of substances, particulates and biological molecules within the Earth's atmosphere. It is a major environmental concern, especially in urban agglomerations and the compounds associated with it can be emitted by either natural or anthropogenic sources. Natural sources make the greatest contribution to air pollution. These include crustal material, emissions from volcanoes or wildfires, biogenic emissions, sea spray and organic matter from biogenic sources (Seinfeld et al., 2012). Despite their great contribution in many cases, natural emissions are rather inactive and in general evenly distributed.

Anthropogenic sources include pollutants that originate from combustion sources, agricultural and industrial activities, waste treatment, as well as other sources associated with urban activities (such as brake and tyre wear, road erosion and resuspension etc.). Due to their nature, they are unevenly distributed and are found in great concentrations within urban agglomerations or industrial areas. Despite their more local nature though, with long range transportation they may affect remote areas (Wang, 2007).

1.2 Aerosols in the atmosphere

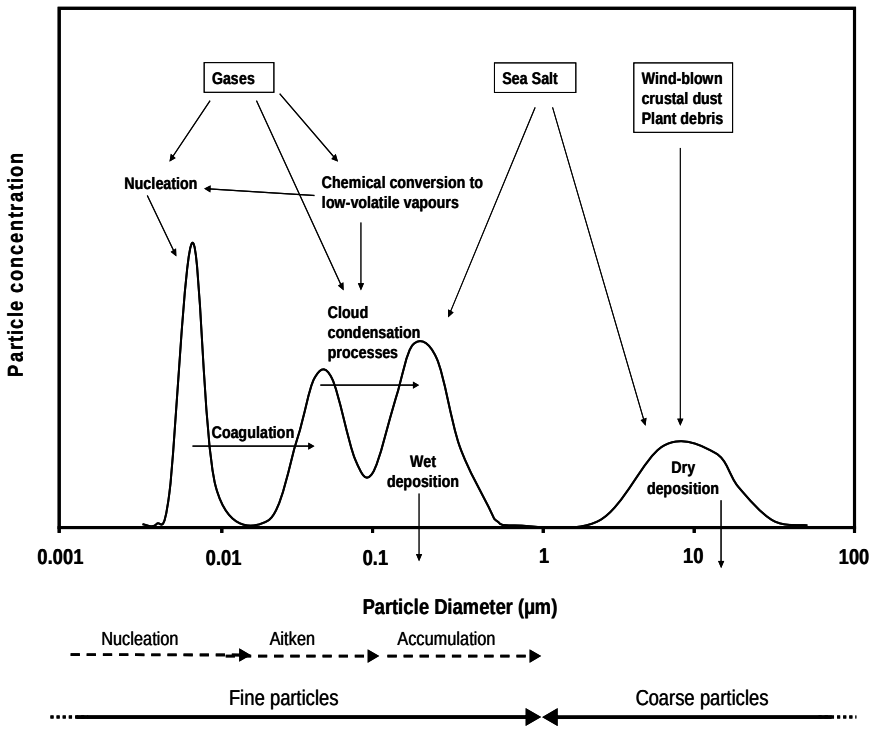
Atmospheric aerosols are particles that can either be solid or liquid and come in different sizes, which usually associates them to different sources. The sizes of particles range from few nanometres up to the size of several micrometres. These particles though are not restricted to one size and may grow or reduce in size with different mechanisms. A general size classification as proposed by Seinfeld et al. (2012) separates particles as (Figure 1):

- Particles of size smaller than 20 nm. This group of particles is characterised as **nucleation** mode and in many cases, especially in urban environments, contains the largest number of particles in the atmosphere. These particles have the shortest life span in the atmosphere and as a result they are usually not found far from their sources. These particles are usually the result of gas-to-particle formation and thus they are considered as secondary particles, though they can originate from primary emissions from combustion sources within the urban environment (Rönkkö et al., 2017). Their main removal mechanisms in the atmosphere are condensation or coagulation (Mészáros, 1999).
- Particles of size 20 to 100 nm. These particles are characterised as **Aitken** mode and they can originate by either primary sources or growth of smaller particles and are the most common size of particles outside urban areas when NPF events do not occur. The larger particles of this mode (>50 nm), as well as those of the accumulation mode discussed later, can act as cloud condensation nuclei on which water vapour can condense and form cloud droplets (Anttila et al., 2009; Komppula et al., 2005). Particles of nucleation and accumulation mode are also characterised as ultrafine particles.
- Particles of size 100 nm to 1 μm. These particles are characterised as **accumulation** mode and originate from sources associated with the coagulation between other particles, or the condensation of semi-volatile compounds on their surface (Seinfeld et al., 2012). They usually have small number concentrations, but due to their larger size they make a large contribution to particle mass and volume. Their number concentrations are present with small variations and are less reactive than smaller sized particles. This group of particles is dominated by secondary species (NO_3^-, SO_4^{2-} NH_4^+ as well as organic compounds) and emissions from combustion processes. The

removal mechanisms of these particles are less effective compared to smaller or larger sized particles and as a result they have longer residential times in the atmosphere and can be transported over longer distances.

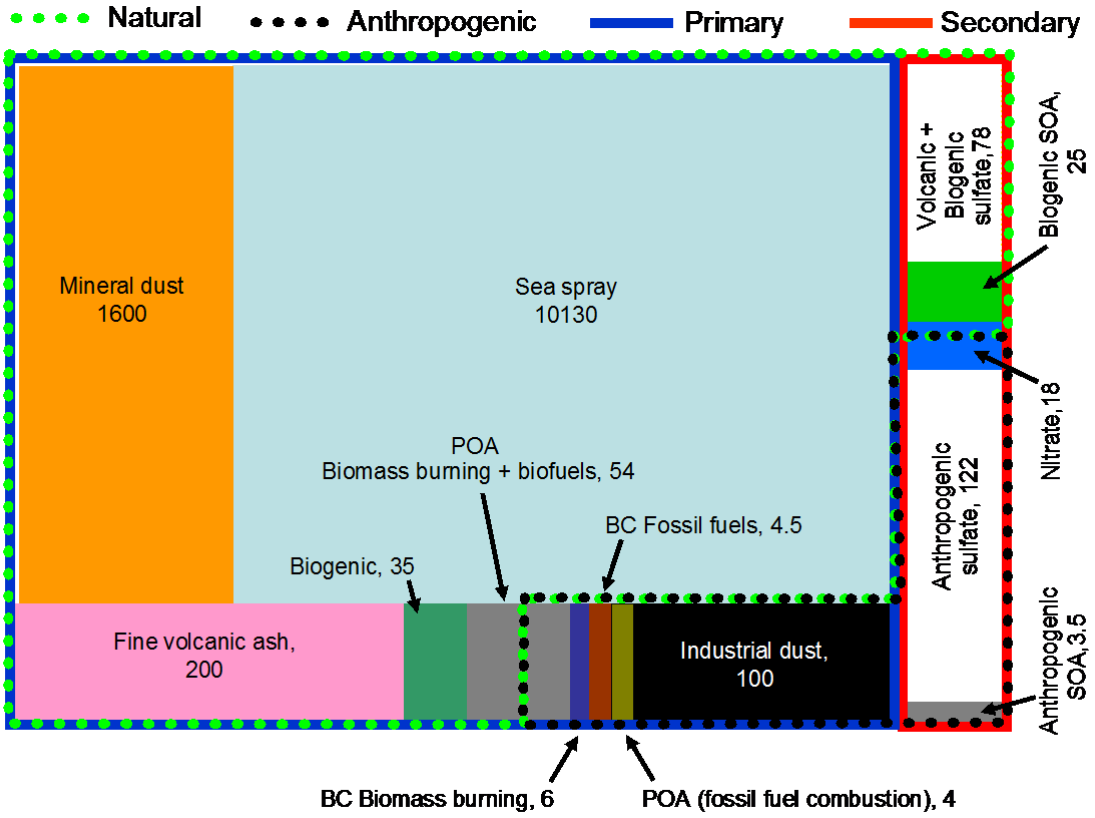
- Particles larger than 1 μm. This mode of particles is characterised as **coarse** and, in most cases, it originates from mechanical processes, such as resuspension or abrasion, or can be of marine origins (It should be noted that for the health and regulatory communities the coarse particle fraction is considered above particle diameters of 2.5 μm, while particles below that size are considered as fine). Secondary coarse particles can also occur though when gases react with pre-existing coarse particles (Querol et al., 1998). Due to their larger size and mass the dominant removal mechanism is gravitation settling. Particles of this size range are regulated as their effects on public health are well documented (WHO, 2013).

Figure 1: A simplified schematic illustration of atmospheric aerosols, including sources transformations and sinks (adapted by Cussack, 2013) from original figure by Viana, 2003)



As mentioned earlier, the particles can also be separated between primary and secondary. **Primary** particles are those that are directly emitted in the atmosphere, while **secondary** particles are those which are formed through physicochemical transformations of primary gaseous emissions. Figure 2 shows the sources of primary and secondary atmospheric particles, also separating them between natural and anthropogenic sources.

Figure 2: Sources of primary and secondary atmospheric PM expressed in teragram per year and shown as a fraction of the area of a rectangle. POA = primary organic aerosol; SOA = secondary organic aerosol; BC = black carbon. Taken from Gieré et al., (2010)



1.3 The effects of particles

1.3.1 Health effects and air quality

Though ultrafine particles are not yet regulated, many epidemiological studies have been carried out about their detrimental effect on the air quality and public health, associating them with human health deterioration (Anderson, 2009) and premature mortality (Dockery et al., 1993; Lelieveld et al., 2015; Schwartz et al., 1996). A correlation was found between the concentrations of ultrafine particles and alterations in morbidity and mortality indices because of cardiac effects in the elderly groups, as well as with an increased proportion of asthma episodes and hospital admissions. Additionally, due to their small size they are able to penetrate to the blood stream as well as the brain and central nervous system (Maher et al., 2016; Politis et al., 2008). Ultrafine particles were found to enter the human respiratory system (Vu et al., 2016) and cause greater inflammation to the lungs compared to fine particles, as the toxicity per unit mass increases as size decreases (Davidson et al., 2005; Kim et al., 2015; MacNee and Donaldson, 2003; Penttinen et al., 2001; Valavanidis et al., 2008), while connections between fine particles and lung cancer have also been found (Pope III et al., 2002). Apart from their effect on the respiratory system they were also found to have a detrimental effect on the nervous system (Kilian et al., 2018; Shiraiwa et al., 2017; Zhang et al., 2017), as well as to affect the heart function and contribute to cardiovascular disease (Kaiser, 2005; Oberdurster, 2000) even on younger adults (Samoli et al., 2016). Finally, UFP can deteriorate the visibility and breathability within the urban environment directly or by participating in haze formation (Buccolieri et al., 2010; Guo et al., 2014).

1.3.2 Effects on the atmosphere

Ultrafine particles while making a small contribution to the total particle volume and mass, have a dominant contribution in particle number concentrations (Harrison et al., 2001; Németh et al., 2018). New Particle Formation (NPF) events specifically, are one of the most important sources of nucleation mode particles in the atmosphere and can increase their number up to 10 times on the day of events (Cheung et al., 2013); the number of very small particles (of diameter smaller than 6 nm) in some areas is negligible on non-event days (Salma et al., 2011). While the relative importance of nucleation on particle composition decreases as the anthropogenic influence increases (Salma et al., 2014), NPF events are still a significant source of ultrafine particles even in the urban environment, comparable to that of direct emissions (Rahman et al., 2017; Zhou et al., 2019). As newly formed particles grow they can affect the optical (Quaas et al., 2009) and radiative properties (Bryant et al., 2006; Myhre, 2009) of the atmosphere. Aerosol particles are capable of absorbing and scattering incident radiation (direct radiative forcing). By affecting the optical properties of the atmosphere, they may also affect vegetation by altering the photosynthesis process (Gu et al., 2002), as well as have an effect on the general ecosystems by altering incoming solar radiation and evaporation (Wang et al., 2008). Due to the complexity of these mechanisms, the exact extend of their effect on climate and environment, on the direct radiative forcing is yet not well defined. Additionally, a significant increase in the number of Cloud Condensation Nuclei (CCN) has been associated with NPF events (Merikanto et al., 2009; Dameto de España et al., 2017; Kalkavouras et al., 2017), though the extent of this effect upon the indirect forcing (Wang et al., 2009) and climate change (Arneth et al., 2009) are still unknown. Nucleation has a very strong effect on the Earth's climate total forcing, maybe greater than that of greenhouse gases (Makkonen et al., 2012), and future improvements in air quality can further affect global warming by reducing the negative radiative forcing (Brasseur et al., 2005).

1.4 Particle formation and New Particle Formation events

"The tube being again exhausted, the mixed air and vapour were allowed to enter it in the dark. The slightly convergent beam of the electric light was then sent through the tube from end to end. For a moment the tube was optically empty, nothing whatever was seen within it; but before a second elapsed a shower of liquid spherules was precipitated on the beam, thus generating a cloud within the tube. This cloud became denser as the light continued to act, showing at some places a vivid iridescence."

John Tyndal, On a New Series of Chemical Reactions produced by Light, 1868,
Proceedings of the Royal Society of London

Particle formation was observed hundreds of years ago, with the magicians of that era using this mechanism as means to impress their audience by filling, what was considered as an empty tube, with a blueish nebula of particles with only the application of light. The mechanism of particle formation was scientifically first observed by Tyndall (1868), experimenting with the effect of light applied to tubes containing organic vapours. Aitken (1897) was the first to report particle formation in the atmosphere, though little progress was made for many years after his findings. Many theoretical approaches have been attempted since then to explain the mechanisms of particle formation, initially focusing on the production of submicrometer particles from the oxidation of terpenes (Went, 1960). Some years later the first theory of binary nucleation of sulphuric acid droplets being formed from the vapour system H_2O – H_2SO_4, was proposed by Junge (1963) and Kiang et al. (1973), shifting the interest from terpenes to sulphate aerosol (Jaecker-Voirol et al., 1989; Mirabel et al., 1974). Middleton et al. (1978) considered the limitations of nucleation in the real atmosphere due to the possible impinging of sulphuric acid vapours on ("foreign" as mentioned in the text) pre-existing aerosols, which may consume the sulphuric acid by condensation, providing one of the first references for the condensation sink, a major factor in the occurrence of NPF events. Due to limitations on measuring instruments though, it took more than 20 years for the binary theory to be validated, as methods to measure H_2SO_4

developed. Both laboratory experiments (Berndt et al., 2005; Viisanen et al., 1997) as well as Weber et al. (1997), using data from a continental site at Idaho Hill in the U.S.A., found a correlation between particle formation rates and sulphuric acid concentrations, though in much lower concentrations of sulphuric acid than expected. The discrepancy found was that for the given concentrations of sulphuric acid in the atmosphere, the formation and growth rates of particles in the atmosphere were too high compared to what was expected by the binary theory, a find that was already speculated some years before (Wyslouzil et al., 1991), though it might be a more relevant mechanism at higher altitudes in the troposphere (Laaksonen et al., 2008a).

In the light of the discrepancy found, the possibility of more compounds participating and enhancing the nucleation procedure was discussed for some years. The ternary system of H_2O-H_2SO_4-NH_3 was considered, though Stelson et al. (1984) used an ammonium sulphate compound instead of NH_3. The model of the ternary nucleation, with the inclusion of NH_3 as a stabilising factor was first proposed by Korhonen et al. (1999) and further improved by Napari et al. (2002), providing results that were closer to those measured in the atmosphere. This was confirmed by Gaydos et al. (2005) who showed that ternary nucleation could explain the occurrence of new particle formation in north-eastern U.S.A., as well as in the Cosmics Leaving Outdoor Droplets (CLOUD) experiment (Dunne et al. 2016). Both the binary and ternary theories appear to describe reasonably well the nucleation rates in the free troposphere (Spracklen et al., 2008), but they cannot reproduce the expected linear correlation between the nucleation rate with the sulphuric acid concentration in the atmospheric boundary layer (Sihto et al., 2009). Binary and ternary theories suggests that the critical cluster contains no more than two sulphuric acid molecules. However, the atmospheric observations showed this relationship to be equal or greater to the power of two. As of this, the kinetic (barrierless) nucleation theory (McMurry et al., 1979) and the

cluster activation mechanism (Kulmala et al., 2004b, 2006) were considered and empirical coefficients were calculated, attempting to reproduce the nucleation rates found in the atmospheric boundary layer.

As the concentrations of sulphuric acid in the atmosphere were not enough to explain the growth rate of the newly formed particles, additional compounds that would participate in the growth procedure were considered. Volatile Organic Compounds (VOCs) both biogenic (BVOCs) and anthropogenic (AVOCs) can be transformed when oxidised to Highly Oxygenated Molecules (HOMs), which due to their low volatility can either form clusters of purely BVOCs (Bianchi et al., 2017; Kirkby et al., 2016; Rose et al., 2018) or participate as condensable compounds during the growth procedure (Donahue et al., 2005; Ehn et al., 2012; Jokinen et al., 2015; Tröstl et al., 2016). Additionally, the possibility that AVOCs or their oxidation products participate in the formation and growth procedure is considered, and laboratory studies are now trying to identify those that have the properties needed to do so (Molteni et al., 2018; Wang et al., 2019).

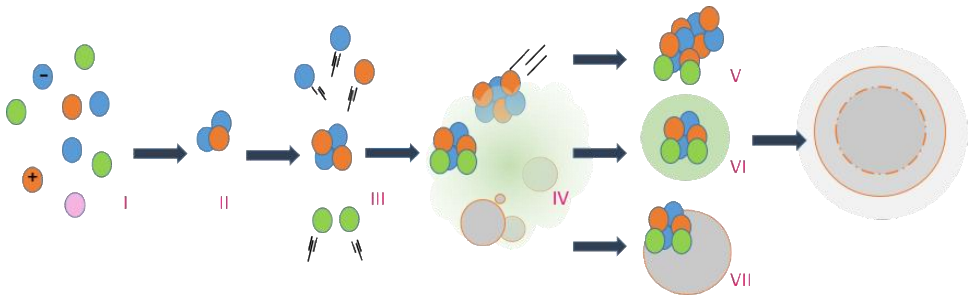
"**Highly Oxygenated Organic Molecules (HOMs)** are formed in the atmosphere via autoxidation involving peroxy radicals arising from Volatile Organic Compounds (VOCs)" (Bianchi et al., 2019), and have lower volatilities than their predecessors. Due to that they have higher tendency to condense on pre-existing particles. The VOCs that can participate in this procedure may be from either biogenic or anthropogenic sources. The increased abundance of anthropogenic VOCs within the urban environment from the activities that take place there (traffic, heating, cooking etc.) provide a greater abundance of HOMs thus increasing the rate at which particles are formed and grow.

Apart from the aforementioned theories and components that participate in the particle formation process, additional theories, pathways and components have been proposed. Ion-induced nucleation has been proposed as a possible mechanism of particle formation, though its contribution to the lower troposphere is probably limited. This includes the formation of ions from the impact of ionising radiation, such as cosmic or gamma rays, on an air molecule. Ions may also be associated with other sources like thunderstorms or vehicle engines. These ions may transfer their charge to trace gases, clusters or pre-existing particles

forming charged compounds, which later continue to grow via condensation. In such a case, the charge acts as the stabilising factor in the initial clusters or small particles. With this, nucleation of pure biogenic particles can be achieved in the absence of H_2SO_4 (Kirkby et al., 2016). Additionally, other components were also proposed to participate in the nucleation procedure, such as iodine from biogenic emissions (O'Dowd et al., 2002), methasulphonic acid (MSA) or molecular iodine (Dall´Osto et al., 2018), as well as nanoparticles directly emitted from combustion engines, which can grow in later stages, resembling new particle formation events (Olin et al., 2019; Rönkkö et al., 2017).

So, what is the nucleation in the atmosphere in simple words? Nucleation is the formation of particles from gaseous precursors (figure 3). Vapours of low volatility are combined into clusters initially containing a few molecules of sulphuric acid (in most cases). These clusters further combine with other similar compounds in the atmosphere in a repeating process to form a cluster of size about 1.5 nm, at which point a phase transition occurs from gas to particle. During these initial steps of nucleation, the clusters or small particles are very unstable and other compounds (such as water vapour, ammonia etc.) or a charge may act as a stabilising factor, reducing the possibility of their loss via evaporation. For these particles to survive and continue growing more condensable compounds are needed. These compounds should also be of low volatility, and low volatility organic compounds are the ones considered to have the biggest contribution in this stage. As the size grows, compounds of higher volatility may also condense onto the new particles.

Figure 3: The steps of nucleation. Neutral and charged clusters (I-II), condense and evaporate constantly (III) until they overcome the losses (IV) and nucleate into particles (V-VI). These clusters can either continue growing (V-VI) or condense onto a larger particle. Taken from Buenrostro Mazon, (2019).



New Particle Formation events are the result of very complicated interactions between several chemical compounds within air masses with unique mixtures of conditions. The constant presence of clusters of size smaller than 1.5 nm has been confirmed in a number of studies (Riipinen et al., 2007; Schobesberger et al., 2015). The numerous combinations of conditions and exact compounds that participate in the process though, that either lead to or limit the activation of these clusters and their growth to sizes that can be atmospherically relevant are yet to be explained. In the studies of NPF, an event is defined as the appearance of a mode of particles in the lower detection limit, which in most cases is between 3 – 15 nm, and is growing to greater sizes, two prerequisites that should be met for an event to be considered. The ability to measure particles as they are formed was impossible until a few years ago (and still measurements are subject to high uncertainties), which practically mean that the studies did not consider nucleation of new particles rather than the aforementioned mode of particles and its consequent growth. Nucleation and growth of particles in the atmosphere is a competition between the survival of new particles by growing in size through the condensation of other compounds and the mechanisms that lead to their loss,

such as evaporation or coagulation onto larger particles. While research into atmospheric nucleation has come a long way since John Tyndal's observations, there is still a long road ahead until we can fully understand the effect of all the variables that participate in the process.

1.5 The conditions favouring NPF

The occurrence of NPF events is affected by a complex combination of different atmospheric variables. The study of the conditions that affect NPF events, though not being new, has developed significantly over the last few decades as measuring instruments have developed. Many meteorological variables and chemical compounds appear to influence the occurrence of NPF events. NPF as studied, is the formation and growth of particles from secondary formation. In atmospheric conditions, the variables mentioned later were studied in most cases for their effect in the occurrence (the frequency) of the events as a whole and not for their effect on the specific mechanisms (formation and growth of the particles) which are governed by different processes. Laboratory studies are the ones that are more focused on explaining the exact effect on each one of the mechanisms that are considered when studying the events and these usually provide results associated to them, though in a very controlled environment where only specific variables are modified.

1.5.1 Meteorological conditions

The intensity of the solar radiation is one of the most important factors in the occurrence of NPF events. The role of the solar radiation and its contribution to many photooxidation processes that lead to particle formation has been underlined in many studies (Cheung et al., 2011; Pikridas et al., 2015). Photooxidation of SO_2 is considered as one of the major

mechanics of particle formation (Cheung et al., 2013) as it leads to the formation of H_2SO_4, which is considered an important component in new particle formation (Weber et al., 1995). Additionally, solar radiation may cause vertical mixing which can also promote particle formation by helping the thermodynamics of the nucleation process as well as the subsequent condensation, or by introducing active precursor gases to different heights of the boundary layer (Mäkelä et al., 2000). Higher solar radiation has been reported on NPF event days by many studies (Jeong et al., 2010; Minguillón et al., 2015; Rimnácová et al., 2011; Salma et al., 2011; Wang et al., 2014; Wonaschütz et al., 2015), solar radiation is found to correlate with NPF events (Kürten et al., 2016) and in general, NPF events are more frequent on sunny days (Stanier et al., 2004). The importance of solar radiation in the strength of NPF events (as defined by the formation and growth rate of the particles) has been underlined (Größ et al., 2018), having a positive effect on the formation and growth rate of the particles (Cheung et al., 2011; Kopanakis et al., 2018). While solar radiation is considered as a prerequisite for nucleation (Shi et al., 2001), reports of night-time events (Kalivitis et al., 2018; Man et al., 2015; Ortega et al., 2012; Salimi et al., 2017) may indicate alternative pathways to particle formation.

Temperature is another variable that plays a significant role in NPF. Laboratory studies have shown that lower temperatures increase the nucleation rates of H_2SO_4 (Kirkby et al., 2011; Yu et al., 2017), as well as affect particle growth, either by increasing the yield of HOMs from autooxidation (higher temperatures) or by reducing the volatility of the compounds that participate in the procedure (lower temperatures). A laboratory study reports that the growth rate is higher at lower temperatures at a given reaction rate (Stolzenburg et al., 2018). In atmospheric conditions though, increased temperatures are associated with a greater abundance of BVOCs, which are considered to participate in the growth procedure. Due to the variable effect of temperature on particle formation there is discrepancy on its

effect, with studies reporting either higher (Sogacheva et al., 2007; Wang et al., 2014; Woo et al., 2001) or lower temperatures on NPF event days (Jeong et al., 2010), demonstrating the complexity of the conditions occurring on NPF events.

The presence of water vapour is important in the nucleation process, acting as a stabilisation agent of the critical nucleus of H_2SO_4 (Weller et al., 2015). A laboratory study has reported that relative humidity did not affect the presence and abundance of HOMs and did not have a constant effect on Secondary Organic Aerosols (SOA) formation up to 60%, which play an important role in the NPF process. For higher values of relative humidity though, a reduction was found in particle number concentrations by a factor of 2-3 (Li et al., 2019). Water in the atmosphere may suppress the formation of low volatility compounds by the ozonolysis of biogenic hydrocarbon compounds and thus suppress NPF events (Bonn et al., 2003). Another laboratory study reported that the power dependency of the number of particles formed on the concentrations of H_2SO_4 reduces with increasing relative humidity values (Zollner et al., 2012). The majority of the field studies have reported lower relative humidity during NPF events (Brines et al., 2015; Jeong et al., 2010; Kürten et al., 2016; Park et al., 2015; Rimnácová et al., 2011; Wang et al., 2014; Wonaschütz et al., 2015) and the formation rate J was found to be reversely correlated to relative humidity (Kopanakis et al., 2018), while to the author's knowledge only one study reported higher values of relative humidity during NPF events in California, U.S.A. (Setyan et al., 2014).

The origin of the incoming air masses is crucial for the occurrence of NPF events. Depending on the path the incoming air masses followed, they carry different properties that may affect all characteristics of new particle formation (Komppula et al., 2006), with Shen et al. (2018) reporting that air masses from more polluted areas resulted on events with higher formation and growth rates. Harrison et al. (2000) reported less events in at Weybourne, UK with incoming air masses from the arctic or mainland Europe compared with other origins, while

Nilsson et al. (2001) and Sogacheva et al., (2005) reported that new particle formation occurred preferably with polar and arctic maritime air masses in Hyytiälä, Finland. Rahman et al. (2017) on the other hand, found that NPF events were linked to the transport of pollutants from the airport, the oil refinery and the port in Brisbane, Australia.

The possible effect of wind speed on NPF events seems to be more complicated as reports provide contradictory results. Reports of a positive effect (Brines et al., 2015; Németh and Salma, 2014; Pushpawela et al., 2019; Wang et al., 2014), negative effect (Kopanakis et al., 2018; Rimnácová et al., 2011), or no effect (Siakavaras et al., 2016) of higher wind speeds on NPF events provide with a mixed picture of the effect of this variable, which makes it possible that either the wind speed does not affect NPF events or the possible effect this variable has depends heavily on local conditions.

In literature, other meteorological factors were also reported to possibly affect new particle formation in the atmosphere. High pressure is usually associated with good weather conditions and stability in the atmosphere, and Shen et al. (2018) reported more NPF events under such conditions. On the other hand turbulence in the atmosphere, which is usually associated with low pressure systems, may cause supersaturation of precursor gases and promote NPF (Wehner et al., 2010), and the mixing of two air parcels with different properties may favour particle formation (Charron et al., 2003; Nilsson et al., 1998).

Finally, some studies considered other variables, with Park et al. (2015) reporting lower cloud coverage on NPF events, while Charron et al., (2007), Creamean et al., (2011) and Kavouras et al., (1998) reported particle formation starting right after the end of rain, which might be due to the lowering of the pre-existing particle surface resulting from it. As the studies for these variables are few, no generalised conclusions can be made for these conditions.

1.5.2 Atmospheric composition

Apart from the effect of synoptic conditions, the composition of the atmosphere also plays a very important role in the NPF process. Many chemical compounds are considered to participate in both the formation of new particles and their growth by condensation. Sulphuric acid due to its low volatility and relative abundance in the atmosphere, as mentioned earlier, is one of the main constituents of the initial clusters that lead to particle formation, and it is suggested that the freshly formed particles contain 1 to 2 H_2SO_4 molecules (Sipila et al., 2010). Many studies have underlined the importance of H_2SO_4 in the occurrence of NPF events (Kalkavouras et al., 2017; Kürten et al., 2016; Siakavaras et al., 2016; Stanier et al., 2004; Wang et al., 2014). H_2SO_4 is found to be a key component in the formation of new particles (Berndt et al., 2005; Cai et al., 2017; Dai et al., 2017; Wang et al., 2011) in both polluted and clean environments (Boy et al., 2005), as well as in the initial growth of the newly formed particles (Birmili et al., 2003; Boy et al., 2005; Fiedler et al., 2005; Iida et al., 2008; Paasonen et al., 2009). Its role in the growth of particles is variable with size (Kulmala et al., 2005; Keskinen et al. 2013), becomes smaller as size increases and negligible above 10 nm (Meng et al., 2015; Xiao et al., 2015). H_2SO_4 is formed in the atmosphere from the oxidation of sulphuric compounds, mainly SO_2, with the hydroxyl radical; the latter is directly associated with the presence of solar radiation. As a result, the presence of SO_2 in the atmosphere has an effect in the occurrence of NPF events (Laaksonen et al., 2008b). Reports of both higher (Betha et al., 2013; Chandra et al., 2016; Wonaschütz et al., 2015; Woo et al., 2001) and lower concentrations of SO_2 (Alam et al., 2003) were presented in studies of NPF events. SO_2 seems to have a variable role depending on the local conditions (Dall'Osto et al., 2013) and it is considered to play a different role, either assisting or inhibiting NPF events, depending on its concentrations (Dall'Osto et al., 2018).

According to the ternary theory, ammonia and other bases are also participating in the nucleation process as stabilising factors. While the role of ammonia is well established in enhancing the nucleation process (Dunne et al., 2016), no significant variations were found in its concentrations on NPF events when studied (Riipinen et al., 2007). Xiao et al., 2015 found a strong correlation of the formation rate of new particles with ammonia concentrations in Beijing. Contrary to this, Kürten et al. (2016) found lower concentrations of ammonia on events in Vielbrunn, Germany, without though excluding its participation in the particle formation process, thus explaining the lower concentrations found. Ammonia in the atmosphere is in most cases in sufficient concentrations for it not be, according to the ternary theory (Korhonen et al., 1999), a limiting factor in the nucleation process. Li et al. (2018) in a laboratory study, found that ammonia has the potential of also enhancing the growth rates of particles in the presence of aromatic hydrocarbons (toluene and xylenes) with the formation of ammonium nitrate. Apart from ammonia, amines are also found to enhance the nucleation process. Methylamine, dimethylamine and trimethylamine, though in limited concentrations in the atmosphere, are also found to enhance the formation of new particles more compared to ammonia (Kürten et al., 2014; Zollner et al., 2012; Almeida et al., 2013), and the simultaneous presence of these compounds with ammonia had a higher potency in particle formation synergistically (Glasoe et al., 2015).

For many years sulphuric acid was considered as the main compound participating in both the formation and growth of particles. Its concentrations though in the atmosphere were not enough to fully explain the growth of the particles. Organic compounds with low volatility were later suggested to participate in the condensational growth of particles (O'Dowd et al., 2002; Stanier et al., 2004; Zhang et al., 2004), which was later confirmed by a number of studies (Dall'Osto et al., 2018; Mentel et al., 2009; Wang et al., 2015; Tröstl et al., 2016). Until recently, NPF research was mainly focused on background sites, thus the

oxidation products of BVOCs, especially of isoprene and α-pinene which are the ones in greatest abundance in the atmosphere, were considered to contribute in the nucleation process by stabilising the initial clusters and assist in the growth of particles by condensation (Mohr et al., 2017; Qi et al., 2018; Riccobono et al., 2014); reduced concentrations of these had a restrictive effect on NPF events (Laaksonen et al., 2008b; Yli-Juuti et al., 2011), though laboratory experiments showed that isoprene specifically has a limiting effect as it appears to limit the formation of SOA, though being a source of SOA itself (Kiendler-Scharr et al., 2009; Lee et al., 2016; McFiggans et al., 2019). Bianchi et al. (2016) and Rose et al. (2018) suggested that in clean and remote areas where sulphuric acid concentrations are low, organic compounds and HOMs are important in both the formation and growth of the particles, though the presence of H_2SO_4 is still needed (Riccobono et al., 2014; Schoebesberger et al., 2013). This was further confirmed by Wiedensohler et al. (2019) who reported very few events in Siberia, where there is abundance of monoterpenes but very low concentrations of H_2SO_4. On recent years, studies for the role of VOCs that originate from urban activities in the nucleation process were undertaken. Molteni et al. (2018) found that anthropogenic VOCs when oxidised can produce HOMs similar to those from BVOCs, which may potentially participate in particle formation, though not all of them are capable to do so (Wang et al., 2019). Similarly Zhao et al. (2014) suggested that Criegee intermediates, resulting from the oxidation of alkenes (many of which are of anthropogenic origin) may also participate in the formation and growth of particles, providing more possible pathways for the formation of HOMs which may participate in the growth of particles within urban areas.

The condensation sink (CS) is a metric of pre-existing aerosol surface area in the atmosphere. As the number of pre-existing particles in the atmosphere increases, the possibility of newly formed particles to condense on them and fail to further grow as individual particles

becomes higher. Lower CS was found to be one of the deciding factors in the occurrence of NPF events in many studies (Alam et al., 2003; Cai and Jiang, 2017; Park et al., 2015; Salma et al., 2011; Wang et al., 2014), as well as restricting the growth of new particles on events from reaching greater sizes in more polluted areas (Dall'Osto et al., 2012; Jayaratne et al., 2017). Additionally, Cheung et al. (2011) have reported that while conditions were favourable for consecutive NPF events to occur, the increased CS due to the presence of Aitken mode particles from the event of the previous day restricted events on subsequent days. Especially within the urban environment though, the CS may be associated with greater abundance of condensable compounds that may speed up the growth procedure and increase the chances of new particles to survive and further grow (Kulmala et al., 2004a, 2017).

Apart from the increased CS in the urban environment, higher concentrations of NO_x, associated with anthropogenic activities can also affect NPF events. NO_x suppress NPF events as they participate in the formation of organonitrates which react with HOMs, thus indirectly suppressing NPF (Lehtipalo et al., 2018). NO_2 specifically reacts faster with OH to form HNO_3 than SO_2 to form H_2SO_4, reducing its formation rate (Rodhe et al., 1981). As a result, NO_x was found to have lower concentrations on NPF events (Brines et al., 2015; Wang et al., 2014). Nitrate though, while associated to urban and long-range pollution, in the form of ammonium nitrate, is considered to participate in later stages of particle growth (Bzdek et al., 2012; Ristovski et al., 2010).

Another important compound that appears to affect the nucleation process is ozone. Ozone participates in the oxidation of a range of compounds associated with the nucleation procedure, and studies have shown its enhanced presence on NPF events (Cheung et al., 2013; Wonaschütz et al., 2015; Woo et al., 2001). While its association with the nucleation process is a given, it should not be overlooked that its presence is associated with many other

conditions that are favourable for NPF events (increased solar radiation, lower concentrations of NO_2, higher OH etc.) and thus the range of its importance in the process is not fully elucidated yet. Additionally, ozone has been considered to participate in the oxidation of organic compounds during night-time particle formation (Kammer et al., 2018; Lauros et al., 2011).

Finally, a handful of reports have studied the possible effect of larger particulate matter (PM) on NPF events. Though a higher PM concentration is usually associated with a higher condensation sink, thus being a restrictive factor for the events (Stanier et al., 2004), Wonaschütz et al., (2015) found no relationship between the concentration of $PM_{2.5}$ and NPF events, while Dupart et al., (2012) and Nie et al., (2014) in their studies in China, found that NPF events were observed on dust-storm events, linking them to increased production of OH by dust-induced photolytic reactions.

1.6 Temporal variation of NPF events

Long-term data studies provide insight in the temporal variation of NPF events. Interestingly, there is great variability in the seasonal variation of NPF events (Dall'Osto et al., 2018; Kulmala et al., 2004a) even for sites of relatively close proximity (Wang et al., 2017). While synoptic conditions are more favourable for NPF events to take place in the local summer (high insolation and temperatures, greater abundance of BVOCs, low relative humidity etc.), local conditions may alter the seasonality of events. For example, Park et al. (2015) reported higher frequency of NPF in spring in Korea, due to it being a drier season compared to the local summer, (Kalivitis et al., 2012; Vratolis et al., 2019; Ždímal et al., 2011) reported almost a lack of NPF events in Greece during summer due to lower concentrations of atmospheric ions and Zhu et al. (2017) reported greater frequency of NPF events in winter than spring in

Beijing, though without including summer data. The reports for the seasonality of the growth rate of particles though present more uniform results, showing a summer peak in most cases (Kulmala et al., 2004a; Nieminen et al., 2018; Riipinen et al., 2007; Yli-Juuti et al., 2011). As for the weekly variation of NPF events, to the author's knowledge there is only one study reporting differences in weekday/weekend variation, with Brines et al. (2015) reporting that the nucleation cluster was found more frequently on weekends in an urban background site in Barcelona. Finally, Kalivitis et al., 2018 and Saha et al., 2018 report a decreasing frequency of NPF events using datasets of more than a decade. This is probably associated with the regulation of pollutants that led to the reduction of SO_2 and the number of pre-existing aerosols associated with pollution, as well as other pollutants that may be associated with NPF processes.

1.7 Variation of NPF events with the type of environment

The occurrence and development of NPF events is the result of a complex combination of atmospheric conditions. The variation found in the conditions, frequency and characteristics of NPF events is mainly the result of the different mixture of conditions found in different areas. Differences in the frequency, seasonality and characteristics of NPF events which are anticipated in larger spatial ranges (Dall'Osto et al., 2018; Reche et al., 2011), can also be found even at sites of very close proximity (Pikridas et al., 2015), as even when synoptic conditions are favourable, local conditions may limit the occurrence of NPF events and vice versa (Berland et al., 2017). Additionally, there can be great variability in the characteristics of NPF that take place simultaneously on nearby sites (regional events), due to the differences in geographical, environmental and to a lesser extent meteorological conditions (Dai et al., 2017; Hussein et al., 2009). The studies of events at sites in close proximity and

regional NPF events provided results that further confirm the effect of the local conditions. Most studies agree that the formation and growth rates are higher within more polluted environments on sites of relatively close proximity in many areas (Chu et al., 2018; Cusack et al., 2013; Kulmala et al., 2005; Lyu et al., 2018; Nieminen et al., 2018; Pierce et al., 2014; Salma et al., 2016; Wang et al., 2017; Wehner et al., 2007), while the frequency of NPF events had different variations (Ma et al., 2015; Németh et al., 2014; Peng et al., 2017), probably as a result of different sets of sites. Similarly, the contribution of NPF events to the ultrafine particle concentrations is different between nearby sites of different land use, being higher for the rural sites compared to urban sites (Ma et al., 2015; Salma et al., 2014, 2017). This further extends to different land uses, such as remote arctic environments, forests, mountainous or coastal sites, though the findings between such sites vary depending on the local conditions. Additionally, direct comparison between sites studied in different studies should be done with great care, as there can be differences between the methods used and the specific conditions found on each site.

1.8 Research gaps and objectives

As pointed out earlier, the studies of the conditions and the effect of each of the variables that are associated with the nucleation and growth of the new particles has not provided with clear answers on the role of each variable. Despite the fact that laboratory studies have provided insight into the effect of those variables in controlled environments, these results do not appear to be easily applied to atmospheric conditions. Additionally, the results from studies in different areas seem to provide variable results and variable effects of the conditions and chemical compounds associated with NPF events. The present study attempts to:

- Determine the atmospheric conditions that are favourable for NPF events in a range of areas and land uses with different characteristics throughout Europe.
- Interpret the effect of the origin of incoming air masses on NPF events in a given area.
- Highlight the variability of the characteristics of NPF events between different areas.
- Identify the positive or negative effect of meteorological and atmospheric variables in the occurrence and characteristics of NPF events, as well as their variability among the different areas studied.
- Reflect the effect of local conditions and type of environment on NPF events.
- Quantify the effect of NPF events on the ultrafine particle composition in a given area.
- Quantify the extent of the effect of NPF events depending on the type of environment.

1.9 Structure of the thesis

Following the introduction, the methodology section provides a brief description of the sites studied, as well as the available data for each one of them. Additionally, in the methodology section there is an explanation of the data treatment and the different metrics used in the analysis of NPF events.

The results section consists of three parts:

1. Analysis of the conditions and development of local and regional NPF events in three sites of different land use in southern U.K. Apart from the study of the meteorological and atmospheric conditions, the effect of the origin of incoming air masses in the area on NPF events is also studied, as well as the effect of the events in the particle

composition at each site. As the sites of the study are in close proximity, the effect of the local environment in the development of the events is also considered **(Paper 1)**.

2. Analysis of the conditions, development and variability of NPF events in 13 sites throughout Europe. This section will focus more on the spatial variability of NPF events, across sites with different characteristics, ranging from almost pristine rural backgrounds at the northern and southern parts of Europe to busy roadsides and backgrounds in northern, central and southern Europe. While the synoptic conditions of NPF events are studied, the effect of local conditions and their detrimental role in NPF events' occurrence is also underlined, as well as the effect of NPF events upon the local environment. **(Paper 2)**.
3. Elucidation of the specific role of meteorological and atmospheric composition in the characteristics of NPF events. Using an extended dataset of more than 85 years with 1950 NPF events, the importance and effect of 11 atmospheric variables on the occurrence of NPF events, as well as the formation and growth rate is studied, using a custom linear regression **(Chapter 3.3)**.

Chapter 4 is a conclusions part which summarises the findings from the three sections of the results chapter, followed by chapter 5 with suggestions and ideas for future work. The thesis is completed with the list of the literature references used.

2. Methodology

2.1 Types of environments

In the present study, three types of environment are considered, Rural Background (RU), Urban Background (UB) and Roadside or kerbside sites within an urban area (RO). Strict definitions of these types of environments do not exist. According to the Department of Environment Food and Rural Affairs (DEFRA) in the U.K. (DEFRA, 2020) a rural site is "sited more than 20 km away from agglomerations and more than 5 km away from built-up areas, industrial installations or motorways or major roads, so that the air sampled is representative of air quality in a surrounding area of at least 1000 km^2". According to the same source, an urban area is defined as "a continuously built-up urban area, meaning complete (or at least highly predominant) building-up of the street front side by buildings with at least two floors or large detached buildings with at least two floors". A background station is defined as "a location such that its pollution level is not influenced significantly by any single source or street, but rather by the integrated contribution from all sources upwind of the station i.e. by all traffic, combustion sources etc. upwind of the station in a city, or by all upwind source areas (cities, industrial areas) in a rural area)". Finally, roadside sites or Traffic stations as referred in the same source are "located such that their pollution level is determined predominantly by the emissions from nearby traffic (roads, motorways, highways)".

2.2 Monitoring sites, measurements and data availability

In the present study, particle number distribution data from 16 sites in six European countries are analysed in the size range 3 nm < D_p < 1000 nm (Fig. 1 in Paper 3). For ease of understanding the sites were named after an abbreviation of the name of the country

followed by the type of the site. In **Papers 1 and 2**, average meteorological conditions and atmospheric composition for each site are provided:

UKRU: Rural background site located at Harwell in Oxfordshire, 80 km west of the greater London area (51° 34' 15" N; 1° 19' 31" W). For this site, a Scanning Mobility Particle Sizer (SMPS) (EC, TSI Model 3080 with a Condensation Particle Counter (CPC), TSI Model 3775) data covering the range 16.6 – 604 nm for 7 years (2009 – 2015) was available in 15-minute averages, providing 46930 hours of data (76.5% coverage). A detailed description for this site is given by Charron et al. (2013). Chemical composition data was also available from this site for the same period (NO, NO_2, NO_x, O_3, SO_2, minerals, organic, elemental and black carbon, particulate nitrate, sulphate and ammonium aerosol, VOCs, gaseous ammonia, $PM_{2.5}$ and PM_{10}); meteorological data was available from the same site, while global solar irradiance (in kJ m^{-2}) data was available from the nearby Little Rissington station measured by the Met Office.

UKUB: Urban background site located at North Kensington in London, UK, 4 km west of the city centre of London (51° 31' 15" N; 0° 12' 48" W). For this site, SMPS (EC, TSI Model 3080 with CPC, TSI Model 3775) data covering the range 16.6 – 604 nm for 7 years (2009 – 2015) was available in 15-minute averages, providing 51059 hours of data (83.3% coverage). A detailed description for this site is given by Bigi et al., (2012). Chemical composition data was also available from this site for the same period (NO, NO_2, NO_x, O_3, SO_2, minerals, organic, elemental and black carbon, particulate nitrate, sulphate and ammonium aerosol, $PM_{2.5}$ and PM_{10}); meteorological and global solar irradiance (in kJ m^{-2}, measured by the Met office) data was available from Heathrow airport 15 km to the west.

UKRO: Kerbside site located at London's city centre at Marylebone Road (51° 31' 21" N; 0° 9' 16" W). For this site, SMPS (EC, TSI Model 3080 with CPC, TSI Model 3775) data covering the range 16.6 – 604 nm for 7 years (2009 – 2015) was available in 15-minute averages,

providing 45562 hours of data (74.3% coverage). A detailed description for this site is given by Charron and Harrison, (2003). Chemical composition data was also available from this site for the same period (NO, NO_2, NO_x, O_3, SO_2, organic, elemental and black carbon, particulate nitrate, sulphate and ammonium aerosol, $PM_{2.5}$ and PM_{10}); meteorological and global solar irradiance data (in kJ m^{-2}, measured by the Met Office) was available from Heathrow airport about 15 km to the west.

At all three UK sites, along with the SMPS a free-standing CPC (TSI Model 3022A) also operated for most of the years of the survey, providing an estimate of particles in the 7 – 16.6 nm range by difference from the SMPS.

DENRU: The measuring station for the rural background in Denmark was located at Lille Valby (55° 41' 41" N; 12° 7' 7" E) for the period 2008 until 6/2010 and at Risø (55° 38' 40" N; 12° 5'19" E) for the period 7/2010 until 2017, both about 25 km west of Copenhagen, Denmark. For these sites, DMPS (Differential Mobility Analyser with TSI Model 3010 CPC) data covering the range 5.8 – 700 nm was available for 9 years (2008 – 2017), providing 51447 hours of data (65.4% coverage). A detailed description for this site is given by Ketzel et al. (2004). Chemical composition data was also available from that site for the same period (NO, NO_x, O_3, minerals, organic and elemental carbon, nitrate, sulphate and ammonium); meteorological data was available from the Ørsted – Institute site in Copenhagen.

DENUB: Urban background site located at the Ørsted - Institude, 2 km northeast of the city centre in Copenhagen, Denmark (55° 42' 1" N; 12° 33' 41" E). For this site, DMPS (Differential Mobility Analyser with TSI Model 3010 CPC) data covering the range 5.8 – 700 nm was available for 9 years (2008 – 2017), providing 46148 hours of data (61.4% coverage). A detailed description for this site is given by Wang et al. (2010). Meteorological and chemical composition data (NO, NO_x, O_3, minerals and elemental carbon) was also available from this site for the same period.

DENRO: Urban roadside located at H.C. Andersens Boulevard in Copenhagen, Denmark (55° 40' 28" N; 12° 34' 16" E). For this site, DMPS (custom built type including TSI CPC Model 3010) data covering the range 5.8 – 700 nm was available for 9 years (2008 – 2017), providing 49721 hours of data (65% coverage). A detailed description for this site is given by Wang et al. (2010). Chemical composition data was also available from that site for the same period (NO, NO_x, SO_2, O_3, minerals, organic and elemental carbon, nitrate, sulphate and ammonium); meteorological data was available from the nearby Ørsted – Institute site.

GERRU: Rural background site located at Melpitz, 40 km northeast of Leipzig, Germany (51° 31' 31.85" N; 12° 26' 40.30" E). Twin DMPS-IfT (TSI Models 3010 and 3025) covering the range 4.8 – 800 nm (Birmili et al., 1999) was available for 3 years (2008 – 2011), providing 22924 hours of data (87.2% coverage). A detailed description of the site is given by Engler et al. (2007). Meteorological and chemical composition data was also available from this site for the same period.

GERUB: Urban background site, part of the Tropos network located 3 km northeast from the city centre of Leipzig, Germany (51° 21' 9.1" N; 12° 26' 5.1" E). Twin DMPS-IfT (TSI Models 3010 and 3025) data covering the range 3 – 800 nm was available for 3 years (2008 – 2011), providing 23142 hours of data (88% coverage). A detailed description for this site is given by Costabile et al. (2009). Meteorological data was also available from this station for the same period.

GERRO: Urban roadside site located at Eisenbahnstraße, at the city of Leipzig, Germany (51° 20' 43.80" N; 12° 24' 28.35" E). Twin DMPS-IfT (TSI Models 3010 and 3025) data covering the range 4 – 800 nm was available for 3 years (2008 – 2011), providing 16943 hours of data (64.4% coverage). A detailed description for this site is given by Birmili et al., (2016). Meteorological data was also available from the nearby Tropos station (at GERUB mentioned later) for the same period.

FINRU: Rural background site located at Hyytiälä, 250 km north of Helsinki, Finland (61° 50' 50.70" N; 24° 17' 41.20" E). TDMPS with CPC (TSI Models 3010 and 3025) data was available for this site for 6 years (2008 – 2011 and 2015 – 2018), providing 50579 hours of data (96.1% coverage). A detailed description of the site is given by Aalto et al. (2001). Meteorological and chemical composition data (NO, NO_x, SO_2, O_3, CO, CH_4, volatile organic compounds and sulphuric acid) was also available from this site for the same period.

FINUB: Urban background site located at the Kumpula Campus 4 km north of the city centre of Helsinki, Finland (60° 12' 10.52" N; 24° 57' 40.20" E). TDMPS with CPC (TSI Models 3010 and 3025) data covering the range 3.4 – 1000 nm was available for 6 years (2008 – 2011 and 2015 – 2018), providing 51769 hours of data (98.4% coverage). A detailed description of the site is given by Järvi et al. (2009). Meteorological data was also available from this site for the same period.

FINRO: Urban roadside site located at Mäkelänkatu street, at the city of Helsinki, Finland (60° 11' 47.57" N; 24° 57' 6.01" E). DMPS (Airmodus CPC A20 and TSI CPC Model 3776) data covering the range 6 – 800 nm was available for 3 years (2015 – 2018, providing 22728 hours of data (90% coverage)). A detailed description of the site is given by Hietikko et al. (2018). Chemical composition data was available from this site for the same period (NO, NO_2, NO_x, SO_2, O_3, and black carbon); meteorological data for the same period was available from the nearby Pasila station (meteorological data from Mäkelänkatu station was also available but it is heavily biased by the street canyon vortex at the site).

SPARU: Rural background site located at the mountain Montseny (about 700 meters above sea level), 50 km north-northeast of the city of Barcelona, Spain (41° 46' 45" N; 2° 21' 29" E). For this site, SMPS (DMA TSI Model 3071 and CPC TSI Model 3785) data covering the range 9 – 856 nm was available for the 3 years (2012 – 2015), providing 10001 hours of data (53.7% availability). A detailed description of the site is given by Dall'Osto et al. (2013).

Meteorological and chemical composition data (NO, NO_2, SO_2, O_3, organic carbon and sulphate) was also provided from that site for the same period.

SPAUB: Urban background site located at Palau Reial, at the city of Barcelona, Spain (41° 23' 14" N; 2° 6' 56" E). For this site, SMPS (DMA TSI Model 3071 and CPC TSI Model 3022) data covering the range 10.9 – 478 nm was available for 3 years (2012 – 2015), providing 12652 hours of data (67.9% coverage). A detailed description of the site is given by Dall'Osto et al. (2012). Meteorological and chemical composition data (NO, NO_2, SO_2, O_3, CO, black carbon, $PM_{2.5}$, PM_{10}) was also available from that site for the same period.

GRERU: Rural background site located at Finokalia, 70 km east of Heraklion, Greece (35° 20' 16.8" N; 25° 40' 8.4" E). SMPS (TSI Model 3034) data covering the range 8.77 – 849 nm was available for 6 years (2012 – 2018), providing 34844 hours of data (83.6% coverage). A detailed description of the site is given by Kalkavouras et al. (2017). Meteorological and chemical composition data (NO, NO_2, O_3, organic and elemental carbon) was also available from this site for the same period.

GREUB: Urban background site located in the fields of the National Scientific Research Centre "Demokritos", about 12 km northeast from the city centre of Athens, Greece (37° 59' 41.96" N; 23° 48' 57.56" E). SMPS (TSI Model 3034) data covering the range 10 – 550 nm was available for 3 years (2015 – 2018), providing 20791 hours of data (79% coverage). A detailed description of the site is given by Vassilakos et al. (2005). Meteorological data was also available from this site for the same period.

2.3 Data treatment

2.3.1 Negative and zero value treatment

In a number of cases pollutant concentrations had negative or zero values in the raw data. Negative values were found in the datasets of a number of chemical compounds' (NO, NO_x, nitrate, sulphate and some VOCs in some rural sites), in most cases though only for a small number of observations. These values are the result of noise which can erroneously present negative values when the concentrations are very low. As they are still though representative of the variation of the concentrations of these compounds they were included, and the statistics were calculated using these as well. Zero values as minimum, were found in some SO_2 datasets for the sites of two areas (namely UK and Finland), the complete lack of which is not possible in the atmosphere, and as a result keeping these values as zero would be non-sensical. For this reason, zero values for the concentrations of SO_2 were set as half of the lowest detection limit of the instrument (as provided by the lowest concentration found in the dataset).

2.3.2 Statistical metrics and tests used in the thesis

For the analysis done in the present study a number of statistical methods were used. These included:

Standard Deviation: The standard deviation is a measure of the amount of dispersion of a set of values. A low standard deviation indicates that the values tend to be close to the mean, while a high standard deviation indicates that the values are spread out over a wider range from the mean value. It is calculated as

$$\sigma = \sqrt{\frac{1}{N}\sum_{i=1}^{N}(x_i - \mu)^2},$$

where N is the population of the sample, x_i is the value and μ is the mean.

Standard Error: The standard error $\sigma_{\bar{x}}$ of a parameter is the standard deviation of its sampling distribution (for example if the parameter is the mean then it is called the standard error of the mean). It is calculated by the standard deviation (σ) of the given parameter divided by the square root of the population of the sample (N).

$$\sigma_{\bar{x}} = \frac{\sigma}{\sqrt{N}},$$

One-way ANOVA test: The Analysis Of Variation is a statistical model used to analyse the differences between group of means developed by the British statistician Ronald Fischer. ANOVA provides a statistical test of whether two or more population means are equal. In the present study this metric was used to test whether two means of a given variable present differences that are statistically significant (meaning that the differences between the means of two conditions are important or not). In the present study the One-way ANOVA test was conducted, providing the level of significance between two conditions (eg. the significance between the conditions when an NPF event occurs or not). The level of significance is given by a p-value which points the level at which the different conditions are significant. For the present study a p-value smaller than 0.005 was considered to characterise the difference between two conditions as significant. As the normality of the variables is required for such an analysis, the Shapiro-Wilk test was used to assess the normality and the vast majority of the variables were found to have $p > 0.05$ and thus were considered as normal. This is probably due to the removal of the extreme values (as mentioned in section 2.2.3, for the calculations 90% of each dataset was kept removing the extremely high and/or low values and the possible outliers included in them). While this was not done to promote the normality of the populations but to reduce the bias from extreme values, it indirectly assisted in making the distributions normal. For the remaining (e.g. the growth rates associated with SO2 concentrations for UKRO in the analysis in

chapter 3.3) for which normality was not present, the square root of the values of the variable were considered to achieve normality and proceed to the ANOVA test.

2.3.3 Air mass back trajectory treatment and clustering

The characteristics of incoming air masses in a given area play a very important role in the occurrence and development of NPF events. Air mass back trajectories provide insight into the origin of incoming air masses, as well as their route for several days until these reach the area of interest. Air mass back trajectory data provided by the National Oceanic and Atmospheric Administration (NOAA), using the HYSPLIT model (Draxler et al., 1998), was used in **Paper 1**. The air masses are provided in a three-hour interval and calculate the route the incoming air masses followed for a period of four days before reaching the sites of study. The back trajectories were clustered in group according to the similarity of the route followed using the clustering method described in Carslaw, (2015). The initial clustering of the incoming air mass trajectories ended up with an optimal solution of nine clusters. As the clustering method takes into account only the geographic origin of the air masses, some of the clusters formed had very similar characteristics and origin. Reducing the number of clusters removed groups of incoming air masses with different characteristics, thus making the distinction of unique sources harder. As a result, using additional meteorological data, the method of merging groups of incoming air masses with similar characteristics was chosen. As described on **Paper 1**, this method resulted in four groups of air masses with unique origin and different characteristics. In the majority of the cases, the incoming air masses during a day were attributed to the same group, and that was the group assigned for the given day. In the cases where the incoming air masses were not from one group during the day, as NPF events start and develop during the morning hours, the group in which the

air masses were attributed between 6:00 and 12:00 was chosen. If the groups of incoming air masses in this time window were more than one, the day was not assigned to any of the groups.

In chapter 3.1.2 of **Paper 1**, the origin of the air masses between regional and local events at the background sites in the U.K. was studied. As the sites of this study were in a close proximity (less than 80 km) there was minimal variation on the origin of the air mass back trajectories between the two sites. As a result, the back trajectories for the two sites on local event days were extracted, as well as the back trajectories for the days of regional events for one of the sites (the site chosen was the UKRU, as for the majority of the NPF event days the wind direction in southern U.K. was southwestern, making UKRU the site from which the air masses passed before reaching the UKUB). The group of back trajectories that resulted was clustered using the same method as with the previous analysis and the results of this analysis were presented in **Paper 1.**

2.4 NPF event selection

NPF event extraction for all the sites studied was done following the method proposed by Dal Maso et al. (2005), which is the most widely used method at a wide range of sites. A set of criteria should be met for a day to be classified as an NPF event. The criteria are:

- A distinct new mode of particles must appear in the nucleation mode
- The new mode of particles should prevail for some hours
- The new mode of particles must show signs of growth

The dataset is visually inspected on a day-to-day basis by at least 2 individuals, and using these criteria NPF events are extracted and classified depending on their characteristics to three classes:

- Class I, containing days when formation and growth of new particles can be determined with good confidence. Class I is further divided into subclasses Ia, containing NPF events with very clear formation and growth and relatively little to no pre-existing particles (though this was not a given at the roadside sites), while Ib contains the rest of the Class I NPF events.
- Class II, containing days when the formation and/or growth of new particles was questionable
- Undefined, containing days when there is uncertainty whether they should be characterised as event or non-event days.

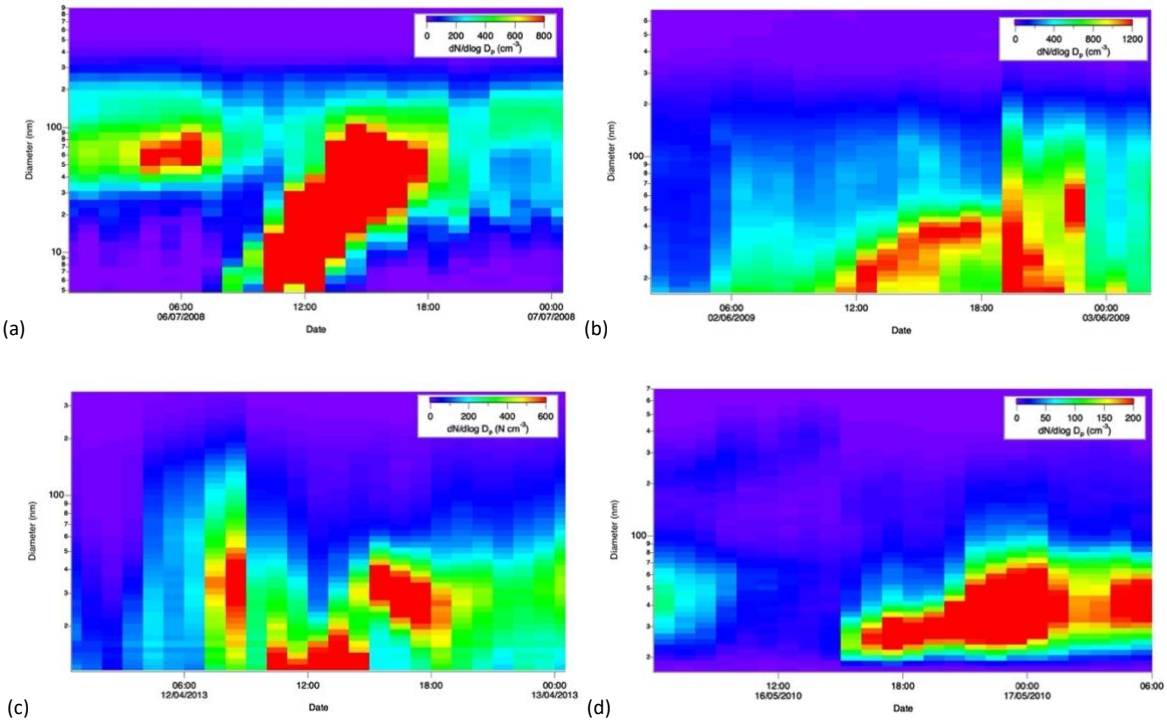
According to the authors of the method, these criteria ensure that no point sources of particles would be included (ex. traffic or heating), as well as explicitly state that NPF events should always involve particle growth, excluding short lived particle formation with no growth, such as that observed in Mace Head, Ireland (O'Dowd et al., 2002).

In the present study, only Class Ia events were considered as there is minimum uncertainty in their occurrence, making them the most suitable for modelling case studies. While the growth period suggested by the method is one hour, in the present study a three-hour growth period of at least 1 nm h^{-1} is required, in order to include only strong and clear events. Especially for UK sites, due to the relatively large size of the smallest size bin available, additional to the use of SMPS data, Condensation Particle Counter (CPC) data was also used, providing a particle count between to 7 - 16 nm. NPF events at these sites were considered when an increase in the particle number concentration appeared at this size range as well. Furthermore, data for gaseous pollutants, aerosol constituents as well as the changes in the condensation sink was also considered to identify and exclude pollution events.

Figure 5 shows examples of events that were either included or excluded. (a) presents an NPF event at GERRU and (b) at UKRO. The formation (c) was not included in the group of NPF

events due to having very short and slow growth of the new particles. As the minimum particle diameter available for the specific site is relatively high, there is a possibility that growth has taken place at the size range below of that available. Due to this uncertainty though, formations like this were not included. Formation (d) fulfils all the criteria set to be considered as an NPF event, though without the new mode of particles appearing from the minimum available size, which probably means that this mode of particles may either be associated to other sources or it was advected to the site from a different area. As of this, while such formations were extracted and characterised as "advected", they were not considered in any of the studies.

Figure 5: Examples of NPF events at (a) GERRU and (b) UKRO. (c) is a rejected formation due to lack of growth at SPAUB, while (d) is rejected as particles are probably advected to the site at UKRU.



Finally, the term "regional" events is used for NPF events that occurred at the same day on both background sites and were studied in **Papers 1 and 2**. These events may have different characteristics and development between the sites due to the differences on the conditions

between them, but due to the close proximity of the sites, meteorological conditions can be considered as similar. This study was done for the background sites in the UK, Germany, Denmark and Finland (with lesser confidence as distance was about 200 km), while the background sites in Greece were excluded due to the large distance between them. Roadside sites were not considered, as the local conditions at this type of sites play a crucial role in the occurrence of NPF events and differentiate them from the background sites. An example of a regional event in the UK is found in figure S1 in **Paper 1.**

Apart from the method used here, alternative approaches have been attempted in other studies. Beddows et al. (2015), Dall'Osto et al. (2012) and Vu et al. (2016) used Positive Matrix Factorisation (PMF) and identified a nucleation factor at urban background sites in London and Barcelona. Similarly, Pey et al., (2009) found a nucleation component using Principal Component Analysis (PCA) in Barcelona. Beddows et al. (2009, 2015) and Brines et al. (2014), used clustering methods to identify the occurrence and effect of nucleation in particle size distributions, while Dall'Osto et al. (2018) studied the variation of NPF events throughout Europe using the k-means clustering method. Finally, Joutsensaari et al. (2018) and Zaidan et al. (2018) used machine learning to identify NPF events within large datasets. While these methods look promising, they are yet to be widely used as they are still in development.

It should be noted that k-means clustering was also attempted in the present study for NPF event identification. While the extraction of NPF events was reasonable at background sites, this method was unable to separate NPF events from direct emissions within the urban environment and especially at roadsides, due to the similarity of the particle size distribution profiles. As the aim of this work was to provide uniform and comparable results among sites of different land use the use of the k-means clustering was abandoned.

2.5 Calculation of the Condensation Sink, Growth Rate, Urban Increment, Formation Rate, Nucleation Strength Factor, P parameter and NPF probability

For the analysis of the conditions, characteristics and effect of the NPF events several metrics were used. The aerosol Condensation Sink (CS) is a metric of the speed at which molecules will condense onto pre-existing aerosols (Lehtinen et al., 2003). It is one of the most important and widely used metrics of the atmospheric conditions used in the studies of NPF events, as it provides a measure of the pre-existing particles in the given atmosphere, which is crucial for the occurrence of the events. A small number of studies have used the Fuchs surface area instead (Cai et al., 2017; Westervelt et al., 2014). In the present study the condensation sink was used, not only because the Fuchs surface area is proportional to the CS, but it is the most widely metric used for this analysis. Higher values of the CS are usually found in more polluted areas, while lower ones are usually found in cleaner areas. This metric depends on both size distribution of the aerosols as well as the meteorological conditions and provides. As proposed by Kulmala et al. (2001), it is calculated as:

$$\mathrm{CS} = 4\pi \mathrm{D}_{vap} \sum \beta_{\mathrm{M}}\ \mathrm{r}\ \mathrm{N}$$

where r and N are the radii and number concentrations of the aerosols at the given time for the particle size bins with available data and D_{vap} is the diffusion coefficient of the condensing gas calculated for T = 293 K and P = 1013.25 mbar (Poling et al., 2001) as:

$$\mathrm{D_{vap}} = 0.00143 \cdot \mathrm{T}^{1.75} \frac{\sqrt{\mathrm{M_{air}^{-1}} + \mathrm{M_{vap}^{-1}}}}{\mathrm{P}\left(\mathrm{D_{x,air}^{\frac{1}{3}}} + \mathrm{D_{x,vap}^{\frac{1}{3}}}\right)^2}$$

where M and D_x are the molar mass and diffusion volume for air and the condensable vapour, in this case sulphuric acid. Sulphuric acid is chosen as it is widely accepted as the

main vapour of which the initial clusters form in the nucleation procedure. β_M is the Fuchs correction factor calculated as Fuchs et al. (1971):

$$\beta_M = \frac{1 + K_n}{1 + \left(\frac{4}{3a} + 0.377\right) K_n + \frac{4}{3a} {K_n}^2}$$

K_n is the Knudsen number, a dimensionless quantity for characterising the boundary conditions of fluid flow, calculated as $K_n = 2\lambda_m/d_p$. λ_m is the free path of the gas which is dependent to the atmospheric pressure and temperature (Kulkarni et al., 2001).

The growth rate (GR) is a metric of the speed at which the newly formed particles grow. This metric is providing information on the development of the NPF events and the efficiency of the given environment in both sustaining the newly formed particles and increasing their size through condensation. As proposed by Kulmala et al. (2012) the growth rate is calculated as:

$$GR = \frac{d_{P_2} - d_{P_1}}{t_2 - t_1}$$

where d_{Px} is the geometrical mean diameter (GMD) of the particle size distribution at the time t_x. In the present study, due to differences in the size availability between the sites, the growth rate was calculated up to the size of 50 nm for the UK sites (as the minimum size available was 16.6 nm), and up to the size of 30 nm for the rest of the sites (as the minimum available size was a lot lower in most cases). To avoid any misinterpretations caused by this difference, the results were not directly compared among the sites in different countries but only among sites in the same country, where datasets were more uniform. For the calculation of the growth rate, the time frame considered was from the start of an NPF event until one of the following conditions was met: a) growth stopped, b) GMD reached the upper limit set or c) the day ended (midnight is considered as the end of the day).

The Urban Increment (U.I.) was a metric proposed in **Paper 1** to provide with a measure of the effect of the urban environment on the number of particles formed from an NPF event. The development of NPF events depends on both meteorological and local environmental conditions. As the background sites in the UK are in a relatively close distance, meteorological conditions could be considered as almost uniform. By using the U.I. on two sites of close proximity but different land use, the effect of the different type of environment can be studied. The U.I. is defined as the ratio of the difference in the number concentration of particles below 20 nm (this size range was chosen as both sites are background and thus sources of particles other than NPF event are negligible) on event days with the average for the urban background site to the same difference at the rural background site and it is calculated as:

$$U.I. = \frac{\mathrm{UB}_{\mathrm{Nuc\,Max}} - \mathrm{UB}_{\mathrm{Bg}}}{\mathrm{RU}_{\mathrm{Nuc\,Max}} - \mathrm{RU}_{\mathrm{Bg}}}$$

where $\mathrm{UB}_{\mathrm{Nuc\,Max}}$ is the maximum concentration of particles below 20 nm found in the diurnal cycle on event days at the urban background site and $\mathrm{UB}_{\mathrm{Bg}}$ is the average mean concentration at the same time (similar for the rural background site for RU). The average used in the specific study is between April and October, which is the period when the majority of NPF events took place at both sites. The time chosen for the comparison was 13:00, as it was the time when maximum particle concentrations at this size range are found according to the diurnal cycle at both sites. The U.I. was chosen over the formation rate for this study due to its ability to include a comparison with the background conditions at the given time, as well as exclude the effect of any other source of particles in that size range.

The formation rate J is a metric proposed by Kerminen et al. (2002) to associate the formation rate from observations to the “real” atmospheric nucleation rate, which at that

point was impossible due to instrument limitations. The method was further adjusted and according to Kulmala et al. (2012) it is calculated as:

$$J_{d_p} = \frac{dN_{d_p}}{dt} + CoagS_{d_p} \times N_{d_p} + \frac{GR}{\Delta d_p} \times N_{d_p} + S_{losses}$$

where dN_{dp} is the difference in number concentration of particles of diameter d_p for time dt, GR is the growth rate of the particles, $CoagS_{dp}$ is the coagulation rate of particles of diameter d_p, calculated as (Kerminen et al., 2001):

$$CoagS_{d_p} = \int K(d_p, d'_p)\, n(d'_p) dd'_p \cong \sum_{d'_p=d_p}^{d'_p=max} K(d_p, d'_p)\, N_{d_p}$$

where $K(d_p, d'_p)$ is the coagulation coefficient of particle sizes d_p and d'_p. The S_{losses} accounts for additional losses (ie. losses to chamber walls), which cannot be considered for atmospheric conditions. In the present study, the formation rate for particles with diameter of 10 nm was calculated as the majority of the sites had data of this size. As the initial particle formation occurs at about 1.5 nm, the formation rate calculated in this study refers to the particles associated with NPF that survived and grew up to that size, thus this metric is affected by the survival and growth of the particles up to that size. Specifically for the UK sites, due to data limitations the size chosen was 16.6 nm as this was the minimum available. Due to the differences in the available data, the formation rates were not directly compared between the sites in this study.

The Nucleation Strength Factor (NSF) is a metric proposed by Salma et al. (2014) and provides information about the contribution of NPF events to the number concentration of ultrafine particles. There are two metrics proposed, being the NSF_{NUC} calculated as:

$$NSF_{NUC} = \frac{\left(\frac{N_{\text{smallest size available}-100}}{N_{100-\text{largest size available}}}\right)_{\text{nucleation days}}}{\left(\frac{N_{\text{smallest size available}-100}}{N_{100-\text{largest size available}}}\right)_{\text{non-nucleation days}}}$$

which provides a measure of the concentration increment which can be attributed to NPF events on event days, and the NSF_{GEN} proposed by Salma et al. (2017) and calculated as:

$$NSF_{GEN} = \frac{\left(\frac{N_{\text{smallest size available}-100}}{N_{100-\text{largest size available}}}\right)_{\text{all days}}}{\left(\frac{N_{\text{smallest size available}-100}}{N_{100-\text{largest size available}}}\right)_{\text{non}-\text{nucleation days}}}$$

which expresses the same contribution but on a longer time span (season, year etc.). Apart from providing information about the effect of NPF events on the ultrafine particles, these metrics may also be used as an indicator for the effect of the events on CCN formation, as the particle size range considered includes those that have reached the critical size for CCN formation. The metric as is though cannot give direct information about this effect as it includes particles with sizes a lot smaller as well. In the present study, the effect of NPF events on CCN was not considered and the metric was used as proposed.

The dimensionless survival parameter P, which is proportional to the L parameter proposed by McMurry et al. (1979), was proposed by Kulmala et al., (2017). It is a metric that classifies sites using data from previous studies and is used to investigate the conditions in which NPF events take place. It is calculated as:

$$P = \frac{CS'}{GR'}$$

where CS' = CS/ (10^{-4} s^{-1}) using the average condensation sink and GR' is the average hourly growth rate in the given site. In the aforementioned study the parameter is calculated for many sites and it was found to vary between 2 in very clean and remote areas up to 2000 for very polluted urban sites in large Asian megacities. As mentioned in the aforementioned study, the P parameter needs to be smaller than about 50 for NPF events to take place. It was found though that even at sites with parameter values a lot higher NPF events still take place which contradicts what would be expected on the basis of theoretical arguments. This parameter was calculated in depth in **Paper 1**, in which the seasonal and incoming air mass

variation was studied and a possible connection between its variation and the development of NPF events was investigated. The P parameter's values can be biased by the differences in the available sizes in datasets, it was not used in the later studies, as they included datasets with variable particle sizes.

The NPF probability is a simple metric for the probability of an NPF event within a certain group of days. It is calculated as:

$$\mathrm{NPF}_{\mathrm{probability}} = \frac{\mathrm{N}_{\mathrm{NPF\ event\ days\ for\ group\ of\ days\ X}}}{\mathrm{N}_{\mathrm{days\ with\ available\ data\ for\ group\ of\ days\ X}}}$$

where group of days X, depending on the analysis refers to a year, season, group of days of the week or wind direction sector. This removes the bias caused by data availability. It should be noted that the frequency of NPF events reported for each site is calculated as the number of NPF events to the total number of days with available data and as a result it is biased by the temporal data availability.

2.6 Calculation of the gradients and intercepts for the variables studied in chapter 3.3

Due to the large datasets available and the great spread of the values, a direct comparison between a given variable and any of the characteristics associated to NPF events (NPF probability, growth rate and formation rate) always provided results with low significance. As a result, in **Chapter 3.3** an alternative method which can provide a reliable result without the noise of the large datasets was used in the present study, to investigate the relationships between the variables which are considered to be associated with the NPF procedure. For this, a timeframe which would be more directly associated with the NPF events was chosen. For NPF probability and GR the timeframe between 5:00 to 17:00 was chosen, which is

considered the time when the vast majority of NPF events take place and develop. For the formation rate a smaller timeframe was chosen, 9:00 to 15:00 which is $\pm$ 3 hours from the time of the maximum formation rate found for almost all sites (12:00). This was done to exclude as well as possible the effect of the morning rush have at the roadsides, as well as only include the time when the formation rate is mostly relevant to NPF events (negative values resulting from particle removing mechanisms that are more probable outside this timeframe would bias the result). Specifically for the condensation sink, the timeframe 5:00 to 10:00 was chosen. This was done to avoid including the direct effect of the NPF events as well as to provide with results for the conditions which either promote or suppress the characteristics studied, which specifically for the condensation sink are more important before the start of the events.

In general, the extreme values which bias the results only carrying a very small piece of information were then removed, though at least 90% of the available data was used for all the variables. The data left was separated into smaller bins and a minimum of 10 bins was required for each variable (for example if the difference between the minimum and the maximum relative humidity is 70%, then 14 bins of a range 5% were formed). The variables of interest were then averaged for each bin and plotted, and a linear relationship was considered for each one of them. The variables of interest were then averaged for each bin, plotted using running averages which makes the interpretation of the relationships and trends easier and discussed in the Results section (chapter 3.3.3).

Finally, a linear relationship was considered for each one of the meteorological and atmospheric variables with the variables studied (NPF probability, growth and formation rate). While it is evident that many of these relationships are not linear, the specific type was chosen in the present analysis because the aim was to elucidate the general positive or negative effect of the variables studied. Furthermore, the effect of many variables appears

to vary between sites with great differences (either geographical or type of land use) and the choice of a single method to describe these relationships ensures the uniformity of the results, as it appears to better describe them in most cases.

3. Results

3.1 Analysis of new particle formation (NPF) events at nearby rural, urban background and urban roadside sites

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Analysis of new particle formation (NPF) events at nearby rural, urban background and urban roadside sites

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Abstract. New particle formation (NPF) events have different patterns of development depending on the conditions of the area in which they occur. In this study, particle size distributions in the range of 16.6–604 nm (7 years of data) were analysed and NPF events occurring at three sites of differing characteristics – rural Harwell (HAR), urban background North Kensington (NK), urban roadside Marylebone Road (MR), London, UK – were extracted and studied. The different atmospheric conditions in each study area not only have an effect on the frequency of the events, but also affect their development. The frequency of NPF events is similar at the rural and urban background locations (about 7 % of days), with a high proportion of events occurring at both sites on the same day (45 %). The frequency of NPF events at the urban roadside site is slightly less (6 % of days), and higher particle growth rates (average 5.5 nm h^{-1} at MR compared to 3.4 and 4.2 nm h^{-1} at HAR and NK respectively) must result from rapid gas-to-particle conversion of traffic-generated pollutants. A general pattern is found in which the condensation sink increases with the degree of pollution of the site, but this is counteracted by increased particle growth rates at the more polluted location. A key finding of this study is that the role of the urban environment leads to an increment of 20 % in $N_{16–20\,nm}$ in the urban background compared to that of the rural area in NPF events occurring at both sites. The relationship of the origin of incoming air masses is also considered and an association of regional events with cleaner air masses is found. Due to lower availability of condensable species, NPF events that are associated with cleaner atmospheric conditions have lower growth rates of the newly formed particles. The decisive effect of the condensation sink in the development of NPF events and the survivability of the newly formed particles is underlined, and influences the overall contribution of NPF events to the number of ultrafine particles in an area. The other key factor identified by this study is the important role that pollution, both from traffic and other sources in the urban environment (such as heating or cooking), plays in new particle formation events.

1 Introduction

Ultrafine particles (particles with diameter smaller than 100 nm) typically make the greatest contribution in the total particle count, especially in urban environments (Németh et al., 2018), but a very small contribution to total volume and mass (Harrison et al., 2000). Research studies have indicated that ultrafine particles can cause pulmonary inflammation, may contribute to cardiovascular disease (Oberdörster, 2000) and have an increased possibility to penetrate the brain and central nervous system (Politis et al., 2008) compared to fine and coarser particles. Since some studies report that toxicity per unit mass increases as particle size decreases (Penttinen et al., 2001; MacNee et al., 2003; Davidson et al., 2005), it is considered possible that particle number concentrations may be a better predictor of health effects than mass concentrations (Harrison et al., 2000; Atkinson et al., 2010; Kelly et al., 2012; Samoli et al., 2016). Additionally, new particle for-

mation (NPF) events have an impact on climate (Makkonen et al., 2012) either by increasing the number of cloud condensation nuclei (Spracklen et al., 2008; Merikanto et al., 2009; Dameto de España et al., 2017; Kalkavouras et al., 2017) or directly affecting the optical properties of the atmosphere (Seinfeld and Pandis, 2012).

The sources of ultrafine particles in urban areas can either be primary particles from emission sources such as traffic (Shi et al., 1999; Harrison et al., 2000), airports (Masiol et al., 2017) and other combustion-related processes (Keuken et al., 2015; Kecorius et al., 2016), or by NPF from gaseous precursors. New particle formation as described by Kulmala et al. (2014) is the process of production of low-volatility vapours, clustering of these vapours, nucleation, activation of the clusters with a second group of vapours and condensational growth to larger sizes. This process can occur both locally or on a larger scale; in the latter case the events are characterized as regional. Regional events have been found to take place on a scale of hundreds of kilometres (Németh and Salma, 2014; Shen et al., 2018), without being affected by air mass advection (Salma et al., 2016). NPF is one of the main contributors of particles in the atmosphere (Spracklen et al., 2010; Kulmala et al., 2016; Rahman et al., 2017) and this relative contribution increases when moving from a kerbside to a rural area (Ma and Birmili, 2015). While NPF events in rural and remote areas have been widely studied for many years (O'Dowd et al., 2002; Dal Maso et al., 2005; Ehn et al., 2010; Dall'Osto et al., 2017; Kalkavouras et al., 2017), in urban areas intensive studies have started mainly in recent years (Jeong et al., 2010; Minguillón et al., 2015; Peng et al., 2017; Németh et al., 2018). Early studies in Birmingham, UK, highlighted the connection of NPF events with solar radiation (Shi et al., 2001) and a low condensation sink (Alam et al., 2003), a measure of pre-existing aerosol loading (Dal Maso et al., 2002). The importance of a low condensation sink was further underlined by later studies as being one of the most influential variables in the occurrence of NPF in all types of environment (Wehner et al., 2007; Park et al., 2015; Pikridas et al., 2015). An important contributor to many NPF pathways is SO_2 (Woo et al., 2001; Berndt et al., 2006; Laaksonen et al., 2008), which in the presence of solar radiation forms H_2SO_4, often the main component of the initial clusters (Kuang et al., 2008; Kulmala et al., 2013; Bianchi et al., 2016; Kirkby et al., 2016). Dall'Osto et al. (2013) pointed out that the role of SO_2 is less significant in urban areas compared to rural and background areas. SO_2 concentration variability in urban areas was found to have a small impact on the frequency of NPF events (Alam et al., 2003; Jeong et al., 2010), though it can have an effect on the number of particles formed (Charron et al., 2007). Furthermore, Dall'Osto et al. (2018) in their research at 24 sites in Europe pointed out the different role SO_2 seems to play depending on its concentration, and that of other species. Jayaratne et al. (2017), however, found that in the heavily polluted environment of Beijing, China, NPF events were more probable in sulfur-rich conditions rather than sulfur-poor conditions. Apart from its role in the initial formation of the clusters, H_2SO_4 seems to participate in the early stages of growth of the newly formed clusters (Kulmala et al., 2005; Iida et al., 2008; Xiao et al., 2015). In later stages of growth, low or extremely low volatility organic compounds (O'Dowd et al., 2002; Laaksonen et al., 2008; Metzger et al., 2010; Kulmala et al., 2013; Tröstl et al., 2016; Dall'Osto et al., 2018) appear to be more important, while the role of ammonium nitrate in particle growth is also considered (Zhang et al., 2017). While in rural areas the organic compounds are mainly of biogenic origin (Riccobono et al., 2014; Kirkby et al., 2016), in urban areas they mainly originate from combustion processes (Robinson et al., 2007; Gentner et al., 2012). Many comparative studies have reported higher growth rates in urban areas compared to background sites (Wehner et al., 2007; Jeong et al., 2010; Salma, et al., 2016; Wang et al., 2017), as well as greater particle formation rates (Salma, et al., 2016; Nieminen et al., 2018) and a higher frequency of NPF events (Peng et al., 2017), which was attributed to the higher concentration of condensable species. Salma et al. (2014), however, reported fewer NPF events in the city centre of Budapest compared to the urban background, due to the higher condensation sink. Due to the complexity of the conditions and mechanisms within an urban area (Harrison, 2017), NPF events are harder to study and factors that drive them are harder to attribute. Increased concentrations of particles in the size range 1.3–3 nm were measured at a kerbside site when downwind from the road, following the trends in traffic-related nucleation mode particles, associating them with traffic emissions and thus not resulting from homogeneous nucleation mechanisms (Rönkkö et al., 2017; Hietikko et al., 2018); studies in Barcelona, Spain (Dall'Osto et al., 2012; Brines et al., 2014), and Leicester, UK (Hama et al., 2017), attributed a larger portion of nucleation mode particles to vehicular emissions compared to photochemically induced nucleation. As the condensation sink is higher within an urban environment, NPF events are less favoured. Their occurrence is attributed to either ineffective scavenging or the higher growth rate of the newly formed particles (Kulmala et al., 2017). The latter may occur when sufficient concentrations of precursors are present in the atmosphere (Fiedler et al., 2005), as particle formation was found to take place on both event and non-event days with variable intensity. Particle formation though is not always followed by survival or growth of the newly formed particles and in such cases does not qualify as a NPF event (Riipinen et al., 2007).

In this study, NPF events in three areas of different land use in the southern UK are analysed. Studies for NPF events have been conducted in the past for Harwell, Oxfordshire (Charron et al., 2007, 2008), and the effect of NPF upon particle size distributions was also considered for North Kensington, London (Beddows et al., 2015). A combined study including all three sites has also been conducted, but in the aspect of ultrafine particle variation (Von Bismarck-Osten

Figure 1. Map of the measuring stations.

et al., 2013). The present study is the first to use a combined long-term database for all three sites, focusing on the trends and conditions of NPF events at these sites, as well as the first which identifies NPF events at the high-traffic Marylebone Road site, as up to this point ultrafine particles were attributed only to traffic (Charron and Harrison, 2003; Dall'Osto et al., 2011). As in this study a rural and an urban background area are studied alongside a kerbside site in the city of London in close proximity, the conditions and development of NPF events in a mid-latitude European region are discussed in relation to the influence of different local environments.

2 Data and methods

2.1 Site description and data availability

This study analysed NPF events in three areas in the southern United Kingdom (Fig. 1). Harwell in Oxfordshire is located about 80 km west of the greater London area. The site is in the grounds of the Harwell Science Centre in Oxfordshire (51°34′15″ N, 1°19′31″ W) and is representative of a rural background area; a detailed description of the site was given by Charron et al. (2013). North Kensington is a suburban area in the western side of London, UK, 4.5 km west of Marylebone Road. The site is located in the grounds of Sion Manning School (51°31′15″ N, 0°12′48″ W) and is representative of the urban background of London. A detailed description of the site was given by Bigi and Harrison (2010). Marylebone Road is located in the centre of London, UK. The site is located on the kerbside of Marylebone Road (51°31′21″ N; 0°9′16″ W), a very busy arterial route within a street canyon. A more detailed description of the area can be found in Charron and Harrison (2003).

At all three sites, 7 years (2009–2015) of particle number size distributions in the range of 16.6–604 nm have been measured and recorded as 15 min averages, using a scanning mobility particle sizer (SMPS), comprised by an electrostatic classifier (EC, TSI model 3080) and a condensation particle counter (CPC, TSI Model 3775), operated on behalf of the UK Department for Environment, Food and Rural Affairs (DEFRA). At all sites the inlet air is dried, and the operation is in accordance with the EUSAAR/ACTRIS protocol (Wiedensohler et al., 2012). These 15 min measurements were averaged to an hourly resolution. In Harwell there were 46 930 h of available SMPS data (76.5 % coverage), in North Kensington 51 059 (83.3 % coverage) and at Marylebone Road 45 562 (74.3 % coverage). Detailed data availability is found in Table S1 in the Supplement. A freestanding CPC (TSI model 3022A) was also operated alongside the EC for most of the years of the survey and was used to give an estimate of particles in the 7–16.6 nm range by difference from the SMPS.

Additionally, air pollutants and other gas and particle chemical composition data (NO_x, SO_2, SO_4^{2-}, Cl, Na, Mg, gaseous ammonia and volatile organic compounds – VOCs) were extracted from the DEFRA website (https://uk-air.defra.gov.uk/, last access: 14 May 2018); daily measurements of particulate organic carbon (OC) were also extracted from the DEFRA website which are determined using the method described in the Annual report of the National Physical Laboratory (Beccaceci et al., 2015). Meteorological data for Harwell and Heathrow airport (used for North Kensington and Marylebone Road) were available from the Met Office, while solar radiation data from Benson station (for Harwell) and Heathrow airport (for North Kensington and Marylebone Road) were extracted from the Centre for Environmental Data Analysis (CEDA) site (http://www.ceda.ac.uk, last access: 16 May 2017). Back trajectory data calculated using the HYSPLIT model (Draxler and Hess, 1998) were extracted by the NOAA Air Resources Laboratory (https://ready.arl.noaa.gov/READYtransp.php, last access: 8 December 2017) and were processed using the Openair package for R (Carslaw and Ropkins, 2012).

2.2 Methods

2.2.1 NPF events selection

The identification of the NPF event days was made by visual inspection of SMPS data, supplemented with the use of CPC data to confirm the formation of a new mode of particles, using the criteria set by Dal Maso et al. (2005). NPF events are considered when a distinctly new mode of particles, which appears in the size distribution at nucleation mode size, prevails for some hours and shows signs of growth. Using these criteria, NPF events are classified into two classes, I and II depending on the level of certainty. Class I events are further classified to Ia and Ib, with class Ia containing very clear and strong particle formation events, while Ib contains less clear events. In this study only the events of class Ia are consid-

ered to be the most suitable for analysing case studies of NPF events (Fig. S1 in the Supplement). At this point it should be mentioned that due to the particle size range available, NPF events in which newly formed particles failed to grow beyond 16.6 nm (if any) could not be identified. Bursts of new particles in the size range < 16.6 nm that were identified using the CPC data but did not appear in the SMPS dataset were ignored as their development was unknown. This type of development was rare and mainly found at the rural background site, occurring on a few days per year mainly in summer. Its main feature was the short duration of the bursts compared to event days. In the urban sites, this type of development was almost non-existent. High time resolution data for gaseous pollutants and aerosol constituents were used to identify pollution events affecting particle concentrations and these were removed from the data analysis. This analysis took account of the fact that nanoparticle emissions from Heathrow Airport affect size distributions at London sites (Harrison et al., 2018), and such primary emission influences were not included as NPF events.

2.2.2 Calculation of the condensation sink and growth rate

For the calculation of the condensation sink the method proposed in Kulmala et al. (2001) was used in which the condensation sink is calculated as

$$\mathrm{CS} = 4\pi D \sum \beta_M r N, \tag{1}$$

where r is the radius of the particles and N is the number concentration of the particles. D is the diffusion coefficient calculated (for $T = 293$ K and $P = 1013.25$ mbar) according to Kuuluvainen et al. (2010):

$$D_{\mathrm{vap}} = 0.00143 \cdot T^{1.75} \frac{\sqrt{M_{\mathrm{air}}^{-1} + M_{\mathrm{vap}}^{-1}}}{P\left(D_{x,\mathrm{air}}^{\frac{1}{3}} + D_{x,\mathrm{vap}}^{\frac{1}{3}}\right)^2}, \tag{2}$$

where P is air pressure, M is the molar mass and D_x is the diffusion volume for air and H_2SO_4. β_M is the Fuchs correction factor calculated as follows (Fuchs et al., 1971):

$$\beta_M = \frac{1 + Kn}{1 + \left(\frac{4}{3a} + 0.377\right) Kn + \frac{4}{3a} Kn^2}, \tag{3}$$

where Kn is the relation of the particle diameter and the mean free path of the gas λ_m, called the Knudsen number.

The growth rate of the particles on nucleation event days was also calculated as proposed by Kulmala et al. (2012), using the formula

$$\mathrm{GR} = \frac{D_{P_2} - D_{P_1}}{t_2 - t_1} \tag{4}$$

for the size range 16.6–50 nm. The number of points taken depended on the development of the event and were considered from the start of the event until (a) growth stopped, (b) GMD reached 50 nm or (c) the day ended (this cut-off was chosen as the development of an event in its later stages is heavily biased by the local conditions, especially at the urban sites).

2.2.3 Calculation of the urban increment (UI)

The urban increment (UI) is defined as the ratio of the number concentration of particles below 20 nm for event days to the average (for the period April–October, when the majority of the events take place) for North Kensington to that at Harwell. This provides a measure of the new particles formed in each area in comparison to the average conditions, and is calculated by

$$\mathrm{UI} = \frac{\mathrm{NK}_{\mathrm{Nuc\,Max}} - \mathrm{NK}_{\mathrm{Avg}}}{\mathrm{HW}_{\mathrm{Nuc\,Max}} - \mathrm{HW}_{\mathrm{Avg}}}, \tag{5}$$

where $\mathrm{NK}_{\mathrm{Nuc\,Max}}$ is the maximum concentration of particles below 20 nm found in the diurnal cycle on event days (found at 13:00 GMT) and $\mathrm{NK}_{\mathrm{Avg}}$ is the average mean concentration at the same time (same for Harwell in the denominator).

2.2.4 Calculation of nucleation strength factor (NSF) and the *P* parameter

The nucleation strength factor (NSF) was proposed by Salma et al. (2014) as a measure of the effect nucleation events have in the composition of ultrafine particles in an area. Two factors were proposed. First is the $\mathrm{NSF}_{\mathrm{NUC}}$. This is calculated as

$$\mathrm{NSF}_{\mathrm{NUC}} = \frac{\left(\frac{N_{\text{smallest size available}-100}}{N_{100-\text{largest size available}}}\right)_{\text{nucleation days}}}{\left(\frac{N_{\text{smallest size available}-100}}{N_{100-\text{largest size available}}}\right)_{\text{non}-\text{nucleation days}}} \tag{6}$$

and provides of a measure of the concentration increment on nucleation days exclusively caused by NPF. The second factor is $\mathrm{NSF}_{\mathrm{GEN}}$ calculated as

$$\mathrm{NSF}_{\mathrm{GEN}} = \frac{\left(\frac{N_{\text{smallest size available}-100}}{N_{100-\text{largest size available}}}\right)_{\text{all days}}}{\left(\frac{N_{\text{smallest size available}-100}}{N_{100-\text{largest size available}}}\right)_{\text{non}-\text{nucleation days}}} \tag{7}$$

and gives a measure of the overall contribution of NPF over a longer time span (Salma et al., 2017).

The dimensionless survival parameter P, as proposed in Kulmala et al. (2017), was calculated as

$$P = \frac{\mathrm{CS}'}{\mathrm{GR}'}, \tag{8}$$

where $\mathrm{CS}' = \mathrm{CS}/(10^{-4}\,\mathrm{s}^{-1})$ and $\mathrm{GR}' = \mathrm{GR}/(1\,\mathrm{nm\,h}^{-1})$. CS and GR values used were calculated with the methods mentioned at Sect. 2.2.2. An increased P parameter is an indication that a smaller percentage of newly formed particles

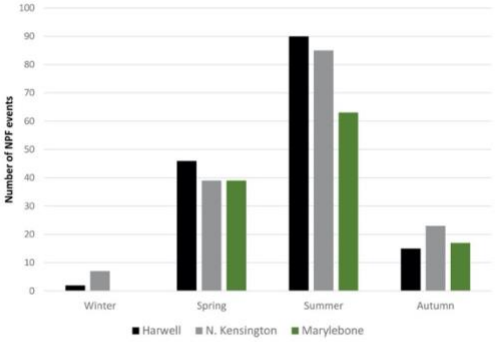

Figure 2. Number of NPF events per season for all 7 years of the present study (winter – DJF; spring – MAM; summer – JJA; autumn – SON) at Harwell (rural), North Kensington (urban background) and Marylebone Road (urban roadside).

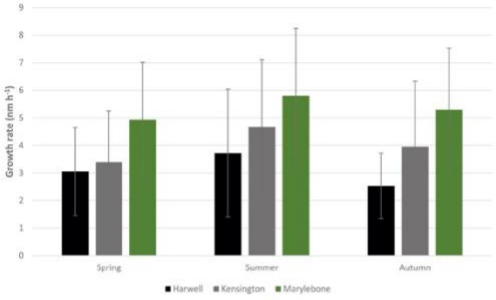

Figure 3. Growth rate per season at the three sites.

will survive to greater sizes. Hence this is the inverse of particle survivability, and values of $P < 50$ are typically required for NPF in clean or moderately polluted environments, although higher values of P are observed in highly polluted atmospheres (Kulmala et al., 2017).

3 Results and discussion

3.1 NPF events in the background areas

3.1.1 Conditions and trends of NPF events

The number of NPF event days for each site per year, those that took place simultaneously at both urban and rural background sites, and those events that took place at all three sites simultaneously appear in Table 1. Given that overall data recovery was in the range of 74 %–83 %, results from individual years are unreliable, but the 7-year runs should average out most of the effects of incomplete data recovery. The number of events is similar for Harwell and North Kensington, with a frequency of about 7 % of all days with data. There is a clear seasonal variation favouring summer and spring (Fig. 2) for both areas of the study. A similar pattern of variation was found for North Kensington by Beddows et al. (2015). In general, higher solar radiation, lower relative humidity, low cloud cover and higher pressure conditions, lower concentrations of pollutants, and lower condensation sinks are found when NPF events took place in all areas (Fig. S2), as was also reported by Charron et al. (2007) for Harwell. While SO_2 is one of the main factors for NPF events to occur, concentrations are lower when events take place. This is indicative that SO_2 concentrations in these areas are sufficient for events to take place, and higher concentrations are likely to be associated with higher pollution and a higher condensation sink. The proxy for $[H_2SO_4]$ was calculated for the background sites using the method outlined in Petäjä et al. (2009) and was found to be higher on event days for both background sites (results not included). This indicates the possible positive effect of increased concentrations of H_2SO_4 in the occurrence of NPF events as well as, since SO_2 concentrations were found lower, the increased role of either the solar radiation (via the formation of OH radical) or the reduced condensation sink to its formation. For the case of gaseous ammonia (results not included) for Harwell where data were available, as there was no distinct variation found between event and non-event days, but as the concentration of ammonia in the UK is in the range of few parts per billion (Sutton et al., 1995), it is sufficient according to ternary nucleation theory (Korhonen et al., 1999) for NPF events not to be limited by ammonia. The average growth rate for Harwell was found to be $3.4\,nm\,h^{-1}$, within the range given by Charron et al. (2007), and higher at North Kensington at $4.2\,nm\,h^{-1}$, a trend found for all seasons (Fig. 3). The increased growth rate in the urban area can be related to the greater presence of organic matter and other condensable species. In both areas NPF events had higher growth rates in summer than in spring, as was also found in previous studies (Kulmala et al., 2004; Nieminen et al., 2018). This may be associated with the higher concentration of organic compounds emitted by trees during summer (Riipinen et al., 2007), or faster oxidation rates due to higher concentrations of hydroxyl radical and ozone (Harrison et al., 2006).

About 45 % of the events took place simultaneously in both background areas. These events are characterized as regional, as NPF took place on a larger scale, regardless of the local conditions of the given area. In this case, meteorological conditions were even clearer, indicative of the greater dependence of regional events on synoptic conditions rather than local. While most chemical constituents were also lower in concentration during regional events, different patterns were found for organic compounds and sulfate for each background area. In Harwell sulfate was higher during regional events, while in North Kensington organic compounds were

Table 1. Number of NPF events per site (in parenthesis the number of days with available data).

	Harwell	N. Kensington	Marylebone Road	Regional (background sites)[1]	Regional (all 3 sites)[2]
2009	9 (210)	0 (332)	4 (290)	0	0
2010	29 (262)	22 (310)	22 (292)	11	9
2011	15 (291)	10 (300)	23 (284)	4	1
2012	8 (334)	28 (303)	12 (140)	3	0
2013	25 (328)	23 (342)	27 (334)	13	11
2014	29 (324)	34 (330)	13 (314)	18	6
2015	25 (282)	22 (314)	18 (338)	11	10
Overall	140 (2031)	139 (2231)	119 (1993)	60	37

[1] Refers to events occurring simultaneously at Harwell and North Kensington. [2] Refers to events which occur simultaneously at all three sites.

higher during regional events. This may be indicative of the variable role that specific chemical species have in condensational nanoparticle growth (Yue et al., 2010). In all cases though, the concentrations of these species were lower compared to the average conditions. Despite these differences, the growth rate of particles was found to be higher for local events in North Kensington (4.4 nm h^{-1}) compared to regional events (3.9 nm h^{-1}), though within the margin of uncertainty. In Harwell, no difference was found in the growth rate between regional and local events.

3.1.2 Urban increment and particle development

The urban environment, depending on the conditions, may have a positive or negative effect in the number of the particles formed and their consequent survival and growth. Both Harwell and North Kensington are in background areas, rural and urban respectively. As a result, while the concentrations of pollutants are higher in North Kensington than Harwell, their effect is smaller compared to that of Marylebone Road. A comparison of the particles smaller than 20 nm gives insight into the formation and survival of the newly formed particles in the early stages. Calculating the urban increment (Eq. 5) using the two background sites showed around 20 % more particles of size 16–20 nm in North Kensington than Harwell for event days, an increment that is even stronger when solely local events are considered (Fig. 4). As the sizes of the particles in the calculation are relatively large and due to the higher condensation sink found in North Kensington, this increment is expected to be larger for smaller size particles. A possible explanation for this result may be the greater concentration of organic compounds which is observed in North Kensington, as discussed earlier, which leads to more rapid formation of secondary condensable species that enhances the nucleation process in the more polluted area.

Considering the local events, most of the pollutant concentration data available appear to be higher, which is reflected in the condensation sink as well. The role of the polluted background appears to be decisive in the further growth of

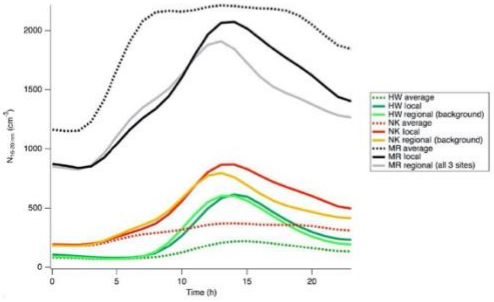

Figure 4. Diurnal variation of $N_{16–20\,nm}$ at each site: annual mean and NPF event days.

the newly formed particles, especially for Harwell. At both sites, this causes the number of particles of greater size to be smaller for the later hours in the days of local events (Fig. S3). Another possible reason for this difference in the larger size ranges could be the higher concentration of organic content on the days of regional events at North Kensington (as discussed earlier). On the other hand, for Harwell all hydrocarbons with available data are lower throughout the day (apart from ethane) during regional events. Unlike North Kensington, at Harwell particles smaller than 20 nm as well as the growth rate of the newly formed particles are almost the same for regional and local events.

The calculation of the increment on Marylebone Road provided negative results; particles smaller than 20 nm were less abundant on event days compared to the average, throughout the day. This is due to the fact that Marylebone Road is heavily affected by traffic pollution and, on average, conditions do not promote NPF events due to the high condensation sink, unless clear conditions prevail, which are also associated with a low particle load.

3.2 NPF events at Marylebone Road

For many years, NPF events were thought not to take place in heavily polluted urban areas, as the effect of the increased condensation sink was considered crucial in suppressing the formation and growth of new particles. Recent long-term analyses have shown this is not the case and nowadays an increasing number of studies confirm the occurrence of NPF events in urban areas. In this study, for the same period of 7 years as for the two background areas, NPF events were found to occur for 6.1 % of days at Marylebone Road, lower than in the background areas. Though due to the particle size range available there cannot be a definitive answer to whether the formation of the particles takes place in the specific locality of the sampling site, due to the observed increase in particle concentrations in the range 7–16 nm (provided by the CPC data) and the increased growth rates found in urban areas in general, it can be assumed that the formation takes place either in the area of the measuring site or in its close vicinity, while the growth of the particles persists in the area for several hours, despite the high condensation sink. Seasonal variation is similar to that at the background sites, but day-of-the-week variation is stronger at Marylebone Road, further favouring weekends (Fig. S4), as on these days traffic intensity is lower.

In general, similar conditions found to affect NPF events at the background sites are also found at Marylebone Road, despite a much larger condensation sink (Fig. S2). As a result, less particles of size smaller than 20 nm were found on NPF event days than the average for the site, as the sum of background particles plus those formed on these days were less than that on an average day. The growth rate of the newly formed particles (5.5 nm h^{-1}) is higher than that of the background sites, which is in agreement with the findings in the study of the background areas on the possible role of the condensable species, the concentrations of which are even greater at the urban kerbside. About 15 % of NPF event days at Marylebone Road presented particle shrinkage after the initial growth; the study of these cases though is outside of the context of the present work. At Marylebone Road, the number of NPF days which were common with the background sites was fewer, as local conditions (high condensation sink) are detrimental to the occurrence of NPF events and thus the days of regional events including Marylebone Road were separately studied for this site. There were 37 regional event days that were common for all three sites (31 % of events at Marylebone Road) (Table 1). As with the other two areas, the growth rate is higher during local events, but the conditions are mixed, with lower concentrations of sulfate and organic compounds but higher SO_2, NO_x and elemental carbon. The relationship with higher wind speed (mainly western; Fig. S6), solar radiation (which results in greater H_2SO_4 formation) and lower relative humidity indicates the stronger relation of the regional events with synoptic conditions than the local events in the heavily polluted environment of Marylebone Road.

3.3 Connection of NPF events with incoming air masses

3.3.1 Air mass back trajectory clustering and connection with NPF events

The origin of the air masses plays a very important role in the occurrence of NPF events. Air masses of different origins have different characteristics. Back trajectories provide excellent insight into the source of the air masses. Air mass back trajectories were calculated both for all days and for NPF event days for each site separately. This analysis gives a view of the frequency of NPF events within different air mass types. The initial air mass back trajectory clustering ended up with an optimal solution of nine clusters of different air masses. As many of these clusters had similar characteristics and origin, solutions with fewer clusters were attempted. As the number of clusters was decreasing clusters became a mixture of different origins, thus making the distinction of different sources harder. As a result, the method chosen was to merge clusters of similar origin and characteristics, which kept the detail of the large number of clusters and made the separation of the different origins more distinct.

The resulting four merged clusters (Fig. 5), using the characterisation proposed by McIntosh et al. (1969), are as follows:

- *An arctic cluster, which originates mainly from the northerly sector.* It occurs about 10 % of the time and consists of cold air masses, which either passed over northern parts of the UK or through the Irish Sea.

- *A tropical cluster, which originates from the central Atlantic.* It occurs 25 % of the time and contains warmer air masses. A small percentage of this cluster contains masses that have passed over countries south of the UK. Even though these days were more polluted, the clustering method was unable to clearly distinguish these days as it does not take into account particle numbers or composition, even when the nine-cluster solution was applied.

- *A polar cluster, which originates from the north Atlantic.* It is the most common type of air mass arriving in the areas of study and occurs about 40 % of the time, bringing fast-moving, "clean" air masses with increased marine components (Cl, Na, Mg) from the west. This cluster also contains air masses that have passed through Ireland, though an effect on particle size and chemical composition is not distinct.

- *A continental cluster, which originates from the east.* It occurs about 25 % of the time and consists mainly of slow-moving air masses, originating from the London

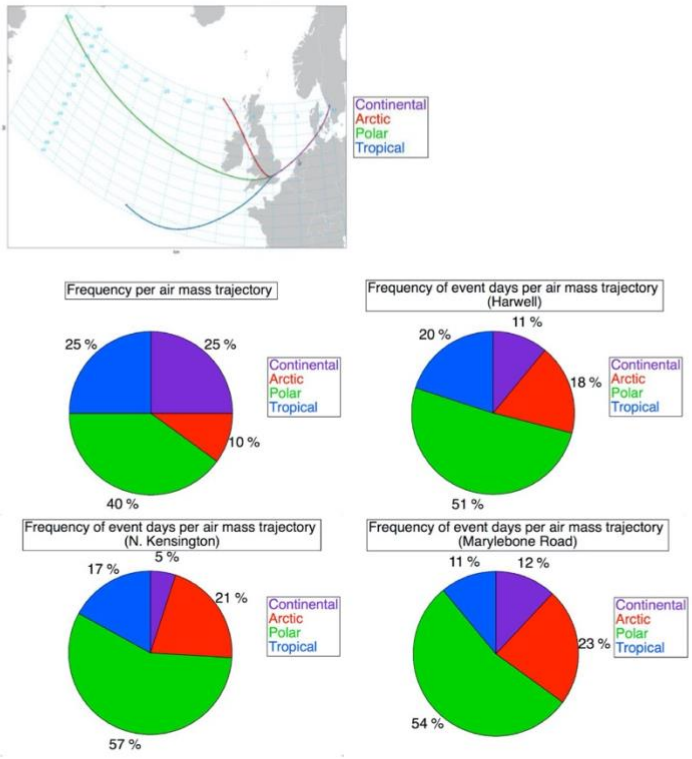

Figure 5. Map and frequency of incoming air mass origin – average and for NPF events per site.

area (for the background areas) and/or continental Europe. It has higher concentrations of most pollutants as well as the highest condensation sink.

The occurrence of each air mass class for average and event days for Harwell and London (both sites) can also be found in Fig. 5, while their main characteristics for each site can be found in Table S2. Though in this case the air mass grouping for each site was done in a different analysis, the resulting groups are almost identical in their characteristics and frequency, as the sites are close to each other.

The polar cluster is the one prevailing on both average and event days. This consists of clean fast-moving air masses originating mainly from middle and high latitudes of the Atlantic, and this cluster presents favourable conditions for NPF events. The association of NPF events with air masses from the mid-Atlantic at North Kensington was also found by Beddows et al. (2015). Cool Arctic air masses on average are not clean as they may have passed over the northern UK. The event days associated with this air mass type have the lowest concentrations of the pollutants within available data for all areas. The increased percentage of events with this air mass at all sites indicates that lower temperatures, in a clear atmosphere with sufficient solar radiation, are favourable for NPF events as found in previous studies (Napari et al., 2002; Jeong et al., 2010; Kirkby et al., 2011). A similar trend of increased probability with polar and arctic maritime air masses was also found for Hyytiälä, Finland, by Nilsson et al. (2001). Tropical air masses have a lower probability for NPF events, which is associated with the fact that a number of these days are associated with air masses which have passed from continental areas south of the UK (France, Spain etc.). Specifically for Marylebone Road the NPF probability is a lot lower (11 % versus 17 % for North Kensington and 20 % for Harwell). This is due to the fact that these air masses are more related to southerly winds which on Marylebone Road are associated with a street canyon vortex which causes higher pollutant concentrations at this site. Finally, the Continental cluster presents the lowest probability for NPF events. The air masses in this group originate from continen-

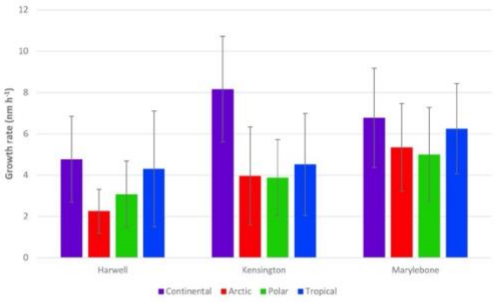

Figure 6. Growth rate per incoming air mass origin at each of the sites.

tal Europe and for the background areas in most cases have passed over the London region as well. This results in both a higher condensation sink and concentration of pollutants, which limits the number of days with favourable conditions for NPF events. The growth rate for all sites though appears to be higher for air masses originating from more polluted areas (Fig. 6), which appear to enhance the growth process due to containing a higher concentration of condensable species (after oxidation).

3.3.2 Variability of the origin of the air masses on NPF events

As both background sites are relatively close to each other (about 80 km) and had a similar number of event days, a combined clustering of back trajectories for the event days (only) in these two areas was attempted. This would provide an insight into the origin of air masses for local and regional events, as well as the conditions for these air masses. The data for local North Kensington events and both local and regional events in Harwell were clustered together and the results along with the characteristics of the air mass clusters are found in Fig. S5.

Cluster C3, which is placed between C2 and C4 among those originating from the Atlantic Ocean, has the highest percentage for both area-specific and regional events. Specifically, for regional events the percentage is over 35 %, much higher compared to all others, showing a clear "preference" of regional events for cleaner and faster-moving air masses from mid-latitudes of the Atlantic Ocean. This "preference" explains the lower production and growth rate of the new particles found for regional events, compared to local ones, as for air masses from this area lower organic carbon and SO_2 concentrations were found at both sites in this study. Cluster C5, originating straight from the north but representing air masses that have crossed the Irish Sea and have not extensively gone over land, presents a similar case. These cold and clean air masses are associated with a low growth rate and consequently low survivability of the newly formed particles. Local events for both sites apart from those in Cluster C3 are highly associated with Clusters C1 and C2. C1, which contains slow and polluted air masses, presents the highest growth rate and as a result high particle survivability, as given by the P parameter (see later). On the other hand, C2, which consists of warm and moist air masses from lower latitudes, is the least common for regional events and presents a high growth rate and survival probability of the particles. Apart from the weak relation found with particulate organic carbon concentrations and growth rate (Fig. S5), there appears to be an inverse relation between the temperature and survivability of the particles. Warmer air masses seem to be related to higher particle survival probability, which may be attributable to greater growth rates as temperature increases (Yli-Juuti et al., 2011).

Table 2. Annual and seasonal NSF for all areas of study.

	Harwell	N. Kensington	Marylebone Road
NSF_{NUC} (spring)	2.04 ± 0.69	2.03 ± 0.51	1.20 ± 0.25
NSF_{NUC} (summer)	2.01 ± 0.85	1.72 ± 0.52	1.26 ± 0.36
NSF_{NUC} (year)	2.25 ± 0.85	1.86 ± 0.56	1.26 ± 0.31
NSF_{GEN} (spring)	1.10 ± 0.64	1.07 ± 0.59	1.02 ± 0.29
NSF_{GEN} (summer)	1.18 ± 0.72	1.11 ± 0.55	1.01 ± 0.25
NSF_{GEN} (year)	1.10 ± 0.61	1.06 ± 0.54	1.02 ± 0.27

3.4 Nucleation strength factor (NSF)

The NSF (Eqs. 6 and 7) is used to describe the effect nucleation events have on the number of particles at a site. The values of NSF for each site and for spring and summer are shown in Table 2. The decrease in the contribution of NPF events to particle number, moving from the rural area to the kerbside was also found in previous studies (Salma et al., 2014, 2017). This is explained by the increased contribution to the particle number concentrations of other sources, mainly combustion in the urban environment, compared to rural areas. Apart from this trend, in the background areas the increase in $N_{16–100}$ was greater in spring than summer. This effect seems stronger in the urban background area compared to the rural, as in that area the variability of $N_{16–100}$ is greater for event days compared to that of the rural area. On the other hand, the contribution of NPF events in the longer term, as is illustrated by the NSF_{GEN}, appears to favour summer for all areas, showing the increased formation and survivability of particles in this season.

For Marylebone Road the result for the increase in the $N_{16–100}$ is greater in summer than in spring, in contrast to what was found for the background sites. This is due to the fact that in summer the traffic intensity is decreased, giving the contribution from NPF events a stronger effect compared to the other sources. The very small increase found in NPF

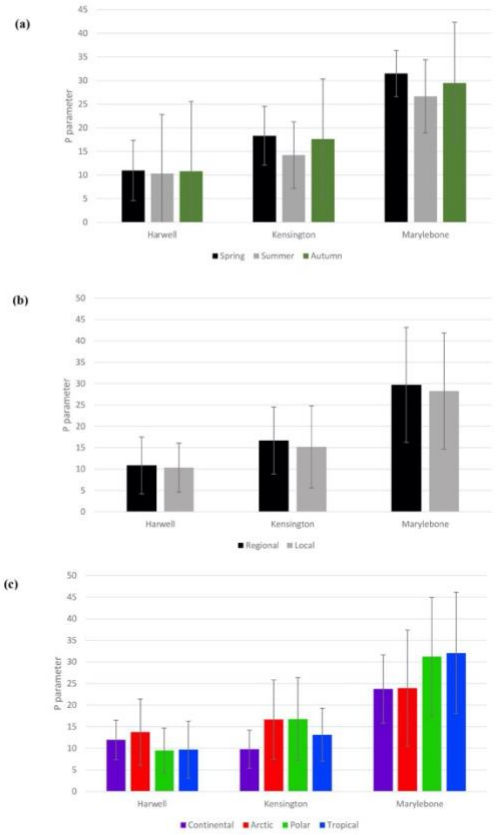

Figure 7. Survival parameter P **(a)** per season, **(b)** for regional and local events (for Marylebone Road regional is for all three sites), and **(c)** by incoming air mass origin.

events on Marylebone Road, with a factor of just 1.26, a lot lower than that found in the urban area of Seoul, South Korea (Park et al., 2015), is indicative of the reduced effect of NPF events in an area which is heavily affected by traffic, as also pointed out by Von Bismarck-Osten et al. (2013) in their study on particle composition on Marylebone Road.

3.5 The survival parameter P

The average values of the P parameter for each of the areas of this study are 10.5 for Harwell, 15.8 for North Kensington and 28.9 for Marylebone Road. The values found place Marylebone Road at the upper end of heavily polluted areas in Europe and North Kensington at the same level as many other urban areas in Europe, while Harwell somehow had higher values compared to other rural background areas in Europe, as calculated by Kulmala et al. (2017). The seasonal, air mass origin and local-versus-regional variations can be found in Fig. 7 (winter is excluded due to very low number of events). While the increasing trend of the P parameter as we move from rural background to kerbside was expected, it can be seen that there is a clear seasonal pattern in all three areas, with summer having the lowest P parameter (greatest survivability) compared to the other two seasons. This is associated with the higher growth rate found in summer for all areas of this study, as the differences in the condensation sink on event days are negligible between seasons. The case is similar for regional and local events. The result per air mass origin is related to the different conditions and parameters of each incoming air mass in each area. For example, the higher P parameter for tropical air masses at Marylebone Road is associated with the higher condensation sink found for this kind of air mass, due to the street canyon effect which is specific for Marylebone Road for southerly wind directions with which these air masses are mainly related, while the higher values for the rather clean Arctic air masses for the other two areas are associated with the lower growth rates found for this kind of air mass in these areas. The more polluted continental air masses seem to have a different effect for rural and urban areas. Their higher condensation sinks and concentrations of pollutants have a negative effect on P values for the rural site and a positive effect at the urban sites. The exact opposite is found for the cleaner air masses of the Polar cluster, which appear to result in reduced P values of the newly formed particles at the urban sites. This is related to the lower condensation sink associated with this air mass type.

4 Conclusions

A total of 7 years of particle size distributions in the range 16.6–604 nm and other meteorological and chemical composition data from three distinct areas (regional background, urban background, kerbside) in the southern UK were analysed and the conditions associated with NPF events were studied. NPF events were found to occur on about 7 % of days at background sites and less at the kerbside site. The conditions on event days for all three areas were similar, with clear atmospheric conditions and a lower condensation sink. While the condensation sink appears to be the most important factor limiting NPF events at the kerbside site, SO_2 was found to have smaller concentrations on event days for all areas, which indicates that either on average it is in sufficient concentrations for NPF events to occur, or that other variables that participate in the production mechanism of H_2SO_4 are more important. The growth rate of the newly formed particles increases from the rural site to the kerbside and is greater in summer compared to other seasons for all three sites. Almost half of the NPF events at the rural and urban background sites were found to happen simultaneously. In

these cases, the atmospheric conditions were cleaner, which resulted in slower growth rates. While most of the chemical species available were at lower concentrations in regional events, a difference in the behaviour with respect to sulfate and organic compounds was found between the two background site types.

The prevailing origin of air masses in the southern UK is from middle and high latitudes of the Atlantic Ocean. These fast-moving air masses present an increased probability for NPF to occur. The case is similar for the cooler and cleaner arctic air masses, while air masses from the tropics and continental Europe, having greater pollutant content, have decreased NPF probability, but a higher growth rate of particles when NPF events occurred. Regional events appear to be more associated with cleaner air masses, presenting a smaller growth rate and condensation sink compared to local events. The difference in growth rate is probably related to the greater content of condensable species; a positive relation of particle survival probability with temperature was also found.

Comparing the background areas in this study, particles of 16–20 nm were found to be about 20 % greater in concentration (above long-term average) on NPF event days at the urban background site compared with the rural site. This is associated with a higher abundance of condensable species in the urban environment, which enhances the nucleation and growth process. This effect though is limited as particle size increases and NPF events have a greater effect on the overall $N_{<100\,\mathrm{nm}}$ in the rural areas, compared to urban areas, as calculated by the NSF. The effect becomes even smaller at the kerbside as the number of background particles emitted by traffic is a lot greater.

The occurrence of NPF events at the highly polluted Marylebone Road site is at first sight surprising given the elevated condensation sink. This must be counteracted by an abundance of condensable material, which is surprising given the generally modest rate of atmospheric oxidation processes in comparison to residence times in a street canyon (Harrison, 2017). However, Giorio et al. (2015), using aerosol time-of-flight mass spectrometry, reported rapid chemical processes within the Marylebone Road street canyon leading to production of secondary particulate matter from road traffic emissions. They postulated that this resulted from very local gas-to-particle conversion from vehicle-emitted pollutants. Condensation of such reaction products upon pre-existing particles could explain the enhanced particle growth rates observed at Marylebone Road (Fig. 3).

Finally, particle survival probability was found to decrease moving from rural to urban areas. While formation and initial growth of new particles is increased in urban areas, their survivability reduces as their size increases. The probability of particles to survive to greater sizes was found to be increased in summer for all areas, which is also explained by the higher growth rate. The probability is also different depending upon the origin of the air masses and is related to conditions specific for each area.

In the present work, the effects of atmospheric conditions upon the NPF process are studied. NPF is a complex process, highly affected by meteorological conditions (local and synoptic), the chemical composition and the pre-existing conditions in an area. For this reason, the study of NPF events in one area cannot provide safe assumptions for other areas, as the mixture of conditions found in different places is unique and alters the occurrence and development of NPF events. Thus, more studies on the conditions and the trends in NPF events should be conducted to better understand the effect of the numerous variables that affect those processes.

Data availability. Data supporting this publication are openly available from the UBIRA eData repository at https://doi.org/10.25500/edata.bham.00000307 (Bousiotis and Harrison, 2019).

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Competing interests. The authors declare that they have no conflict of interest.

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3.2 A Phenomenology of New Particle Formation (NPF) at Thirteen European Sites

(The Effect of Land Use and Local Conditions in Conjunction with Synoptic Conditions in the Occurrence and Characteristics of New Particle Formation (NPF) at Several European Sites)

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A Phenomenology of New Particle Formation (NPF) at Thirteen European Sites

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ABSTRACT

New particle formation (NPF) events occur almost everywhere in the world and can play an important role as a particle source. The frequency and characteristics of NPF events vary spatially and this variability is yet to be fully understood. In the present study, long term particle size distribution datasets (minimum of three years) from thirteen sites of various land uses and climates from across Europe were studied, and NPF events, deriving from secondary formation and not traffic related nucleation, were extracted and analysed. The frequency of NPF events was consistently found to be higher at rural background sites compared to roadside sites, while the growth and formation rates particles were higher at roadside sites, underlining the importance of the abundance of condensable compounds of anthropogenic origin found there. The growth rate was higher in summer at all rural background sites studied. The urban background sites presented the highest uncertainty due to greater variability compared to the other two types of site. The origin of incoming air masses and the specific conditions associated with them greatly affect the characteristics of NPF events. In general, cleaner air masses present higher probability for NPF events, while the more polluted ones show higher growth rates. However, different patterns of NPF events were found even at sites in close proximity (< 200 km) due to the different local conditions at each site. Region-wide events were also studied and were found to be associated with the same conditions as local events, although some variability

was found which was associated with the different seasonality of the events at two neighbouring sites. NPF events were responsible for an increase in the number concentration of ultrafine particles of more than 400% at rural background sites on the day of their occurrence. The degree of enhancement was less at urban sites due to the increased contribution of other sources within the urban environment. It is evident that, while some variables (such as solar radiation intensity, relative humidity or the concentrations of specific pollutants) appear to have a similar influence on NPF events across all sites, it is impossible to predict the characteristics of NPF events at a site using just these variables, due to the crucial role of local conditions.

Keywords: Nucleation; New Particle Formation; Ultrafine Particles; Roadside; Urban Background; Rural

1. INTRODUCTION

Ultrafine particles (particles with diameter smaller than 100 nm), while not yet regulated, are believed to have adverse effects upon air quality and public health (Atkinson et al., 2010; Politis et al., 2008; Tobías et al., 2018), as well as having a direct or indirect effect on atmospheric properties (Makkonen et al., 2012; Seinfeld and Pandis, 2012). The source of ultrafine particles can either be from primary emissions (Harrison et al., 2000; Masiol et al., 2017), including delayed primary emissions (Hietikko et al., 2018; Olin et al., 2020; Rönkkö et al., 2017), or from secondary formation from gaseous precursors (Brean et al., 2019; Chu et al., 2019; Kerminen et al., 2018; Kulmala et al., 2004a; Yao et al., 2018), which is considered as an important source of CCN in the atmosphere (Dameto de España et al., 2017; Kalivitis et al., 2015; Spracklen et al., 2008). For the latter, while the process of formation of initial clusters that subsequently lead to particle formation has been extensively studied (Dal Maso et al., 2002; Kulmala et al., 2014; Riipinen et al., 2007; Weber et al., 1998), there is no consistent explanation of the factors which determine the occurrence and development of NPF events in the atmosphere. Additionally, events that resemble NPF, with the initial particles deriving from primary emissions, especially close to traffic sources (Rönkkö et al., 2017), have been also reported but these are out of the scope of the present study.

A large number of studies both in laboratories and in real world conditions have been conducted to either describe or explain the mechanisms that drive NPF events. The role of meteorological conditions, such as solar radiation intensity (Kumar et al., 2014; Shi et al., 2001; Stanier et al., 2004) and relative humidity (Li et al., 2019; Park et al., 2015), are well documented, while great diversity was found for the effect of other meteorological factors such as the wind speed (Charron et al., 2008; Németh and Salma, 2014; Rimnácová et al., 2011) or temperature (Jeong et al., 2010; Napari et al., 2002). There are also influences of atmospheric composition, with the positive role of low condensation sink and concentrations of pollutants such as NO_x upon NPF event occurrence being widely agreed upon (Alam et al., 2003; Cheung et al., 2013; Kerminen et al., 2004; Wang et al., 2014; Wehner et al., 2007). Contrary to that, while the indirect role of SO_2 is well established in the nucleation process, via the formation of new clusters of H_2SO_4 molecules (Boy et al., 2005; Iida et al., 2008; Kulmala et al., 2005; Sipila et al., 2010; Xiao et al., 2015), uncertainty exists in the role that different concentrations of SO_2 play in the occurrence of NPF events in real world atmospheric conditions (Alam et al., 2003; Dall'Osto et al., 2018; Wonaschütz et al., 2015; Woo et al., 2001). Ammonia is known to enhance the formation of initial clusters (Korhonen et al., 1999; Ortega et al., 2008; Schobesberger et al., 2015), and volatile organic compounds are regarded as the main drivers of the growth of the newly formed particles (Kulmala et al., 2013; Riccobono et al., 2014; Tröstl et al., 2016). NPF

events in different locations do not appear to follow consistent trends with the concentrations of these compounds and meteorological parameters (McFiggans et al., 2019; Minguillón et al., 2015; Riipinen et al., 2007), though links between NPF events and sulphuric acid vapour concentrations (Petäjä et al., 2009; Weber et al., 1995) and organics (Bianchi et al., 2019; Ehn et al., 2014) have been reported.

It is evident that NPF events and their development are complex, and local conditions play an important role in their variability. Many studies have attempted to explain this variability by analyzing multiple datasets from wider areas. Studies in the UK (Bousiotis et al., 2019; Hama et al., 2017), Spain (Brines et al., 2014; Carnerero et al., 2018; Dall'Osto et al., 2013; Minguillón et al., 2015), Hungary (Németh and Salma, 2014; Salma et al., 2014, 2016), Greece (Kalkavouras et al., 2017; Siakavaras et al., 2016), Germany (Costabile et al., 2009; Ma and Birmili, 2015, Sun et al., 2019) and China (Peng et al., 2017; Shen et al., 2018; Wang et al., 2017) have attempted to explain the differences found in NPF event conditions and variability between different sites in close proximity, while larger scale studies using descriptive (Brines et al., 2015; Hofman et al., 2016; Jaatinen et al., 2009; Kulmala et al., 2005) or statistical methods (Dall'Osto et al., 2018; Rivas et al., 2020) have provided insights into the effect of the variability of parameters that are considered to play an important role in the occurrence and development of NPF events on a broader scale.

The present study, combining thirteen long term datasets (minimum of three years) from five different countries across Europe and combined with the results from a previous study in the UK (Bousiotis, 2019), attempts to elucidate the effect of the local conditions on NPF event characteristics (frequency of NPF events, formation rate and growth rate) both for sites in close proximity (< 200 km), and by intercomparison of sites on a continental scale in order to find general trends of the variables that affect the characteristics and development of NPF events on a larger scale. Finally, the effect of NPF events upon the ultrafine particle number concentrations was calculated, providing insight to the potential of NPF events to influence the local air quality conditions in all areas studied.

2. DATA AND METHODS

2.1 Site Description and Data Availability

In the present study, particle number size distribution data from 13 sites in Europe (Figure 4) are analysed in the size range 3 nm < D_p < 1000 nm. A detailed list of the site locations and the data availability for each is found in Table 1. Average meteorological conditions and concentrations of chemical compounds for all sites are found in Tables S1 and S2 respectively; their seasonal variation is found in Table S3.

Figure 4: Map of the areas of study (Paper 2).

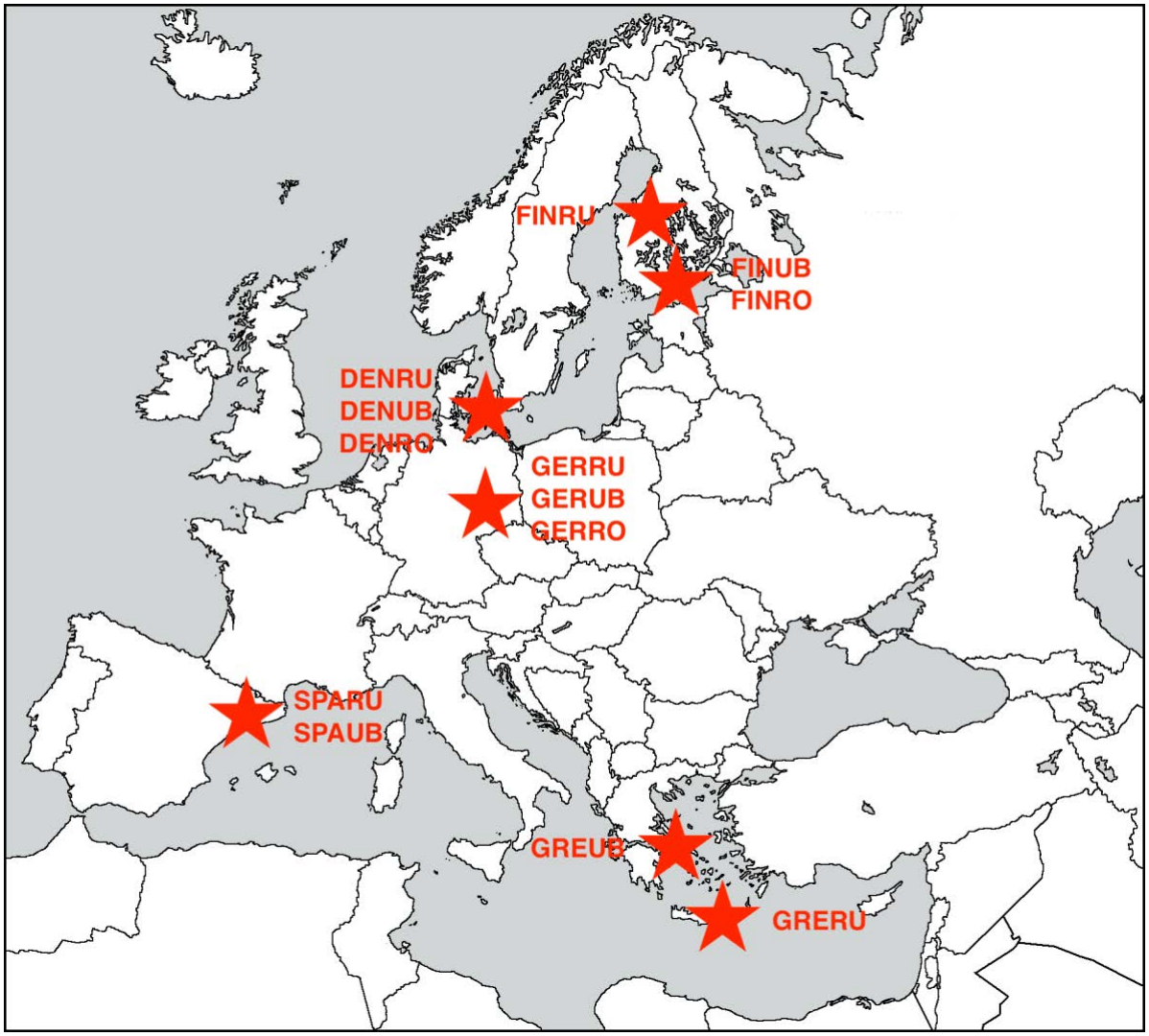


Table 1: Location and data availability of the sites in Paper 2 (RU denotes rural site, UB is urban background and RO is roadside).

Site	Location	Available data	Meteorological data location	Data availability	Reference
DENRU	Lille Valby, 25 km W of Copenhagen, (55° 41′ 41″ N; 12° 7′ 7″ E) (2008 – 6/2010) Risø, 7 km north of Lille Valby, (55° 38' 40" N; 12° 5' 19" E) (7/2010 – 2017)	DMPS and CPC (5.8 - 700 nm, 89.3% availability), NO, NO_x, SO_2, O_3, minerals, OC, EC, NO_3^-, SO_4^{2-}, NH_4^+	Ørsted – Institute station	2008 – 2017	Ketzel et al., 2004
DENUB	Ørsted - Institude, 2 km NE of the city centre, Copenhagen, Denmark (55° 42′ 1″ N; 12° 33′ 41″ E)	DMPS and CPC (5.8 - 700 nm, 61.4% availability), NO, NO_x, O_3, minerals, EC	On site	2008 – 2017	Wang et al., 2010
DENRO	H.C. Andersens Boulevard, Copenhagen, Denmark (55° 40′ 28″ N; 12° 34′ 16″ E)	DMPS and CPC (5.8 - 700 nm, 65.7% availability), NO, NO_x, SO_2, O_3, minerals, OC, EC, NO_3^-, SO_4^{2-}, NH_4^+	Ørsted – Institute station	2008 – 2017	Wang et al., 2010
GERRU	Melpitz, 40 km NE of Leipzig, Germany (51° 31′ 31.85″ N; 12° 26′ 40.30″ E)	TDMPS with CPC (4.8 - 800 nm, 90.4% availability), OC, NO_3^-, SO_4^{2-}, NH_4^+, Cl^-	On site	2008 – 2011	Birmili et al., 2016
GERUB	Tropos, 3 km NE from the city centre of Leipzig, Germany (51° 21′ 9.1″ N; 12° 26′ 5.1″ E)	TDMPS with CPC (3 - 800 nm, 68.3% availability)	On site	2008 – 2011	Birmili et al., 2016
GERRO	Eisenbahnstraße, Leipzig, Germany (51° 20′ 43.80″ N; 12° 24′ 28.35″ E)	TDMPS with CPC (4 - 800 nm, 65.1% availability)	Tropos station	2008 – 2011	Birmili et al., 2016
FINRU	Hyytiälä, 250 km N of Helsinki, Finland (61° 50′ 50.70″ N; 24° 17′ 41.20″ E)	TDMPS with CPC (3 – 1000 nm, 98.2% availability), NO, NO_x, SO_2, O_3, CO, CH_4, VOCs, H_2SO_4	On site	2008 – 2011 & 2015 – 2018	Aalto et al., 2001
FINUB	Kumpula Campus 4 km N of the city centre, Helsinki, Finland (60° 12′ 10.52″ N; 24° 57′ 40.20″ E)	TDMPS with CPC (3.4 - 1000 nm, 99.7% availability)	On site	2008 – 2011 & 2015 – 2018	Järvi et al., 2009
FINRO	Mäkelänkatu street, Helsinki, Finland (60° 11′ 47.57″ N; 24° 57′ 6.01″ E)	DMPS (6 - 800 nm, 90.0% availability), NO, NO_2, NO_x, O_3, BC and SO_2 from Kalio Station	Pasila station and on site	2015 – 2018	Hietikko et al., 2018
SPARU	Montseny, 50 km NNE from Barcelona, Spain (41° 46′ 45″ N; 2° 21′ 29″ E)	SMPS (9 – 856 nm, 53.7% availability), NO, NO_2, SO_2, O_3, CO, OM, SO_4^{2-}	On site	2012 - 2015	Dall'Osto et al., 2013
SPAUB	Palau Reial, Barcelona, Spain (41° 23′ 14″ N; 2° 6′ 56″ E)	SMPS (10.9 – 478 nm, 67.9% availability), NO, NO_2, SO_2, O_3, CO, BC, OM, SO_4^{2-}, $PM_{2.5}$, PM_{10}	On site	2012 – 2015	Dall'Osto et al., 2012
GRERU	Finokalia, 70 km E of Heraklion, Greece (35° 20′ 16.8″ N; 25° 40′ 8.4″ E)	SMPS (8.77 - 849 nm, 85.0% availability), NO, NO_2, O_3, OC, EC	On site	2012 – 2018	Kalkavouras et al., 2017
GREUB	"Demokritos", 12 km NE from the city centre, Athens, Greece (37° 59′ 41.96″ N; 23° 48′ 57.56″ E)	SMPS (10 – 550 nm, 88.0% availability)	On site	2015 – 2018	Vassilakos et al., 2005

2.2 Methods

2.2.1 NPF event selection

The identification of NPF events was conducted manually using the criteria set by Dal Maso et al. (2005). According to these, a NPF event is considered to occur when:

- a distinctly new mode of particles appears in the nucleation range,
- this new mode prevails for some hours,
- the new mode shows signs of growth.

The NPF events extracted using this method are then classified into classes I or II depending on the level of confidence. Class I (high confidence) is further classified as Ia and Ib, with class Ia containing the events that both present a clear formation of a new mode as well as a distinct growth of this mode, while Ib includes those with a less distinct formation and development. In the present study, only the events classified as Ia were used as they are considered as more suitable for study. As the growth criterion is not fully defined, in the present study a minimum growth rate of 1 nm h^{-1} is required for NPF events to be considered. The events found using this method should not be confused with the formation and growth of particles deriving from primary emissions next to pollution sources, such as traffic. While to an extent the particle formation found can be biased by primary emissions (especially at roadside sites), great effort was made using additional

data, such as atmospheric composition data, to not include any incidents of traffic related nucleation.

2.2.2 Calculation of condensation sink, growth rate, formation rate, Nucleation Strength Factor (NSF) and NPF event probability

The calculation of the condensation sink was made using the method proposed by Kulmala et al. (2001). The condensation sink (CS) is calculated as:

$$\mathrm{CS} = 4\pi \mathrm{D}_{vap} \sum \beta_{\mathrm{M}}\ \mathrm{r}\ \mathrm{N}$$

where r and N are the radius and the number concentration of the particles and D_{vap} is the diffusion coefficient, calculated for T = 293 K and P = 1013.25 mbar, according to Poling et al. (2001):

$$\mathrm{D_{vap}} = 0.00143 \cdot \mathrm{T}^{1.75} \frac{\sqrt{\mathrm{M_{air}^{-1}} + \mathrm{M_{vap}^{-1}}}}{\mathrm{P}\left(\mathrm{D_{x,air}^{\frac{1}{3}}} + \mathrm{D_{x,vap}^{\frac{1}{3}}}\right)^2}$$

where M and D_x are the molar mass and diffusion volume for air and H_2SO_4. β_M is the Fuchs correction factor calculated from Fuchs and Sutugin (1971):

$$\beta_M = \frac{1 + K_n}{1 + \left(\frac{4}{3a} + 0.377\right) K_n + \frac{4}{3a} {K_n}^2}$$

K_n is the Knudsen number, defined as $Kn = 2\lambda_m/d_p$, with λ_m being the mean free path of the gas.

The growth rate of the newly formed particles is calculated according to Kulmala et al. (2012), as

$$GR = \frac{d_{P_2} - d_{P_1}}{t_2 - t_1}$$

for the size range between the minimum available particle diameter up to 30 nm. For the calculation of the growth rate, the time considered was from the start of the event until a) growth stopped, b) GMD reached the upper limit set or c) the day ended.

The formation rate J was calculated using the method proposed by Kulmala et al. (2012) in which:

$$J_{d_p} = \frac{dN_{d_p}}{dt} + CoagS_{d_p} \times N_{d_p} + \frac{GR}{\Delta d_p} \times N_{d_p} + S_{losses}$$

where $CoagS_{dp}$ is the coagulation rate of particles of diameter d_p, calculated by:

$$CoagS_{d_p} = \int K(d_p, d'_p)\, n(d'_p) dd'_p \cong \sum_{d'_p = d_p}^{d'_p = max} K(d_p, d'_p)\, N_{d_p}$$

as proposed by Kerminen et al. (2001). $K(d_p, d'_p)$ is the coagulation coefficient of particle sizes d_p and d'_p. S_{losses} accounts for the additional loss terms (i.e. chamber walls), not considered here. Initial particle formation starts at about 1.5 ± 0.4 nm (Kulmala et al., 2012). The formation rate calculated here refers to particles in the atmosphere that reached the diameter of 10 nm during NPF events for uniformity reasons. This means that these particles were formed earlier during the day of the events, survived and grew to this size later in the day. Furthermore, due to the effect of the morning rush which biased the results at roadsides, the averages are calculated for the time window between 9:00 to

15:00 ($\pm$ 3 hours from noon, when J_{10} peaked in the majority of the events). This was done for all the sites in this study for consistency.

The Nucleation Strength Factor (NSF) proposed by Nemeth and Salma (2014) is a measure of the effect of NPF events on ultrafine particle concentration. It can either refer to the effect of NPF events on the day of their occurrence, calculated by:

$$\mathrm{NSF_{NUC}} = \frac{\left(\frac{\mathrm{N_{smallest\ size\ available-100nm}}}{\mathrm{N_{100nm\ -largest\ size\ available}}}\right)_{\mathrm{nucleation\ days}}}{\left(\frac{\mathrm{N_{smallest\ size\ available-100nm}}}{\mathrm{N_{100nm-largest\ size\ available}}}\right)_{\mathrm{non-nucleation\ days}}}$$

or their overall contribution on the ultrafine particle concentrations at a site calculated by:

$$\mathrm{NSF_{GEN}} = \frac{\left(\frac{\mathrm{N_{smallest\ size\ available-100nm}}}{\mathrm{N_{100nm-largest\ size\ available}}}\right)_{\mathrm{all\ days}}}{\left(\frac{\mathrm{N_{smallest\ size\ available-100nm}}}{\mathrm{N_{100nm-largest\ size\ available}}}\right)_{\mathrm{non-nucleation\ days}}}$$

The NPF event probability is a simple metric of the probability of NPF events calculated by the number of NPF event days divided by the number of days with available data for the given group (temporal, wind direction etc.). It should be mentioned that all the results

presented are normalised according the seasonal data availability for each site, based upon the expression:

$$\mathrm{NPF}_{\mathrm{probability}} = \frac{\mathrm{N}_{\mathrm{NPF\ event\ days\ for\ group\ of\ days\ X}}}{\mathrm{N}_{\mathrm{days\ with\ available\ data\ for\ group\ of\ days\ X}}}$$

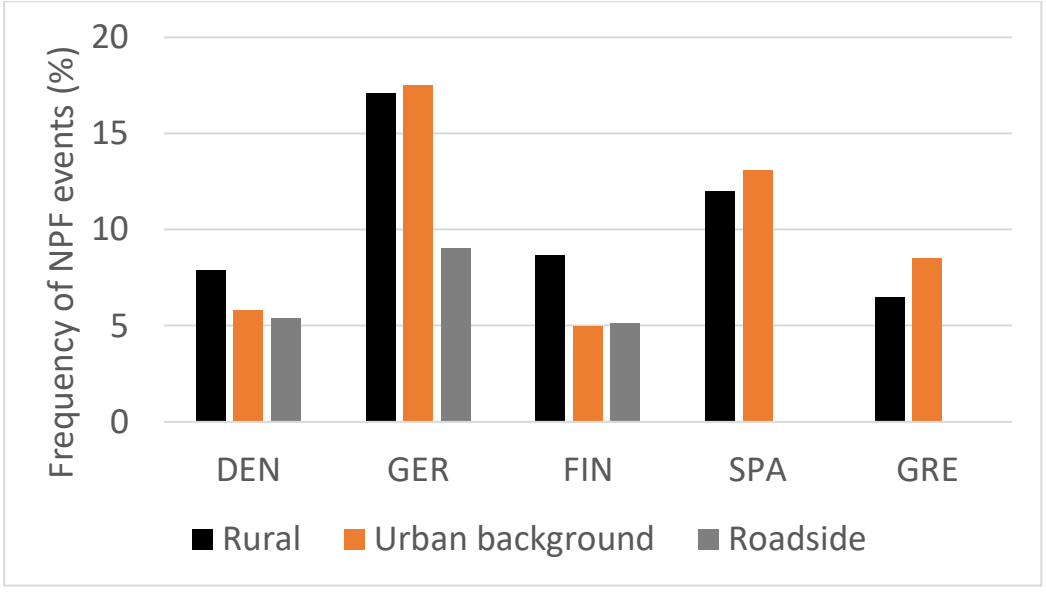
Finally, all the differences found between variable averages were tested for statistical significance using the one-way ANOVA test. Those that were found to be statistically significant ($p < 0.05$) are reported along with their p-value.

3. RESULTS

3.1 Sites in Denmark

NPF events occurred at all three sites with available data with a similar frequency for the urban sites (5.4% for DENRO and 5.8% for DENUB) and higher for the rural DENRU site (7.9%). For the DENRO and DENRU sites the seasonal variation favoured summer, while at DENUB a higher frequency of events was found for spring (Figure 5). The growth rate was found to be higher at the DENRO site at 4.45$\pm$1.87 nm h^{-1} and it was similar for the other two sites (3.19$\pm$1.43 for DENRU and 3.19$\pm$1.45 for DENUB) nm h^{-1} (Figure 6), though the peak was found in different seasons (Figure 7), coinciding with that of the frequency of NPF events (the highest average for DENRO was found for winter but it was only for a single

Figure 5: Frequency (top panel) and seasonal (lower panel) variation of New Particle Formation events (Winter – DJF; Spring – MAM; Summer – JJA; Autumn – SON). For site naming first three letters refer to the country (DEN = Denmark, GER = Germany, FIN = Finland, SPA = Spain, GRE = Greece) while next two to the type of the site (RU = Rural background, UB = Urban background, RO = Roadside)



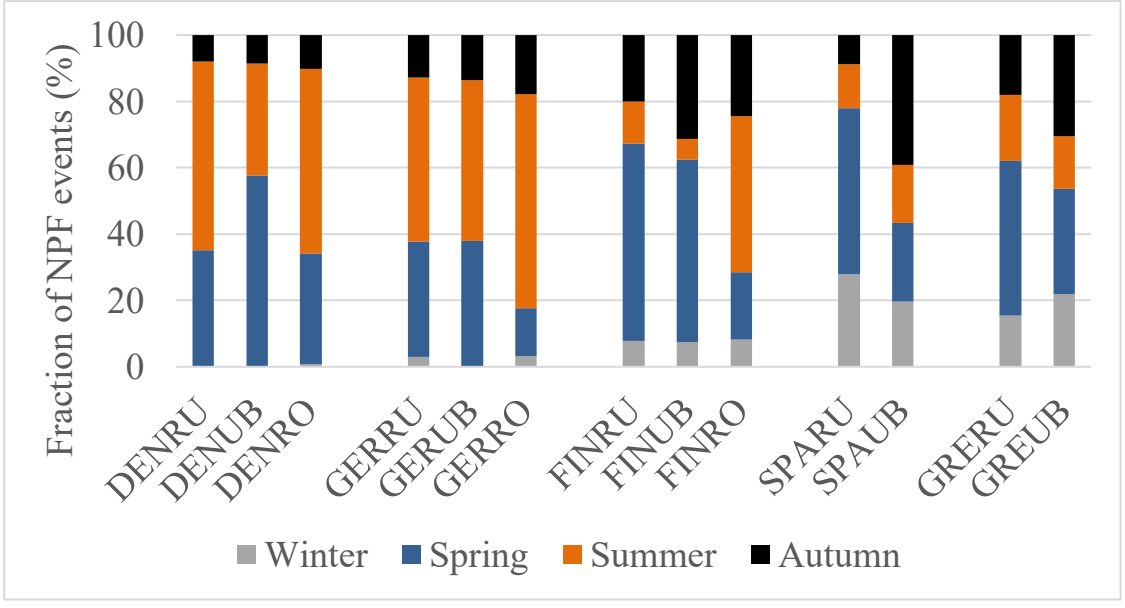


Figure 6: Growth rate of particles up to 30 nm (with standard deviations) during New Particle Formation events at all sites.

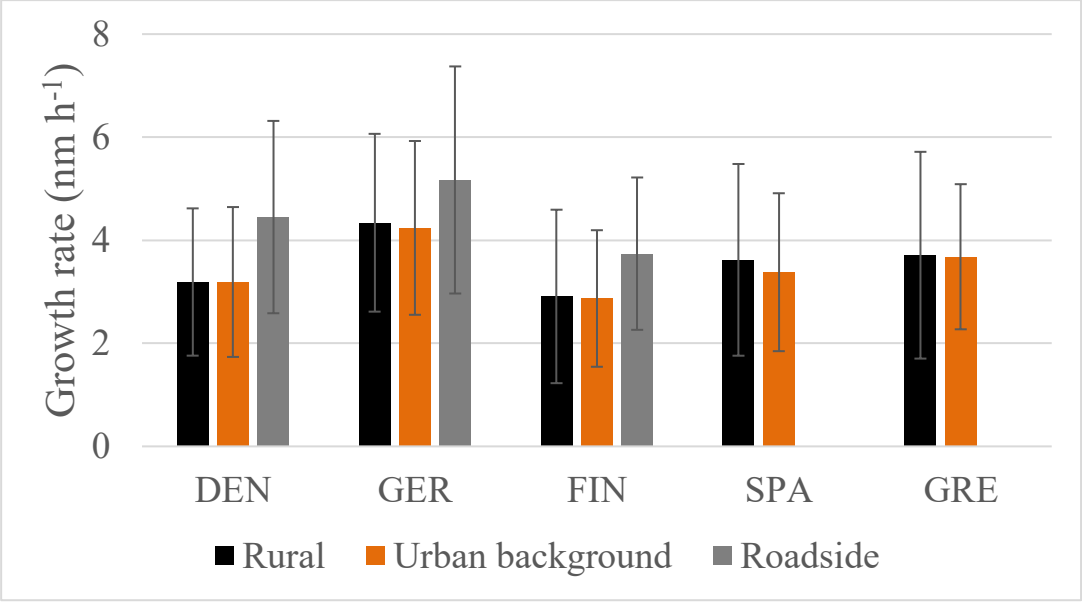


Figure 7: Seasonal variation of growth rate (with standard deviations) on NPF at all sites (top to bottom is for rural, urban background and roadside sites).

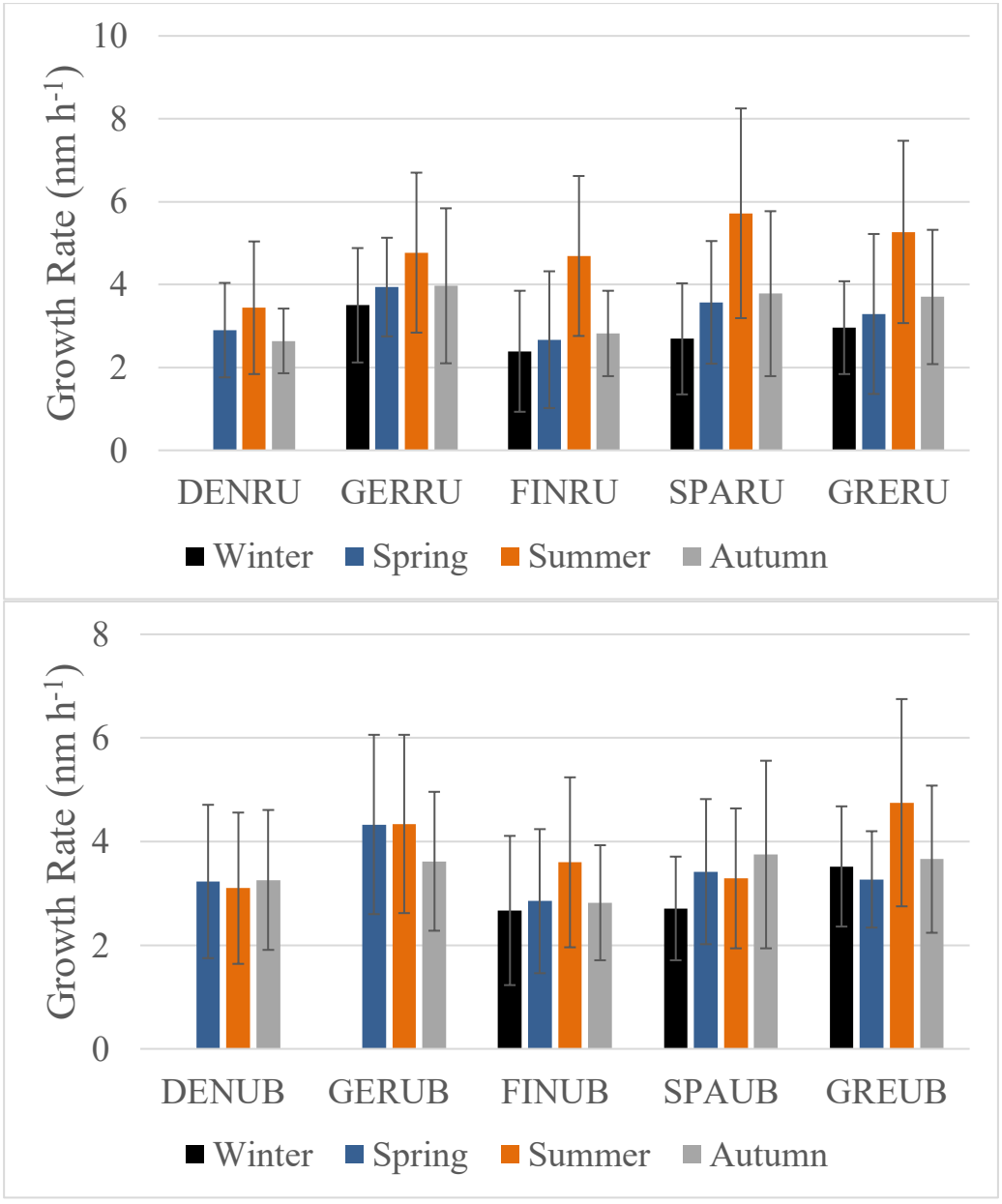
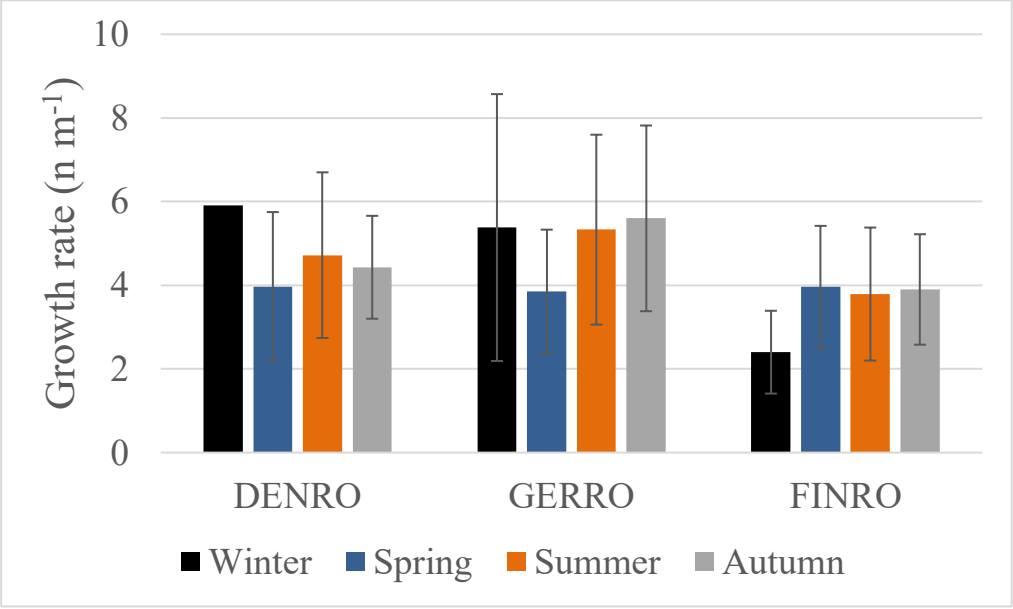


Figure 7 (continued)



event that occurred in that season). As for the within-week variation of the events, there is an increasing probability of NPF events to occur on weekends than weekdays going from the rural background site to the roadside site (Figure 8). Interesting (and probably coincidental) is the increased frequency of NPF events found at all sites on Thursday among the weekdays. J_{10} was found to be broadly similar at the rural and urban background sites and higher at DENRO (Figure 9), favoured by different seasons at each site (summer at DENRU, spring at DENUB though with minimum differences and autumn at DENRO) (Figure 10).

Figure 8: Ratio of New Particle Formation event probability between weekends to weekdays. The greater the ratio the more probable it is for an event to take place during weekends compared to weekdays.

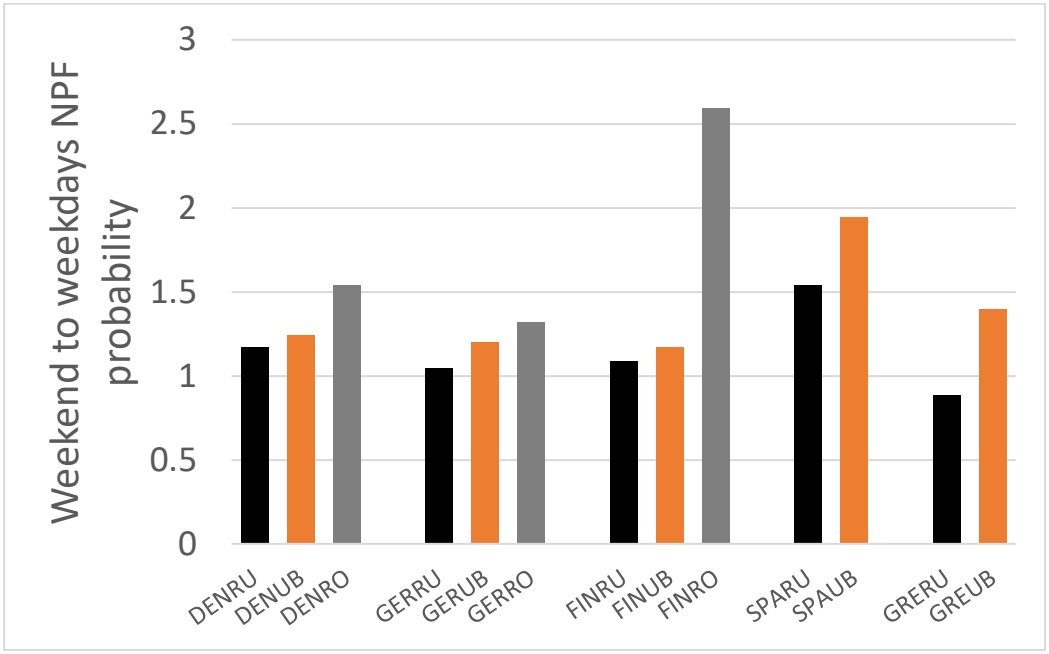


Figure 9: Formation rate of 10 nm particles (J_{10}) (with standard deviations) during New Particle Formation events at all sites.

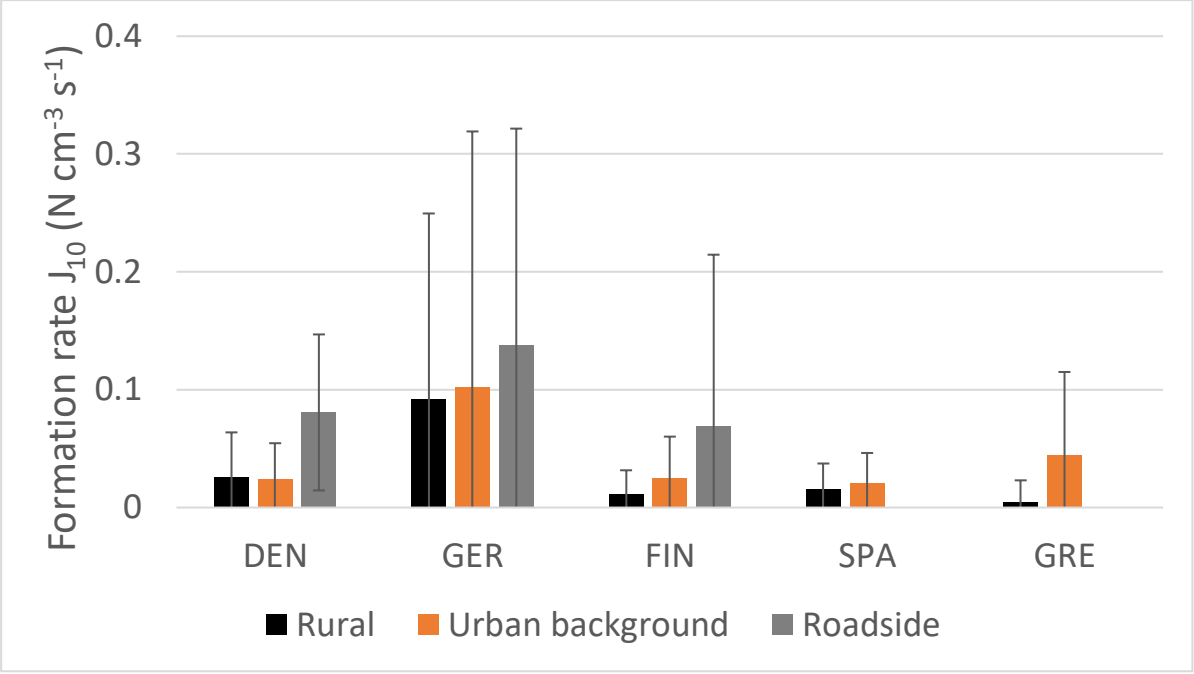


Figure 10: Seasonal variation of formation rate (J_{10}) (with standard deviations) during NPF at all sites (top to bottom is for rural, urban background and roadside sites).

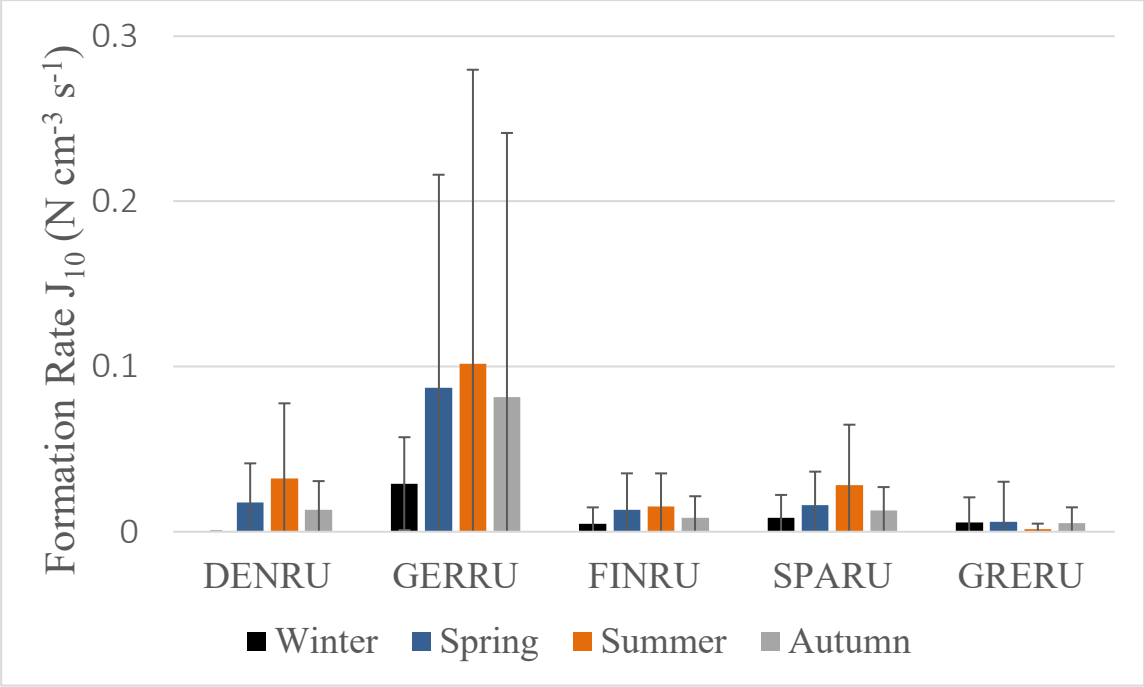
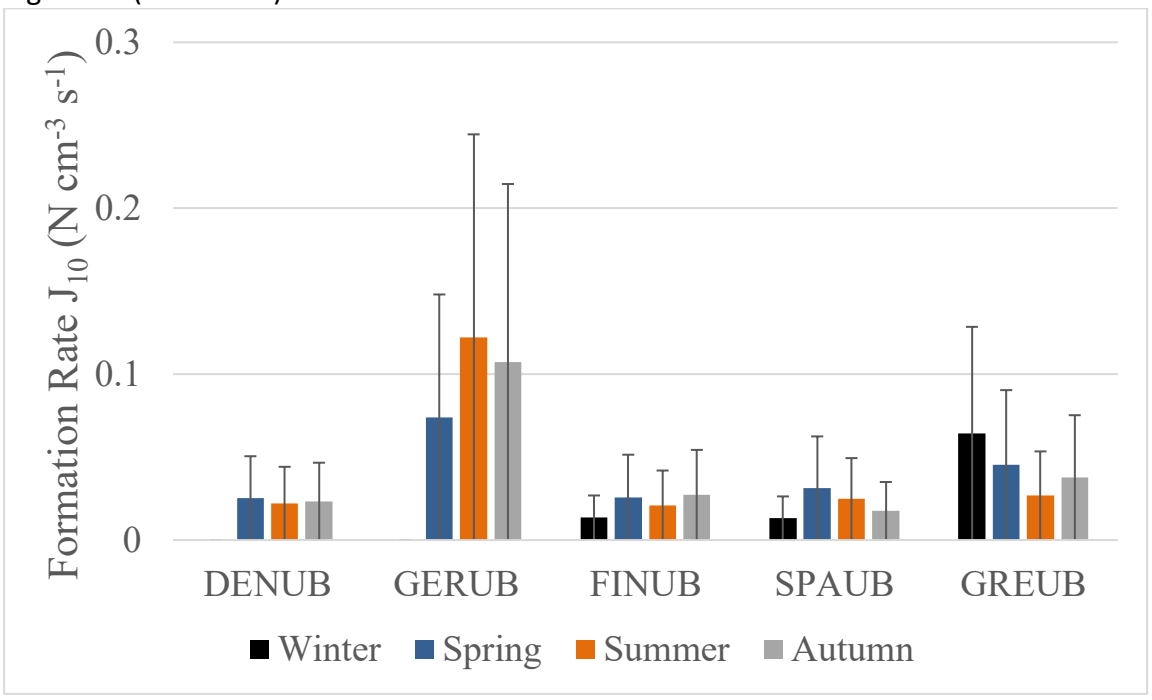
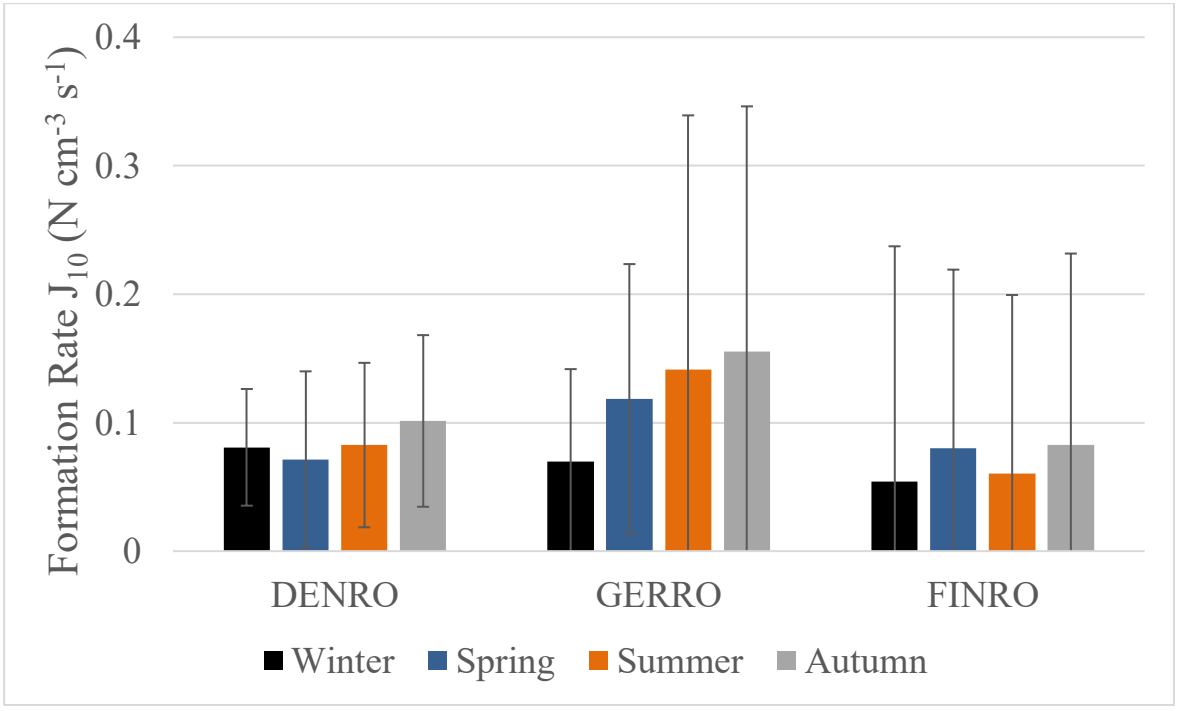


Figure 10 (continued)





In general, pollutant concentrations were found to be lower on event days for all sites (apart from O_3), including the secondary pollutants and minerals (apart from marine related ions like Na^+, Cl^- and Mg^{2+} – data not included) where data was available (Table S2). Among the compounds with lower concentrations on NPF event days was SO_2 (for the sites with available data), possibly due to being in sufficient concentrations for not being a limiting factor in the occurrence of NPF events, while higher concentrations are associated with increased pollution conditions which may suppress the occurrence of the events.

The meteorological conditions that prevailed on NPF event days (Table S1) were higher incoming solar radiation, wind speed and temperature and lower relative humidity compared to average conditions (consistently at all sites and significant for all ($p < 0.001$) except wind speed). As meteorological conditions were available from the urban background site (the variation between the rural and urban sites should not be great since they are about 25 km away from each other), the average conditions for the three sites were almost the same with the only variability being the data availability among the sites. Thus, the more common wind directions in the area are southwesterly; for all sites though the majority of NPF events are associated with direct westerly and northwesterly winds, similar to the findings of Wang et al. (2013) for the same site, which are those with the

lowest concentrations of pollutants and condensation sink for all sites, probably being of marine origin as elemental concentrations showed an increased presence of Na^+, Cl^- and Mg^{2+} (results not included). The wind directions with the highest probability for NPF events present low growth rates and vice versa (Table S4), though it was proposed by Kristensson et al. (2008) that there is a possibility for events observed at the nearby Vavihill site in Sweden with northwesterly winds to be associated with the emissions of specific ship lanes that pass from that area. Wind direction sectors with higher concentrations of OC coincide with higher growth rates at DENRO, while this variability is not found at DENRU, possibly showing that different compounds and mechanisms take part in the growth process of the newly formed particles (Kulmala et al., 2004b).

As mentioned earlier, DENUB although close to the DENRO site has different seasonal variation of NPF events with a marginally lower frequency in summer compared to the other two Danish sites, which have almost the same seasonal variation of NPF events. At DENUB, a strong presence of particles in the size range of about 50 – 60 nm is observed (Figure S1), especially during summer months, increasing the condensation sink in the area (this enhanced mode of particles is visible at DENRO as well, but its effect is dampened due to the elevated particle number concentrations in the other modes). This mode is probably part of the urban particle background. The strongest source of particles though at DENUB

appears to be from the east and consistently appears at both urban sites; this sector is where both elevated pollutant concentrations and condensation sink are found. In this sector, there are two possible local sources, either the port located 2 km to the east or the power plants located at a similar distance (or both). In general, both stations are located only a few kilometres away from the Øresund strait, a major shipping route. Studying the SMPS plots it can be seen that NPF events at DENUB especially in summer tend to start but are either suppressed after the start or have a lifetime of a couple of hours before the new particles are scavenged or evaporate. While this might explain to an extent the frequency and variability of NPF events at this site, the balance between the condensation sink and the concentration of condensable compounds is highlighted. While at DENRO the condensation sink is considerably higher than at DENUB and the effect of the aforementioned mode of particles is present on both, the occurrence and development of NPF events at DENRO are more pronounced in the data due to the higher concentrations of condensable compounds.

3.2 Sites in Germany

A higher frequency of NPF events was found for each type of the sites in Germany compared to the other countries in this study. The background sites had NPF events for more than 17% of the days, while the roadside had a lower frequency of about 9%, with a

seasonal variability favouring summer at all sites (Figure 5). It should be noted though that due to the lack of spring and summer data for the first two years at GERRO, the frequency of events is probably a lot higher and the seasonal variation should further favour these seasons. Similarly, all sites had higher growth rates compared to sites of the same type in other areas of this study, with GERRU showing 4.34$\pm$1.73 nm h^{-1}, GERUB 4.24$\pm$1.69 nm h^{-1} and GERRO 5.17$\pm$2.20 nm h^{-1} (Figure 6). While the difference between GERRU and GERUB is not statistically significant, there is a significant difference for GERRO ($p < 0.005$). Higher growth rates were found in summer compared to spring for all sites (Figure 7). Specifically for the roadside though, the highest average growth rates were found in autumn, which may be either a site-specific feature or an artefact of the limited number of events in that season (total of 11 NPF events in autumn). No substantial within-week variation was found for any of the sites in this country (Figure 8), a feature that is expected mainly at background sites. For GERRO, this may be due to not being as polluted as other sites of the same type, having an average condensation sink comparable to that of urban background sites. The value of J_{10} at the German sites was also the highest among the sites of this study (Figure 9), increasing from the GERRU to GERRO. It was found to be higher in summer for the background sites and in autumn for GERRO (Figure 10).

Compared to the average conditions, a higher temperature and solar radiation were found on NPF event days, while wind speed and relative humidity were lower at all sites (Table S1). The wind sector frequency distribution is different between the urban and the rural sites, with mainly northeasterly and southwesterly winds at the rural site and a more balanced profile for the urban sites. This difference is probably due to differences in the local topography. For the urban sites the majority of NPF events are associated with easterly winds (to a lesser extent westerly as well for GERRO). At GERUB, along with the increased frequency of NPF events the highest average growth rate is also found with easterly wind directions (though the differences are rather small). At GERRO the frequency and growth rate appear to be affected by the topography of the site. Eisenbahnstraße is a road with an axis at almost 90^{o} – 270^{o} and although the H/W ratio (surrounding buildings' height to width ratio) is not high, the effect of a street canyon vortex is observed at the measuring site (Voigtländer et al., 2006). Possibly as a consequence of this, the probability of NPF events is low for direct northerly and southerly winds, although there are high growth rates of the newly formed particles (highest growth rates observed with southerly winds, associated with cleaner air).

At GERRU an increased probability of NPF events and a large growth rate are also found for wind directions from the easterly sector, although these are not very frequent for this site

(Table S4). For this site chemical composition data for $PM_{2.5}$ and PM_{10} are available, and it is found that the generally low (on average) concentrations of pollutants (such as elemental carbon, nitrate and sulphate) in general are elevated for wind directions from that sector. This is also reported for the Melpitz site (GERRU) by Jaatinen et al. (2009) and probably indicates that in a relatively clean area as a rural background site, the presence of low concentrations of pollutants may be favourable in the occurrence and development of NPF events, as in general pollutant concentrations are lower on NPF event days compared to average conditions. Another interesting point is the concentration of organic carbon at the site (average of 2.18 μg m^{-3} in $PM_{2.5}$), having the highest average concentration among the rural background sites studied. As other pollutant concentrations are relatively low at this site, it is possible that a portion of this organic carbon is of biogenic origin, considering also that the area is largely surrounded by forests and green areas, with a minimal effect of marine air masses (as indicated by the low marine component concentrations – data not included) and possibly pointing to increased presence of BVOCs. The increased presence of organic species at GERRU may explain to some extent the increased frequency of NPF events as well as the highest growth and formation rates found among the sites of this study.

3.3 Sites in Finland

NPF events at the sites studied in Finland presented the most diverse seasonal variation, peaking at the background sites in spring and at the roadside in summer (Figure 5). The frequency of NPF events at FINRU was higher (8.66%), while being less at the urban sites (4.97% at FINUB and 5.20% at FINRO). Growth rates were similar at the background sites (2.91±1.68 nm h^{-1} at FINRU and 2.87±1.33 nm h^{-1} at FINUB), peaking in summer months, similar to the findings of (Yli-Juuti et al., 2011), while the peak for FINRO (growth rate at 3.74±1.48 nm h^{-1}) was found in spring, though the differences between the seasons for this site were rather small (Figures 6 and 7). Strong within-week variation favouring weekends is found for the roadside, while no clear variation was found for the other two sites (Figure 8). This may be due to either the higher condensation sink during weekdays that suppresses the events or the dominant impact of the traffic emissions which could make the detection of NPF events harder. J_{10} was the highest at FINRO, peaking in autumn for both urban sites (with small differences with spring), while FINRU presented the highest J_{10} in summer (Figures 9 and 10).

For all sites of this study in Finland, NPF events were consistently associated with lower relative humidity and higher solar radiation (Table S1). At the background sites temperature was found to be lower on NPF event days compared to the average

conditions, whereas it was found higher for FINRO associated with the different seasonality of the events. No significant differences were found for the wind speed on NPF events for all sites. There are though some significant differences in the wind conditions for NPF events compared to average conditions. At FINRU, NPF events were more common with northerly wind directions, as was also found by Nieminen et al. (2014) and Nilsson et al. (2001). This is probably due to the lower condensation sink which can be associated with the lower relative humidity also found for incoming winds from that sector and also explains the lower temperatures found with NPF events at this site (Table 4). Similarly, at FINUB NPF events were favoured by wind directions from the northerly sector, while there is almost a complete lack of NPF on southerly winds. This is due to its position at the north of both the city centre and the harbour, though winds from that sector are not common in general for that site. Finally, the wind sector frequency distribution for NPF events at FINRO also favours northerly winds with an almost complete absence of southerly winds probably due to the elevated pollutant concentrations and condensation sink associated with them.

At all sites, NPF event days had a lower condensation sink compared to the average for the site, as well as lower concentrations of pollutants (apart from O_3) where data was available (Table S2). The seasonal variation of NPF events in Finland favouring spring, was explained by earlier work as the result of the seasonal variation of H_2SO_4 concentrations (Nieminen et

al., 2014), which in the area peak in spring. The variation of H_2SO_4 concentrations is directly associated with SO_2 concentrations in the area, which follow a similar trend. The seasonal variation of NPF events at FINRO though cannot be explained by the variation of H_2SO_4 in the area. SO_2 concentrations, which were available only for the nearby urban background site at Kalio (about 3 km away from FINRO) and may provide information upon the trends of SO_2 in the greater area, peak during January (probably due to increased heating in winter, limited oxidation processes due to lower incoming solar radiation and the effect of shallow boundary layer) and are higher during spring months compared to summer. In general, the variation of pollutant concentrations and the condensation sink is not great for the spring and summer seasons. The only variable out of the ones considered that may explain the seasonality of NPF events at the site is the increased concentrations of PM_{10} found for spring months, which might be associated with road sanding and salting that takes place in Scandinavian countries during the colder months (Kupiainen et al., 2016) and are released in the ambient air during spring months (Stojiljkovic et al., 2019). The source of these particles though is uncertain, as no major differences in the wind roses are found between the two seasons. Another study by Sarnela et al. (2015) at a different site in southern Finland attributed the seasonality of NPF events in Finland to the absence of H_2SO_4 clusters during summer months due to a possible lack of stabilizing agents (e.g. ammonia). This could explain the limited number of small particles (smaller than 10 nm) at

the background sites during summer. In the more polluted environment at a roadside these agents may exist, but such data was unfortunately not available.

Finally, a feature mentioned by Hao et al. (2018) in their study at the site of Hyytiälä, in which late particle growth is observed was also found in this study. This happened on about 20% of NPF days at FINRU (and a number of non-event days) and in most cases in early spring (before mid-April) or late autumn (after mid-September). New particles were formed and either did not grow or grew very slowly until later in the day when growth rates increased (Figure S2). In all these cases, growth started when solar radiation was very low or zero, which probably associates the growth of particles with nighttime chemistry leading to the formation of organonitrates (as found by the same study). A similar behaviour was also found at FINUB but rarely. Particle growth at late hours is not a unique feature, as it was found at all sites studied. What is different in the specific events is the lack or very slow growth during the daytime. Lower temperature (-0.81^{o}C), incoming solar radiation (112 Wm^{-2}) and higher relative humidity (68.4%) occurred on event days with later growth, while no clear wind association was found. Lower concentrations of organic matter and nitrate were found throughout the days with later growth compared to the rest of the NPF days. The very high average particle number concentration in the smaller size bins is due to particles, though not growing to larger sizes for some time, persisting in the

local atmosphere for hours. These results though should be used with caution due to the limited number of observations.

3.4 Sites in Spain

For Spain, data was available for an urban and a rural background site in the greater area of Barcelona. NPF events were rather frequent, occurring on about 12% of the days at the rural site and 13.1% at the urban site. Though the sites are in close proximity (about 50 km), the seasonality of NPF events was different between them, peaking in spring at SPARU and autumn at SPAUB (Figure 5). The frequency of NPF events in winter was relatively high compared to the sites in central and northern Europe and higher than summer for both sites. Similarly, the growth rate was similar for the two sites, being 3.62 ± 1.86 nm h^{-1} at SPARU and 3.38 ± 1.53 nm h^{-1} at SPAUB, again being higher in autumn for the urban site (which appears to be a feature of more polluted sites), while the rural site follows the general trend of rural background sites, peaking in summer (Figure 7). The formation rate J_{10} at SPAUB is comparable to the other urban background sites (apart from GERUB) and it peaked in spring, while once again the peak at SPARU was found in summer, similar to the other rural sites of this study apart from the Greek (Figures 9 and 10). For both sites a higher probability for events was found on weekends compared to weekdays, though this trend is stronger at SPAUB (Figure 8). On the other hand, at the urban site both the growth

and formation rates were higher on weekdays compared to weekends (both $p < 0.001$). While the higher growth rate during weekdays may be associated with the increased presence of condensable species from anthropogenic activities, the higher formation rate might be affected by the increased emissions during these days, which bias to an extend its value.

In general, the atmospheric conditions favouring NPF events at both sites are similar to most other sites, with lower relative humidity and higher solar radiation and wind speed ($p < 0.001$ for wind speed at SPAUB) (Table S1). The wind sector frequency distribution between the two sites is different, with mainly northwesterly and southeasterly winds for SPARU (which seems to be affected by the local topography), while a more balanced profile is found at SPAUB. For both sites, though, increased probability for NPF events is found for westerly and northwesterly winds. For both sites, these incoming wind directions originate from a rather clean area with low concentrations of pollutants and condensation sink (Table S4). At SPARU, incoming wind from directions with higher concentrations of pollutants and condensation sink were associated with lower frequency of NPF events but higher growth rates. At SPAUB, NPF events were relatively rare and growth rates were lower with easterly wind directions, as air masses originating from that section have passed from the city centre and the industrial areas from the Besos River. Due to this, incoming air

masses from these sectors had higher concentrations of pollutants and condensation sink. The concentrations of all the pollutants with available data were lower at SPAUB (apart from O_3 and CO - the results for the latter are not included) on NPF event days (Table S2) as was found by Brines et al. (2015), as were the condensation sink and PM concentrations. At SPARU, the concentrations of the pollutants with available data are rather low and as a result minimal differences were found between event and non-event days.

While NPF events with subsequent growth of the particles were rare during summer, cases of bursts of particles in the smallest size range available were found to occur frequently, especially in August and July (the month with the fewest NPF events, despite the favourable meteorological conditions). In such cases, a new mode of particles appears in the smallest size available, persisting for many hours though without clear growth (brief or no growth is only observed), as reported by Dall'Osto et al. (2012). Due to the lack of growth of the particles these burst events do not qualify as NPF events using the criteria set in the present study. These burst events are associated with southerly winds (known as Garbí-southwest and Migjorn-south in Catalan, which are common during the summer in the area) that bring a large number of particles smaller than 30 nm to the site from the nearby airport (located about 15 km to the southwest) and port (7 km south), as well as

Saharan dust, increasing the concentrations of PM (Rodríguez et al., 2001) and thus suppressing NPF events due to the increased condensation sink.

Finally, the wind sector frequency distribution at SPARU appears to have a daily cycle, with almost exclusively stronger southeasterly winds at about midday (Figure S3), which might be the result of the movement of the air masses due to the increased solar activity during that time (which results in different heating patterns of the various land types in the greater area). These incoming southeast winds are more polluted and have higher condensation sink, which almost consistently bring larger particles at the site during the midday. This may explain to an extent the lowest probability for NPF events from that sector, despite the very high concentrations of O_3 associated to them, with some extreme values well above 100 μg m^{-3} (Querol et al., 2017). The highest average growth rates are also found from that direction.

3.5 Sites in Greece

Data are available for two background sites in Greece, though not in close proximity. While in Greece meteorological conditions are favourable in general for NPF events, with high solar radiation and low relative humidity, their frequency was only about 8.5% for the urban background site in Athens and 6.5% for the rural background site in Finokalia, similar

to the frequency of Class I events in the study by Kalivitis et al. (2019). Most NPF events occurred in spring at both sites, peaking in April (Figure 5). It is interesting that all sites in southern Europe have a considerable number of NPF events during winter, which might be due to the specific meteorological conditions found in this area, where winter is a lot warmer and with higher solar radiation than the sites in northern and central Europe. The growth rate of particles in these events was found to be similar at both sites (3.68±1.41 nm h^{-1} for GREUB and 3.78±2.01 nm h^{-1} for GRERU) and was higher in summer compared to the other seasons (Figures 6 and 7), having a similar trend with the temperature and particulate organic carbon concentrations in the area. J_{10} presented an interesting trend, having high averages in winter for both sites. Interestingly, the lowest average J_{10} was found for summer at both sites (Figure 10).

Similar to all sites, higher solar radiation and lower relative humidity compared to average conditions were found on NPF event days (Table S1). Temperature and wind speed were found to be lower, but the differences are minimal and are associated with the seasonal variability of the events. The wind rose for GREUB mainly consists of northeasterly and southwesterly winds. Due to its position, the site is heavily affected by emissions in Athens city centre with westerly winds, resulting in increased particle number concentrations and condensation sink. Despite this, the highest NPF probability and growth rates were found

with northwesterly wind directions (Table S4). This may be due to them being associated with the highest solar radiation (probably the result of seasonal and diurnal variation) and temperature and the lowest relative humidity, along with the highest condensation sink and particle number concentrations of almost all sizes. Chemical composition data was not available for GREUB, though SO_2 concentrations are rather low in Athens and kept declining after the economic crisis (Vrekoussis et al., 2013). The seasonality of SO_2 concentration in Athens favoured winter months and was at its lowest during summer for the period studied (ΥΠΕΚΑ, 2012) (this trend changed later as SO_2 concentrations further declined), which may also be a factor in the seasonality of NPF events, though this will be further discussed later.

At the GRERU site, the wind rose is dominated by westerly winds, and though it coincides with the most important source of pollutants in the area, the city of Heraklion, its effect while observable is not significant due to the topography in the area. The wind sector frequency distribution for NPF events is similar to the average with significantly higher wind speeds ($p < 0.001$). In general, GRERU has very low pollutant concentrations, with an average NO of 0.073 μg m^{-3}, NO_2 of 0.52 μg m^{-3} and SO_2 in concentrations below 1 ppb (Kouvarakis et al., 2002). Due to this, the differences in the chemical composition in the atmosphere are also minimal (Table S2). For this specific site two different patterns of

development of NPF events were found. In one case, NPF events occurred in a rather clear background, while in the other they were accompanied with an increase in number concentrations of larger particles or a new mode appearing at larger sizes (about a third of the events). No differences were found in the seasonal variation between the two groups; increased gaseous pollutant and particulate organic carbon concentrations were found for the second group (though the differences were rather small) and a wind rose that favoured southwesterly winds (originating from Crete itself) instead of the northwesterly (originating from the sea) ones for the first group. The growth rate for the two groups was found to be 3.56 nm h^{-1} for the first group and 4.17 nm h^{-1} for the second, which might be due to the increased presence of condensable compounds. As the dataset starts from the particle size of 8.77 nm, the possibility that these particles were advected from nearby areas should not be overlooked, though they persisted and grew at the site. Other than that, no significant differences were found for the different wind directions.

As mentioned earlier, both sites had a very low frequency of events and J_{10} in summer similar to previous studies also reporting few or no events during summer (Vratolis et al., 2019; Ždímal et al., 2011), though the incoming solar radiation is the highest and relative humidity is the lowest during that season. This variation was also observed by Kalivitis et al. (2012) who associated the seasonal variation of NPF events at GRERU to the

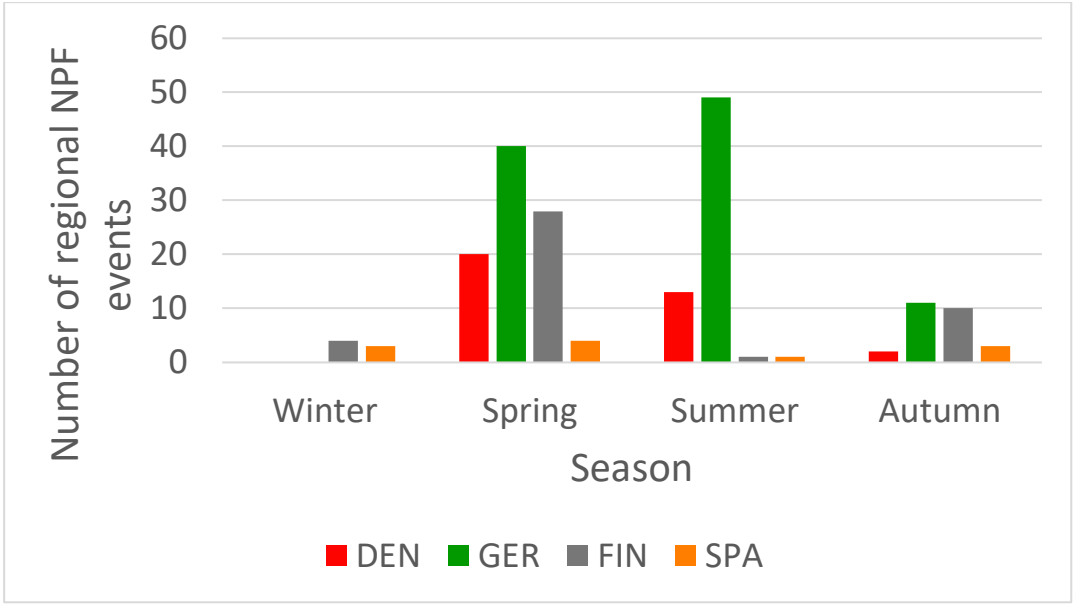
concentrations of atmospheric ions. The effect of the Etesian winds (known as Meltemia in Greek), which dominate the southern Aegean region during the summer months though should not be overlooked. These result in very strong winds with an average wind speed of 8.15 m s^{-1} during summer at GRERU, and increased turbulence found in all years with available data, affecting both sites of this study. During this period, $N_{<30nm}$ drops to half or less compared to other seasons at both sites, while $N_{>100nm}$ is at its maximum due to particle aging (Kalkavouras et al., 2017), increasing the condensation sink, especially in GRERU (the effect in GREUB is less visible due to both the wind rose, blowing from east which is a less polluted area, as well as the reduction of urban activities during summer months in Athens). Both the increased condensation sink and turbulence are possible factors for the reduced number of NPF events found at both sites in summer. Another possible factor is the effect of high temperatures in destabilising the molecular clusters critical to new particle formation.

3.6 Region-Wide Events

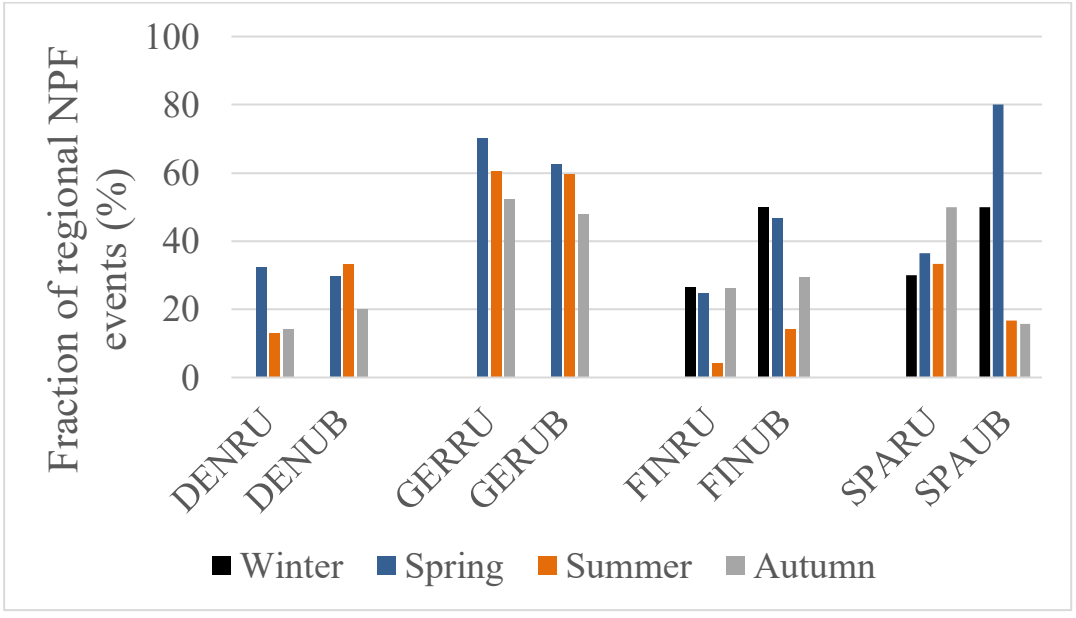
Region-wide events are NPF events which occur over large scale areas, that may cover hundreds of kilometres (Shen et al., 2018). In general, NPF events are large-scale phenomena that can be limited by local conditions (ex. local factors such as pollution sources). In the present study, NPF events that took place on the same day at both

background sites (urban background and rural) and thus were not constrained by local factors, are considered as regional and their conditions are studied (Table S5). The background sites in Greece were not considered due to the great distance between them (about 350 km). There is also uncertainty for the background sites in Finland, where the distance is about 190 km, though a large number of days were found when NPF events occurred on the same day. The number of region-wide events per season (or the fraction of region-wide events to total NPF events) is found in Figure 11 and it appears as if they are more probable in spring at all the sites of the present study (apart from Finland, though the number of events in winter was low), despite the differences found in absolute numbers.

Figure 11: (a) Number of region-wide New Particle Formation events per season and **(b)** fraction of region-wide events to total New Particle Formation events per season for each site. Region-wide events are defined as those that occur on the same day at both background sites (Rural and Urban background).



(a)



(b)

For the Danish sites, about 20% of NPF events in DENRU were regional (the percentage is higher for DENUB due to the smaller number of events, at 29%). The relatively low frequency of region-wide NPF events can be explained by the different seasonal variation of NPF events (region-wide NPF events were more frequent in spring compared to the average due to the seasonality of NPF events in DENUB). Compared to local NPF event conditions, higher wind speed and solar radiation, as well as O_3 and marine compound concentrations (results not included) were found, while the concentrations of all pollutants (such as NO, NO_x, sulphate, elemental and organic carbon) were lower. The exceptions found at DENRU (increased relative humidity and less incoming solar radiation) are probably due to the different seasonality between local and region-wide NPF events at the site, though region-wide events rarely present similar characteristics at different sites even in the same country due to the differences in the initial meteorological and geographical conditions (Hussein et al., 2009). The growth rates of region-wide events were found to be lower than those of local events at both sites, which is probably associated with the limited concentrations of condensable compounds due to the cleaner air masses of marine origin (as confirmed by the higher concentrations of marine compounds).

For the German sites, the majority of NPF events of this study were region-wide (about 60%). Compared to the average, the meteorological conditions found for NPF event days

compared to average conditions were more distinct for the region-wide events, with even lower wind speed and relative humidity and higher temperature and solar radiation, and all of these differences were significant ($p < 0.001$). At GERRU where chemical composition data was available, higher concentrations of particulate organic carbon and sulphate and lower nitrate concentrations were found. The differences are significant ($p < 0.001$) and may explain the higher growth rates found in region-wide events at both sites compared to the average, which is a unique feature. It should be noted that as the majority of NPF events at the German sites are associated with easterly winds, it is expected that in most cases the region-wide events will be associated with these, carrying the characteristics that come along with them (increased growth rates and concentrations of organic carbon, as discussed in Section 3.2).

For the Finnish sites, about a quarter of the NPF event days at FINRU (26%) occurred on the same day as at FINUB (the frequency is a lot higher for FINUB, at 39%). As in Germany, the meteorological conditions found on NPF event days compared to average conditions were more distinct during region-wide events. Thus, for both sites temperature and relative humidity were lower while solar radiation was higher. The different trend found for the wind speed at the two sites (being higher on average NPF days at FINRU and lower at FINUB compared to average conditions) was enhanced as well at the two sites for region-

wide events. At FINRU where chemical composition data was available, NO_x and SO_2 had similar concentrations on region-wide event days, while O_3 was significantly higher ($p < 0.001$). As at most other sites, the growth rate was found lower on region-wide event days compared to the average at both sites.

Finally, for the Spanish sites the datasets of the two sites did not overlap greatly, having only 322 common days. Among these days, 13 days presented with NPF events that took place simultaneously at both sites, with smaller growth rates on average compared to local events (43% of the events at SPARU and 36% of the events at SPAUB in the period 8/2012 to 1/2013 and 2014 when data for both sites were available). Due to the small number of common events the results are quite mixed with the only consistent result being the lower relative humidity and higher O_3 concentrations for regional events at both sites, though none of these differences is significant. The wind sector frequency distribution at SPAUB seems to further favour the cleaner sector, with the majority of incoming winds being from the NW and even higher wind speeds (though with low significance). The result is similar at SPARU, though less clear and with lower wind speeds.

These results are in general in agreement with those found in the UK in a previous study, where meteorological conditions were more distinct on region-wide event days compared

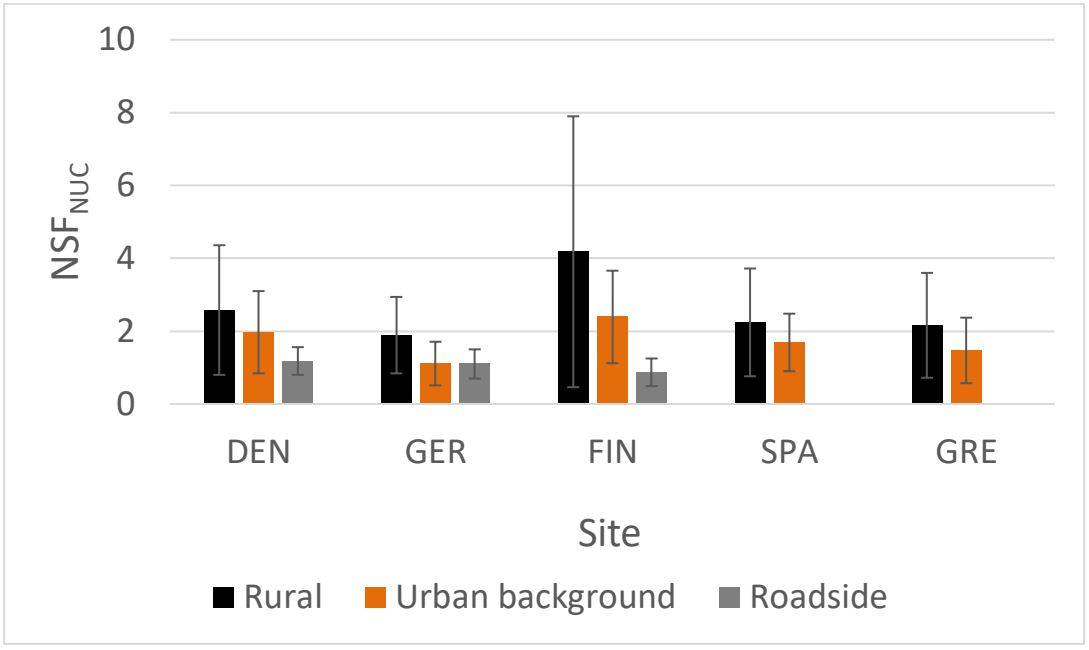
to local NPF events; pollutant concentrations were lower as well as the growth rates of the newly formed particles (Bousiotis et al., 2019).

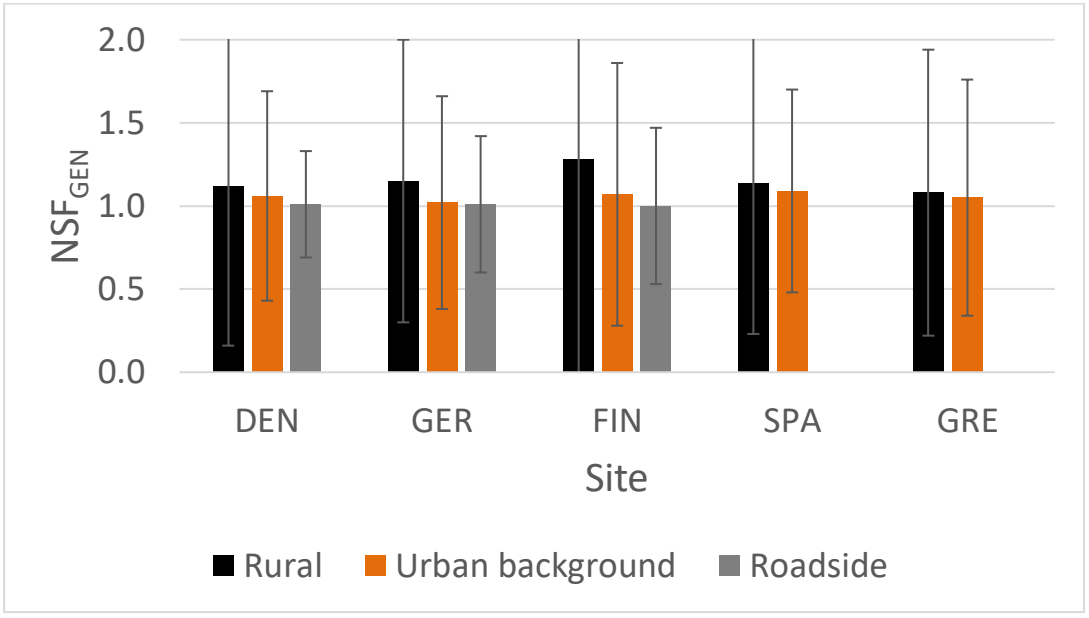
Common events were also found between either of the background sites and the roadside, but they were always fewer in number, due to the difference in their temporal variability compared to the background sites, resulting from the effect of roadside pollution.

3.7 The Effect of NPF Events on the Ultrafine Particle Concentrations

The NSF is a metric of the effect of NPF events upon particle concentrations on either the days of the events or over a larger timescale. Both the NSF_{NUC} and NSF_{GEN} were calculated for all sites of this study and the results are presented in Figure 12. For almost all rural background sites NSF_{NUC}, which indicates the effect of NPF on ultrafine particle concentrations on the day of the event, was found to be above or near 2 (the only exception was GERRU), which means that NPF events more than double the number of ultrafine particles (particles with diameter smaller than 100 nm) at the

Figure 12: NSF_{NUC} (average relative increase of ultrafine particles – particles of diameter up to 100 nm - due to New Particle Formation events on event days) and NSF_{GEN} (average annual relative increase of ultrafine particles due to New Particle Formation events) at all sites.





site on the days of the events, as NPF events are one of the main sources of ultrafine particles in this type of sites, especially below 30 nm. This reaches up to 4.18 found at FINRU (418% more ultrafine particles on the day of the events – 100% being the average), showing the great effect NPF events have on rather clean areas. The long-term effect was smaller, and it was found that at FINRU NPF events increase the number of ultrafine particles by about 130% in general. The effect of NPF events was a lot smaller at the urban sites, though still significant at urban background sites (reaching up 240% at FINUB on the days of events), while roadsides had the smallest NSF compared to their respective background sites. This is because of the increased effect of local sources such as traffic or heating, and the associated increased condensation sink found within these sites, which cause the new particles to be scavenged by the more polluted background.

The calculation of NSF at the sites around Europe showed a weakness of the specific metric, which points to the need for more careful interpretation of the results of this metric, especially at roadside sites. At FINRO, the NSF_{NUC} provided a value smaller than 1, which translates as ultrafine particles are lost instead of formed on NPF event days. This though is the result of both the sharp reduction in particle number concentrations at all modes that are required for NPF events to occur at a busy roadside (much lower condensation sink), as well as a difference in the ratio between smaller to larger particles

(smaller or larger than 100 nm) on NPF event days (favouring the larger particles) at the specific site. Similarly, the long-term effect of NPF events at the site was found to be 1, which means that NPF events appear to cause no changes in the number concentration of ultrafine particles.

4. DISCUSSION

4.1 Variability of the frequency and seasonality of the events

The most consistent result found throughout the areas studied, regardless of the geographical location was the higher frequency of NPF events at rural background sites compared to roadsides. This pattern comes in contrast with what was found for the more polluted Asian cities (Peng et al., 2017; Wang et al., 2017), where NPF events were more frequent at the urban sites. This is probably associated with the even greater abundance of condensable species (which further enhances the growth rate of the particles, thus increasing their chance of survival), deriving from anthropogenic emissions, found in Asian megacities compared to European ones and results in a greater frequency of NPF events in Asian cities, even compared to the most polluted cities in Europe. This contrast emphasises the differences in the occurrence of NPF events between the polluted cities in Europe and Asia, which are associated with the level of pollution found in them, as well as the influence that the level of pollution has on the occurrence of NPF events in general.

The type of site dependence found in Europe together with the average conditions found on NPF event days compared to the average for each site, underline the importance of clear atmospheric conditions (high solar radiation and low relative humidity and pollutant concentrations) at all types of sites in Europe, especially for region-wide events. The temperature and wind speed presented more diverse results which in many cases are associated with local conditions. The origin of the incoming air masses though, appears to have a more important influence upon the NPF events. Cleaner air masses tend to have higher probability for NPF events, a result which was consistent among the sites of this study regardless of their type.

The frequency of NPF events at roadsides peaked in summer in all three countries with available data. Greater variability in the seasonality of NPF events was found at the background sites. The urban background sites presented more diverse results, for both the occurrence and development of NPF events, especially compared to rural background sites. The within-week variation of NPF events was found to favour weekends in most cases, as the pollution levels decrease, due to the weekly cycle, especially at the roadsides. As background sites have smaller variations between weekdays and weekends, the within-week variation of NPF events is smaller at the urban background sites and almost non-existent at the rural background sites.

4.2 Variability and seasonality of the formation and growth rate

The growth rate of the newly formed particles was found to be higher at all the roadsides compared to their respective rural and urban background. The picture is similar for J_{10}, the formation rate of particles with 10 nm diameter (the rate of formed particles associated with NPF events that reached 10 nm diameter), for which urban background sites were between their respective rural background sites and the roadsides with the sole exception of DENUB (the difference with DENRU is rather small though). The growth and formation rate at the rural background sites (apart from the Greek site) were found to be higher in summer than in other seasons. On the other hand, the seasonality of the growth rate at the roadsides is not clear but the formation rate peaks in the autumn at all three roadside sites. While the trend at the rural sites is probably associated with the enhanced photochemistry and increased concentrations of BVOCs during summer, the seasonality of the growth rate at the roadside sites is more difficult to explain and probably shows the smaller importance of the BVOCs compared to the compounds of anthropogenic origin (which are in less abundance in summer) in this type of environment. In general, higher temperatures were associated with higher growth rates. This though applies only for the specific conditions at each site and cannot be used as a general rule for the expected growth rate at a site, as locations with higher temperatures did not present the higher

growth rates. Additionally, the origin of the incoming air masses appears to have an effect on the growth of the particles as well. In most of the sites in this study, incoming air masses from directions associated with higher concentrations of pollutants presented higher growth rates of the newly formed particles. The effect of the different wind directions upon the formation rate was more complex and a definitive conclusion cannot be made.

4.3 Effect of local conditions in the occurrence and development of NPF events

Apart from the general meteorological and atmospheric conditions that affect the occurrence and the metrics of NPF events, conditions with a more local character were found to play a significant role as well. These include synoptic systems, such as the one occurring during the summer at the Greek sites, affecting the frequency and seasonality of the events. As a result, sites or seasons with conditions that favoured NPF presented decreased frequency of events and unexpected seasonality, due to the increased turbulence caused by such pressure systems. Additionally, local sources of pollution can also have a significant impact in the temporal trends and metrics of the events, even for sites of very close proximity. One such example was the urban sites in Denmark, which despite being affected by the same source of pollution (the nearby port) and being only a few kilometres away from each other, they presented different outcomes in the occurrence of the events. This was due to the different atmospheric conditions found

between them, being a background and a roadside site, which led to a different response in that local variable. In this case, the effect of the specific source was more prominent at the urban background site compared to the roadside, resulting in fewer NPF events, as the newly formed particles were more effectively supressed at the urban background site, due to their slower growth.

5. CONCLUSIONS

There are different ways to assess occurrences of new particle formation (NPF) events. In this study, the frequency of NPF events, the formation and growth rate of the particles associated with secondary formation of particles and not primary emissions, at 13 sites from five countries in Europe are considered. NPF is a complicated process, affected by many meteorological and environmental variables. The seasonality of these variables, which varies throughout Europe, results in the different temporal trends found for the metrics studied in this paper. Apart from meteorological conditions though, some of which have a uniform effect (such as the solar radiation intensity and relative humidity), many local variables can also have a positive or negative effect in the occurrence of these events. Sites with less anthropogenic influence seem to have temporal trends dependant on the seasonality of synoptic conditions and general atmospheric composition. The urban sites though and especially those with important sources of pollution in close proximity, present

more complex trends, as the NPF occurrence depend less to favourable meteorological and more to the local atmospheric conditions. As NPF events' occurrence is based on the balance between the rapid growth of the newly formed particles and their loss from processes such as evaporation or coagulation, the importance of significant particle formation, fast growth (which is enhanced by the increased presence of condensable compounds from anthropogenic activities found in urban environments) and low condensation sink is increased within such environments, also affecting the temporal trends of the events, making them more probable during periods with smaller pollution loads (e.g. summer, weekends). This explains the smaller frequency of NPF events at roadside sites compared to their respective background sites despite the greater formation and growth rates observed in them. Consequently, NPF events have a smaller influence on the ultrafine particle load at the urban sites compared to background sites, due to both the increased presence of ultrafine particles from anthropogenic emissions as well as the smaller probability of ultrafine particles to survive in such environments.

Nevertheless, NPF events are an important source of ultrafine particles in the atmosphere for all types of environments and are an important factor in the air quality of a given area. The present study underlines the importance of both the synoptic and local conditions on NPF events, the mix of which not only affects their development but can also influence

their occurrence even in areas of very close proximity. Since the mechanisms and general trends in NPF events are yet to be fully explained and understood, more laboratory and field studies should be undertaken to generate new knowledge.

DATA ACCESSIBILITY

Data supporting this publication are openly available from the UBIRA eData repository at https://doi.org/10.25500/edata.bham.00000467

AUTHOR CONTRIBUTIONS

The study was conceived and planned by MDO and RMH who also contributed to the final manuscript. The data analysis was carried out by DB who also prepared the first draft of the manuscript. AM, JKN, CN, JVN, HP, NP, AA, GK, SV and KE have provided the data for the analysis. FDP, XQ, DCB and TP provided advice on the analysis.

COMPETING INTERESTS

The authors have no conflict of interests.

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3.3 The effect of meteorological conditions and atmospheric composition in the occurrence and development of New Particle Formation (NPF) events in Europe

3.3.1 INTRODUCTION

New Particle Formation (NPF) events are an important source of particles in the atmosphere (Merikanto et al., 2009; Spracklen et al., 2010), which are known to have adverse effects on human health (Schwartz et al., 1996; Politis et al., 2008; Kim, et al., 2015) as well as affecting the optical and physical properties of the atmosphere (Makkonen et al., 2012; Seinfeld and Pandis, 2012). While they occur almost everywhere in the world (Dall'Osto et al., 2018; Kulmala et al., 2017; O'Dowd et al., 2002; Wiedensohler et al., 2019; Chu et al., 2019; Kerminen et al., 2018), great diversity is found in the atmospheric conditions within which they take place. Many studies have been done in a large number of different types of locations (urban, traffic, regional background) around the world and differences were found in both the seasonality and intensity of NPF events. To an extent this variability is due to the mix of conditions that are specific to each location, which blurs the general understanding of the conditions that are favourable for the occurrence of NPF events (Berland et al., 2017; Bousiotis et al., 2020). For example, solar radiation is considered as one of the most important factors in the occurrence of NPF events (Kulmala and Kerminen, 2008; Kürten et al., 2016; Pikridas et al., 2015; Salma et al., 2011), as it is needed for the photochemical reactions that lead to the formation of sulphuric acid (Petäjä et al., 2009; Cheung et al., 2013), which is considered as the main component of the formation and growth of the initial clusters (Iida et al., 2008; Weber et al., 1995); although in many cases, NPF events did not occur in the seasons with the highest insolation (Park et al., 2015; Vratolis et al., 2019). Similarly, higher temperatures are considered favourable for the growth of the newly formed particles as increased concentrations of both Biogenic Volatile Organic Compounds (BVOCs) and Anthropogenic Volatile Organic Compounds (AVOCs) (Yamada, 2013; Paasonen et al., 2013) and their oxidation products (Ehn et al., 2014) are associated with the growth of

the particles. This appears to be true in most cases, as higher growth rates are found in most cases in the local summer (Nieminen et al., 2018), although the actual importance of those VOCs in the occurrence of NPF events is still not fully elucidated. The effect of other meteorological variables is even more complex, with studies presenting mixed results on the effect of the wind speed and atmospheric pressure. Extreme values of those variables may be favourable for the occurrence of NPF events, as they are associated with increased mixing in the atmosphere, but at the same time suppress due to increased dilution of precursors (Brines et al., 2015; Rimnácová et al., 2011; Shen et al., 2018; Siakavaras et al., 2016), or favour them due to a reduced condensation sink (CS).

The effect of atmospheric composition on NPF events is also a puzzle of mixed results. While the negative effect of the increased CS is widely accepted (Kalkavouras et al., 2017 ; Kerminen et al., 2004; Wehner et al., 2007), cases are found when NPF events occur on days with higher CS compared to average conditions (Größ et al., 2018; Kulmala et al., 2005). Sulphur dioxide (SO_2), which is one of the most important contributors to many NPF pathways, in most studies was found in lower concentrations on NPF event days compared to average conditions (Alam et al., 2003; Bousiotis et al., 2019), although there are studies that have reported the opposite (Woo et al., 2001; Charron et al., 2008). Additionally, in a combined study of NPF events in China, events were found to be more probable under sulphur-rich conditions rather than sulphur-poor (Jayaratne et al., 2017). Similar is the case with the BVOCs and AVOCs, which present great variability depending the area studied (Dai et al., 2017), and their contribution in the growth of the particles is not fully understood yet. Until recently, it was considered unlikely for NPF events as they are considered in the present study, deriving from secondary formation not associated with traffic related processes such as dilution of the exhaust, to occur within the complex urban environment

due to the increased presence of compounds, mainly associated with combustion processes, which would suppress the survival of the newly formed particles within this type of environment (Kulmala et al., 2017). Despite that though, NPF events were found to occur within even the most polluted areas and sometimes with high formation and growth rates (Bousiotis et al., 2019; Yao et al., 2018).

It is evident that while a general knowledge of the role of the meteorological and atmospheric variables has been achieved, there is great uncertainty over the extent and variability of their effect (and for some of them even their actual effect) in the mechanisms of NPF in real atmospheric conditions, especially in the more complex urban environment (Harrison, 2017). The present study, using an extensive dataset from 16 sites in six European countries, attempts to elucidate the effect of several meteorological and atmospheric variables not only in general, but also depending on the geographical region or type of environment. While studies with multiple sites have been reported in the past, to our knowledge this is the first study that focuses directly on the effect of these variables upon the probability of NPF events as well as the formation and growth rates of newly formed particles in real atmospheric conditions.

3.3.2 SITE DESCRIPTION AND DATA AVAILABILITY

The present study uses a total of more than 85 years of hourly data from 16 sites from six countries of Europe of various land usage and climates from which 1950 NPF events were extracted and studied. A list of the available data and a brief description for each site is found in Table 2 (for the ease of reading the sites are named by the country of the site followed by the last two letters which refer to the type of site, being RU for rural/regional

background, UB for urban background and RO for roadside), while a map of the sites is found in Figure 13. The NPF frequency and formation rate for each site is found in Table 2.

Figure 13: Map of the sites of the study (Chapter 3.3).

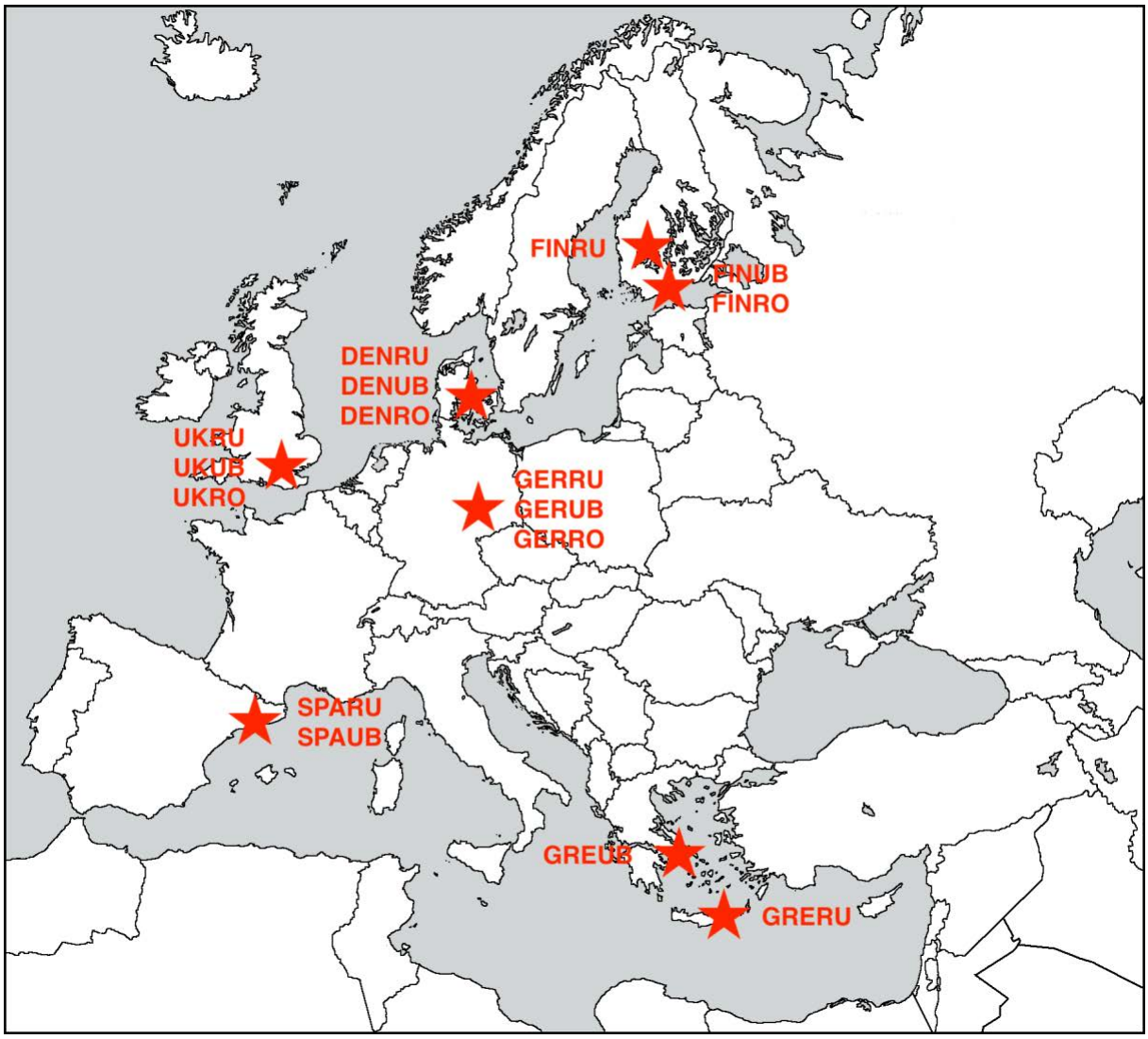


Table 2: Location and data availability of the sites (Chapter 3.3).

Site	Location	Available data	Meteorological data location	Data availability	Reference
UKRU	Harwell Science Centre, Oxford, 80 km W of London, UK (51° 34' 15" N; 1° 19' 31" W)	SMPS (16.6 - 604 nm, 76.5% availability), NOx, SO_2, O_3, OC, SO_4^{2-}, gaseous ammonia	On site	2009 - 2015	Charron et al., 2013
UKUB	North Kensington, 4 km W of London city centre, UK (51° 31' 15" N; 0° 12' 48" W)	SMPS (16.6 - 604 nm, 83.3% availability), NOx, SO_2, O_3, OC, SO_4^{2-}	Heathrow airport	2009 - 2015	Bigi and Harrison, 2010
UKRO	Marylebone Road, London, UK (51° 31' 21" N; 0° 9' 16" W)	SMPS (16.6 - 604 nm, 74.3% availability), NOx, SO_2, O_3, OC, SO42-	Heathrow airport	2009 - 2015	Charron and Harrison, 2003
DENRU	Lille Valby, 25 km W of Copenhagen, (55° 41' 41" N; 12° 7' 7" E) (2008 – 6/2010) Risø, 7 km north of Lille Valby, (55° 38' 40" N; 12° 5' 19" E) (7/2010 – 2017)	DMPS and CPC (5.8 - 700 nm, 68.3% availability), NOx, SO_2, O_3, OC, SO_4^{2-}	H.C. Ørsted – Institute station	2008 – 2017	Ketzel et al., 2004
DENUB	H.C. Ørsted – Institute, 2 km NE of the city centre, Copenhagen, Denmark (55° 42' 1" N; 12° 33' 41" E)	DMPS and CPC (5.8 - 700 nm, 61.4% availability), NOx, O_3	On site	2008 – 2017	Wang et al., 2010
DENRO	H.C. Andersens Boulevard, Copenhagen, Denmark (55° 40' 28" N; 12° 34' 16" E)	DMPS and CPC (5.8 - 700 nm, 65.7% availability), NOx, SO_2, O_3, OC, SO_4^{2-}	H.C. Ørsted – Institute station	2008 – 2017	Wang et al., 2010
GERRU	Melpitz, 40 km NE of Leipzig, Germany (51° 31' 31.85" N; 12° 26' 40.30" E)	TDMPS with CPC (4.8 - 800 nm, 87.2% availability), OC, SO_4^{2-}	On site	2008 – 2011	Birmili et al., 2016
GERUB	Tropos, 3 km NE from the city centre of Leipzig, Germany (51° 21' 9.1" N; 12° 26' 5.1" E)	TDMPS with CPC (3 - 800 nm, 90.4% availability)	On site	2008 – 2011	Birmili et al., 2016
GERRO	Eisenbahnstraße, Leipzig, Germany (51° 20' 43.80" N; 12° 24' 28.35" E)	TDMPS with CPC (4 - 800 nm, 68.3% availability)	Tropos station	2008 – 2011	Birmili et al., 2016
FINRU	Hyytiälä, 250 km N of Helsinki, Finland (61° 50' 50.70" N; 24° 17' 41.20" E)	TDMPS with CPC (3 – 1000 nm, 98.2% availability), NOx, SO_2, O_3, VOCs	On site	2008 – 2011 & 2015 – 2018	Aalto et al., 2001
FINUB	Kumpula Campus 4 km N of the city centre, Helsinki, Finland (60° 12' 10.52" N; 24° 57' 40.20" E)	TDMPS with CPC (3.4 - 1000 nm, 99.7% availability)	On site	2008 – 2011 & 2015 – 2018	Järvi et al., 2009
FINRO	Mäkelänkatu street, Helsinki, Finland (60° 11' 47.57" N; 24° 57' 6.01" E)	DMPS (6 - 800 nm, 90.0% availability), NOx, O_3	Pasila station and on site	2015 – 2018	Hietikko et al., 2018
SPARU	Montseny, 50 km NNE from Barcelona, Spain (41° 46' 45" N; 2° 21' 29" E)	SMPS (9 – 856 nm, 53.7% availability), NO_2, SO_2, O_3	On site	2012 - 2015	Dall'Osto et al., 2013
SPAUB	Palau Reial, Barcelona, Spain (41° 23' 14" N; 2° 6' 56" E)	SMPS (11 – 359 nm, 88.1% availability), NO_2, SO_2, O3	On site	2012 – 2015	Dall'Osto et al., 2012
GRERU	Finokalia, 70 km E of Heraklion, Greece (35° 20' 16.8" N; 25° 40' 8.4" E)	SMPS (8.77 - 849 nm, 85.0% availability), NO_2, O_3, OC	On site	2012 – 2018	Kalkavouras et al., 2017
GREUB	"Demokritos", 12 km NE from the city centre, Athens, Greece (37° 59' 41.96" N; 23° 48' 57.56" E)	SMPS (10 – 550 nm, 88.0% availability)	On site	2015 – 2018	Mølgaard et al., 2013

3.3.3 RESULTS

In this study NPF events are generally observed as particles growing from a smaller size (typically 3-16 nm depending on the size detection limit of instruments used) to 30 nm or larger. They therefore reflect the result both of nucleation, which creates new particles of 1-2 nm (not detected with the instruments used in this study), and growth to larger sizes (the number of NPF events per day of the week and month of the year is found in Table S1). In analysing NPF events, we therefore consider three diagnostic features:

- the frequency of events occurring (i.e. days with an event divided by total days with relevant data),
- the rate of particle formation at a given size (J_{10} in this case), which is also affected to an extent by the survival and growth rate up to the lower particle size available for each site,
- the growth rate of particles from the lower measurement limit to 30 nm (or 50 nm for the UK sites).

From the analysis of the extended dataset a total of 1952 NPF events were extracted and studied. The NPF frequency, growth and formation rate for each site is found in Table 3. The seasonal variation of NPF events is found in Table S1.

Table 3: Frequency (and number of NPF events), growth and formation rate of NPF events.

Site	Frequency of NPF events (%)	GR (nm h-1)	J_{10} (N cm^{-3} s^{-1})
UKRU	7.0 (160)	3.4*	8.69E-03**
UKUB	7.0 (156)	4.2*	1.42E-02**
UKRO	6.1 (120)	5.5*	3.75E-02**
DENRU	7.9 (176)	3.19	2.57E-02
DENUB	5.8 (116)	3.19	2.40E-02
DENRO	5.4 (117)	4.45	8.07E-02
GERRU	17.1 (164)	4.34	9.18E-02
GERUB	17.5 (169)	4.24	1.02E-01
GERRO	9.0 (62)	5.17	1.38E-01
FINRU	8.7 (190)	2.91	1.19E-02
FINUB	5.0 (110)	2.87	2.49E-02
FINRO	5.1 (49)	3.74	6.94E-02
SPARU	12 (68)	3.87	1.54E-02
SPAUB	13.1 (97)	3.71	2.12E-02
GRERU	6.5 (116)	3.68	4.90E-03
GREUB	8.5 (82)	3.4	4.41E-02

* GR up to 50 nm calculated

** J_{16} calculated

3.3.3.1 Meteorological Conditions

The gradients, coefficients of determination (R^2 – the relationships found are characterised as weak for $R^2 < 0.50$, strong for $0.50 < R^2 < 0.75$ and very strong for $R^2 > 0.75$) and the p-values from the analysis of the meteorological variables when linear relationships are considered, as well as the average conditions of these variables are found in Table 4. The results for each site and variable are found in Figures 14-18.

Table 4: Gradients, R^2 and p-values (for values > 0.05) for the relation between meteorological conditions and NPF event variables when linear relationships are considered. Gradients of R^2 > 0.50 are in bold.

Downward shortwave solar radiation K↓ (W m^{-2})										
Site	**a_N**	**R^2**	**p**	**a_G**	**R^2**	**p**	**a_J**	**R^2**	**p**	**Average**
UKRU	**2.89E-04**	0.93	<0.001	1.79E-04	0.04	-	**1.45E-05**	0.89	<0.001	123
UKUB	**1.74E-04**	0.89	<0.001	-5.76E-04	0.31	-	1.51E-06	0.03	-	124
UKRO	**1.85E-04**	0.96	<0.001	8.84E-05	0.01	-	**2.93E-05**	0.73	<0.005	129
DENRU	**1.75E-04**	0.88	<0.001	4.24E-04	0.20	-	**3.55E-05**	0.64	<0.001	115
DENUB	**1.08E-04**	0.91	<0.001	1.47E-04	0.03	-	2.16E-05	0.48	<0.01	115
DENRO	**1.33E-04**	0.95	<0.001	1.27E-04	0.01	-	**5.46E-05**	0.50	<0.005	117
GERRU	**4.91E-04**	0.98	<0.001	**9.88E-04**	0.72	<0.01	**1.33E-04**	0.81	<0.001	130
GERUB	**5.57E-04**	0.97	<0.001	**7.28E-04**	0.51	<0.005	**1.56E-04**	0.69	<0.001	114
GERRO	**2.16E-04**	0.95	<0.001	-5.89E-04	0.09	-	**1.37E-04**	0.59	<0.005	114
FINRU	**2.29E-04**	0.76	<0.001	**1.01E-03**	0.57	<0.01	**2.43E-05**	0.82	<0.001	91.5
FINUB	6.90E-05	0.37	-	1.81E-04	0.08	-	2.24E-05	0.25	-	111
FINRO	**8.98E-05**	0.59	<0.005	9.15E-04	0.34	<0.005	3.09E-05	0.03	-	114
SPARU	4.15E-05	0.35	<0.05	5.68E-04	0.13	-	**3.03E-05**	0.74	<0.001	162
SPAUB	**7.76E-05**	0.58	<0.05	6.98E-04	0.23	-	**3.35E-05**	0.81	<0.001	180
GRERU	**2.67E-05**	0.52	<0.001	**7.14E-04**	0.55	<0.001	-3.09E-06	0.05	-	201
GREUB	2.97E-05	0.31	-	-1.10E-04	0.02	-	3.96E-05	0.34	<0.05	183

Relative Humidity (%)										
Site	**a_N**	**R^2**	**p**	**a_G**	**R^2**	**p**	**a_J**	**R^2**	**p**	**Average**
UKRU	**-4.12E-03**	0.85	<0.001	1.69E-03	0.02	-	**-2.91E-04**	0.85	<0.001	79.7
UKUB	**-2.39E-03**	0.94	<0.001	8.23E-03	0.24	-	-8.04E-05	0.19	-	75.3
UKRO	**-3.10E-03**	0.85	<0.001	7.03E-03	0.25	-	-5.59E-04	0.46	<0.05	74.5
DENRU	**-3.08E-03**	0.95	<0.001	**9.42E-03**	0.74	<0.001	1.40E-05	0.00	-	75.7
DENUB	**-1.82E-03**	0.94	<0.001	3.64E-03	0.06	-	6.17E-05	0.00	-	75.7
DENRO	**-1.97E-03**	0.95	<0.001	-1.21E-02	0.22	-	-3.16E-04	0.10	-	75.7
GERRU	**-8.69E-03**	0.88	<0.001	**-1.30E-02**	0.72	<0.001	**-2.26E-03**	0.91	<0.001	81.9
GERUB	**-9.36E-03**	0.86	<0.001	**-6.34E-03**	0.67	<0.001	**-2.30E-03**	0.86	<0.001	78.7
GERRO	**-2.55E-03**	0.90	<0.001	3.98E-03	0.05	-	**-2.37E-03**	0.81	<0.001	78.7
FINRU	**-3.90E-03**	0.94	<0.001	**-7.07E-03**	0.65	<0.001	**-2.57E-04**	0.87	<0.001	80.1
FINUB	**-2.95E-03**	0.95	<0.001	1.04E-02	0.26	-	-1.62E-04	0.18	-	76.5
FINRO	**-1.70E-03**	0.92	<0.001	-1.47E-03	0.01	-	5.13E-04	0.10	-	71.1
SPARU	**-1.85E-03**	0.90	<0.001	-4.67E-03	0.08	-	-1.10E-04	0.14	-	66.4
SPAUB	**-6.34E-03**	0.93	<0.001	**2.43E+02**	0.50	<0.01	-2.08E-04	0.19	-	69.2
GRERU	-5.02E-04	0.22	-	1.06E-02	0.06	-	-8.97E-04	0.15	-	70.0
GREUB	**-1.21E-03**	0.62	<0.001	2.83E-03	0.06	-	2.14E-05	0.00	-	60.5

Table 4 (continued)

Temperature (°C)										
Site	**a_N**	**R^2**	**p**	**a_G**	**R^2**	**p**	**a_J**	**R^2**	**p**	**Average**
UKRU	**7.70E-03**	0.93	<0.001	**7.85E-02**	0.94	<0.001	**7.58E-04**	0.84	<0.001	10.6
UKUB	**6.33E-03**	0.98	<0.001	**1.39E-01**	0.96	<0.001	**9.00E-04**	0.73	<0.005	11.8
UKRO	**5.01E-03**	0.98	<0.001	**3.51E-02**	0.52	<0.05	1.62E-03	0.44	<0.05	12.1
DENRU	**5.28E-03**	0.83	<0.001	1.54E-02	0.08	-	**1.72E-03**	0.92	<0.001	9.80
DENUB	**1.45E-03**	0.45	<0.05	2.40E-02	0.33	-	7.32E-04	0.45	<0.05	9.82
DENRO	**3.59E-03**	0.88	<0.001	3.51E-03	0.00	-	**2.39E-03**	0.58	<0.005	10.0
GERRU	**1.24E-02**	0.92	<0.001	**5.65E-02**	0.92	<0.001	**4.93E-03**	0.93	<0.001	10.3
GERUB	**1.44E-02**	0.93	<0.001	**3.38E-02**	0.62	<0.001	**4.37E-03**	0.54	<0.005	11.1
GERRO	**4.57E-03**	0.89	<0.001	-3.33E-03	0.00	-	2.22E-03	0.11	-	11.1
FINRU	-1.75E-03	0.17	-	**1.13E-01**	0.79	<0.001	**5.08E-04**	0.72	<0.001	4.79
FINUB	-2.11E-04	0.00	-	**7.42E-02**	0.83	<0.001	4.16E-04	0.28	-	6.52
FINRO	**3.18E-03**	0.65	<0.005	**9.28E-02**	0.87	<0.001	-7.56E-04	0.05	-	7.72
SPARU	-3.01E-03	0.41	<0.05	**1.23E-01**	0.92	<0.001	**1.40E-03**	0.71	<0.001	13.9
SPAUB	-4.49E-04	0.02	-	**6.67E-02**	0.66	<0.005	2.50E-04	0.08	-	18.2
GRERU	**-3.03E-03**	0.75	<0.001	**1.74E-01**	0.75	<0.001	-4.63E-04	0.47	<0.05	18.2
GREUB	-8.50E-04	0.25	-	**4.67E-02**	0.62	<0.005	-1.26E-03	0.20	-	17.6

Wind Speed (m s^{-1})										
Site	**a_N**	**R^2**	**p**	**a_G**	**R^2**	**p**	**a_J**	**R^2**	**p**	**Average**
UKRU	4.00E-03	0.20	-	-3.04E-02	0.07	-	5.97E-05	0.00	-	3.96
UKUB	**1.20E-02**	0.87	<0.001	**-1.91E-01**	0.71	<0.001	5.06E-05	0.00	-	4.16
UKRO	3.87E-03	0.19	-	3.21E-02	0.02	-	2.73E-03	0.45	<0.005	4.14
DENRU	**8.53E-03**	0.88	<0.001	**-2.33E-01**	0.74	<0.001	3.29E-03	0.44	<0.01	4.17
DENUB	**8.70E-03**	0.90	<0.001	-3.33E-02	0.10	-	1.99E-03	0.19	-	4.17
DENRO	**8.91E-03**	0.89	<0.001	-1.51E-01	0.49	<0.001	7.33E-04	0.00	-	4.16
GERRU	**-1.81E-02**	0.57	<0.005	**-2.26E-01**	0.83	<0.001	-4.88E-04	0.00	-	2.58
GERUB	**-2.22E-02**	0.52	<0.01	**-1.41E-01**	0.60	<0.005	-3.39E-03	0.04	-	2.33
GERRO	**-2.16E-02**	0.56	-	-2.54E-01	0.38	-	-1.79E-02	0.22	-	2.33
FINRU	**1.41E-02**	0.63	<0.005	-1.29E-01	0.16	<0.05	9.51E-04	0.07	-	1.31
FINUB	-1.59E-03	0.08	-	7.26E-02	0.20	<0.05	-2.43E-03	0.17	-	3.43
FINRO	**4.40E-03**	0.51	<0.05	-1.60E-01	0.32	<0.05	-1.29E-02	0.32	-	4.26
SPARU	-2.64E-03	0.02	-	3.80E-01	0.31	-	8.84E-04	0.02	-	0.94
SPAUB	**3.80E-02**	0.93	<0.001	7.71E-02	0.24	-	-1.25E-03	0.05	-	2.05
GRERU	**2.84E-03**	0.54	<0.001	1.01E-01	0.36	<0.005	8.48E-06	0.00	-	6.06
GREUB	-9.61E-03	0.47	<0.01	**-1.88E-01**	0.50	<0.005	-1.67E-03	0.01	-	1.87

Table 4 (continued)

Atmospheric Pressure (mbar)										
Site	**a_N**	**R^2**	**p**	**a_G**	**R^2**	**p**	**a_J**	**R^2**	**p**	**Average**
UKRU	**2.98E-03**	0.83	<0.005	**3.93E-02**	0.58	<0.005	2.56E-04	0.47	<0.05	1007.7
UKUB	**1.33E-03**	0.50	-	1.17E-02	0.05	<0.05	5.91E-05	0.04	-	1011.7
UKRO	**3.86E-03**	0.95	<0.001	-1.21E-01	0.40	-	-1.12E-03	0.17	-	1012
GERRU	**8.72E-03**	0.97	-	**8.95E-02**	0.85	<0.001	1.98E-03	0.21	-	1007.0
GERUB	**1.10E-02**	0.97	-	**4.00E-02**	0.76	-	2.04E-03	0.37	<0.05	995.5
GERRO	**4.11E-03**	0.79	-	-9.61E-02	0.43	-	-3.86E-03	0.21	-	995.5
FINRU	**3.01E-03**	0.88	<0.001	**2.90E-02**	0.57	<0.001	1.25E-04	0.14	-	985.1
FINUB	**1.31E-03**	0.55	<0.005	-3.57E-03	0.02	-	1.09E-04	0.05	-	1004.4
FINRO	**2.50E-03**	0.70	-	-2.67E-02	0.17	-	9.92E-04	0.26	-	1008.8
SPARU	-2.42E-03	0.09	-	4.79E-02	0.14	-	4.45E-04	0.08	-	939.3
SPAUB	-3.71E-03	0.44	<0.05	1.86E-02	0.08	-	3.56E-04	0.21	-	1006.3
GRERU	3.90E-03	0.46	<0.001	-1.50E-01	0.73	-	3.99E-04	0.33	-	1014.5
GREUB	8.01E-04	0.10	<0.05	-1.00E-01	0.71	-	6.97E-04	0.04	-	1015.7

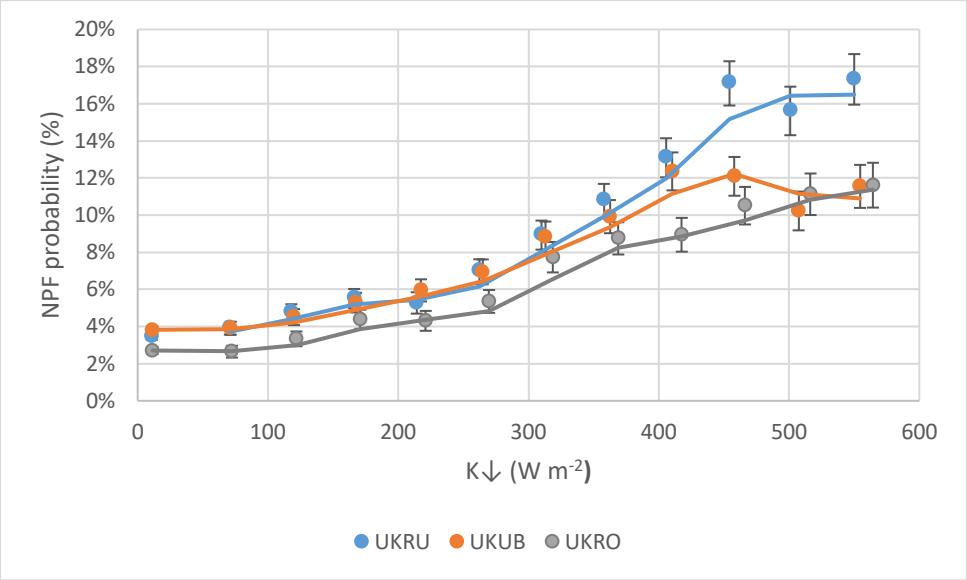
Solar radiation intensity

As mentioned earlier, solar radiation is considered as one of the most important variables in NPF occurrence, as it contributes to the production of H_2SO_4 which is a main component of the initial clusters and participates in the early growth of the newly formed particles. Hidy et al. (1994) reported up to six times higher SO_2 oxidation rates into H_2SO_4 in typical summer conditions compared to winter. For almost all sites this relationship is confirmed with very strong correlations between the intensity of solar radiation and the probability for NPF events when a linear relationship is considered. The correlation between the solar radiation and NPF probability was positive at all sites and only three sites (FINUB, SPARU and GREUB) presented weak correlations (R^2 below 0.40). Weaker correlations were found for the southern European sites, which might be associated with the higher averages for solar radiation, or the interference of other processes (such as coinciding with increased CS by recirculation of air masses (Carnerero et al., 2019), possibly making it less of an important

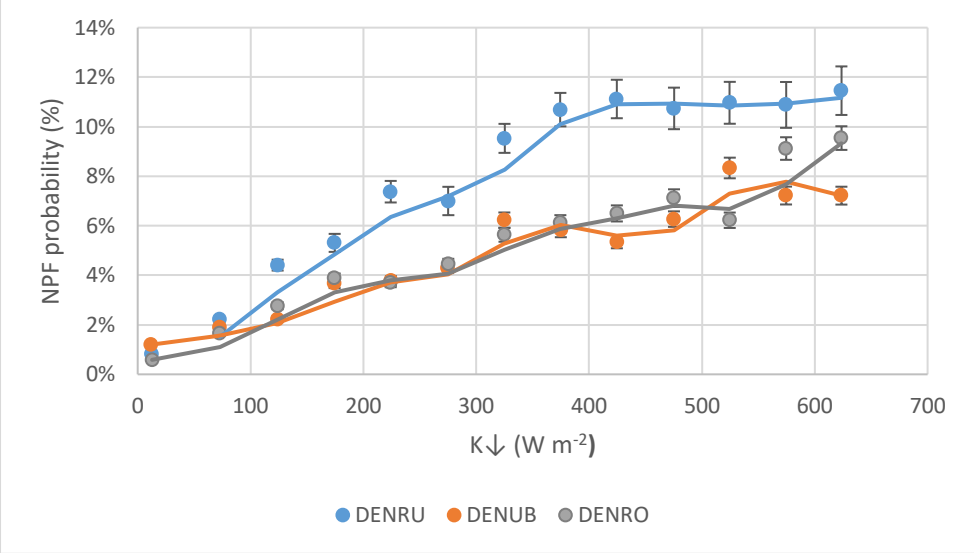
factor for these areas. For most sites this positive relationship tends to flatten or even become negative (such as the Finnish sites) for high solar radiation intensities, making it a less important factor on extreme values.

The relationship of solar radiation to the growth rate was weaker in all cases and did not present a clear pattern which might indicate a non-linear response. A few sites presented a strong correlation when linear relationships were considered, which in all cases were background sites (either rural or urban). The relationship found in most cases was positive apart from two roadsides and GREUB, though due to the low R^2 these results cannot be used with confidence. It seems though that the solar radiation intensity is probably a more important factor at background sites than at roadsides, where possibly local conditions (such as local emissions) are more important. Finally, the formation rate has a positive relationship with the solar radiation intensity, with strong correlations in most areas when linear relationships are considered. The correlations were stronger at the rural background sites compared to the roadsides, which further underlines the increased importance of this factor at this type of site. A negative correlation between the solar radiation intensity and the formation rate was found at the GRERU site when a linear relationship was considered, but the R^2 is very low.

Figure 14: Relationship of solar radiation with NPF variables.



(a)



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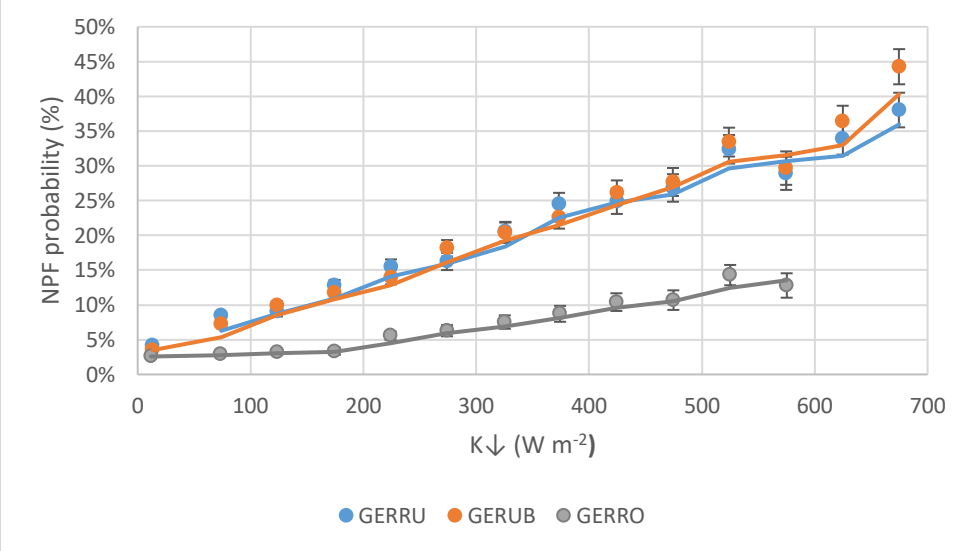

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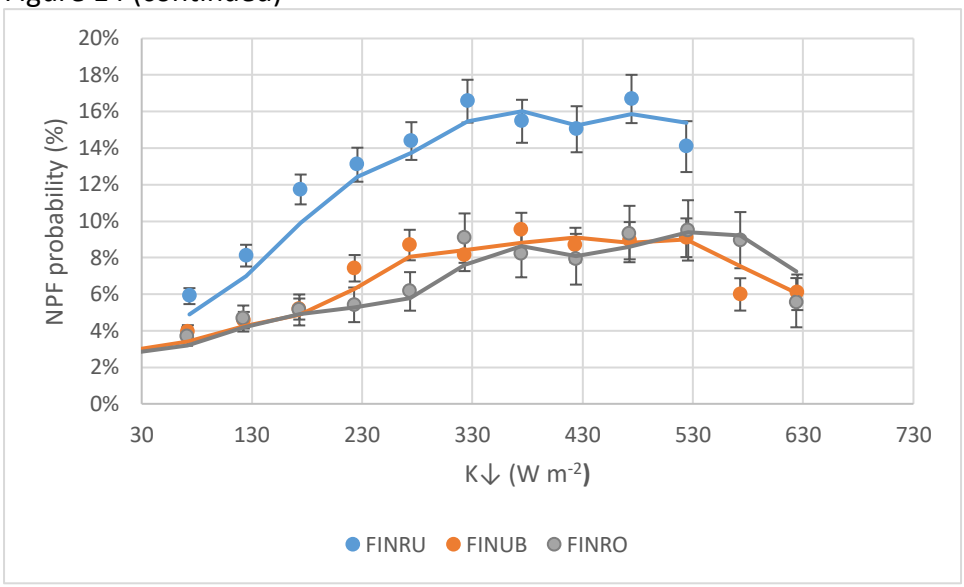

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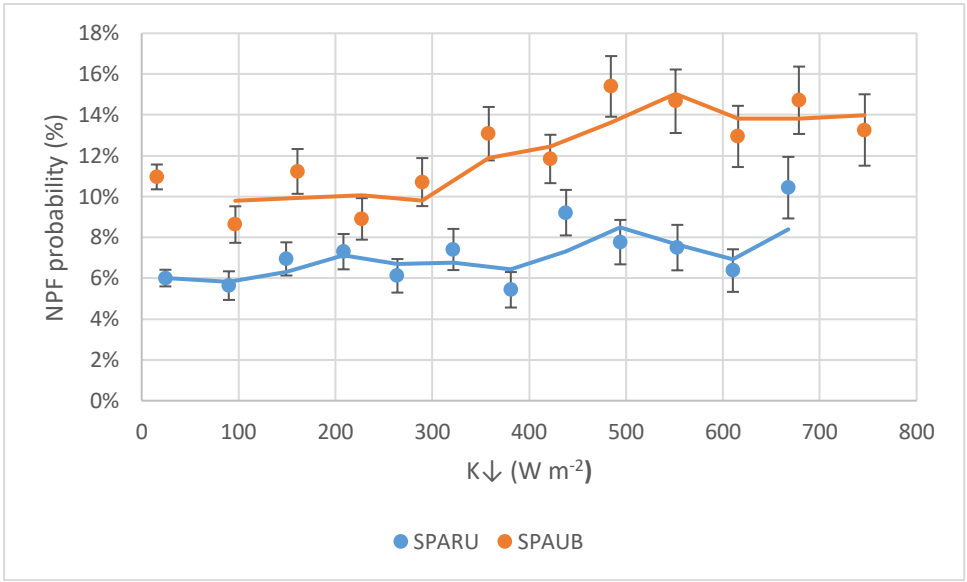

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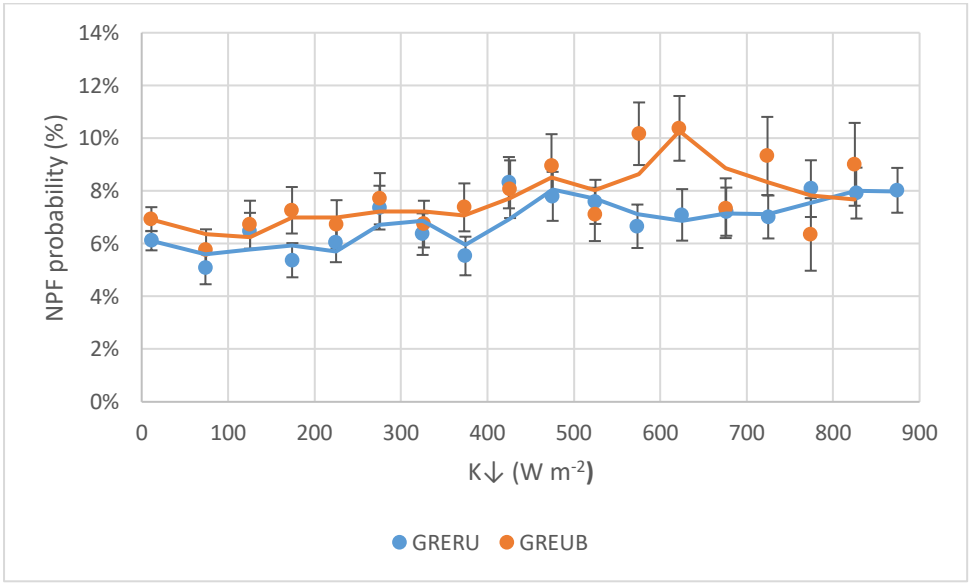

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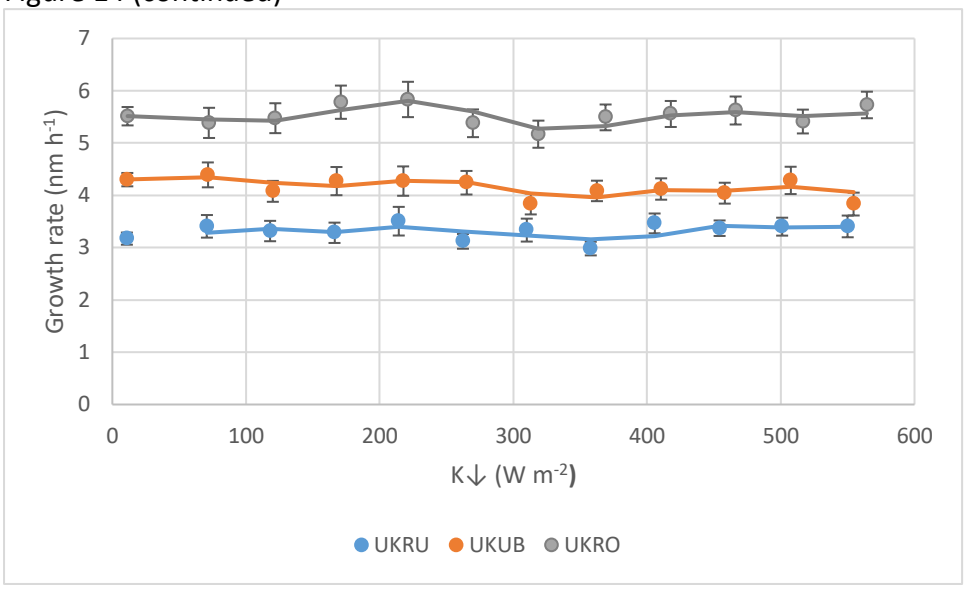

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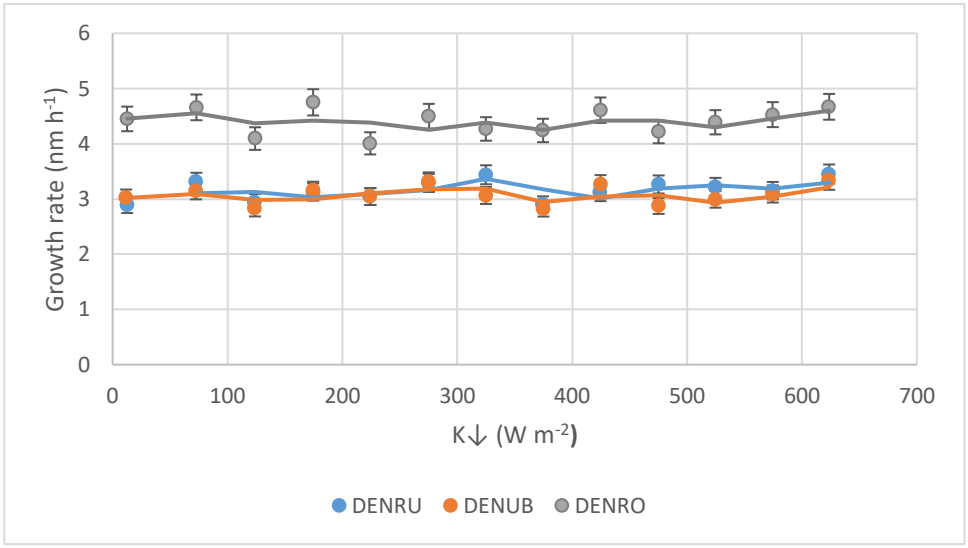

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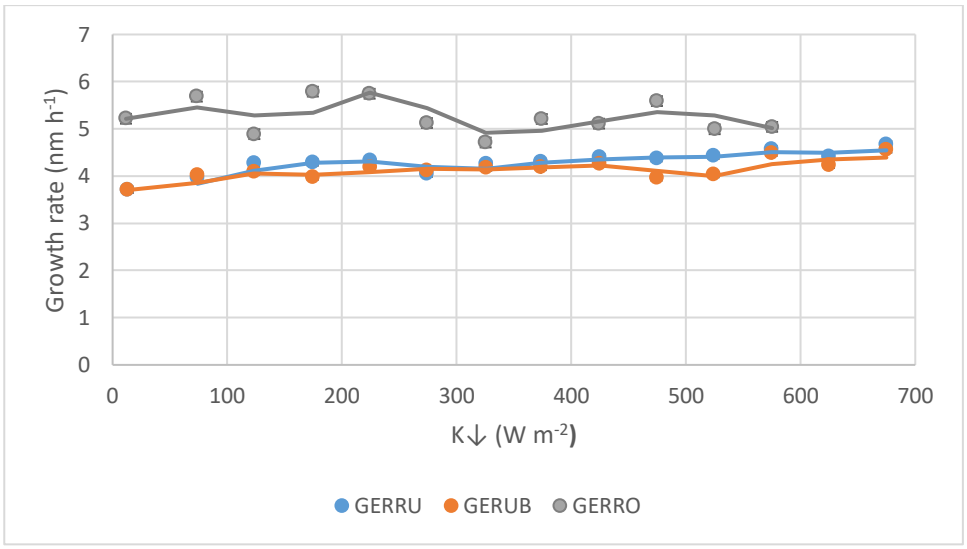

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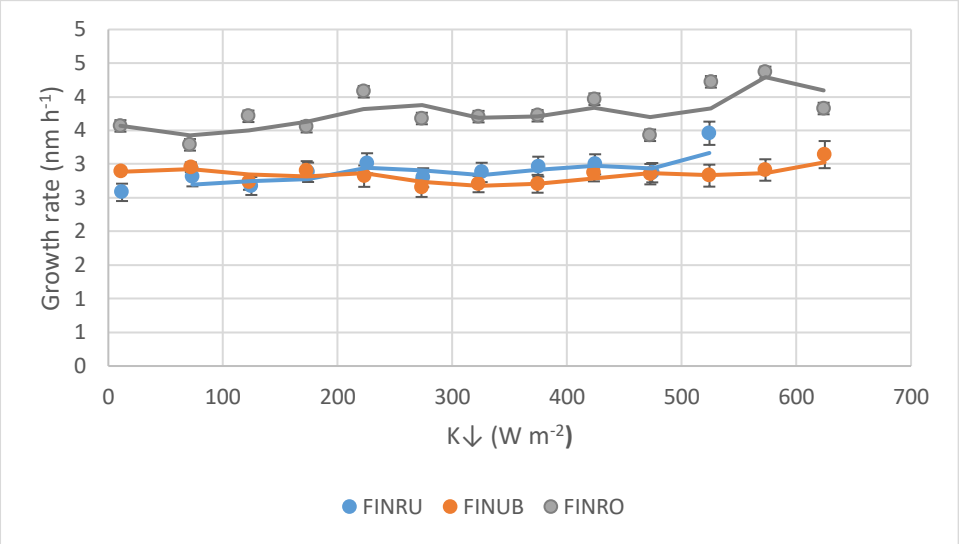

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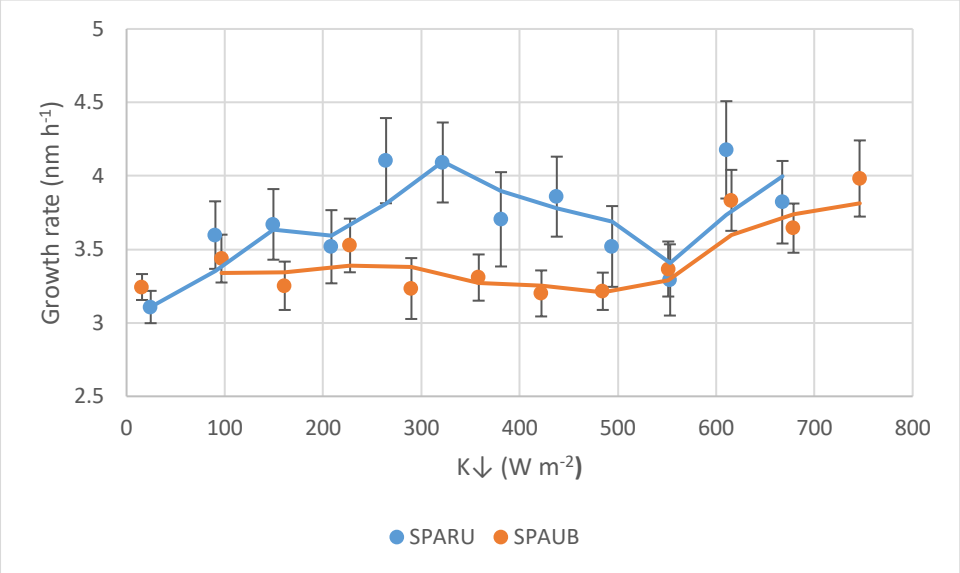

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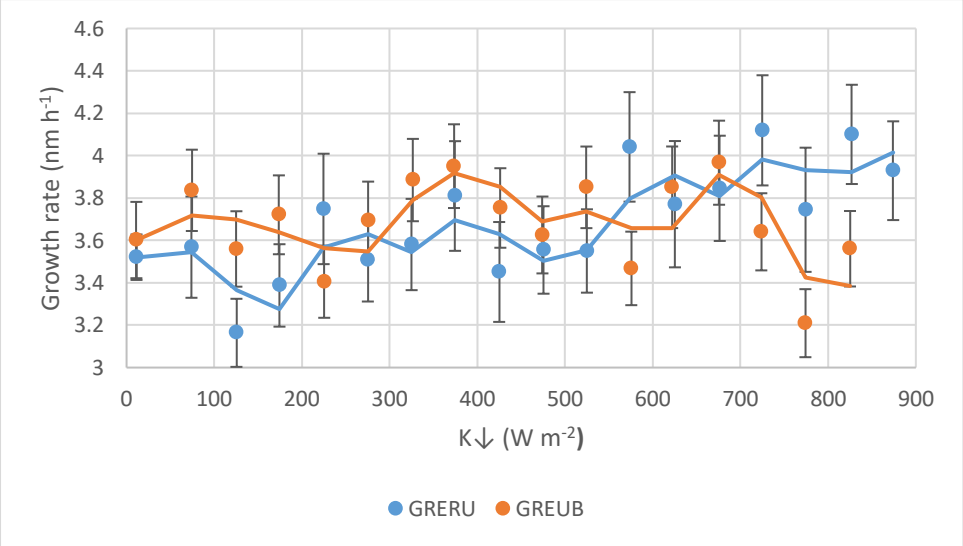

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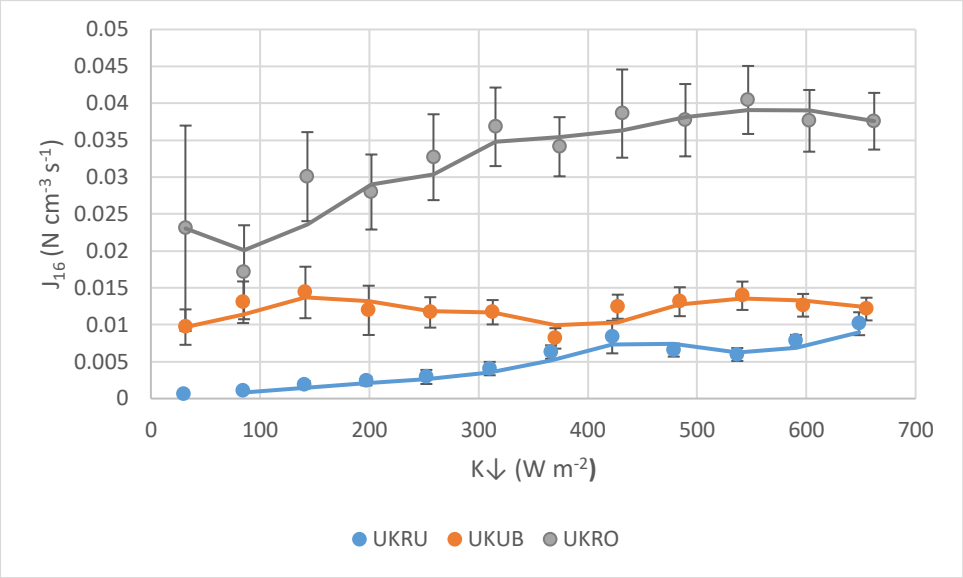

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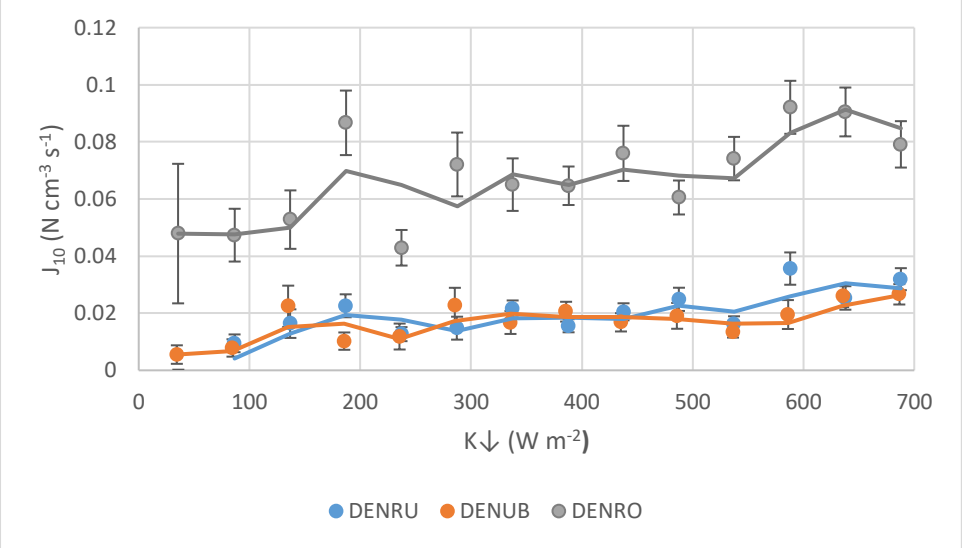

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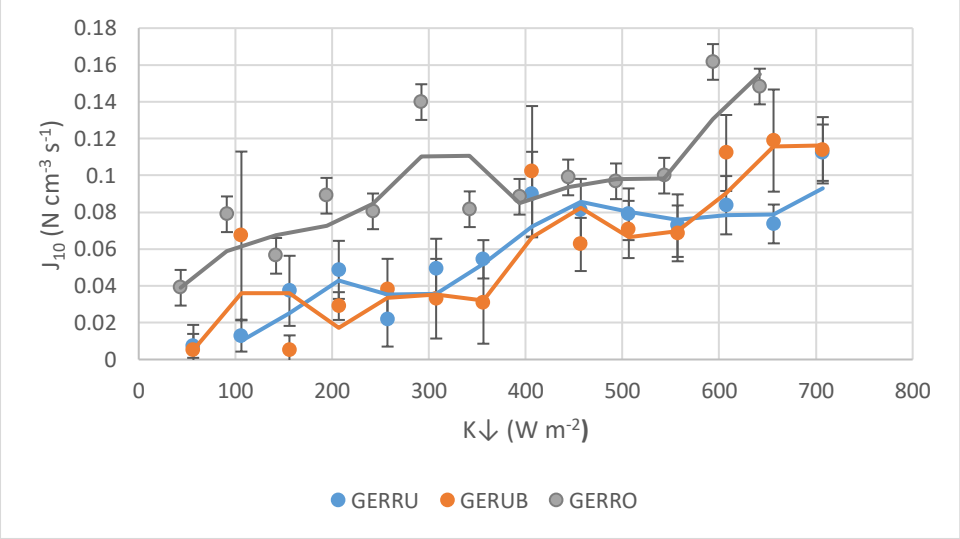
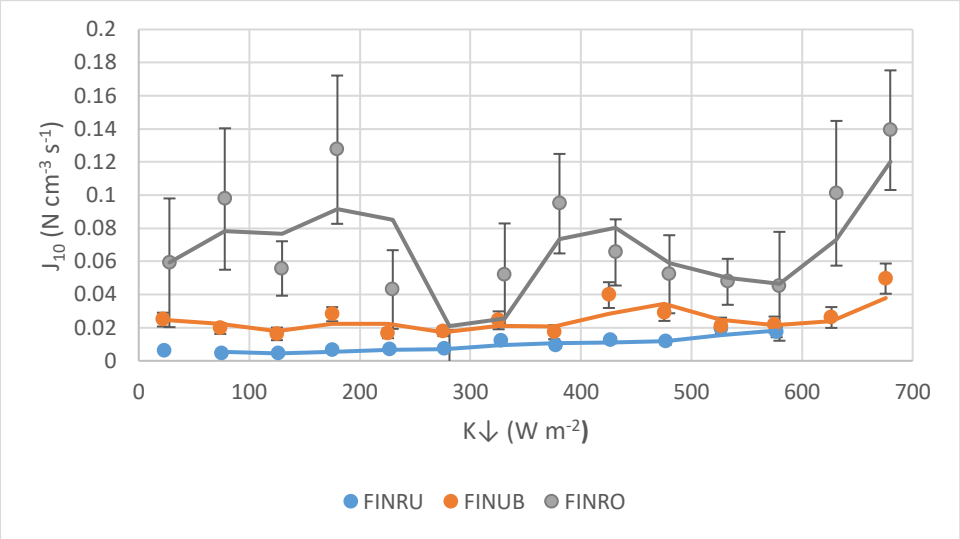


(o)

Figure 14 (continued)



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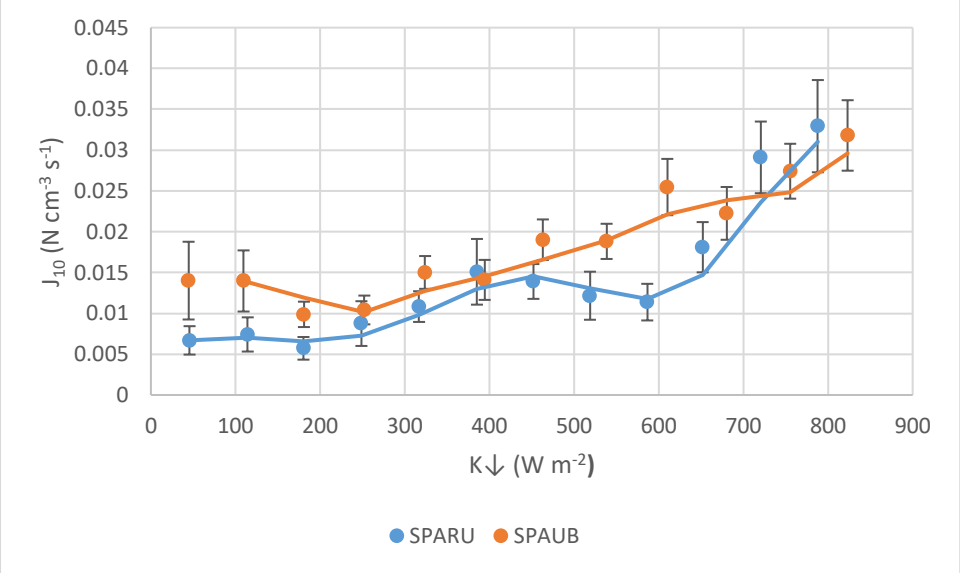
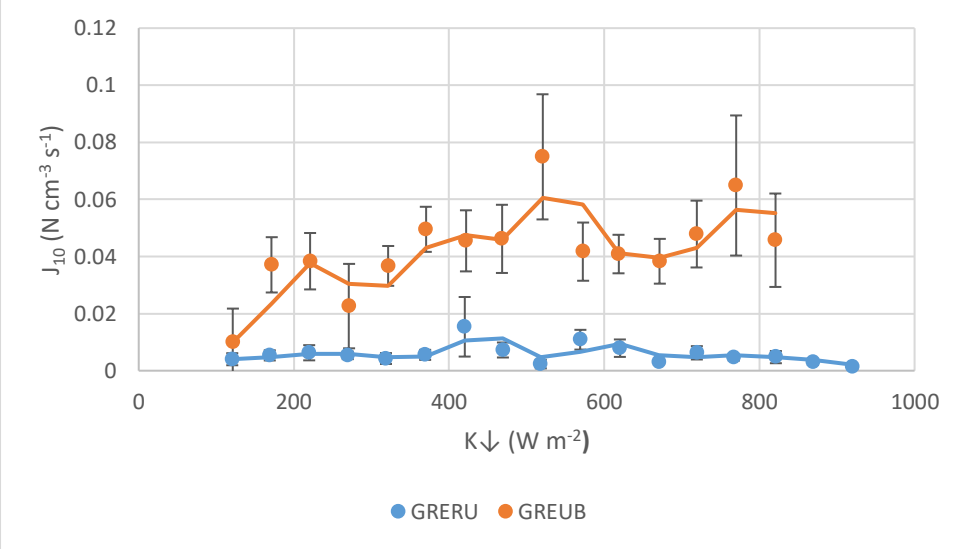


(q)



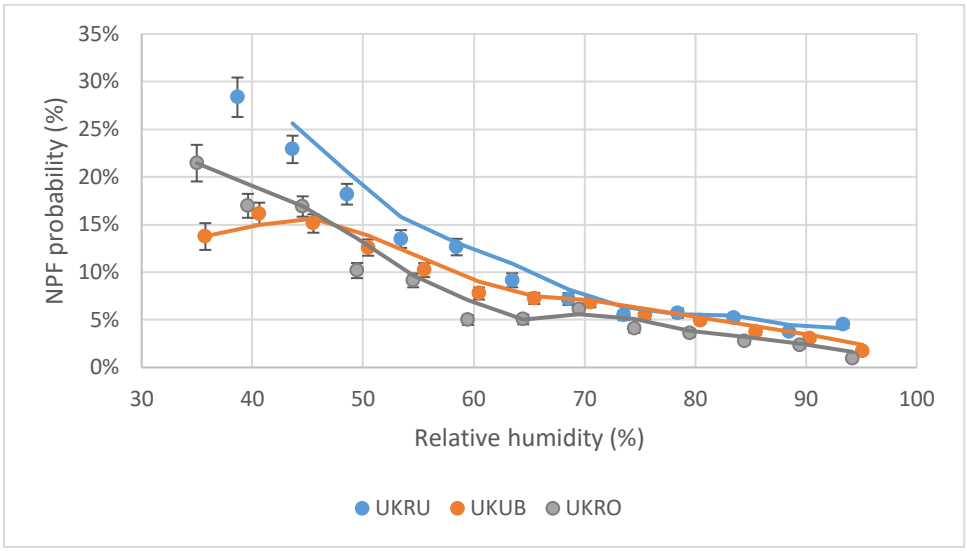
(r)

Relative humidity

Relative humidity is considered to have a negative effect on the occurrence of NPF events (Jeong et al., 2010; Hamed et al., 2011; Park et al., 2015; Dada et al., 2017; Li et al., 2019). While water in the atmosphere is one of the main compounds needed for the formation of the initial clusters both in the binary or ternary nucleation theory (Korhonen et al., 1999; Mirabel and Katz, 1974), in atmospheric conditions it may also play a negative role, suppressing the number concentrations of new particles by increasing aerosol surface area. Consistent with this, a negative impact of RH with NPF probability was found for all the sites of this study with very high R^2 for almost all of them, when linear relationships were considered. This is not simple to interpret as solar radiation, temperature, RH and CS are not independent variables, since an increase in temperature of an air mass due to increased solar radiation will be associated with reduced RH, which in turn affects the CS. The sites in Greece specifically, presented lower R^2 compared to the other sites. This is because both sites in Greece presented a positive relationship for low RH values, which peaked at about 40 – 50 %. For greater values of RH a clear negative relationship is found for the Greek sites as well. Growth rate on the other hand had a variable relationship, either positive or negative, with only a handful of background sites having strong correlations. Among these the German background sites as well as FINRU, which were among the sites with the highest average RH (average RH for GERRU is 81.9%, GERUB is 78.7% and FINUB is 80.1%) presented a negative relationship between the RH and growth rate, while DENRU (average RH at 75.7%) had a positive relationship, which might indicate that the relationship between these two variables may vary depending upon the RH range. Formation rate also appears to have a negative relationship with the RH, though this relationship was significant ($R^2 > 0.40$) for only 6 sites when linear relationships were considered, which once again in most cases are sites with

higher average RH. Along with the results of the growth rate, this might indicate that the RH becomes a more important factor in the development of NPF events as its values increase.

Figure 15: Relationship of relative humidity with NPF variables.



(a)

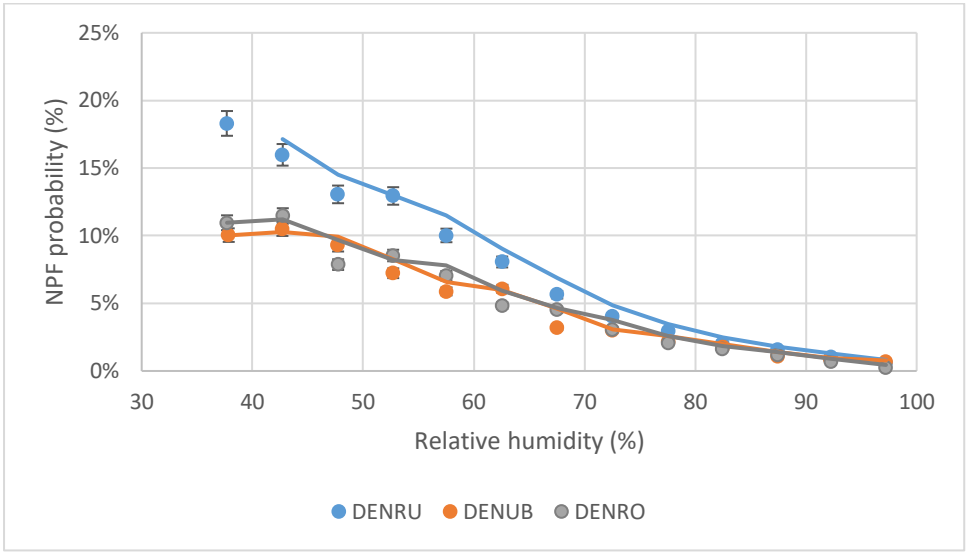


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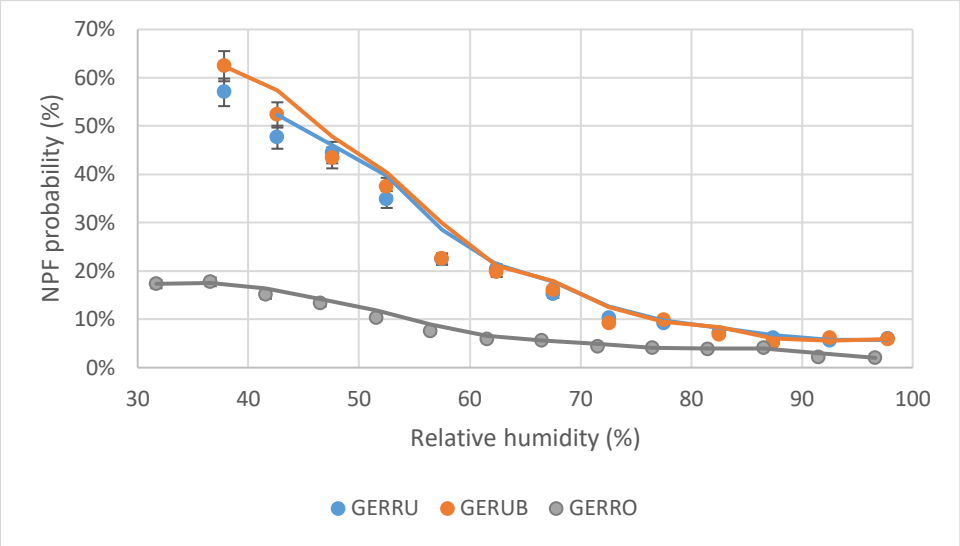
Figure 15 (continued)



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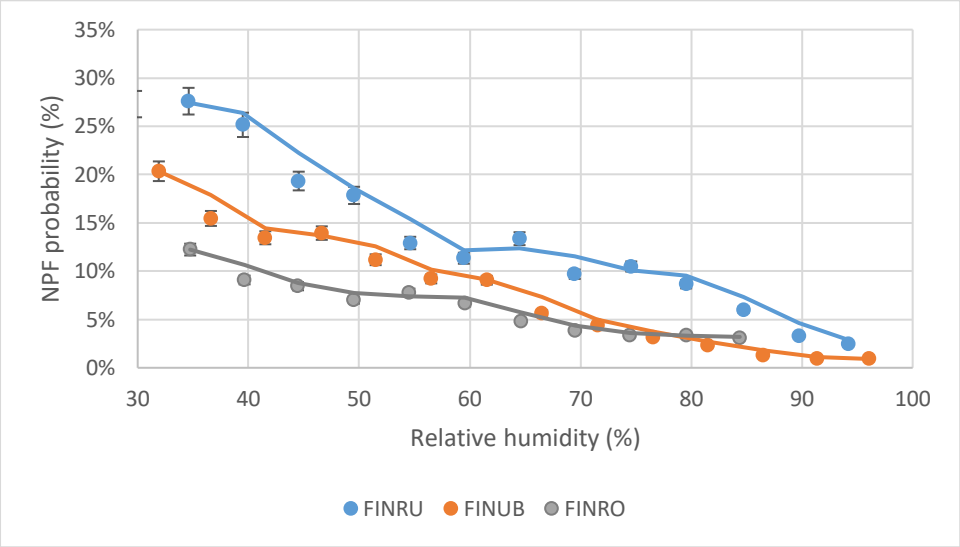

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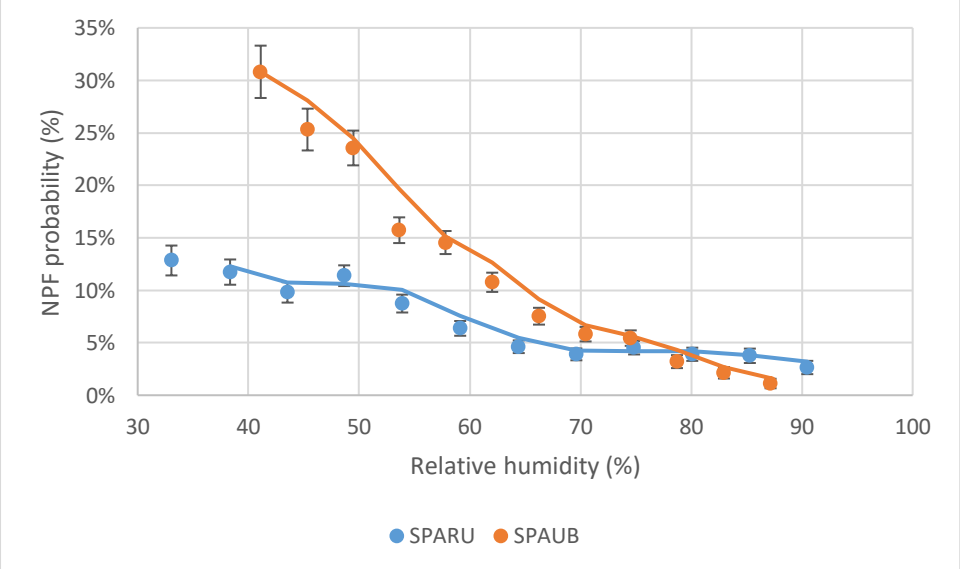
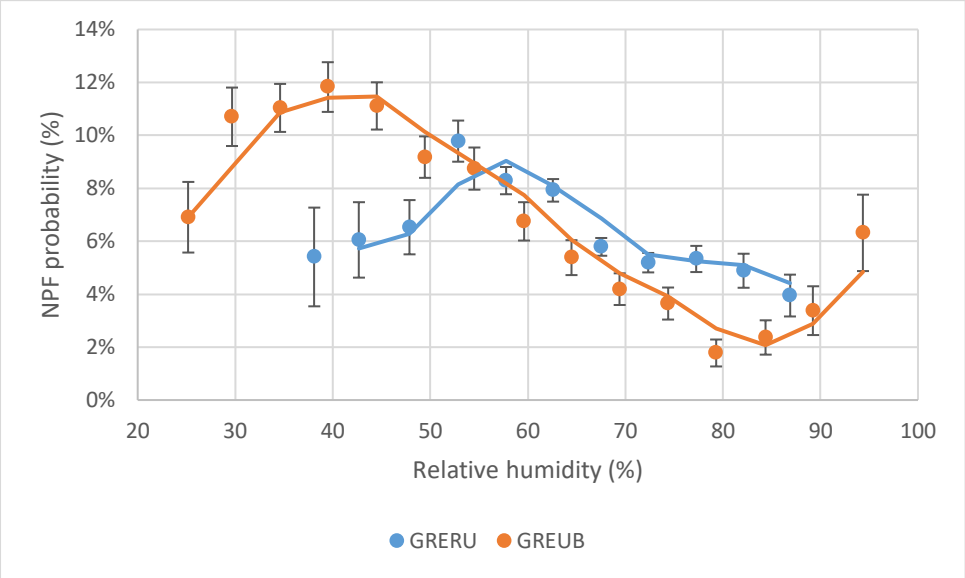


(e)

Figure 15 (continued)



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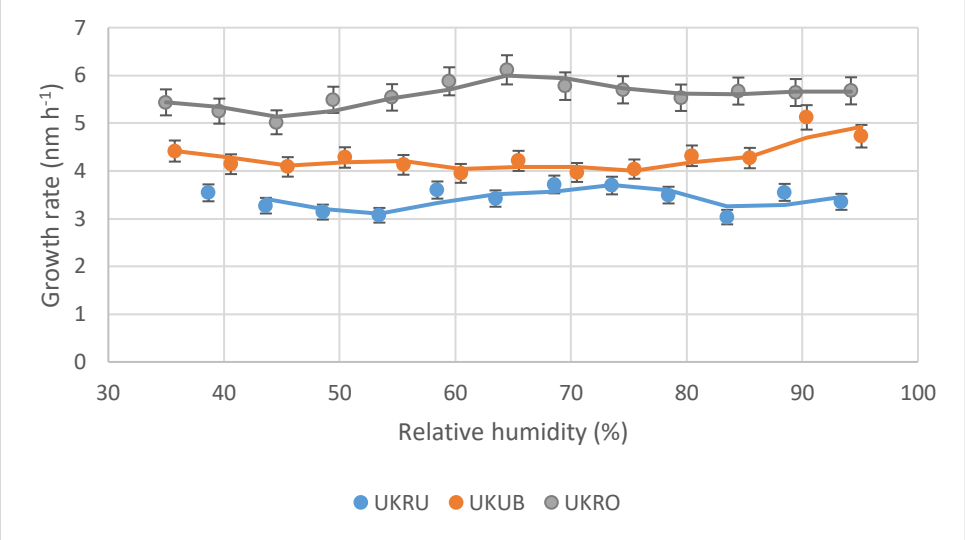

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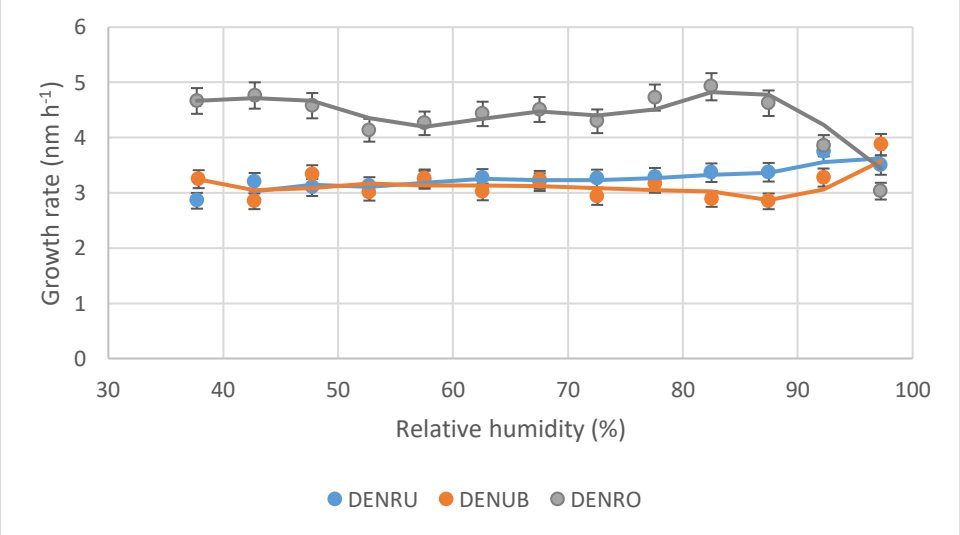

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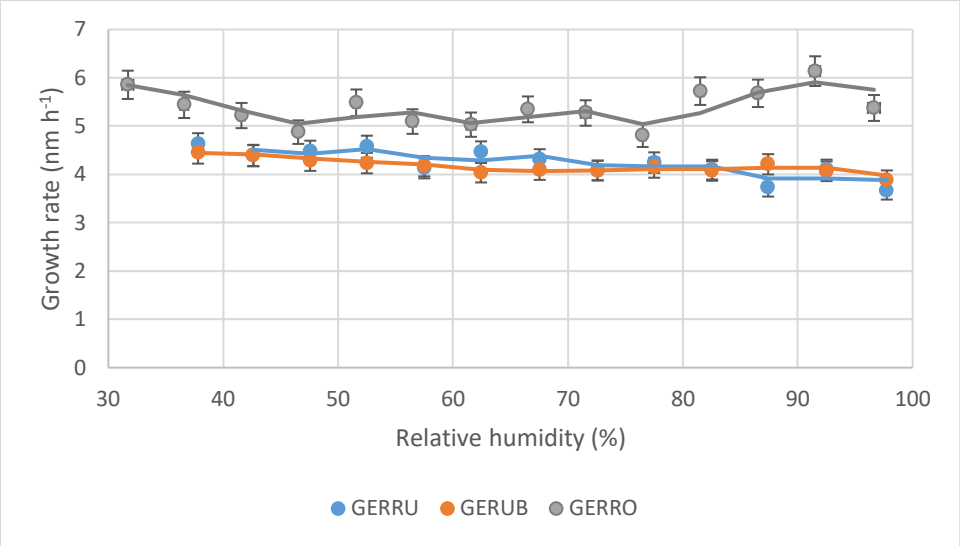

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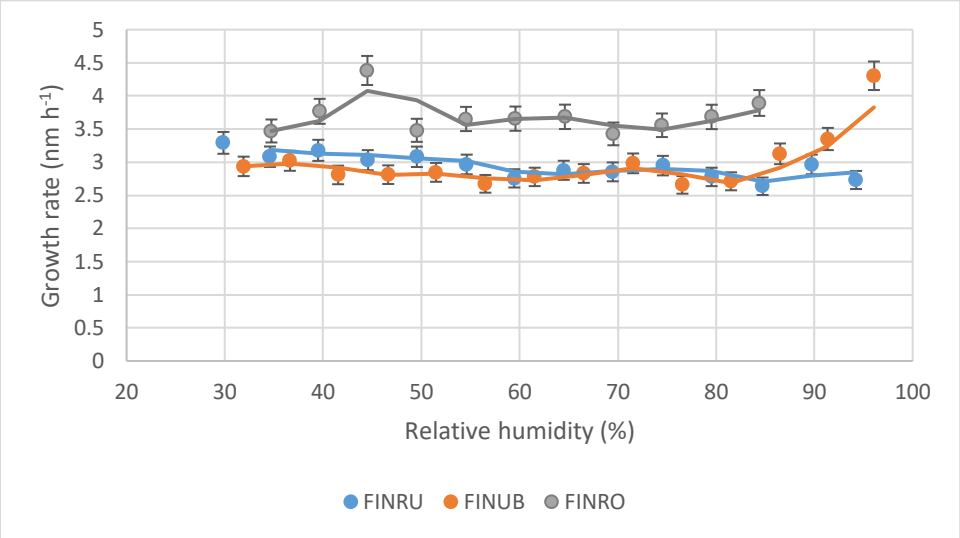

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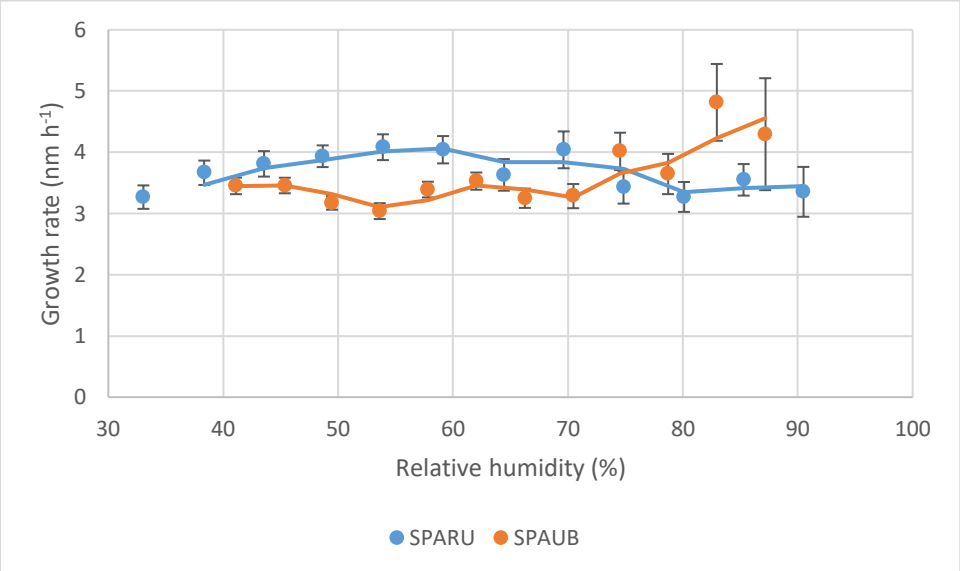

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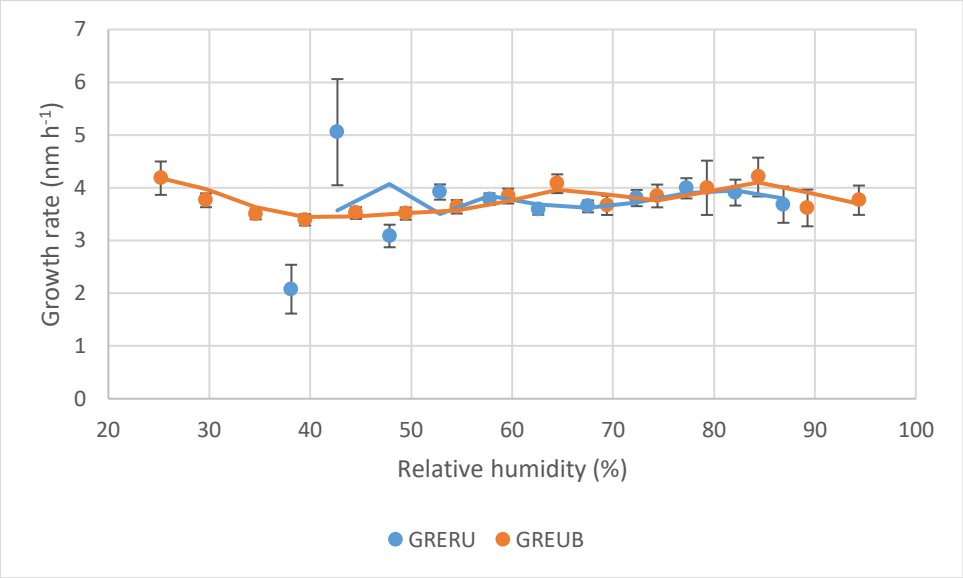

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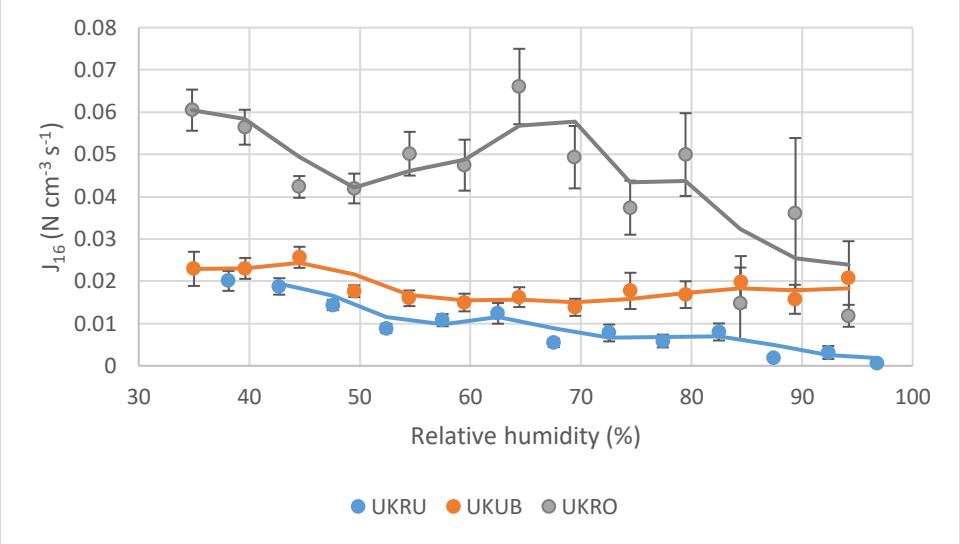

(m)

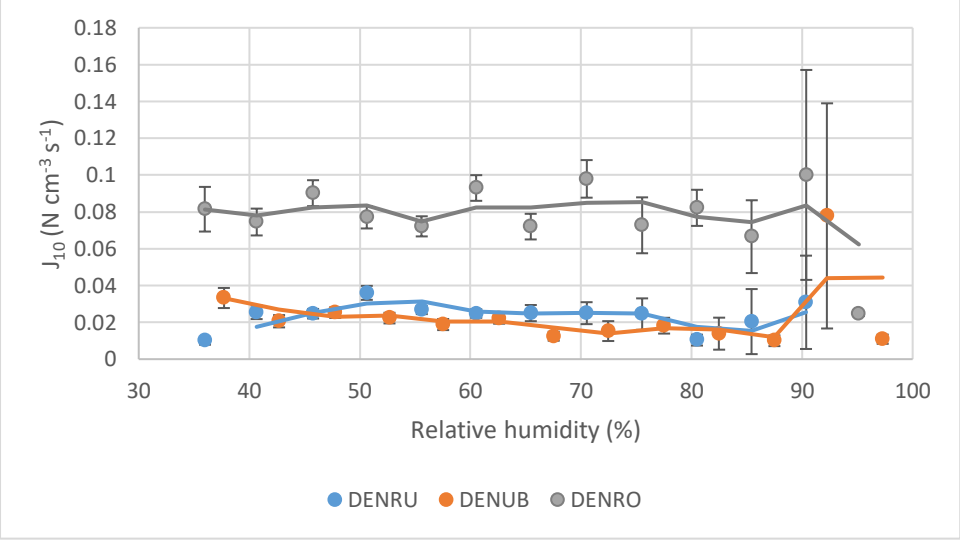

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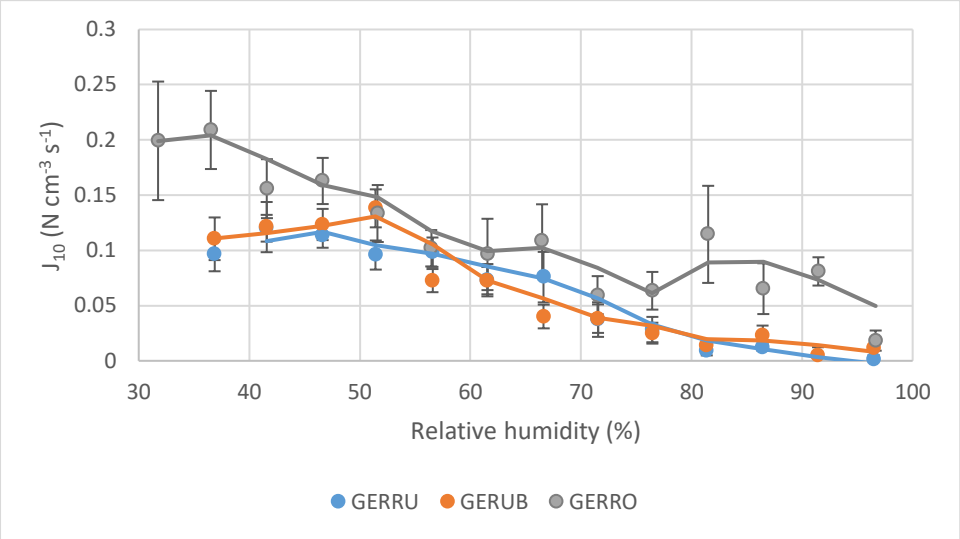
Figure 15 (continued)



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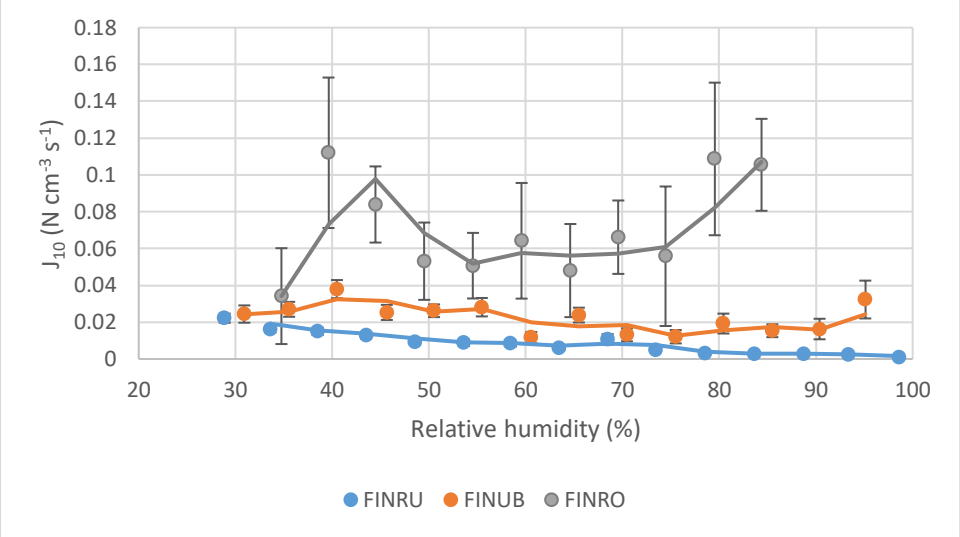

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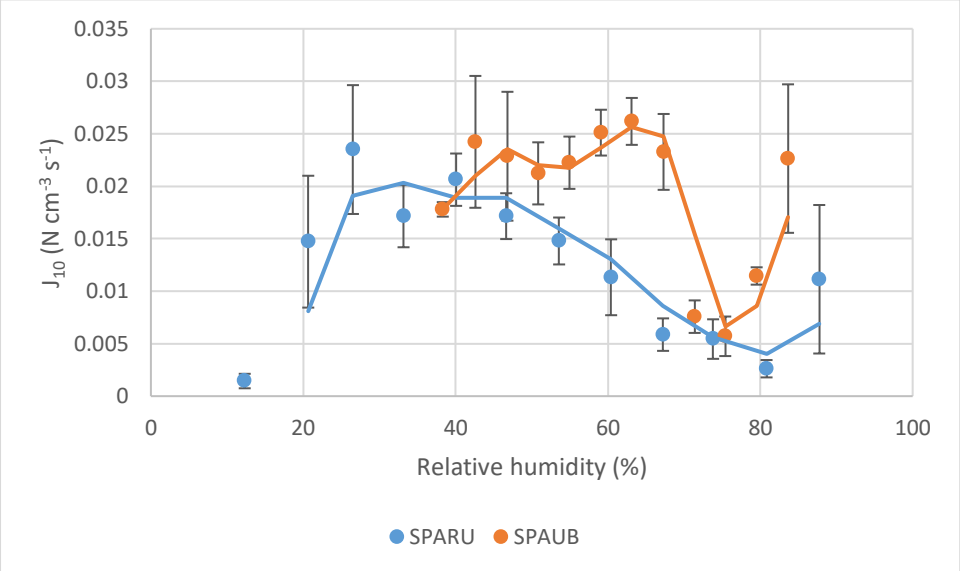

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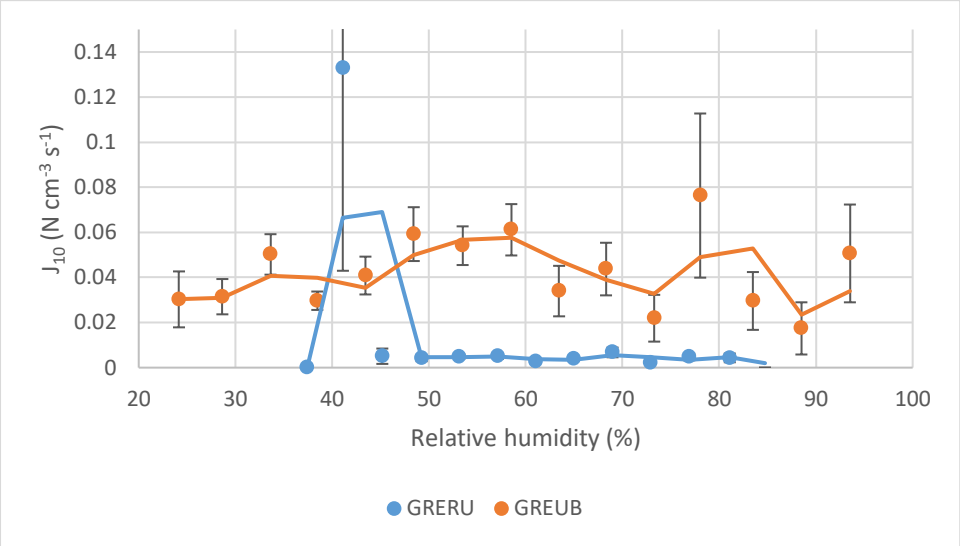
Figure 15 (continued)



(r)

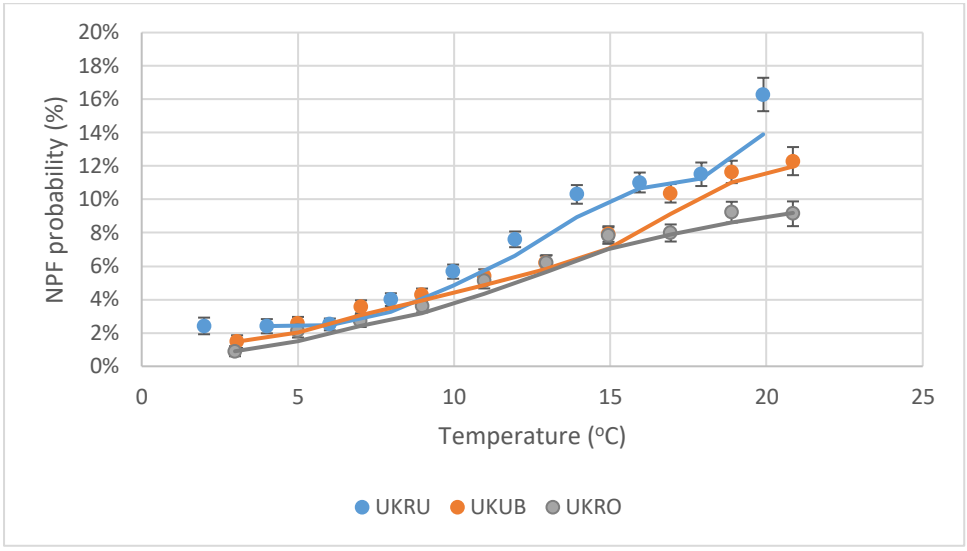
Temperature

Temperature can have both a direct and indirect effect in the development of NPF events, as it is directly associated with the abundance of biogenic volatile carbon which is an important group of compounds whose oxidation products can participate in nucleation itself (Lehtipalo et al., 2018; Rose et al., 2018), as well as in the growth of newly formed particles, while it may affect the particle size distributions or number concentrations through other processes such as particle evaporation. Most of the sites of the present study presented a strong association between NPF probability with temperature, which in most cases was positive, though in many cases (such as the Danish, Finnish and Spanish sites – figures 16b, d and e) there seems to be a peak in the NPF probability at some temperature, after which a decline starts. Sites with smaller R^2 when linear relationships were considered (weaker association with temperature), were mainly those that have a seasonal variation that favoured seasons other than summer as found in the previous studies. These sites not only had weaker relationship of NPF probability with temperature, but in most cases had a negative relationship (background sites in Finland, Spain and Greece). The Finnish sites, having the

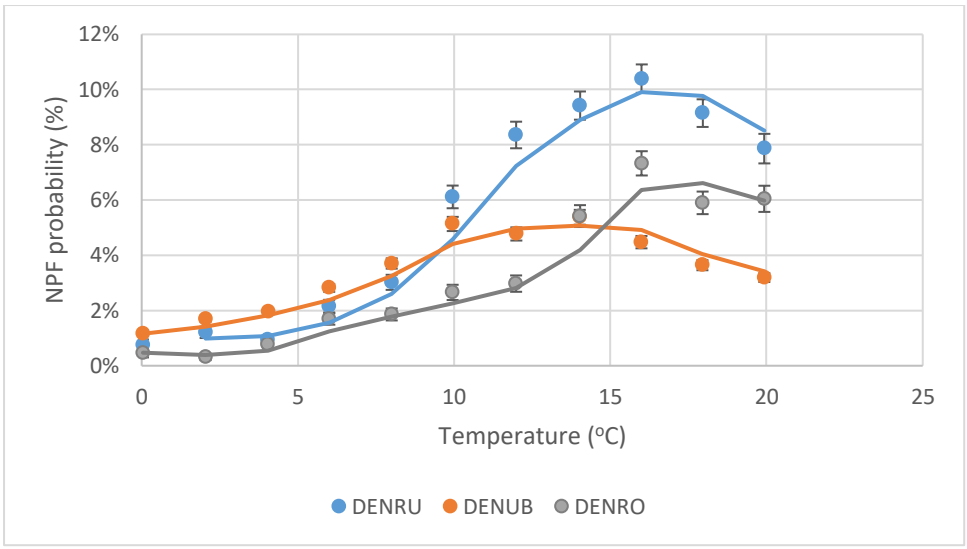
lowest average temperatures and a sufficient amount of data below zero temperature, show at all three sites the possible presence of a peak in the NPF event probability for temperatures below zero (Figure 16d). This seems to be the cause of the weak relationships found there and they seem to be associated with the formation rate J_{10}, which also seems to have an increasing trend below zero degrees (Figure 16p). This may be the result of increased stability of molecular clusters at lower temperatures, as well as the possible enhancement of growth mechanisms in lower temperatures (below 5^{o}C) by other chemical compounds in the atmosphere (i.e. nitric acid and ammonia) as found by Wang et al., (2020). Laboratory experiments show that the characteristics of organic aerosol forming from alpha-pinene is governed by gas phase oxidation (e.g. Ye et al. 2019). In the real atmosphere, the higher temperature enhances the amount of biogenic vapours (e.g. Paasonen et al. 2013), and although the oxidation can be more efficient in higher temperatures, the lower temperatures favour formation of more non-volatile compounds (Ye et al. 2019; Stolzenburg et al. 2018).

Growth rate had a more uniform association, with almost all sites having a positive relationship with temperature (apart from GERRO though with $R^2 = 0.00$) when linear relationships were considered. This relationship was very strong for most sites, which is also confirming the summer peak found for the growth rate at most of these sites. A strong relationship with temperature was also found for the formation rate for most sites and was positive for almost all sites (apart from FINRO with $R^2 = 0.01$ and the Greek sites). As with the NPF probability, in general the sites with a seasonal variation of events that favoured summer had the strongest correlation (high R^2) of the formation rate with temperature, which might indicate that this variable, either through its direct or indirect effect is an important one for the seasonal variability of NPF events in a given area.

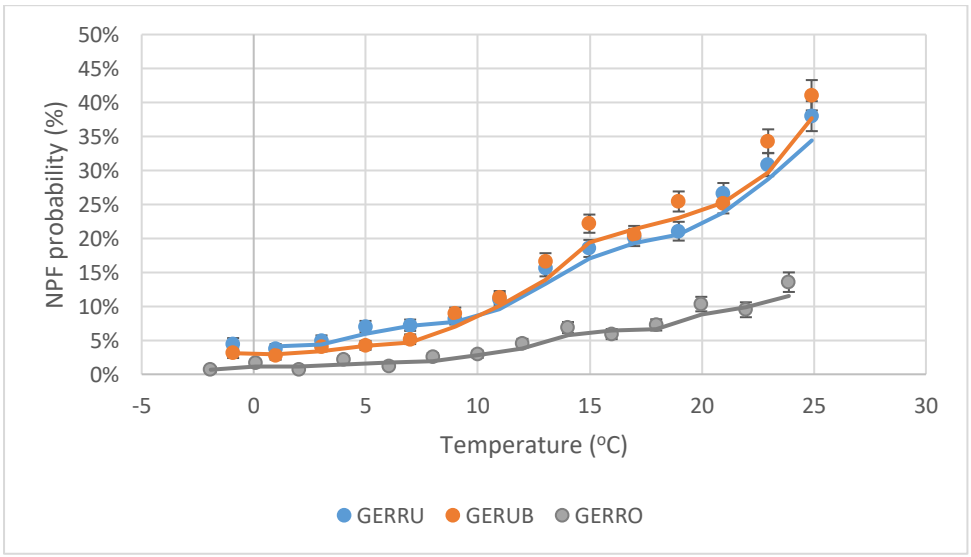
Figure 16: Relationship of temperature with NPF variables.



(a)

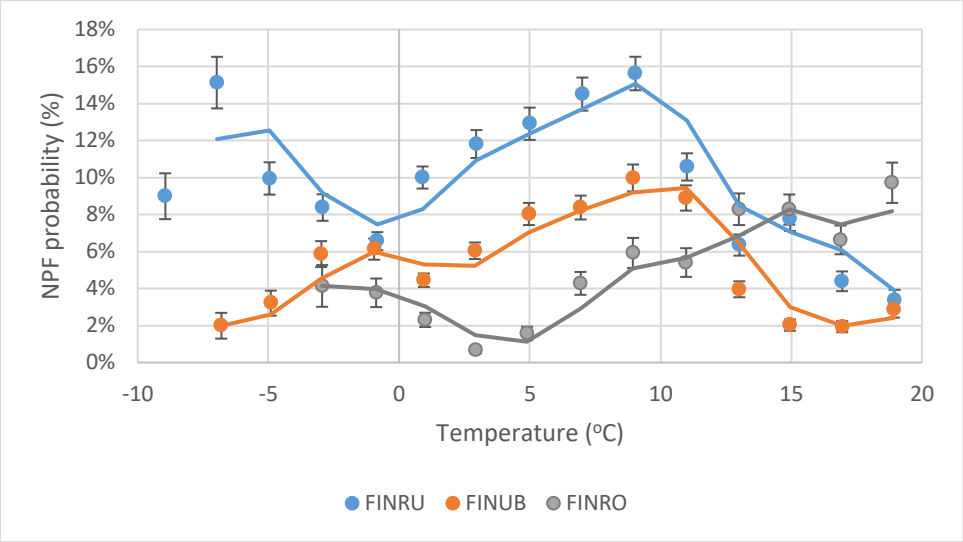


(b)



(c)

Figure 16 (continued)



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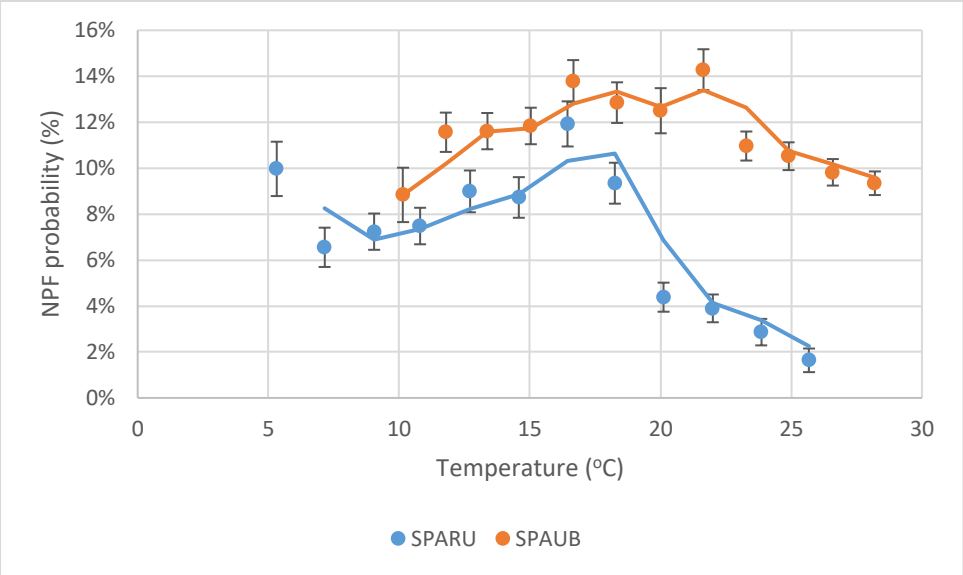

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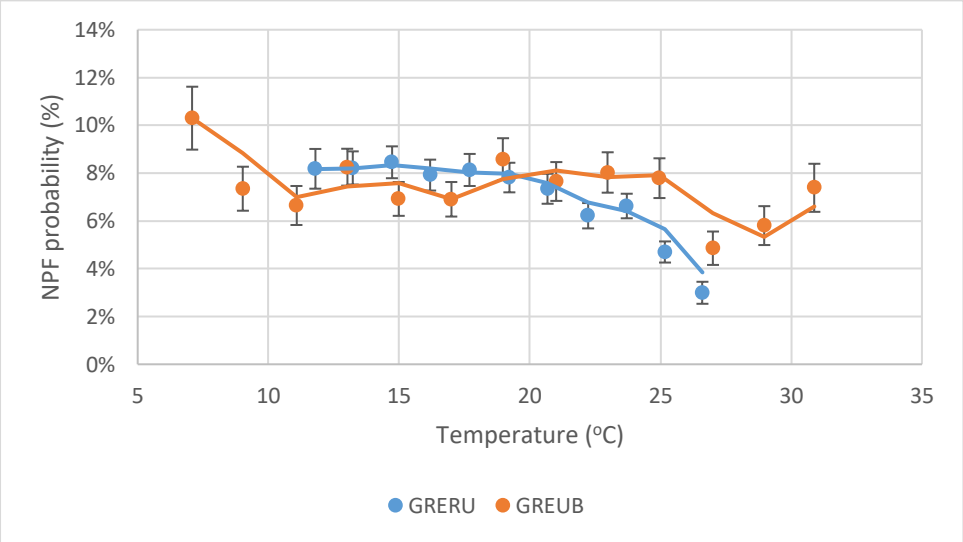

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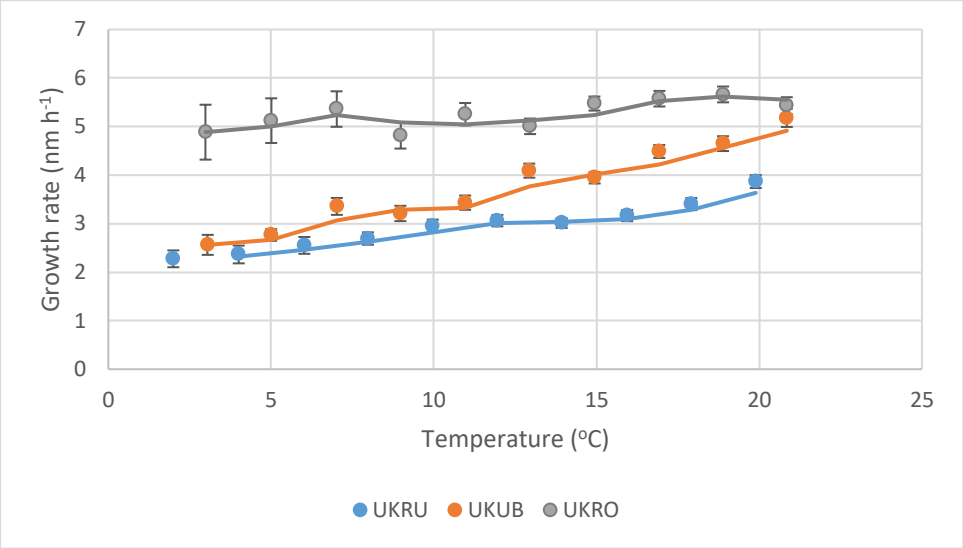

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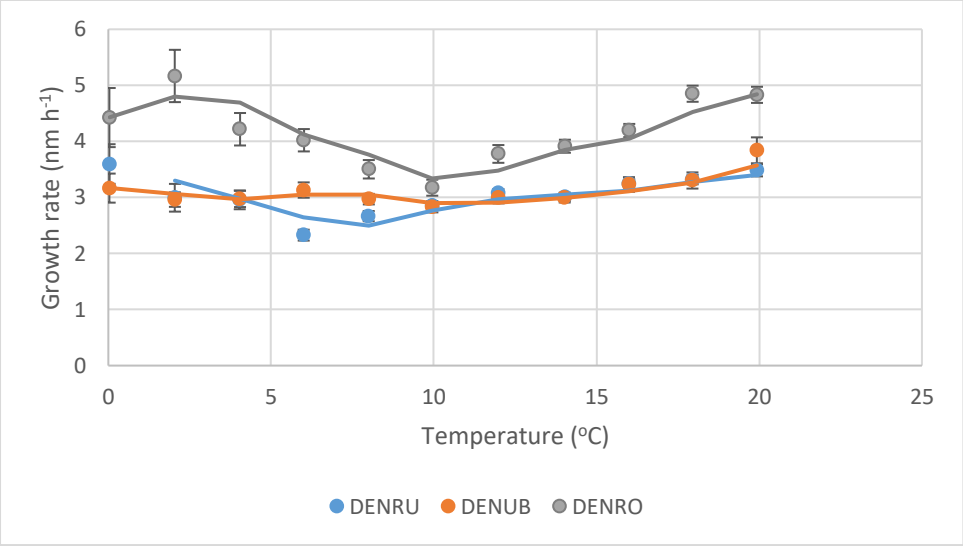
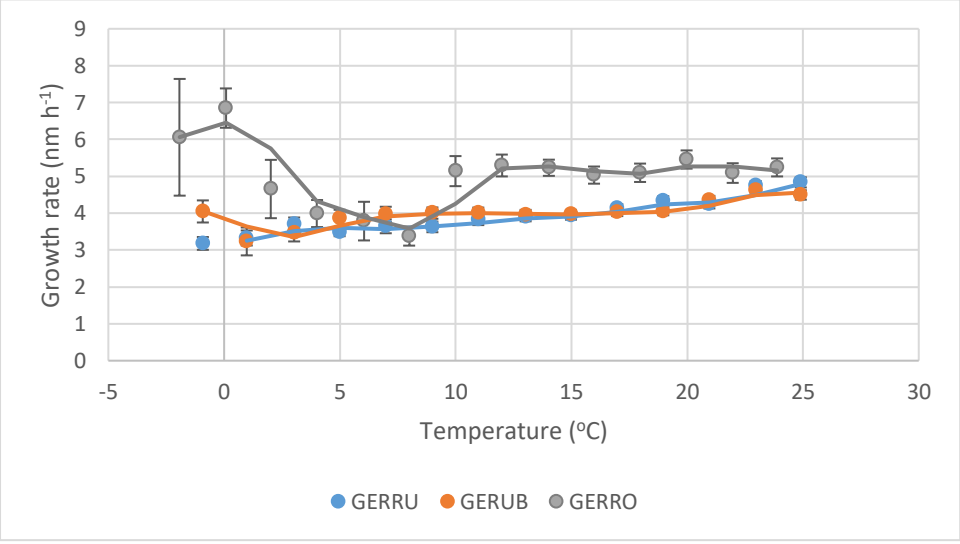


(h)



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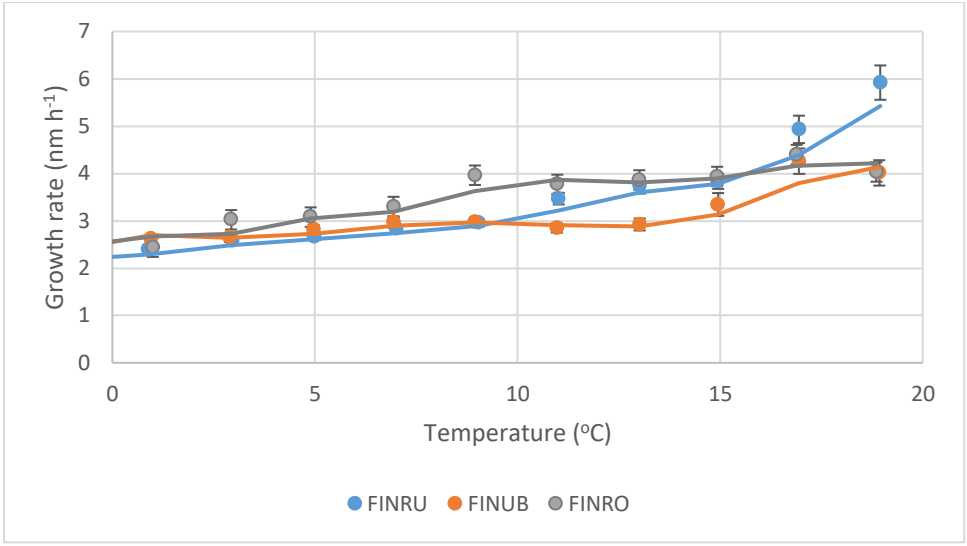

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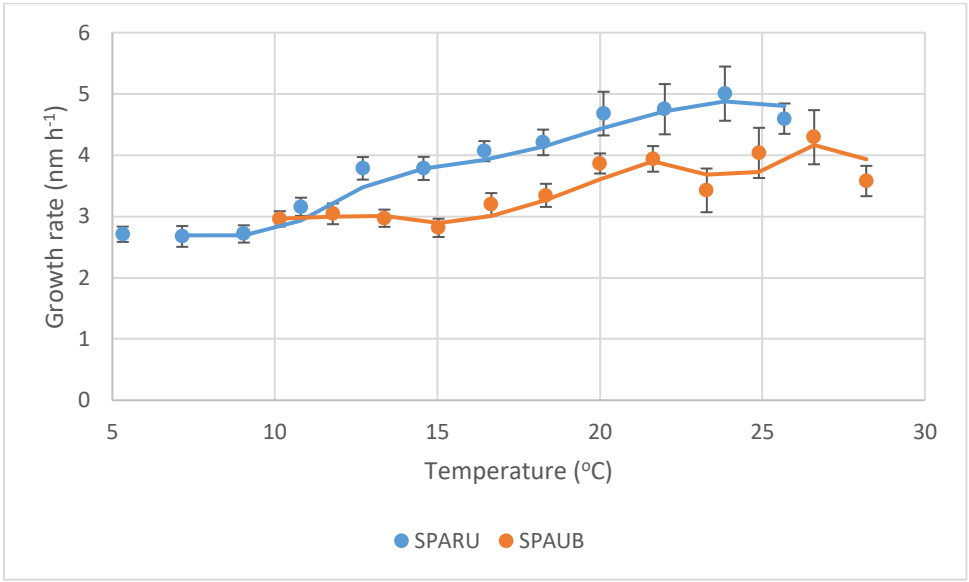

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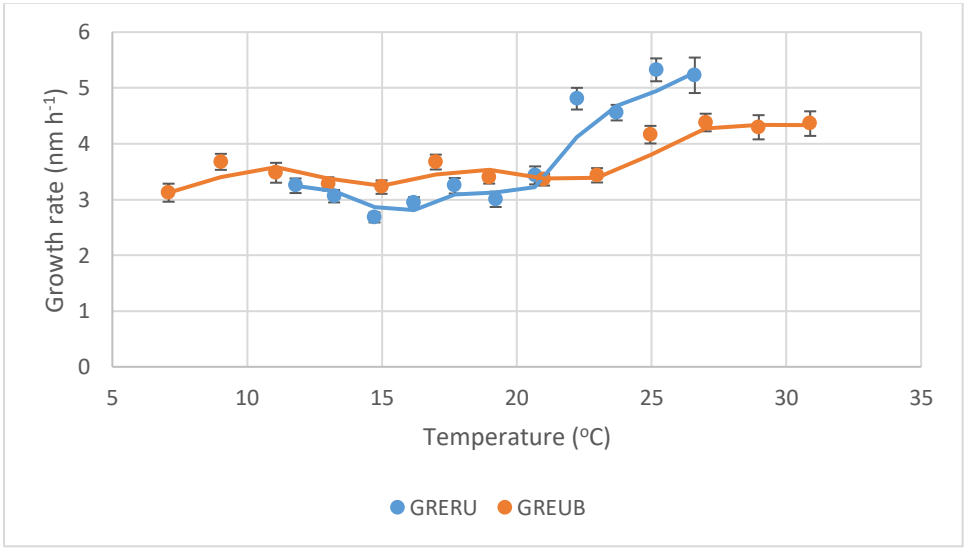

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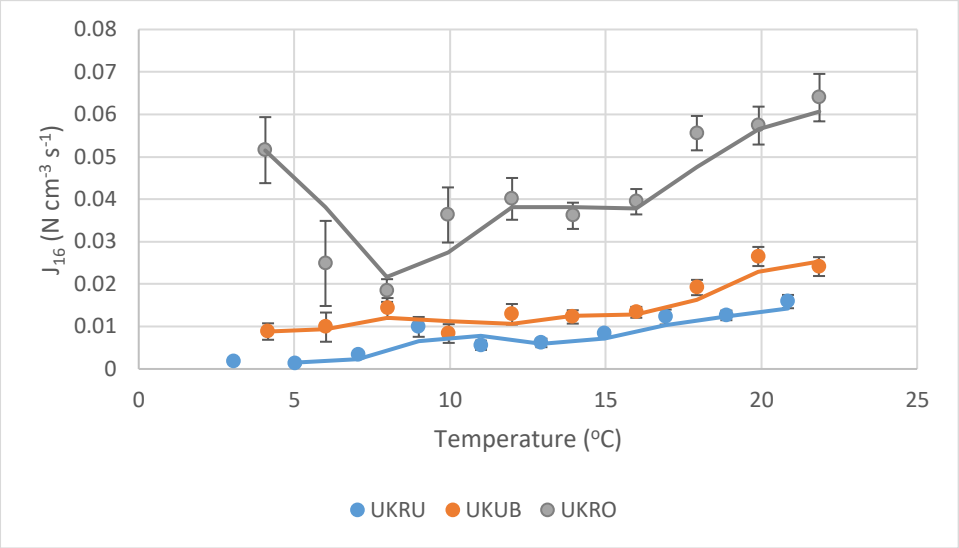

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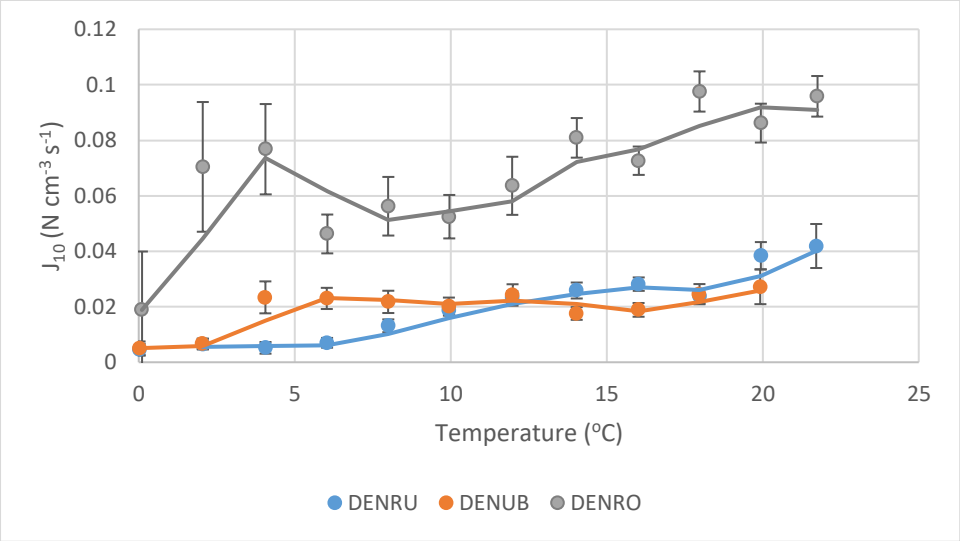

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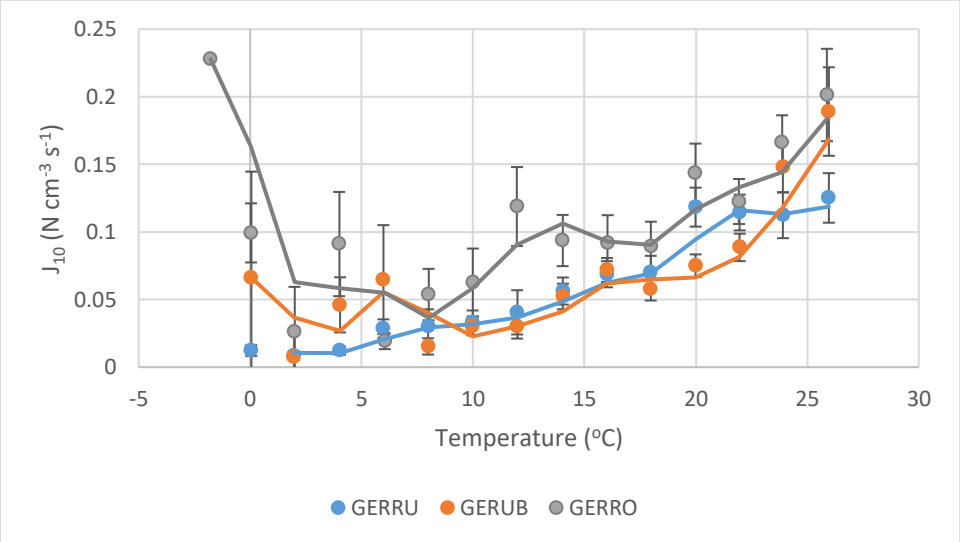

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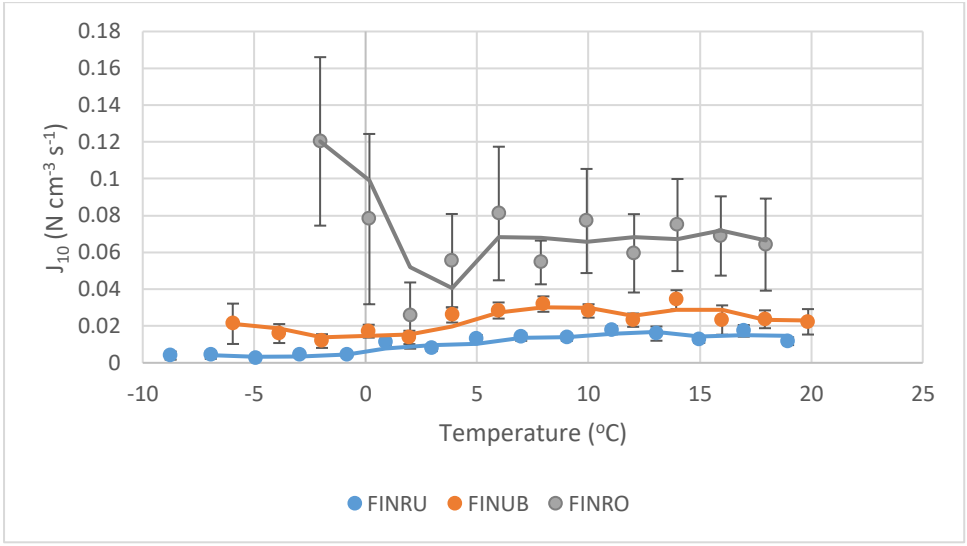

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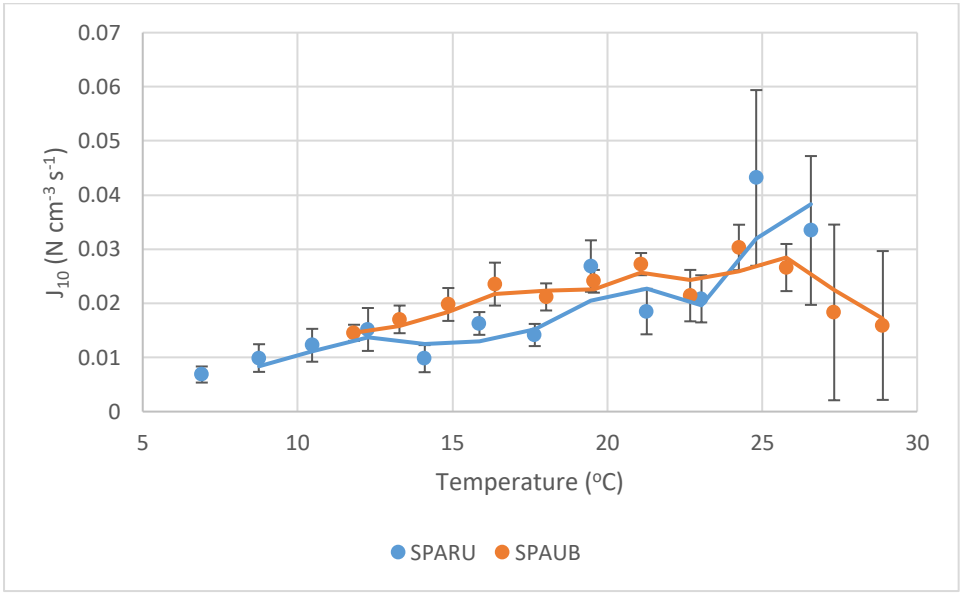

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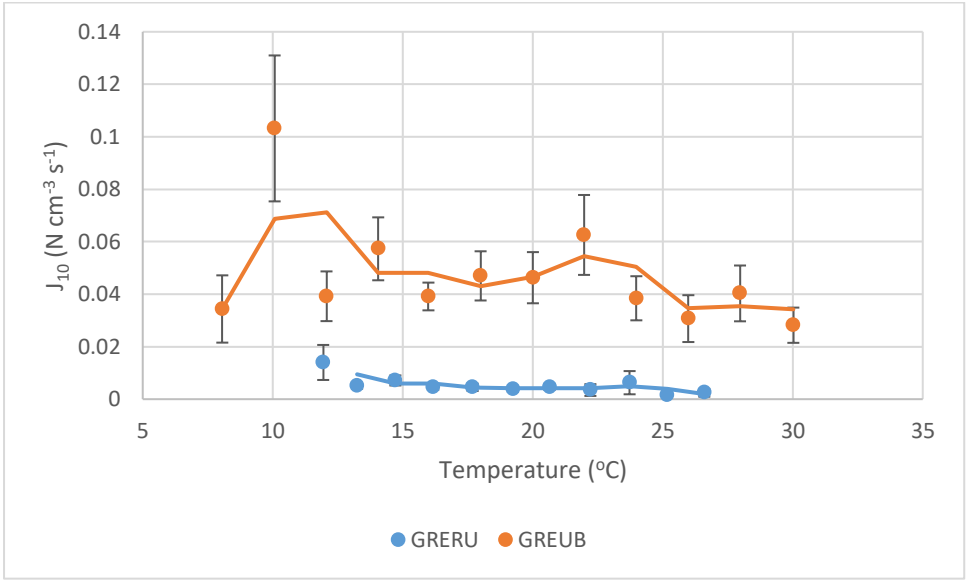


(r)

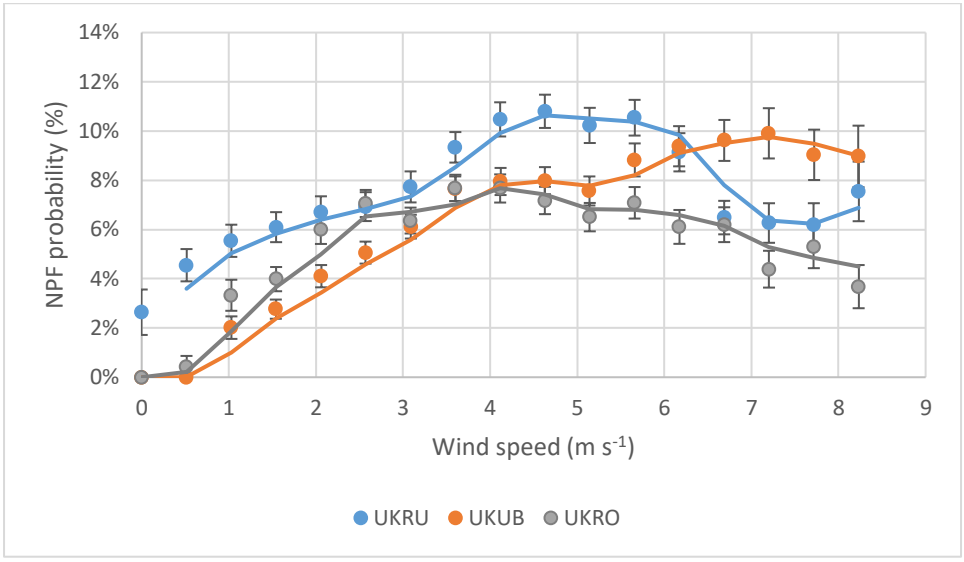
Wind speed

Wind speed may have both a positive and a negative effect on the occurrence of NPF events. On one hand, it may promote NPF events by the increased mixing of the condensable compounds in the atmosphere as well as by reducing the CS, while on the other hand high wind speeds may suppress NPF events due to increased dilution. It should be considered that the variability found is also affected by the specific conditions found at each site as discussed in earlier chapters. The wind speed measurements in many cases, especially in urban sites, can be biased by the local topography or specific conditions found at each site, thus representing the local conditions for this variable rather than the regional ones. Similarly, measurements of wind speed at well sited meteorological stations may be more representative of regional conditions, than of those affecting the sites of nucleation measurement. The sites in this study presented mixed results, both in the importance as well as the effect of the wind speed variability. Three different behaviours were found in the variation of NPF event probability and wind speed which appear to be associated with local conditions as they are almost uniformly found among the sites within close proximity. Some sites presented a steady increase of NPF event probability with wind speed (Danish sites as well as UKUB, FINRU, SPAUB and GRERU), while others were found to steadily decline with increasing wind speeds (German sites – it should be noted that the German sites are the only ones that are located at a great distance from the sea), while some were found to reach a peak and then decline, which also leads to smaller R^2 when linear relationships are considered (UKRU, UKRO, SPARU and to a lesser extent GREUB – figures 17a, e and f). The reasons for these differences between the sites are very hard to distinguish as apart from the wind speed the origin and the characteristics of these air masses play a crucial role. Following this, it appears that NPF probability is very low or zero for wind speeds close to

calm for the sites with an increasing trend (as well as those that have a peak and decline after), while the opposite is observed for the German sites where the maximum NPF probability is found for very low wind speeds.

Similarly, the effect of different wind speeds upon the growth rate also varied a lot, though it was found to be negative in all the cases where R^2 was higher than 0.50 when linear relationships were considered (UKUB, DENRU, DENRO, GERRU, GERUB and GREUB). Finally, the formation rate variable relationships and mixed trends were found. When linear relationships were considered, a significant correlation was only found only at two sites (UKRO and DENRU), probably indicating that the variability of the wind speed either does not affect this variable or its effect is rather small.

Figure 17: Relationship of wind speed with NPF variables.



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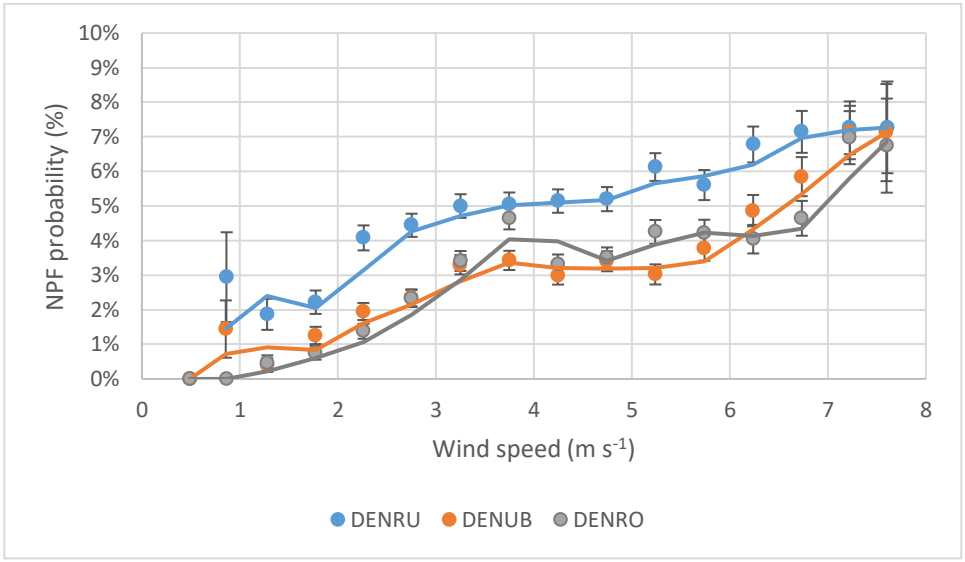

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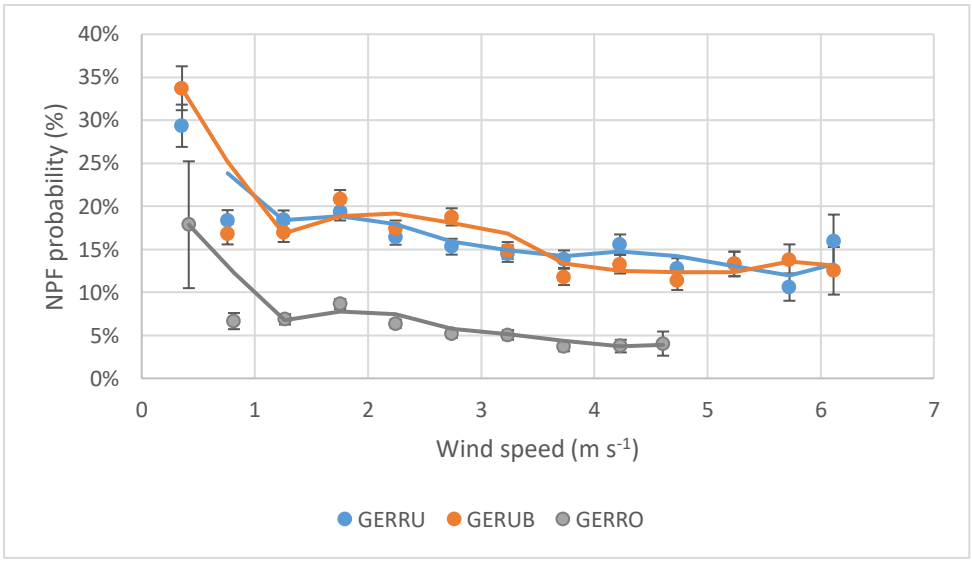

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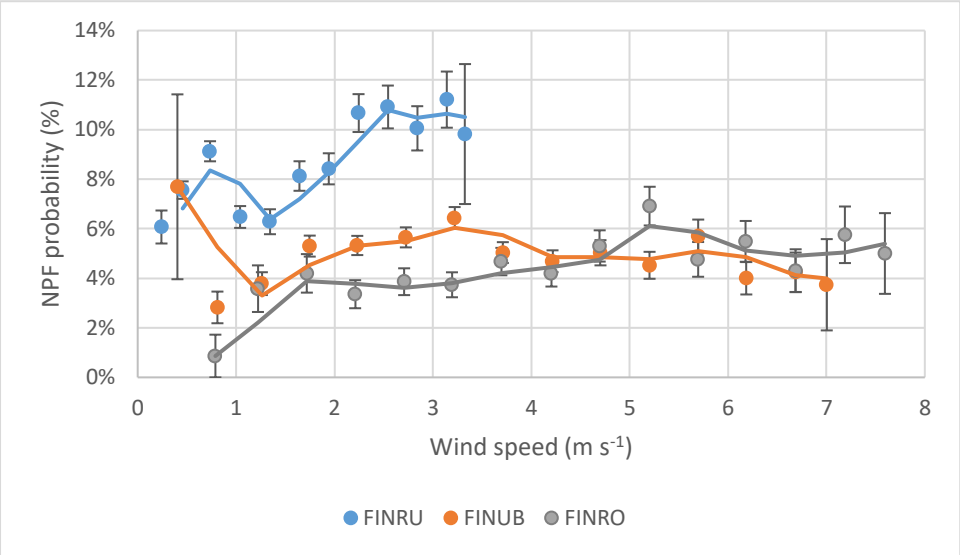
Figure 17 (continued)



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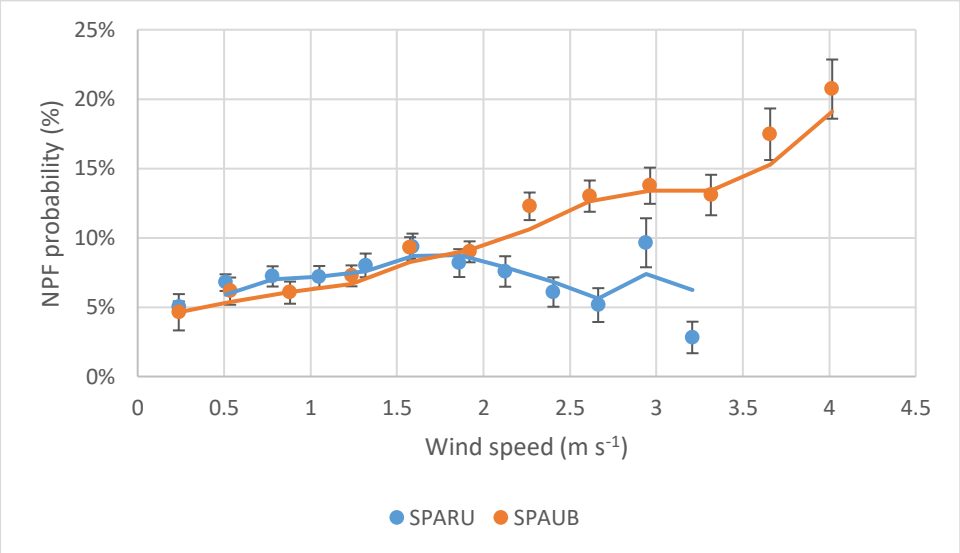

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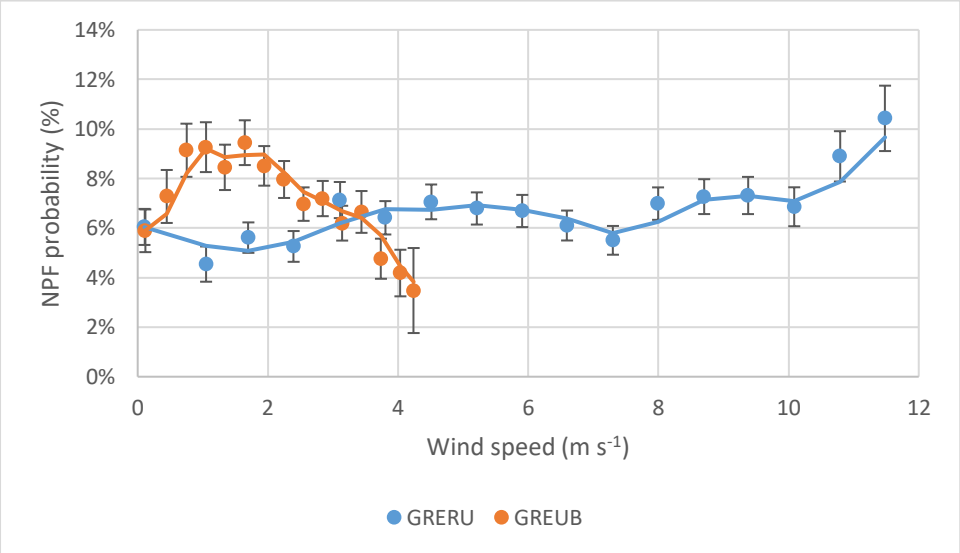

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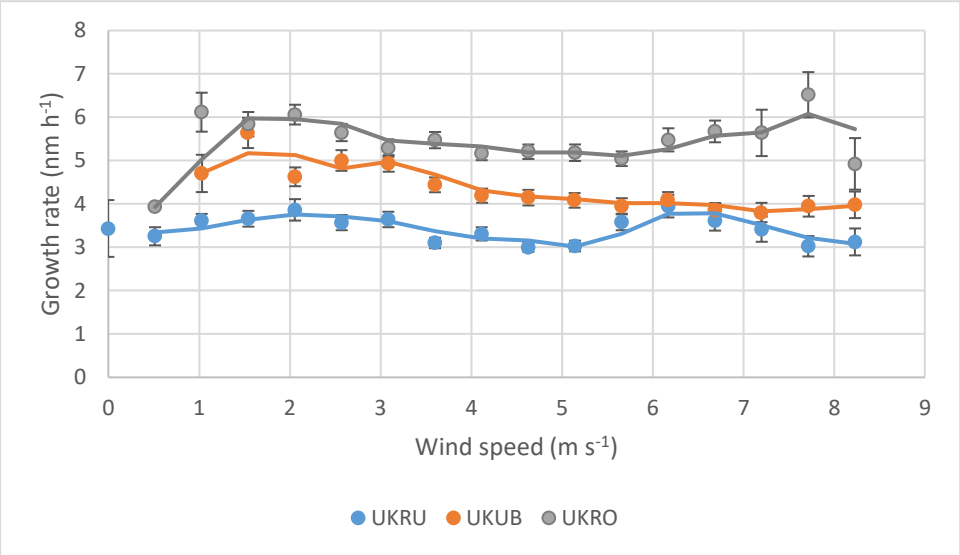

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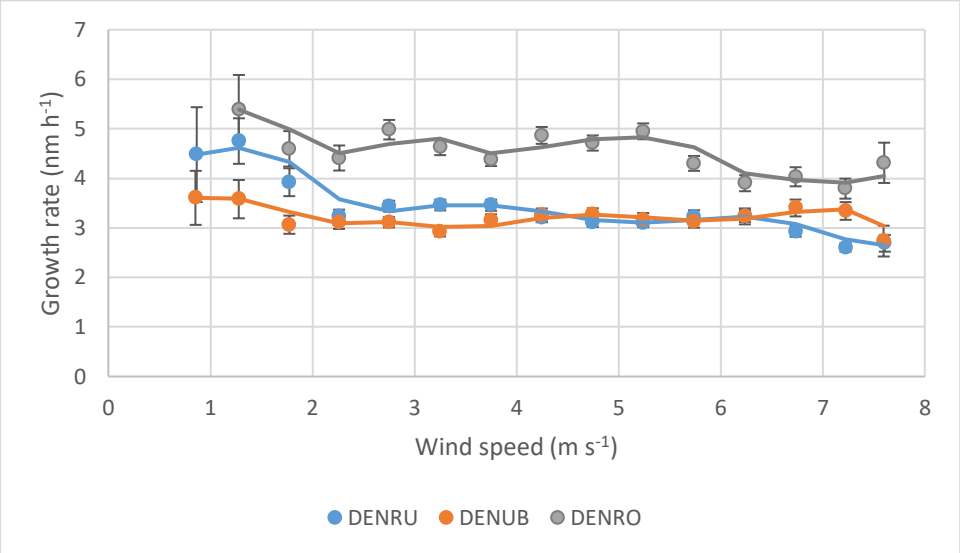

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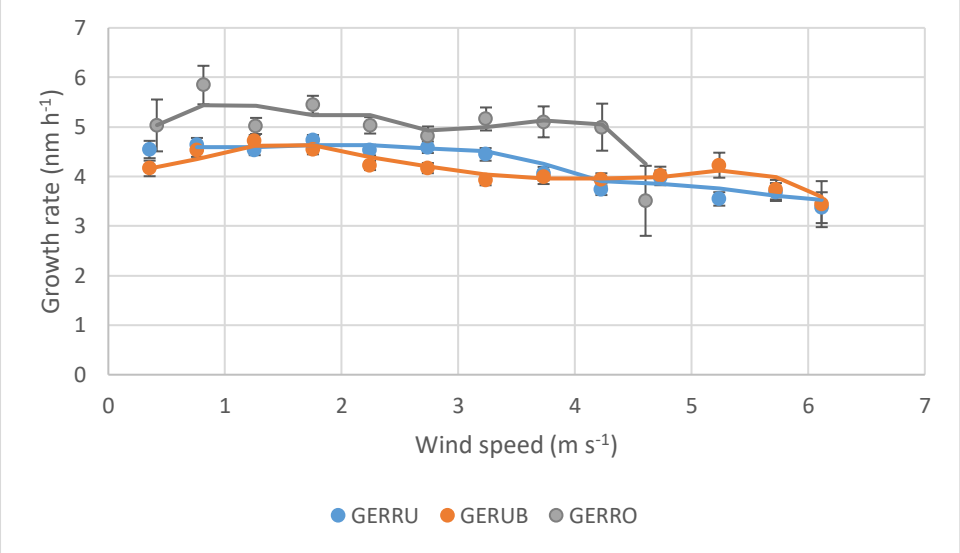

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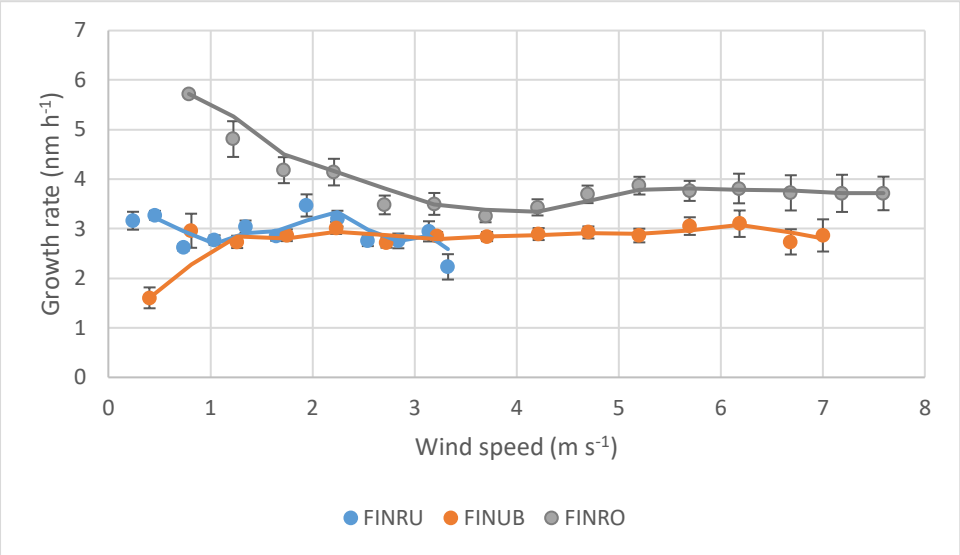

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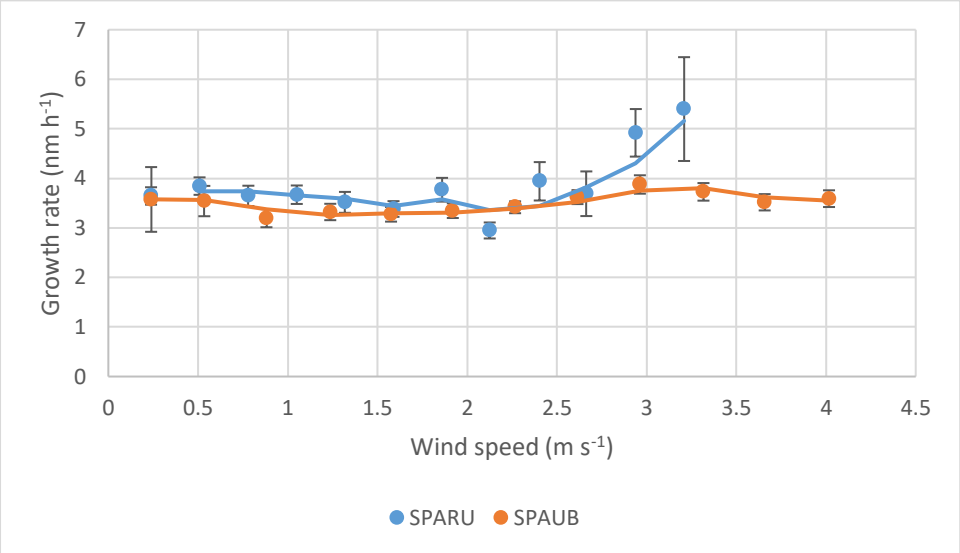

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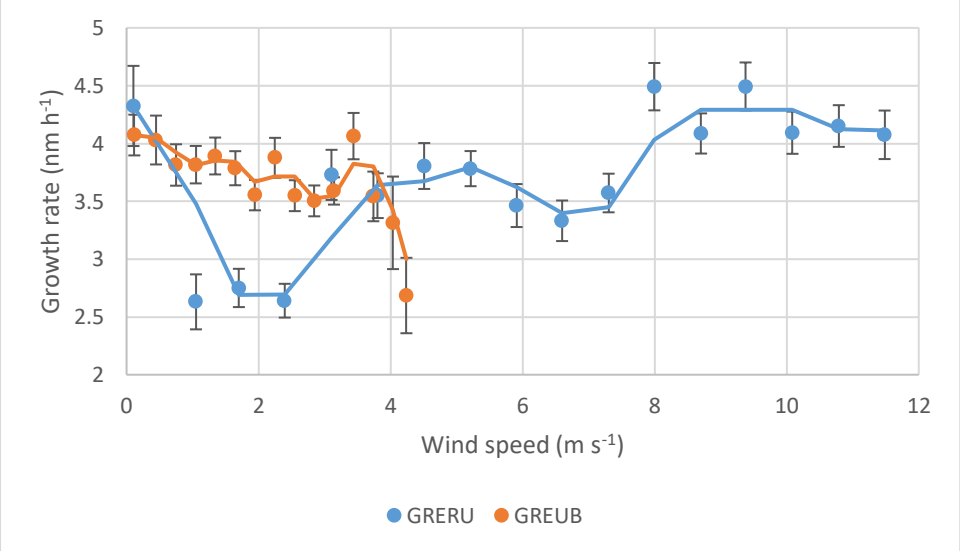

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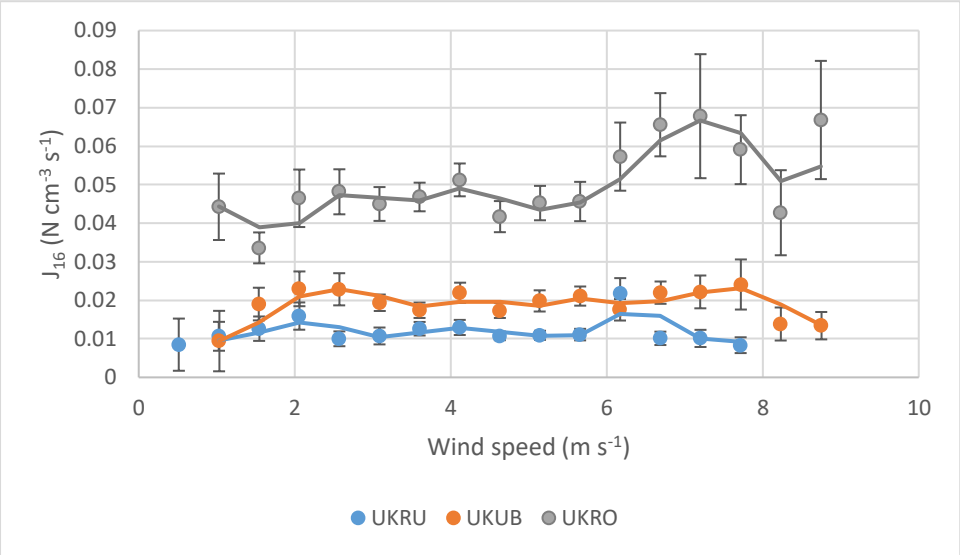

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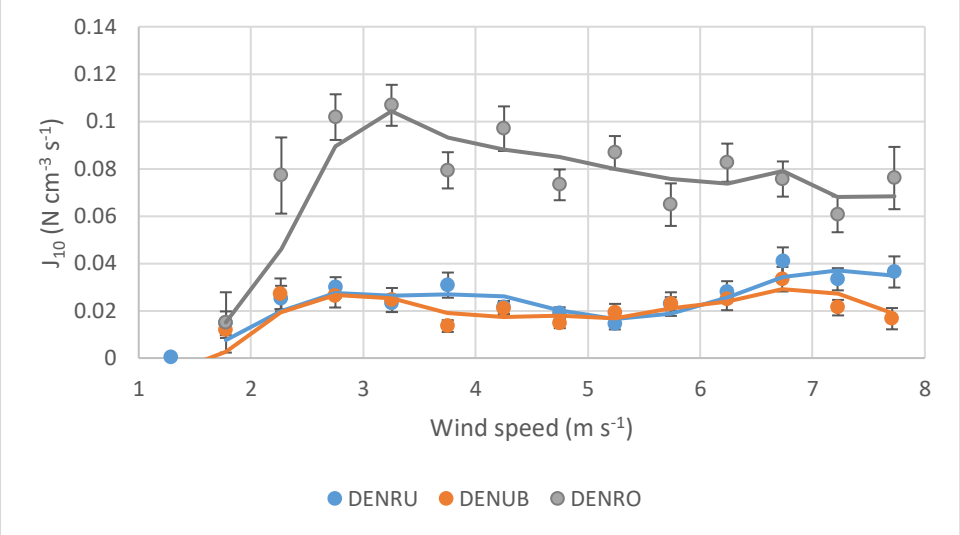

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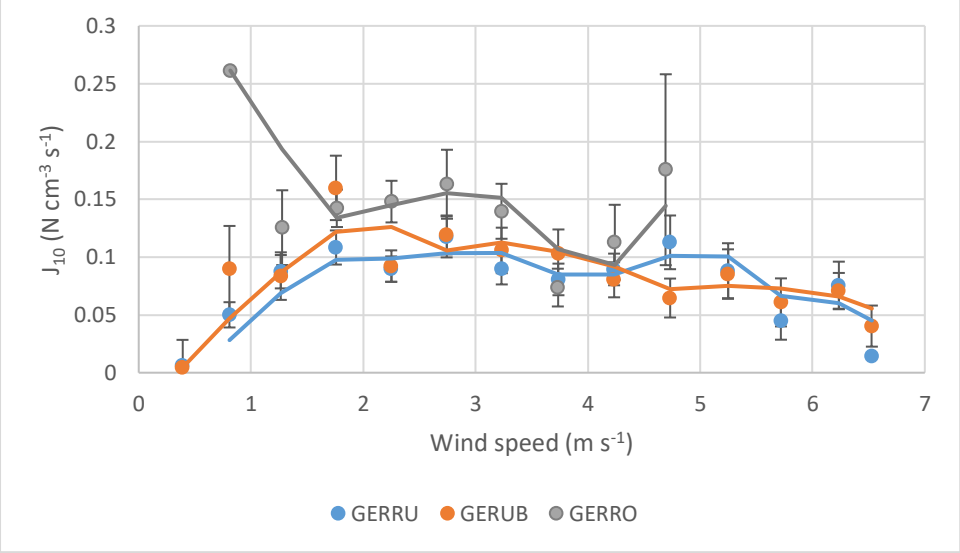

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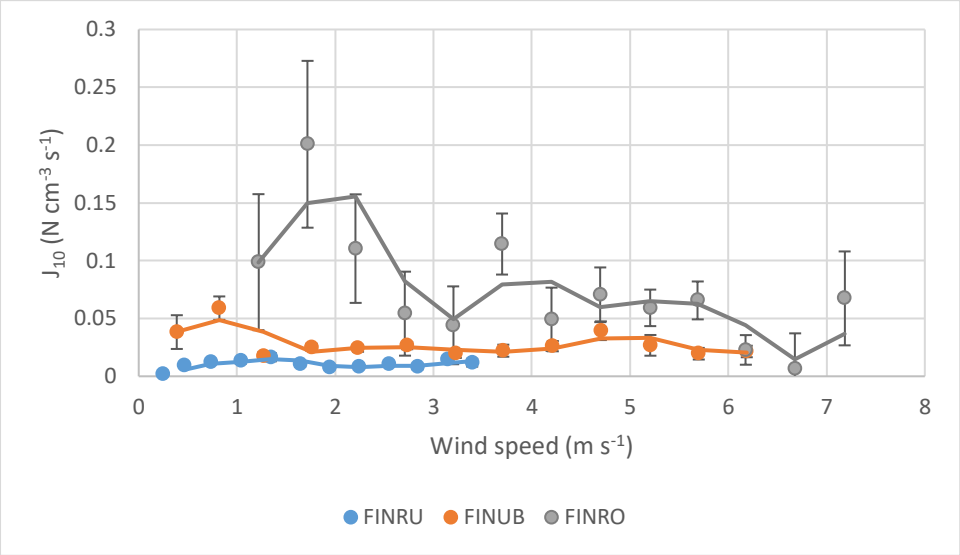
Figure 17 (continued)



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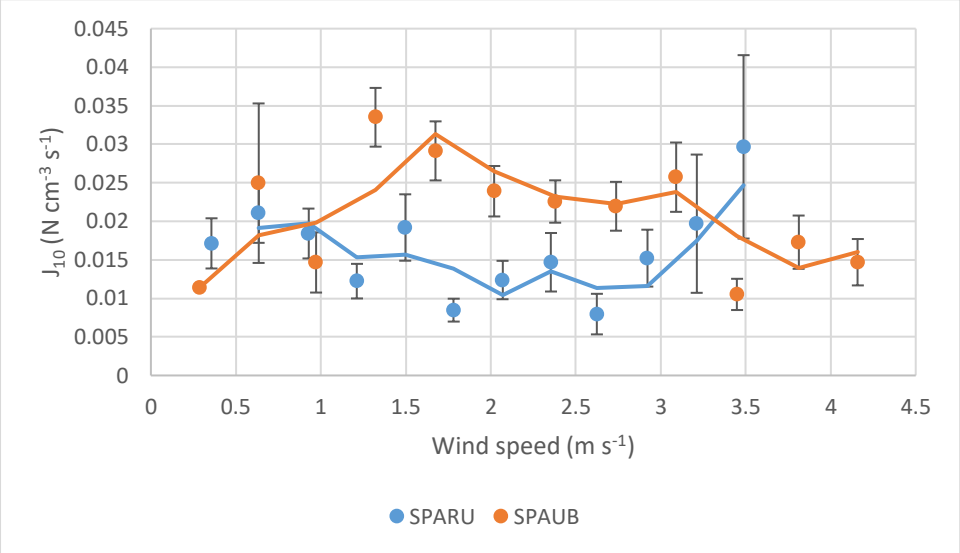

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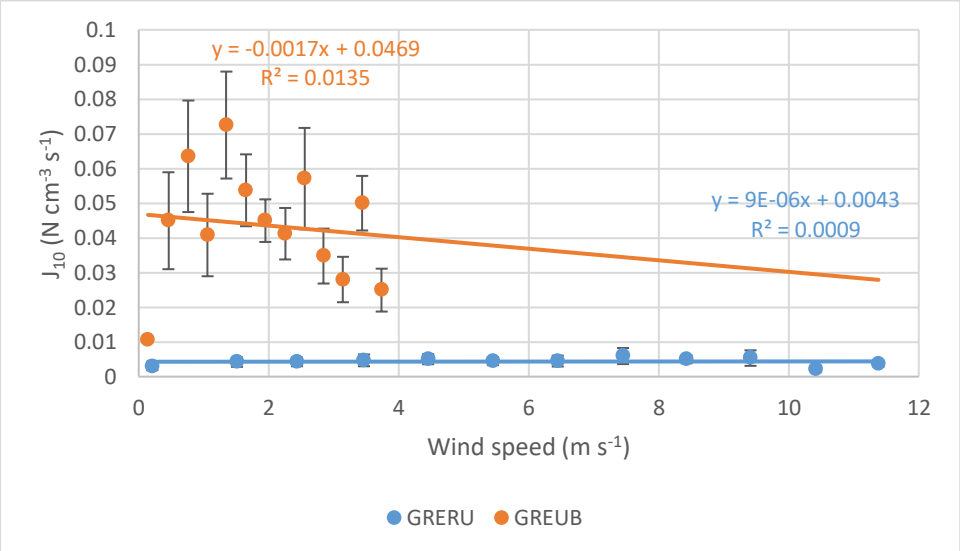
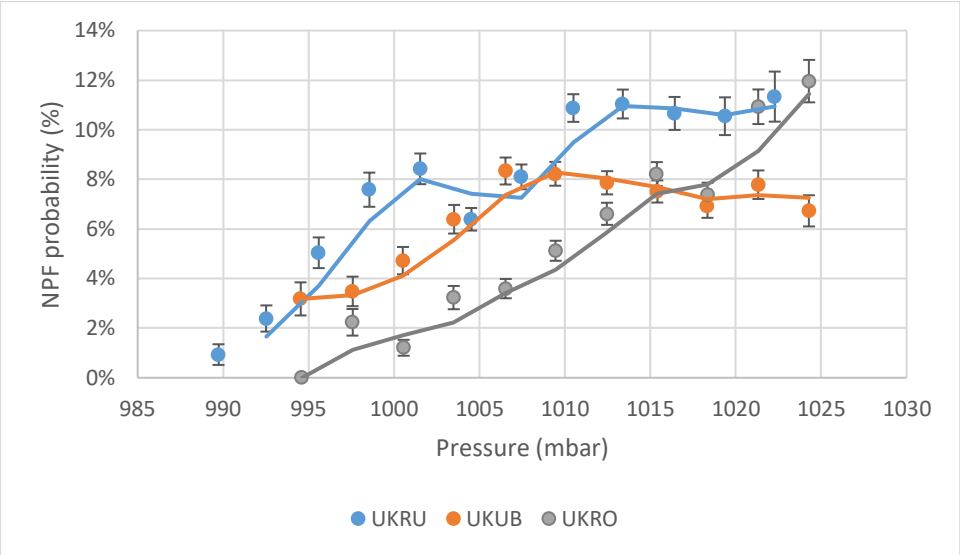


(r)

Pressure

In almost all the sites with available data (apart from the Spanish), the NPF probability presented a positive relationship with high significance at all types of sites when linear relationships were considered. The greater significance found at the rural sites indicates the increased importance of meteorological conditions in the occurrence of NPF events at this type of site. The growth rate also presented a similar picture, with positive relationships at all the background sites of this study except the ones in Greece and FINUB (though with low R^2 at 0.02). This is probably associated with the seasonal variation found in Greece where higher growth rates were found in summer, a period when increased wind speeds and lower atmospheric pressure was found due to the Etesians (Kalkavouras et al., 2017). An interesting finding is the negative gradients found at all the roadsides when linear relationships were considered, though the significance of these results is relatively low (R^2 < 0.43) and always lower compared to the rural sites. The effects of pressure found are not likely to be important. Once again however, this is not an independent variable and higher pressure in summer tends to be associated with higher insolation and temperatures and lower RH. Since most events occur in the warmer months of the year, this is probably the explanation for the apparent effects of pressure. The formation rate presented variable relationships of low significance for the sites of this study as well. Due to this, pressure should not be an important factor for the formation rate at any type of site.

Figure 18: Relationship of atmospheric pressure with NPF variables.



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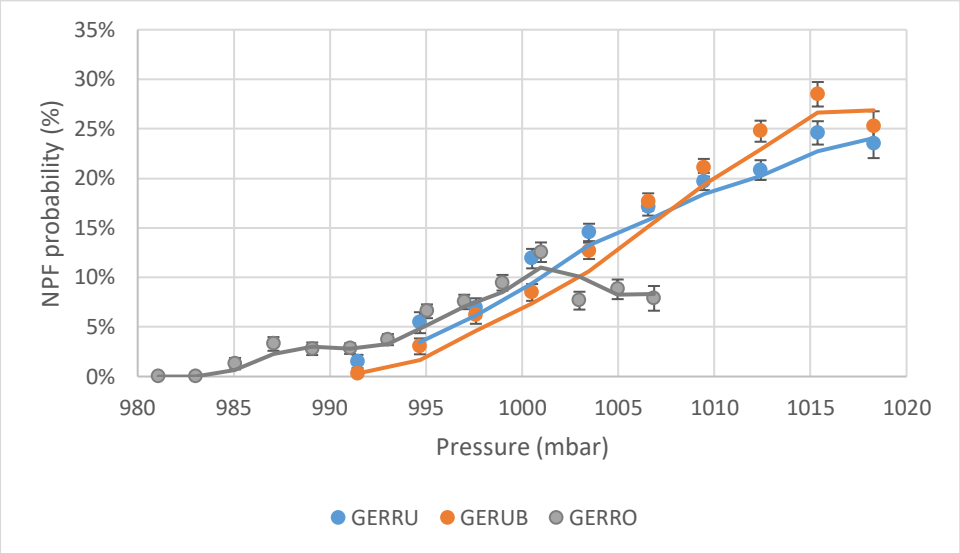

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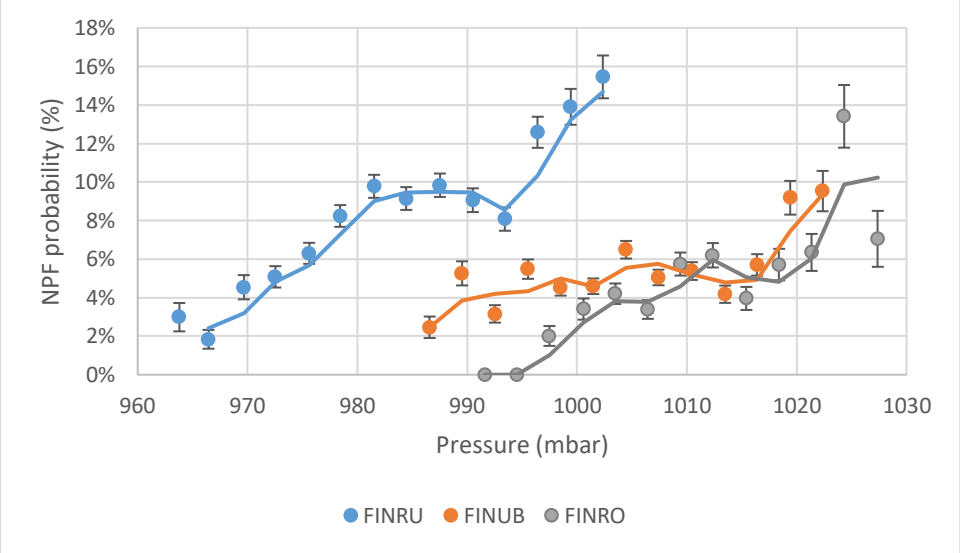

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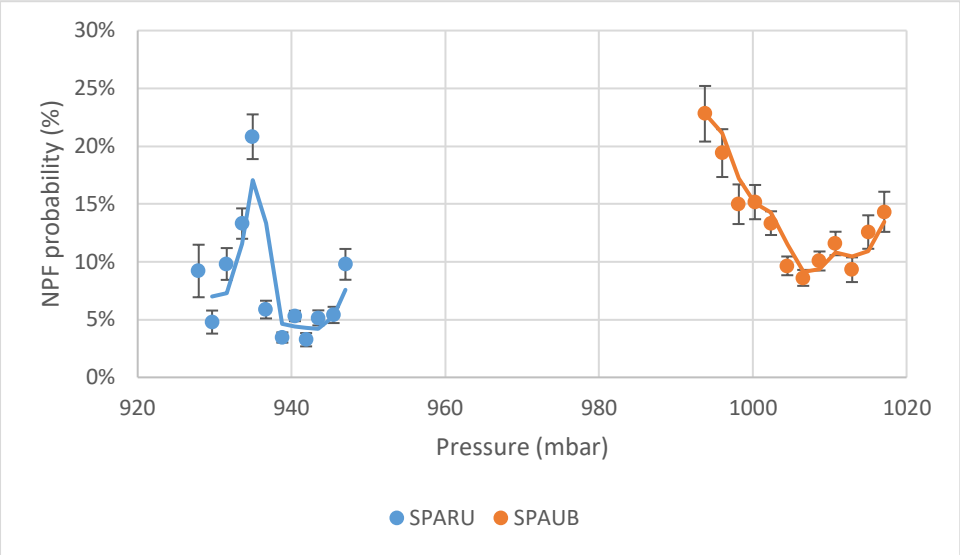

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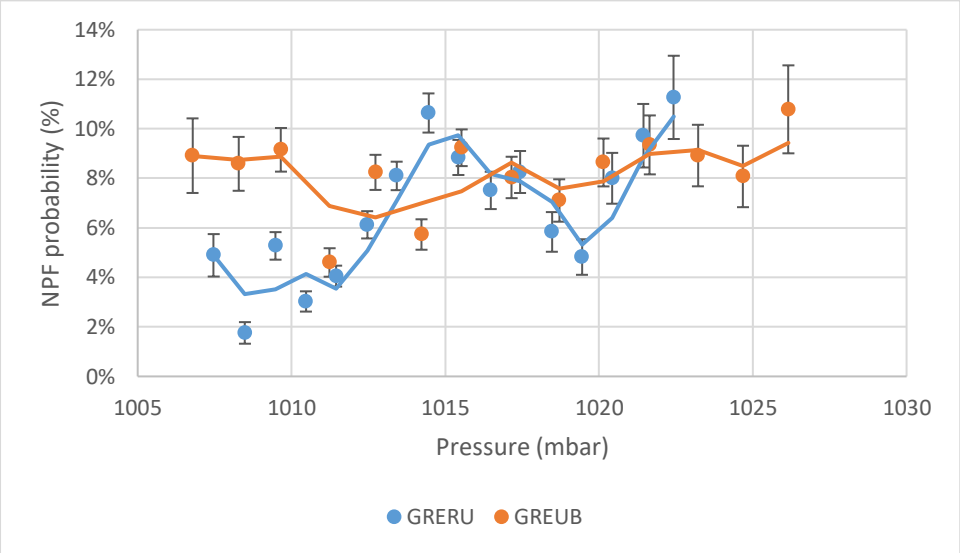

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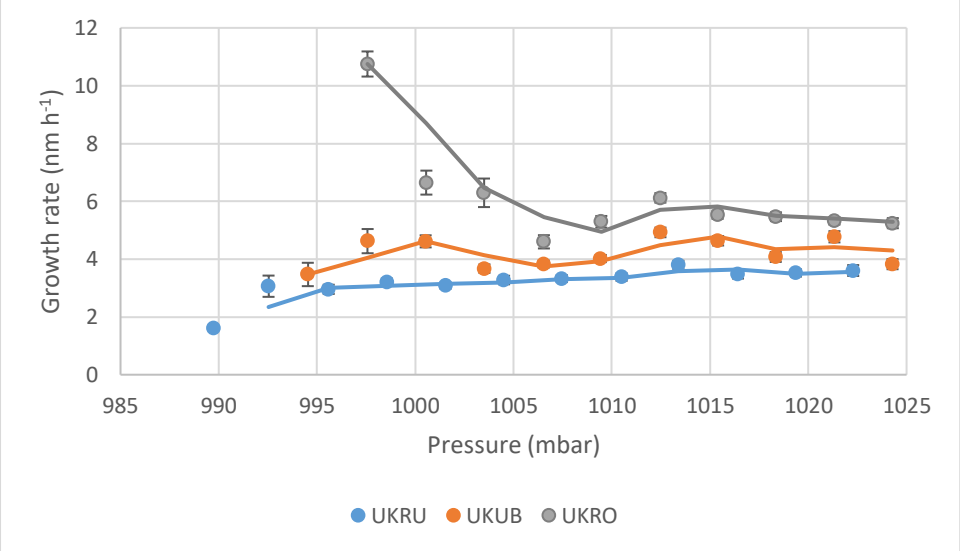

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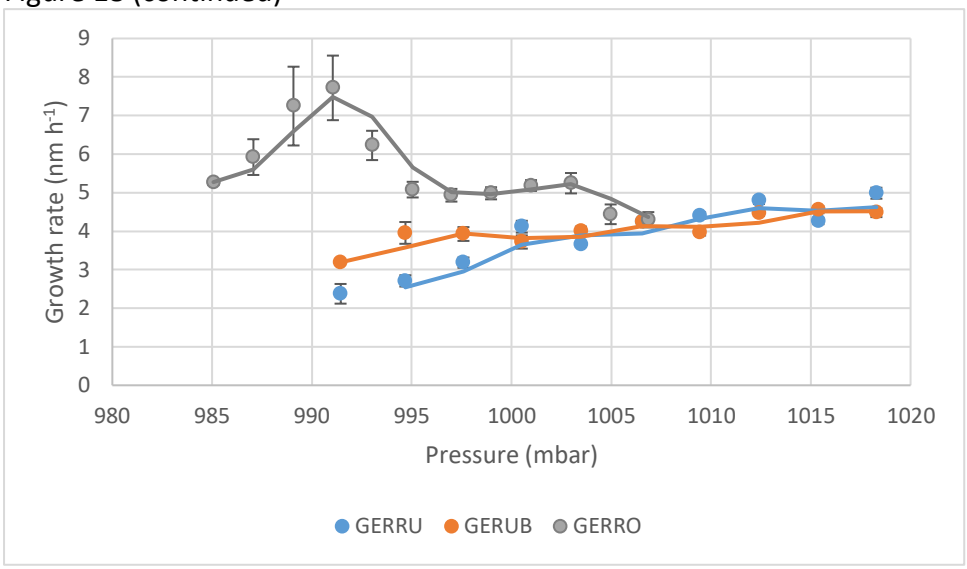

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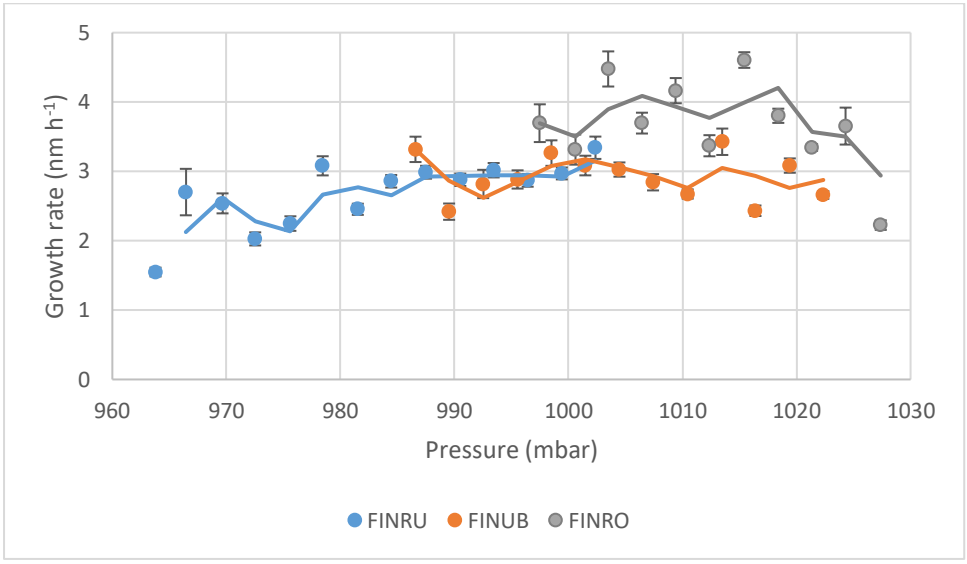

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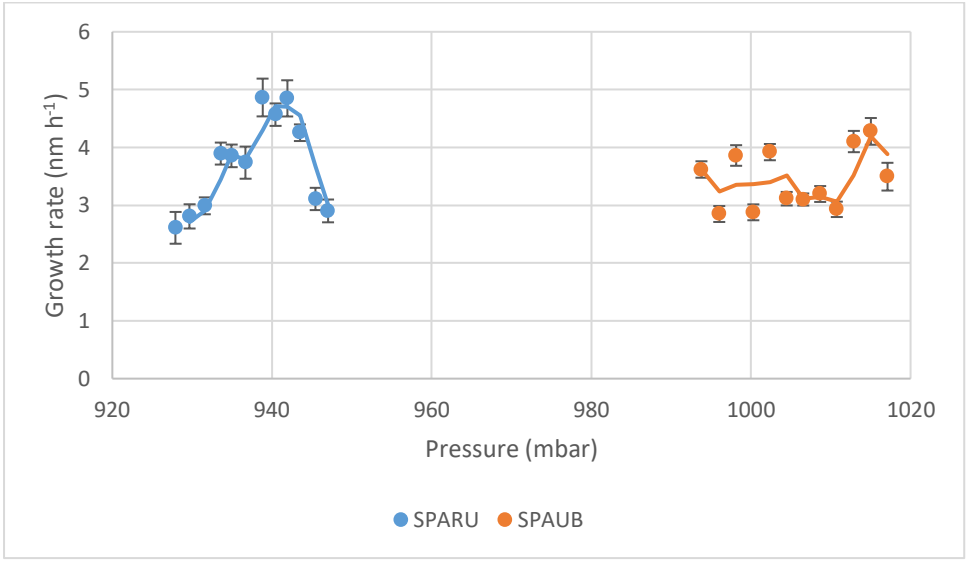

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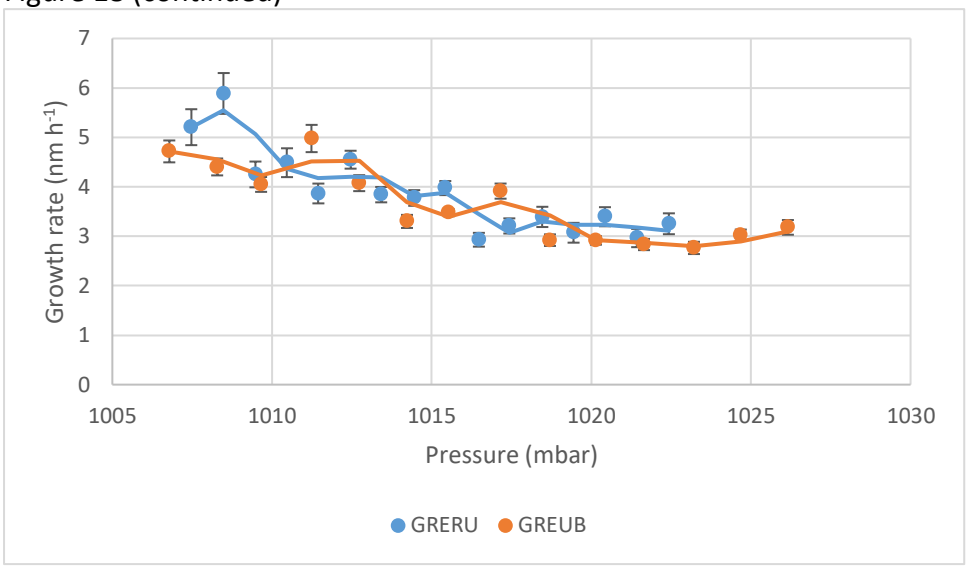

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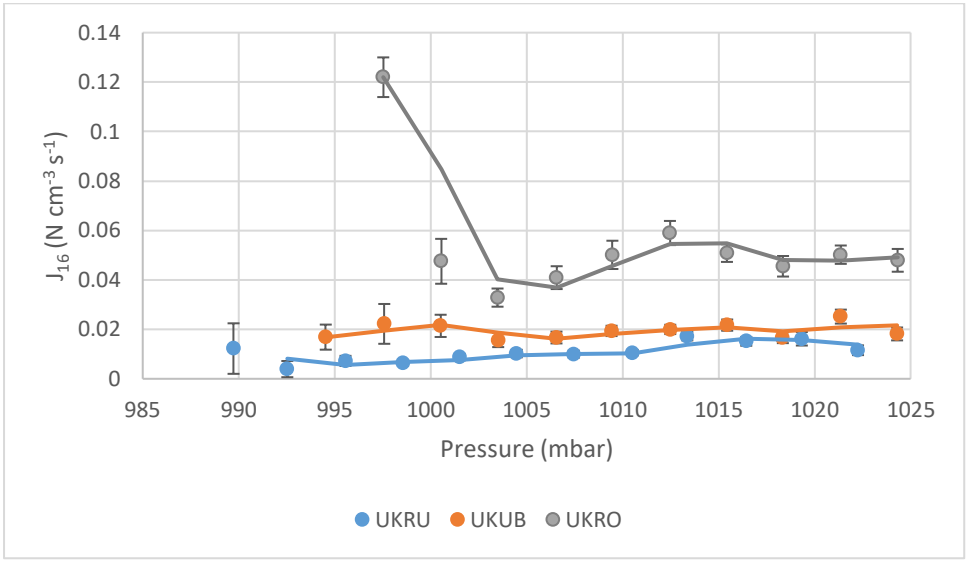

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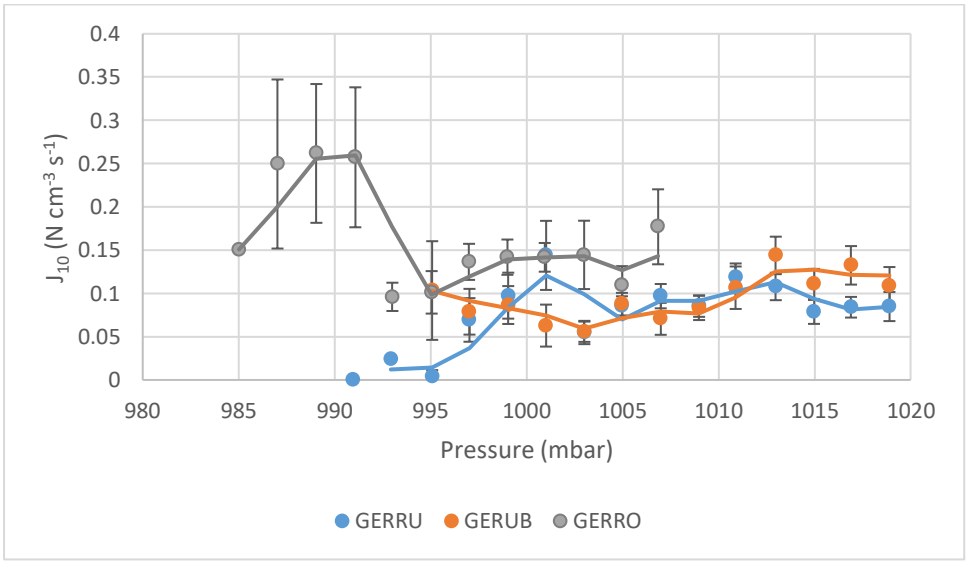

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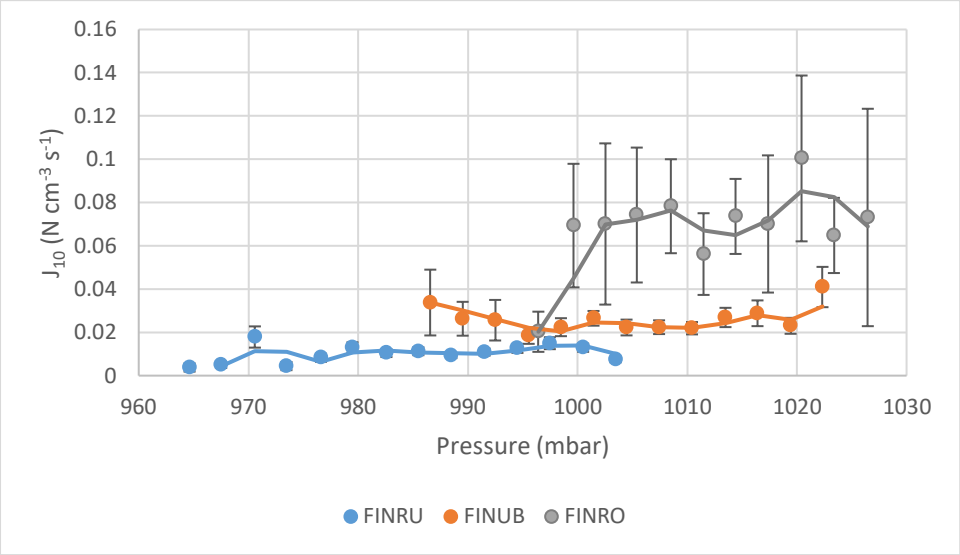

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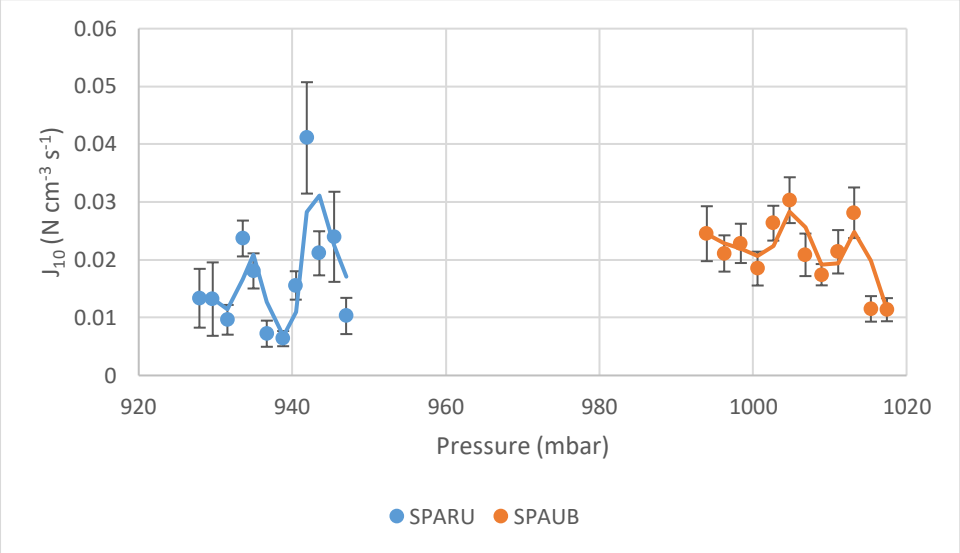

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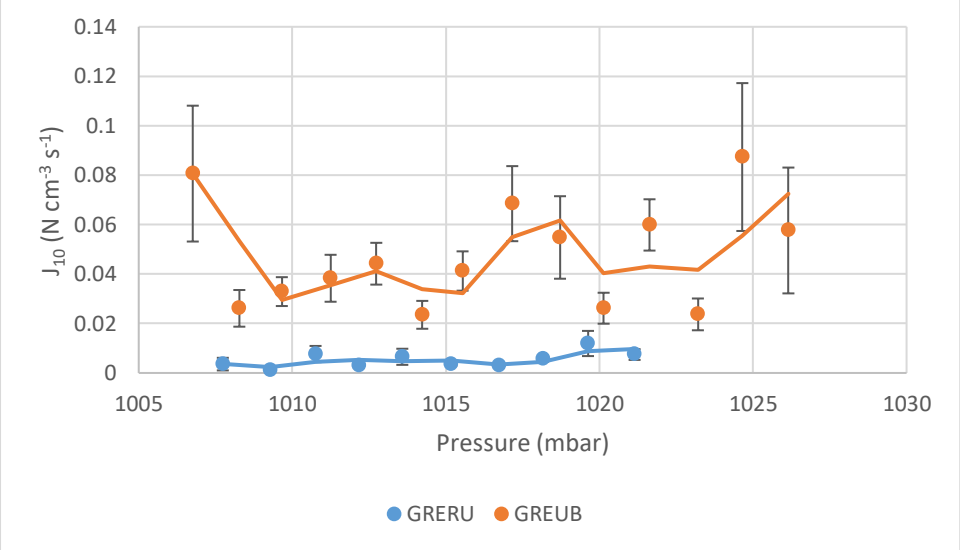


(o)

3.3.3.2 Atmospheric Composition

The gradients and R^2 from the analysis of a number of air pollutants and the condensation sink when linear relationships are considered, as well as the average conditions of these variables are found in Table 5. The results for each site and variable are found in Figure 19 – 25.

Table 5: Gradients, R^2 and p-values (for values > 0.05) for the relation between atmospheric composition variables and NPF event variables when linear relationships are considered. Gradients of R^2 > 0.50 are in bold.

SO_2 (µg m^{-3})										
Site	**a_N**	**R^2**	**p**	**a_G**	**R^2**	**p**	**a_J**	**R^2**	**p**	**Average**
UKRU	-1.38E-02	0.38	<0.05	-6.17E-02	0.02	-	2.87E-03	0.06	-	1.64
UKUB	**-1.80E-02**	0.62	<0.001	1.93E-02	0.00	-	5.94E-03	0.40	-	2.04
UKRO	**-6.28E-03**	0.82	<0.001	6.90E-02	0.34	<0.01	**3.16E-03**	0.77	<0.001	7.46
DENRU	**-7.72E-02**	0.53	<0.05	2.84E+00	0.37	-	1.13E-02	0.09	-	0.52
DENRO	**-2.27E-02**	0.91	<0.001	**6.42E-01**	0.54	<0.005	**4.57E-02**	0.62	<0.001	0.97
FINRU	4.92E-02	0.05	-	-1.42E+00	0.19	-	-7.50E-04	0.00	-	0.09
SPARU	**-4.34E-02**	0.74	<0.001	-1.33E-01	0.02	-	-5.47E-04	0.01	-	0.95
SPAUB	-3.84E-03	0.04	-	**4.12E-01**	0.59	-	2.27E-03	0.29	-	1.99

NO_x or NO_2 (ppb)										
Site	**a_N**	**R^2**	**p**	**a_G**	**R^2**	**p**	**a_J**	**R^2**	**p**	**Average**
UKRU	**-3.49E-03**	0.67	<0.005	**4.52E-02**	0.58	<0.05	**-3.92E-04**	0.70	<0.005	11.7
UKUB	**-6.13E-04**	0.83	<0.001	-3.97E-04	0.00	-	-1.55E-04	0.43	<0.05	53.6
UKRO	**-1.96E-04**	0.72	<0.001	1.44E-03	0.39	<0.05	**8.21E-05**	0.66	<0.001	299
DENRU	-7.43E-03	0.43	<0.005	-4.89E-03	0.00	<0.001	**-1.66E-03**	0.55	<0.01	5.42
DENUB	**-2.89E-03**	0.68	<0.001	2.85E-02	0.26	-	2.05E-05	0.00	-	10.5
DENRO	**-2.75E-04**	0.75	<0.001	**1.10E-02**	0.69	<0.001	**6.72E-04**	0.88	<0.001	68.5
FINRU	**-6.32E-02**	0.54	<0.001	-2.74E-01	0.11	-	2.32E-03	0.05	-	0.72
FINRO	**-3.18E-04**	0.68	<0.001	1.70E-03	0.12	-	2.26E-04	0.03	-	88.1
SPARU*	-1.84E-03	0.05	-	2.54E-02	0.01	-	1.93E-03	0.21	-	3.26
SPAUB*	**-3.39E-03**	0.62	<0.005	**2.23E-02**	0.70	<0.001	5.45E-05	0.01	-	31.4
GRERU*	1.96E-02	0.19	-	**-1.40E+00**	0.75	<0.001	2.56E-03	0.13	-	0.52

***** NO_2 measurements

Table 5 (continued)

O_3 (ppb)										
Site	**a_N**	**R^2**	**p**	**a_G**	**R^2**	**p**	**a_J**	**R^2**	**p**	**Average**
UKRU	**1.59E-03**	0.88	<0.001	**-4.89E-02**	0.53	<0.005	-3.07E-05	0.01	-	54.4
UKUB	**9.59E-04**	0.87	<0.001	**-3.45E-02**	0.68	<0.001	-8.45E-05	0.05	-	39.3
UKRO	**4.55E-03**	0.95	<0.001	-1.06E-02	0.09	-	**-9.15E-04**	0.63	<0.005	16.2
DENRU	**3.93E-03**	0.92	<0.001	-1.32E-02	0.15	-	3.16E-04	0.08	-	30.1
DENUB	**3.39E-03**	0.84	<0.001	**-1.69E-02**	0.58	-	6.65E-04	0.32	<0.05	28.2
DENRO	**3.47E-03**	0.51	<0.05	1.39E-02	0.03	-	**2.61E-03**	0.91	<0.05	31.1
FINRU	**5.88E-03**	0.77	<0.05	**-4.23E-02**	0.60	-	4.66E-04	0.37	<0.05	27.4
FINRO	**1.21E-03**	0.91	<0.001	6.11E-03	0.24	-	-1.27E-03	0.29	-	37.1
SPARU	1.88E-03	0.02	-	4.34E-02	0.11	-	2.02E-04	0.31	-	75.9
SPAUB	**1.05E-03**	0.38	<0.05	-5.83E-03	0.30	-	-2.40E-05	0.01	-	54.9
GRERU	4.91E-04	0.04	-	3.68E-02	0.17	-	-1.47E-04	0.15	-	49.5

Particulate Organic Carbon (µg m^{-3})										
Site	**a_N**	**R^2**	**p**	**a_G**	**R^2**	**p**	**a_J**	**R^2**	**p**	**Average**
UKRU	-2.31E-03	0.00	-	1.13E+00	0.42	<0.005	1.85E-03	0.16	-	1.96
UKUB	**-1.93E-02**	0.59	<0.005	**6.63E-01**	0.58	<0.05	**3.11E-03**	0.55	<0.05	3.63
UKRO	**-2.31E-02**	0.89	<0.001	**8.12E-01**	0.57	<0.005	**1.73E-02**	0.75	<0.001	6.24
DENRU	**-3.51E-02**	0.75	<0.001	2.24E-01	0.11	-	**-8.15E-03**	0.68	<0.01	1.48
DENRO	-4.21E-03	0.11	-	**1.10E+00**	0.77	<0.005	**3.24E-02**	0.81	<0.005	2.59
GERRU	-2.15E-02	0.24	-	1.35E-01	0.09	-	2.88E-03	0.03	-	2.18
FINRU	1.97E-03	0.00	-	**3.39E-01**	0.60	<0.005	-4.12E-03	0.16	-	1.78
GRERU	-1.35E-02	0.11	-	7.87E-01	0.41	<0.05	4.38E-03	0.11	-	1.58

Sulphate (µg m^{-3})										
Site	**a_N**	**R^2**	**p**	**a_G**	**R^2**	**p**	**a_J**	**R^2**	**p**	**Average**
UKRU[1]	**-1.83E-02**	0.57	<0.001	**7.34E-01**	0.77	<0.001	6.94E-03	0.44	<0.05	1.97
UKUB[1]	**-2.50E-02**	0.89	<0.001	9.28E-01	0.44	<0.01	1.38E-02	0.16	-	1.58
UKRO[1]	-3.69E-03	0.24	-	3.04E-01	0.34	<0.05	-2.33E-03	0.04	-	1.98
DENRU[2]	-6.17E-02	0.34	<0.05	**1.02E+00**	0.60	<0.05	**-2.65E-02**	0.63	<0.01	0.52
DENRO[2]	-4.44E-02	0.28	-	1.99E+00	0.22	-	2.28E-02	0.12	-	0.55
GERRU[1]	-5.76E-03	0.00	-	5.89E-01	0.11	-	-4.49E-03	0.01	-	0.92
FINRU[3]	**-1.03E-01**	0.65	<0.001	2.35E-01	0.09	-	-3.01E-03	0.17	-	1.02

[1] Measurements in PM_{10}

[2] Measurements in $PM_{2.5}$

[3] Measurements in PM_1

Table 5 (continued)

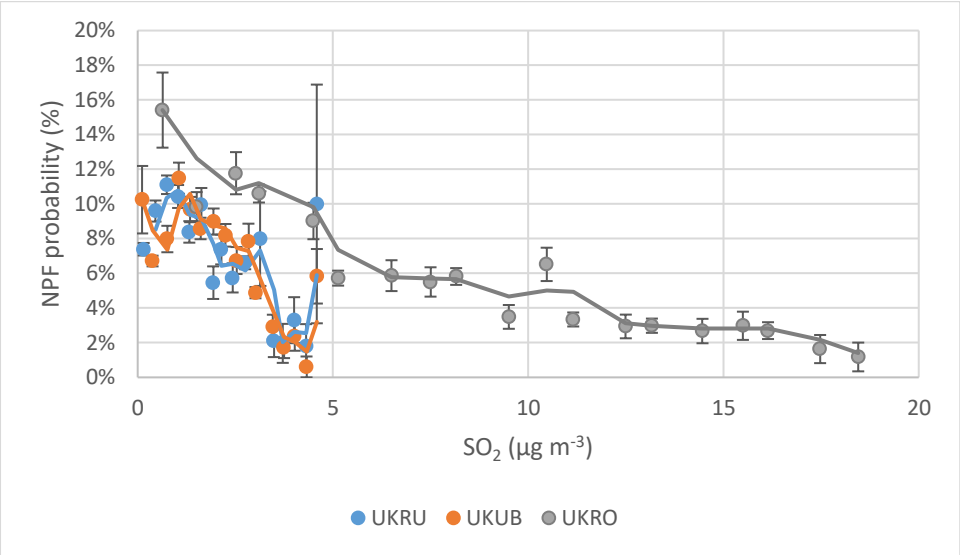
Condensation Sink (s^{-1})										
Site	**a_N**	**R^2**	**p**	**a_G**	**R^2**	**p**	**a_J**	**R^2**	**p**	**Average**
UKRU	**-1.60E+01**	0.72	<0.001	**2.64E+02**	0.60	<0.001	6.59E-01	0.22	-	3.38E-03
UKUB	**-1.16E+01**	0.78	<0.001	2.49E+02	0.41	<0.05	2.46E+00	0.35	<0.05	7.41E-03
UKRO	**-2.46E+00**	0.75	<0.001	2.33E+01	0.18	-	**3.35E+00**	0.91	<0.001	2.12E-02
DENRU	**-3.54E+00**	0.91	<0.001	6.90E+01	0.49	<0.05	1.38E+00	0.24	-	9.46E-03
DENUB	**-2.19E+00**	0.75	<0.001	3.58E+01	0.25	-	**3.72E-01**	0.56	<0.005	1.42E-02
DENRO	**-5.72E-01**	0.73	<0.001	**2.53E+01**	0.56	<0.005	**2.20E+00**	0.79	<0.001	3.10E-02
GERRU	**2.63E+01**	0.86	<0.001	**1.33E+02**	0.56	<0.001	**6.12E+00**	0.63	<0.001	7.02E-03
GERUB	**6.28E+00**	0.56	<0.005	3.63E+01	0.17	-	**4.83E+00**	0.75	<0.001	9.11E-03
GERRO	3.50E+00	0.22	<0.05	-2.21E+01	0.03	<0.005	4.89E+00	0.45	<0.005	1.20E-02
FINRU	**-1.57E+01**	0.59	<0.005	**4.01E+02**	0.74	<0.001	5.93E-01	0.10	-	2.32E-03
FINUB	**-7.55E+00**	0.63	<0.005	8.14E+01	0.31	-	5.00E+00	0.41	<0.05	6.34E-03
FINRO	**-3.56E+00**	0.77	<0.001	-1.56E+01	0.05	-	**1.68E+01**	0.83	<0.001	8.96E-03
SPARU	**-2.58E+01**	0.65	<0.005	1.86E+01	0.00	-	1.32E+00	0.47	<0.05	5.49E-03
SPAUB	**-1.55E+01**	0.65	<0.005	3.74E+01	0.38	<0.05	**2.02E+00**	0.52	<0.01	1.00E-02
GRERU	2.81E-01	0.00	-	**2.86E+02**	0.70	<0.001	**8.67E-01**	0.56	<0.005	4.66E-03
GREUB	**1.39E+01**	0.65	<0.001	9.31E+01	0.28	<0.05	**7.63E+00**	0.83	<0.001	7.55E-03

Sulphur dioxide (SO_2)

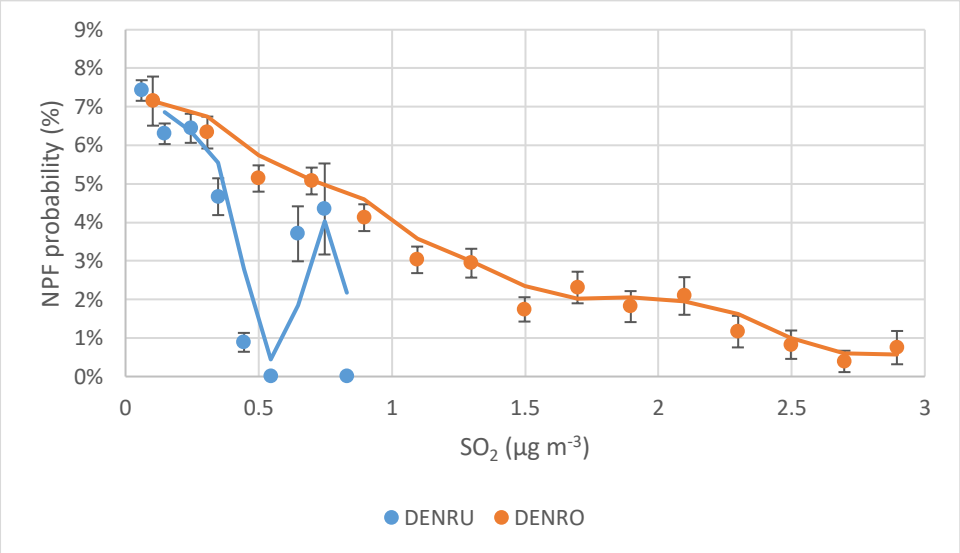
Sulphur dioxide is considered as one of the main components that participate in the NPF process. According to nucleation theories and observations, H_2SO_4 is the most important compound from which the initial clusters are formed, as well as one of the candidate compounds for the initial steps of particle growth (Kirkby et al., 2011; Nieminen et al., 2010; Sipila et al., 2010). As H_2SO_4 in the atmosphere is produced from oxidation reactions of SO_2 it would be expected that increased concentrations of the latter would be associated with increased values for all the variables associated with the NPF process. Contrary to this though, the relationship of SO_2 concentrations with NPF probability was found to be negative at all the sites in this study with available data. This relationship was relatively strong ($R^2 > 0.50$) in most areas with an increased significance at roadsides compared to their respective rural sites. As this is a negative relationship, this may indicate that SO_2 is in

sufficient concentrations for H_2SO_4 formation, thus not suppressing the occurrence of NPF events, as well as showing that in increased concentrations, it is a more important factor (or surrogate for a factor) in preventing the occurrence of NPF events within the urban environment, as probably higher SO_2 is associated with increased co-emitted particle pollution and hence CS. The growth rate on the other hand, presented mixed results and the significance of the relationships is low in most cases, which makes these results untrustworthy. Finally, the relationship of SO_2 concentrations with the formation rate was found to be positive at all sites but SPARU and FINRU (which had the lowest concentrations across the sites of this study). The significance of this relationship was rather low for all but the roadsides. This suggests that higher H_2SO_4 concentrations favour increased formation rates (i.e. more particles can be formed), rather than necessarily promoting nucleation itself because of the competing effect of condensation onto the pre-existing particle population.

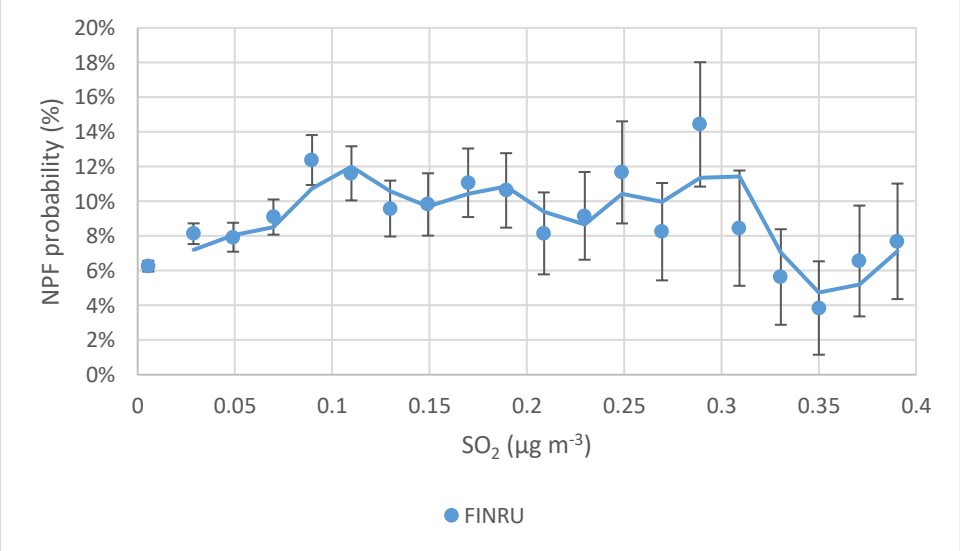
Figure 19: Relationship of SO_2 concentrations with NPF variables.



(a)

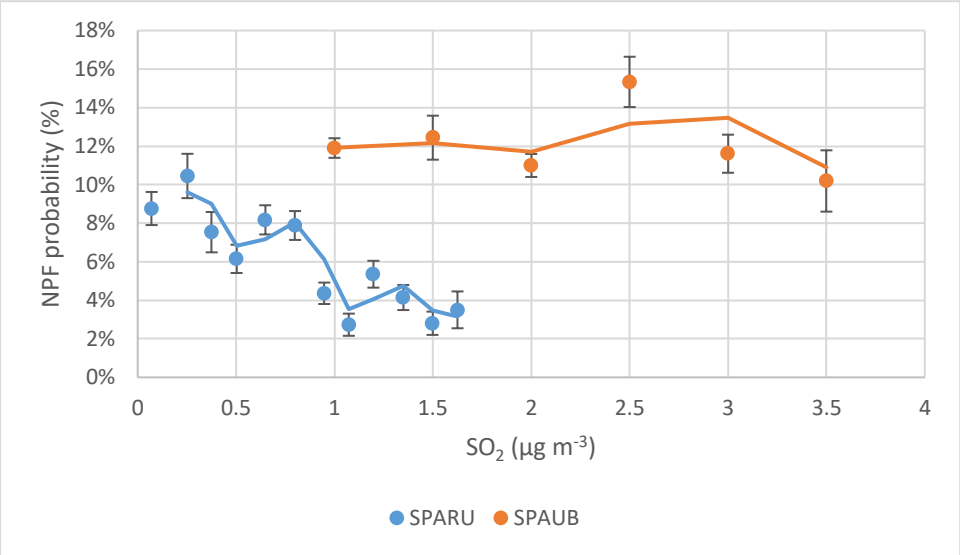


(b)

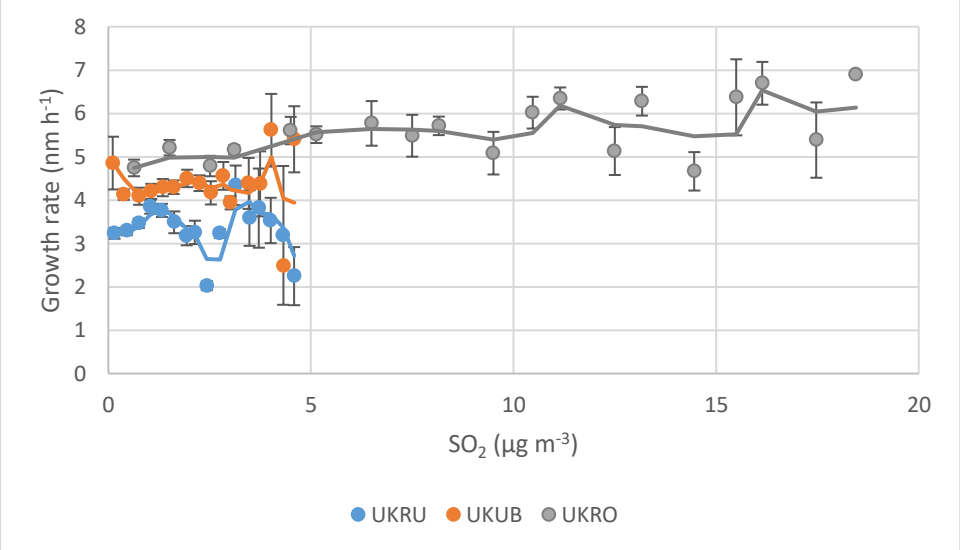


(c)

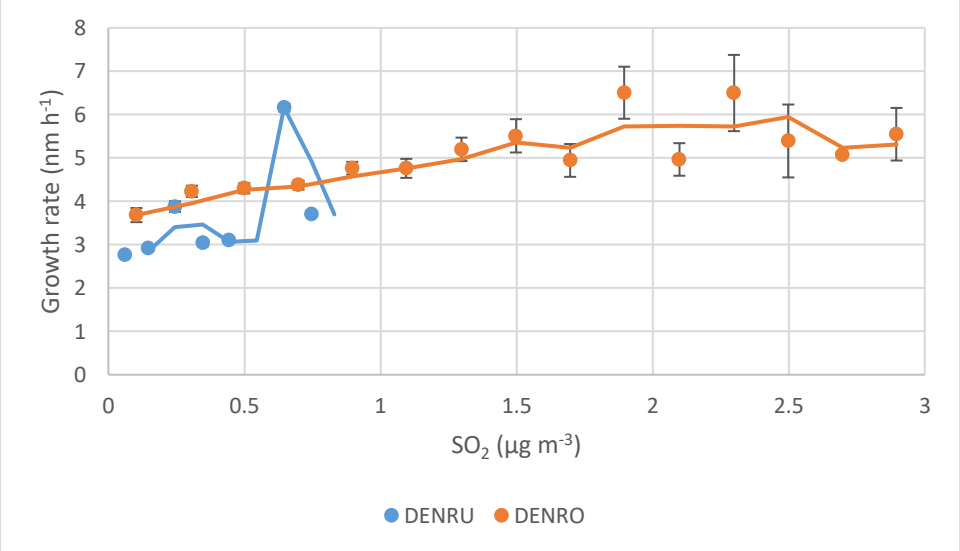
Figure 19 (continued)



(d)

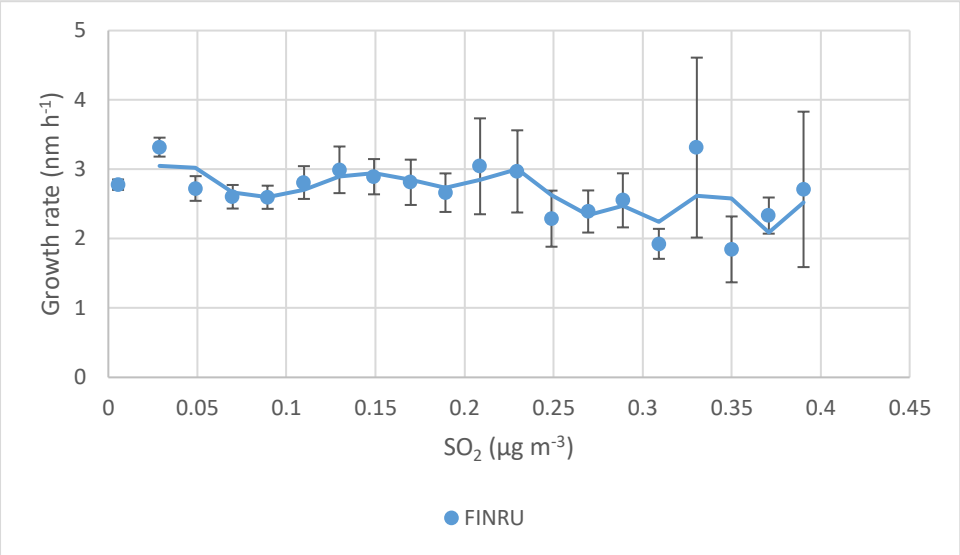


(e)

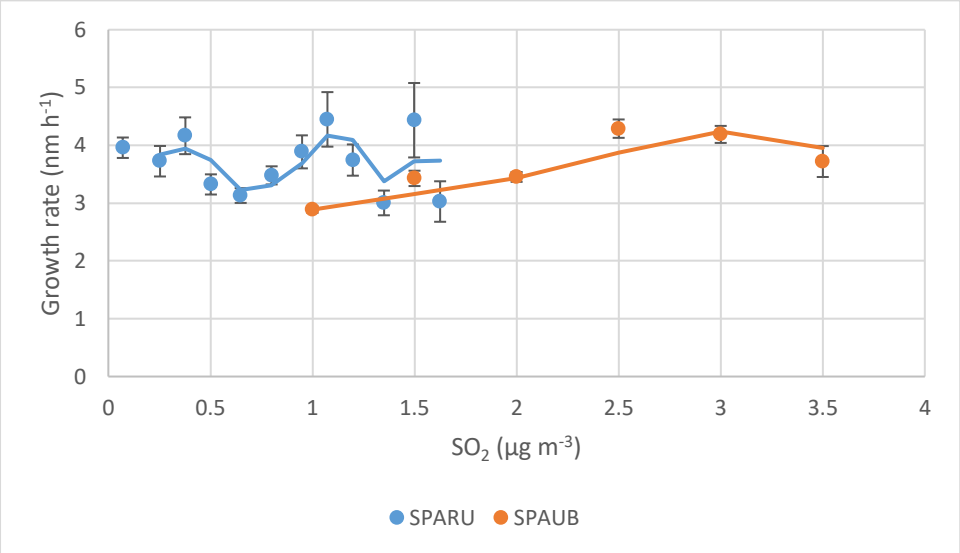


(f)

Figure 19 (continued)



(g)



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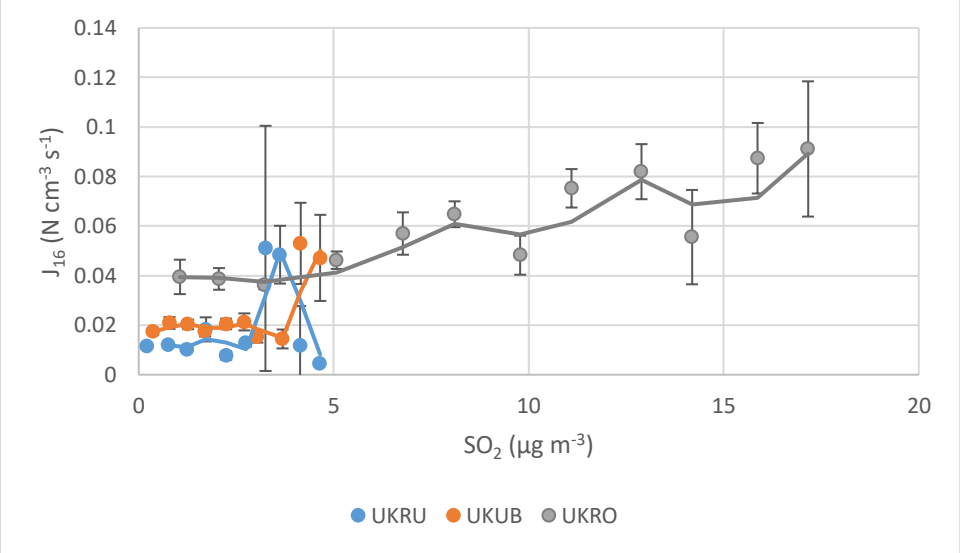
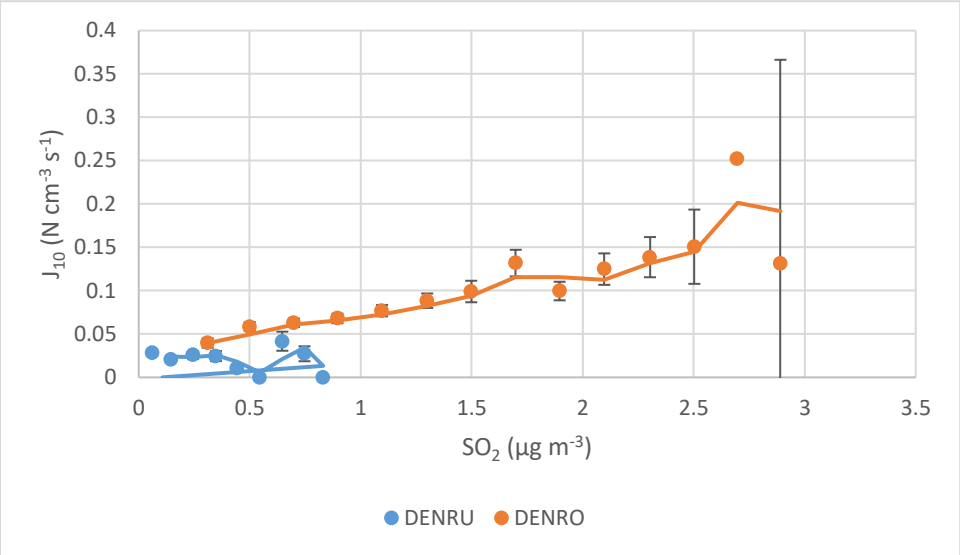
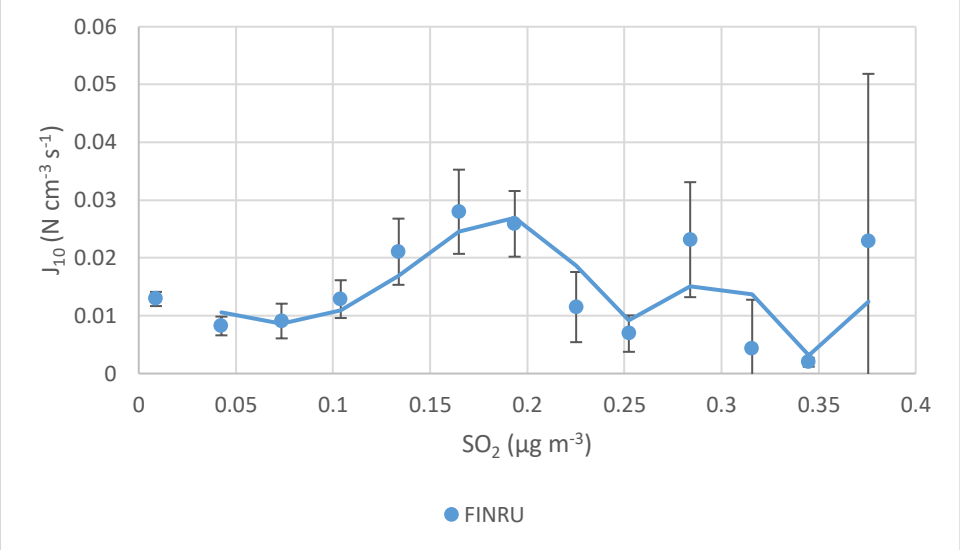


(i)

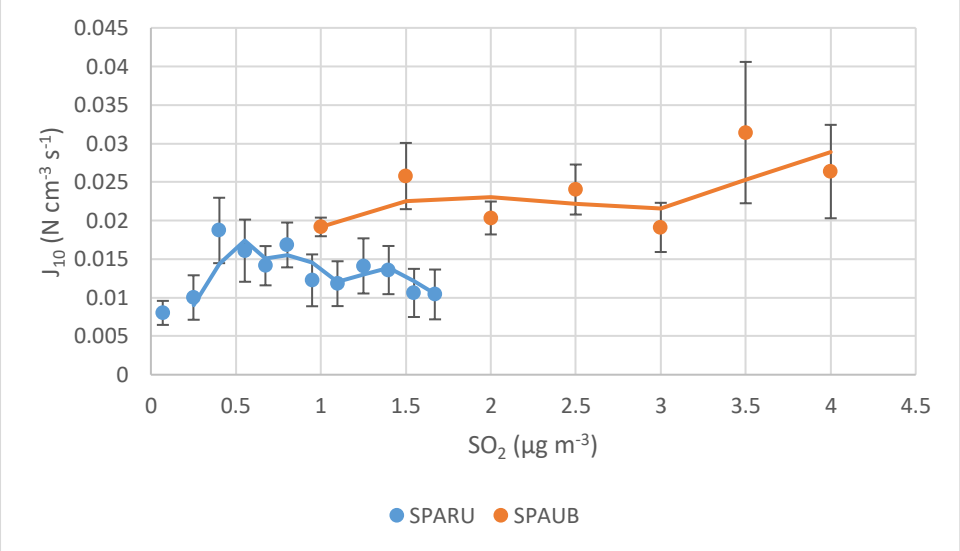
Figure 19 (continued)



(j)



(k)

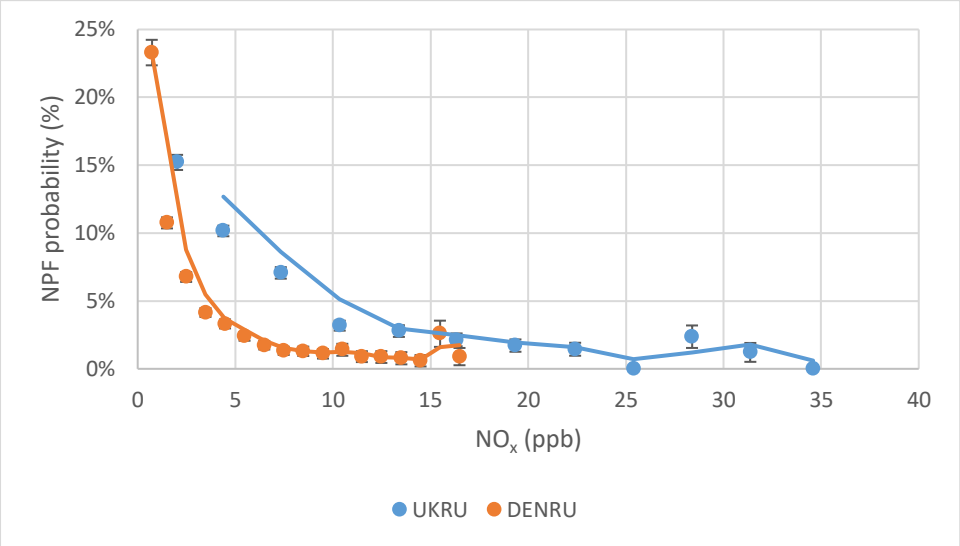


(l)

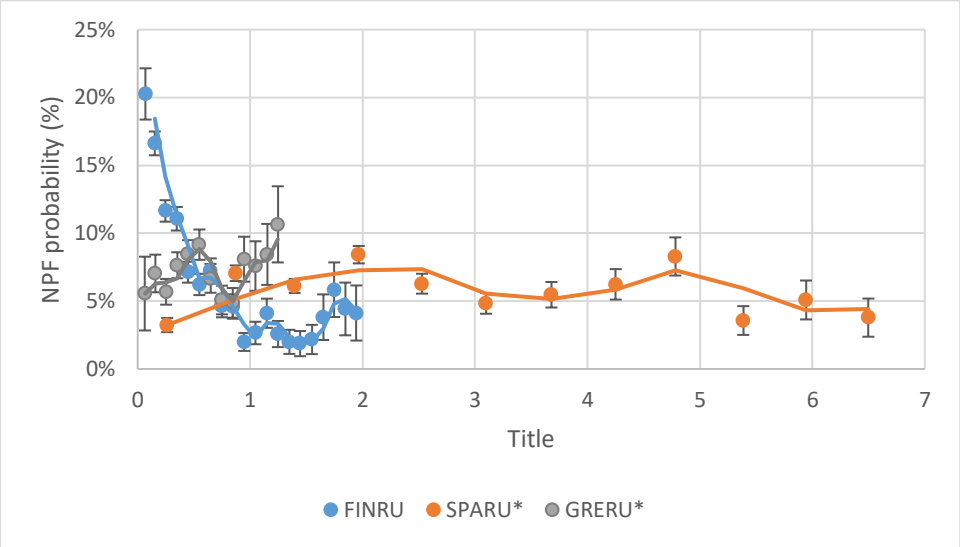
Nitrogen oxides or nitrogen dioxide (NO_x or NO_2)

NO_x and NO_2 are directly associated with pollution, which can be a limiting factor for NPF events as it increases the CS and may suppress the events (An et al., 2015), though with the reduction of SO_2 concentrations achieved the last couple of decades, there is possibility for oxidation products of NO_x to become an important component for NPF (Wang et al., 2020). For almost all sites (apart from GRERU) with available data a negative relationship between the NPF probability and NO_x (or NO_2) concentrations (depending on what data was available) was found when linear relationships were considered. Similarly, for all the sites but SPARU and GRERU, the correlations were strong with $R^2 > 0.43$. The rural background sites had a weaker relationship between the two variables compared to the urban sites, which is probably associated with them having rather low concentrations of NO_x (or NO_2) and variability, making the variations of this factor less important. Growth rate had weaker correlations with NO_x and different trends between the sites, either being positive or negative. The variable effect of NO_X on particle growth, shifting HOMs' volatility, was previously discussed by Yan et al. (2020). While variability was found for the background sites, all roadsides regardless of the strength of the relationship had positive relationships between NO_x and the growth rate. This may indicate the different components associated with the growth process at each type of site which, as found in other studies can be related to compounds associated with combustion processes that take place within the urban environment (Guo et al., 2020; Wang et al., 2017a). The formation rate presents variable trends either positive or negative. While much effort was made to isolate the effect of NPF events by taking a shorter time frame before the event, the effect of local pollution is still included, especially at the urban sites. As a result, no confident conclusions can be made.

Figure 20: Relationship of NO_x / NO_2 concentrations with NPF variables.

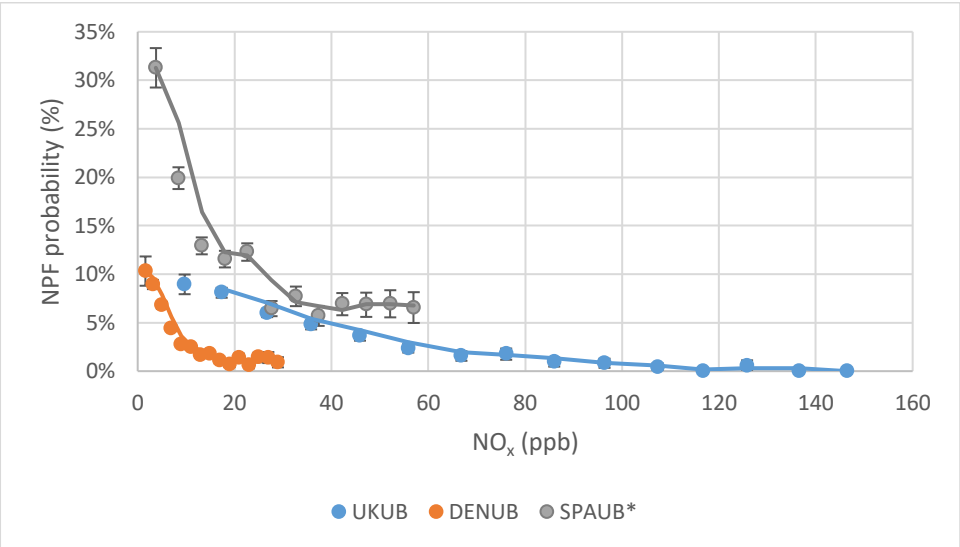


(a)



(b)

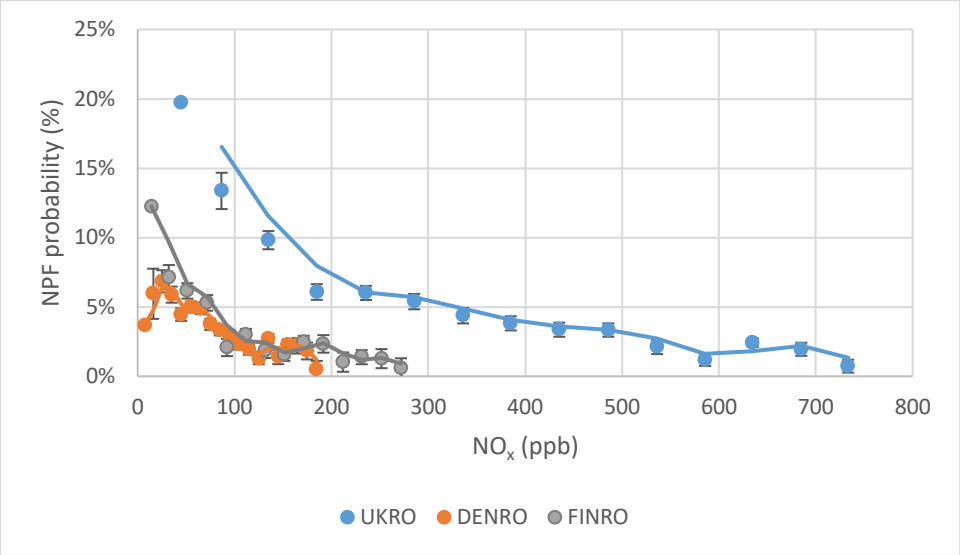
***NO_2 for SPARU and GRERU**



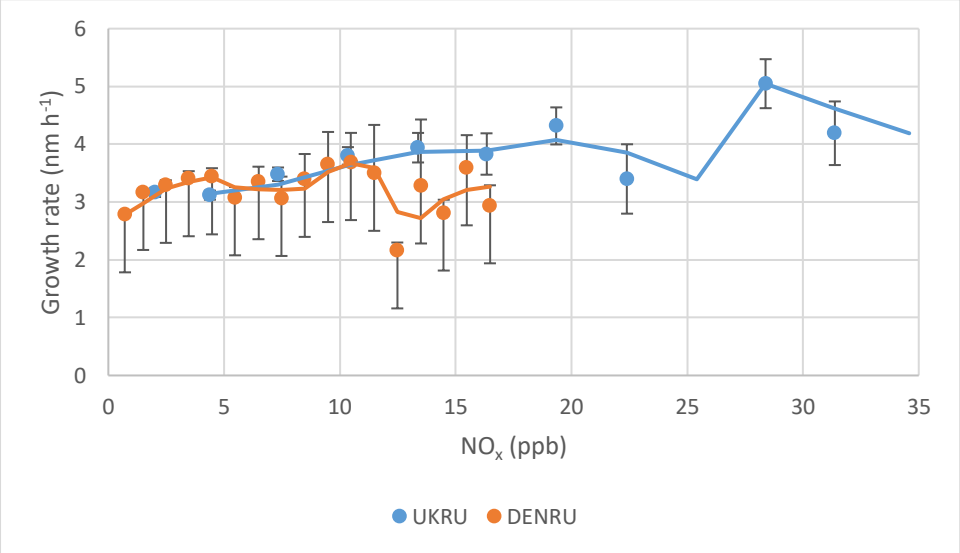
(c)

***NO_2 for SPAUB**

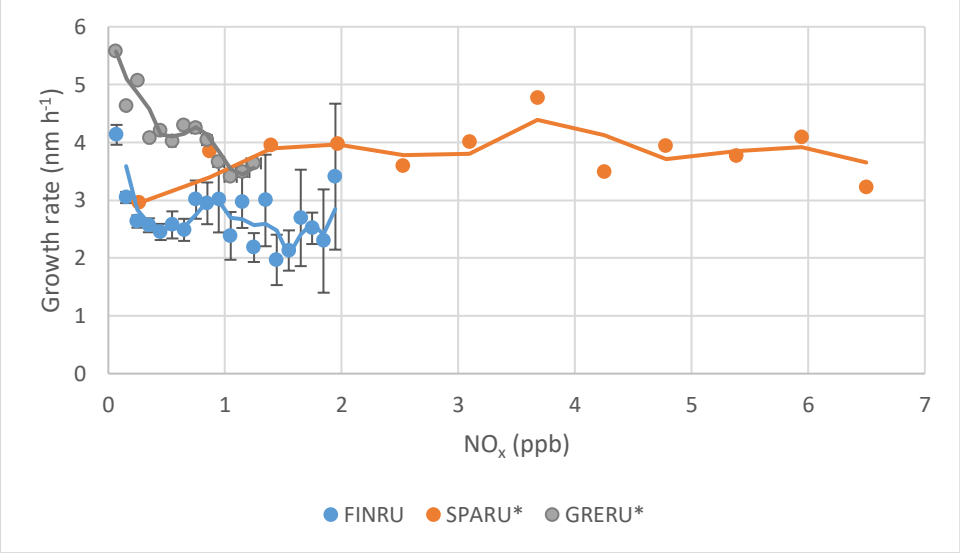
Figure 20 (continued)



(d)



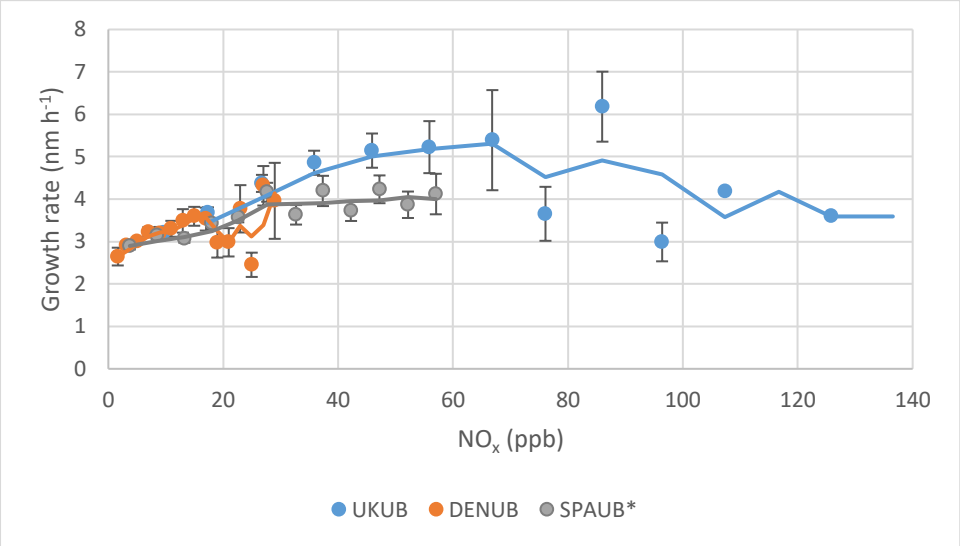
(e)



(f)

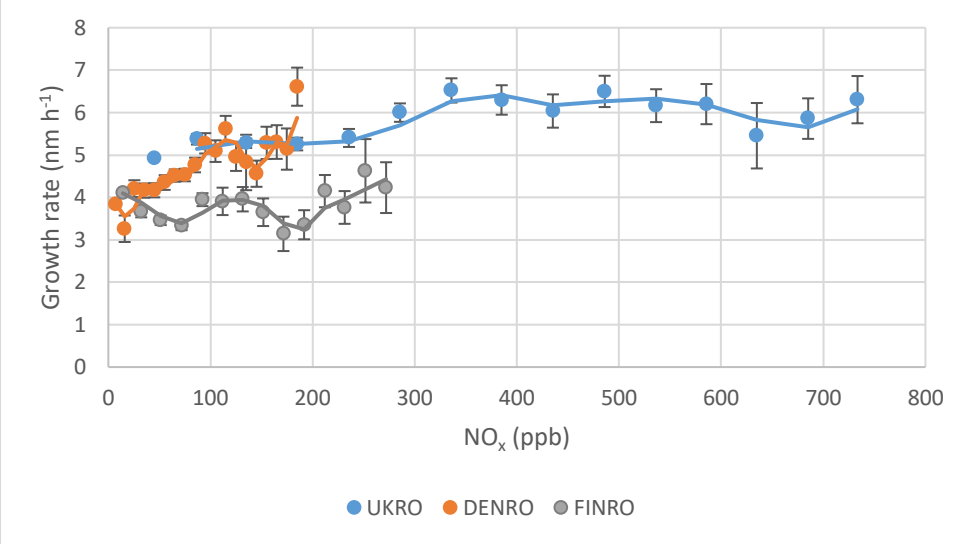
***NO_2 for SPARU and GRERU**

Figure 20 (continued)

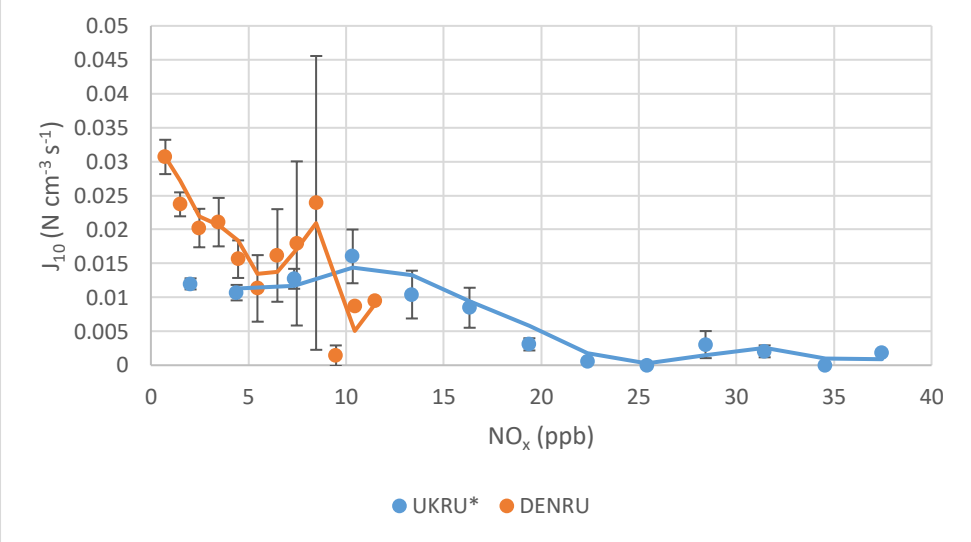


(g)

***NO_2 for SPAUB**



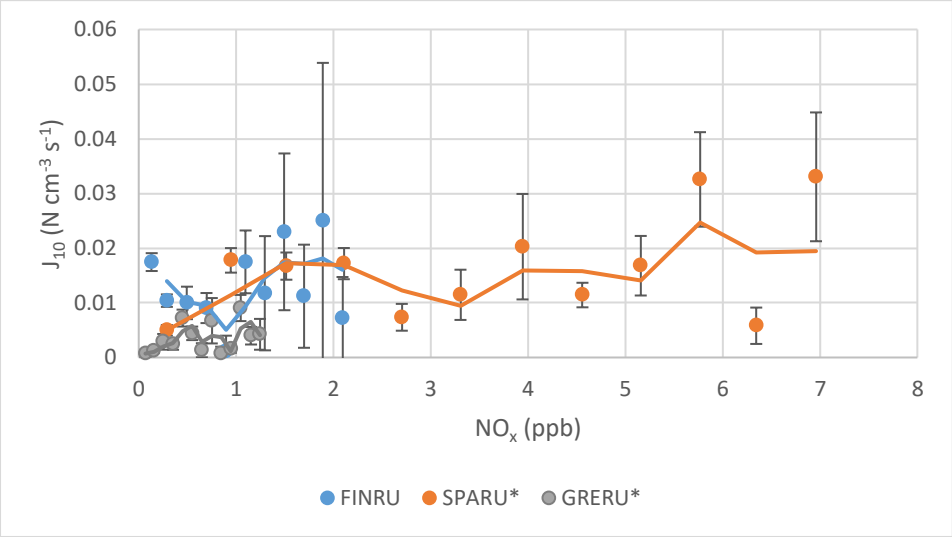
(h)



(i)

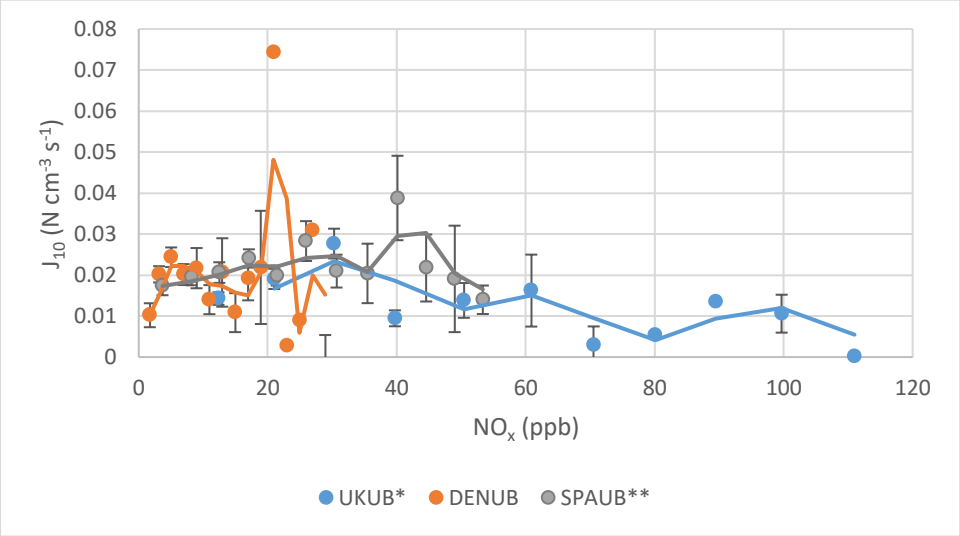
***J_{16} for UKRU**

Figure 20 (continued)



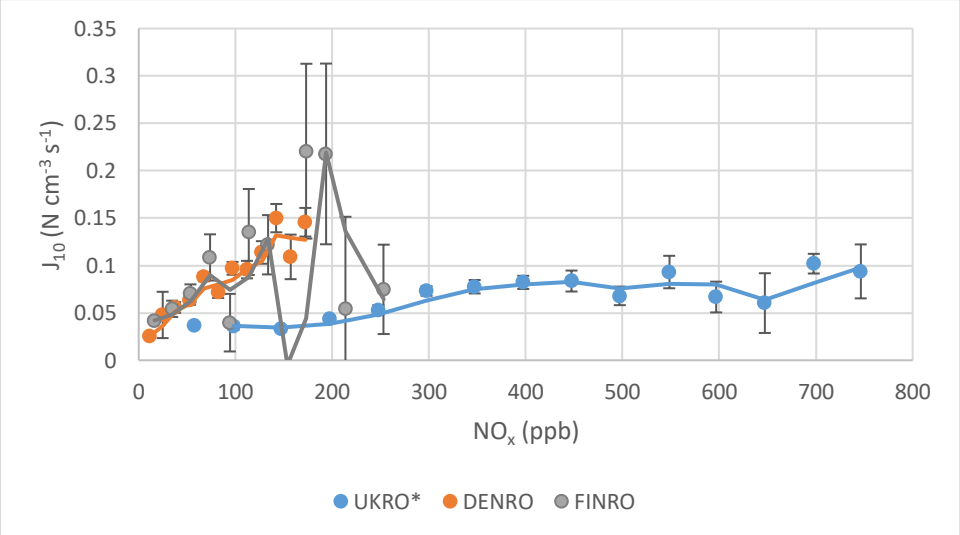
(j)

***NO_2 for SPARU and GRERU**



(k)

***J_{16} for UKUB **NO_2 for SPAUB**



(l)

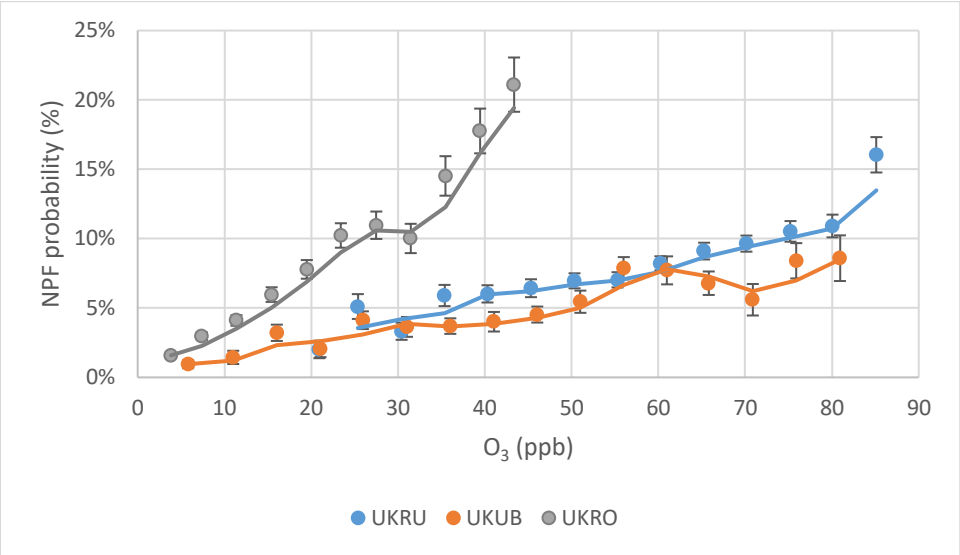
***J_{16} for UKRO**

Ozone (O_3)

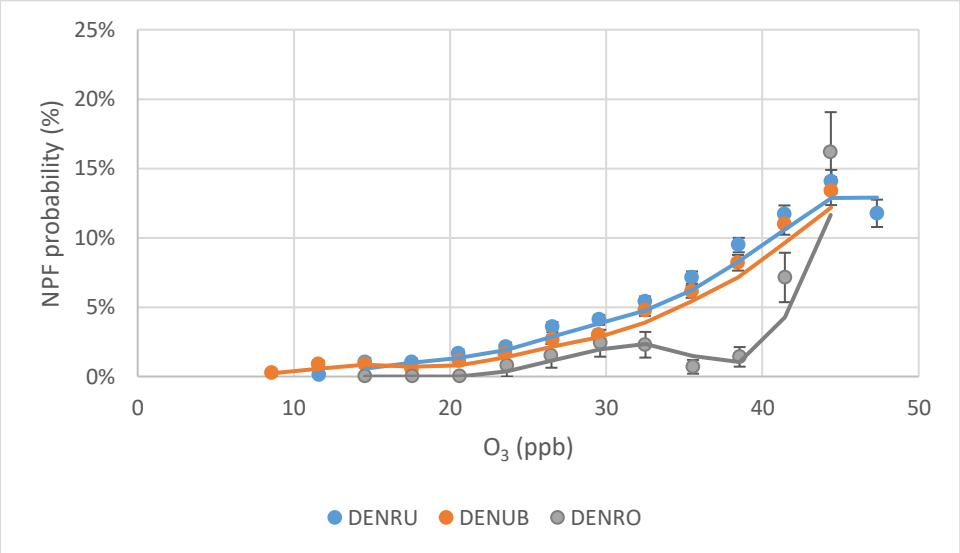
Ozone is typically the result of atmospheric photochemistry and is itself a source of hydroxyl radical through photolysis, or ozonolysis of alkenes both during daytime and night-time (Fenske et al., 2000). It might therefore be expected to act as an indicator of photochemical activity which promotes the oxidation of SO_2 and VOCs. Ozone concentrations may be directly related to the solar radiation intensity (though the correlations were not high for all the sites of the study) as well as the pollution levels in the area studied, and O_3 is considered as a positive factor in the occurrence of NPF events (Woo et al., 2001; Berndt et al., 2006). As for the solar radiation, there is a strong relationship between O_3 concentration and the probability for NPF events. This positive relationship was found to be stronger for the sites in northern Europe, while it was not significant for the sites from southern Europe (Spanish sites and GRERU) when linear relationships were considered, possibly indicating that O_3 is a less important factor at the southern sites. Specifically for the Spanish sites which have the highest average concentrations of O_3 with some extreme values (Querol et al., 2017), the relationship of O_3 concentrations with the NPF probability presents a unique trend (Figure 21d), having a clear peak then a steady decline at both sites (though at different O_3 concentrations). This trend seems to also occur at SPARU for the growth rate and to a lesser extent for the formation rate as well, though for different O_3 concentration ranges – figures 21i and n). The specific variability found at the Spanish sites was also studied by Carnerero et al., (2019). For sites with a marked seasonal variation in ozone, associations with NPF may be artefactual due to correlations with other variables such as temperature, RH and solar radiation.

Unlike the solar radiation though, the growth rate presents a negative relationship at the sites where the relationship between these two variables was significant (UKRU, UKUB, DENUB and FINRU), which might either be an indication of a polluted background that may have a negative effect on the growth of the newly formed particles (though the trends found for NO_x indicate differently) or specific chemical processes which cannot be identified due to the lack of detailed chemical composition data. A clear relationship between O_3 and the formation rate was only found for a few sites (though the trends become a lot clearer if some values are removed from the extreme lower or higher end). This way the relationships become strong, but positive for some areas and negative for others without any clear trend (type or location of the site, O_3 concentrations etc.). When linear relatiosnhips were considered, no clear association between these two variables was found as the sites with strong relationships have both positive and negative relationships and as a result no confident conclusions can be drawn.

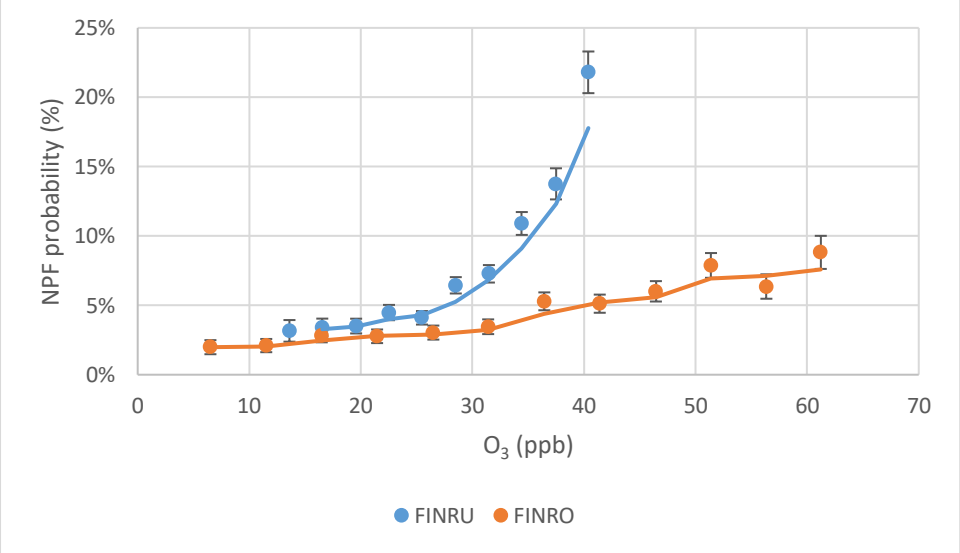
Figure 21: Relationship of O_3 concentrations with NPF variables.



(a)

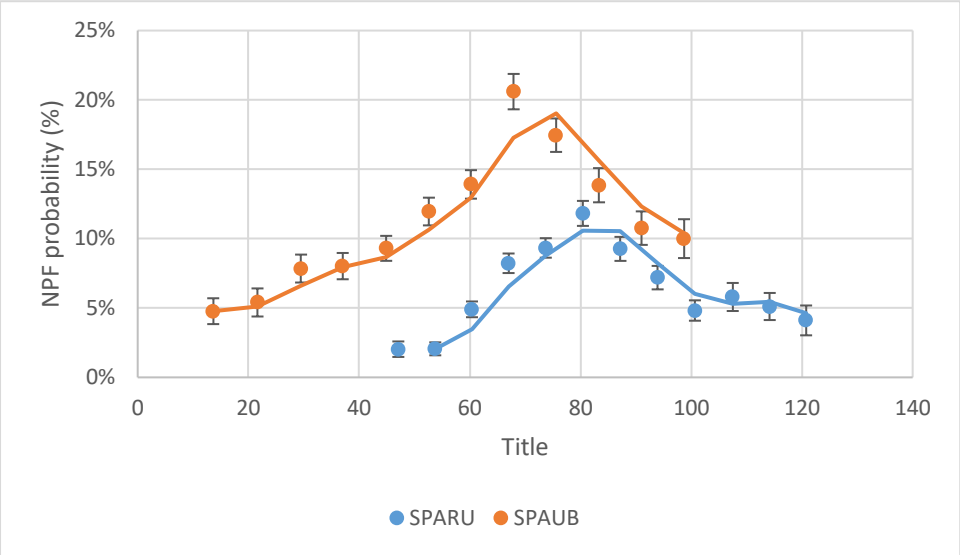


(b)

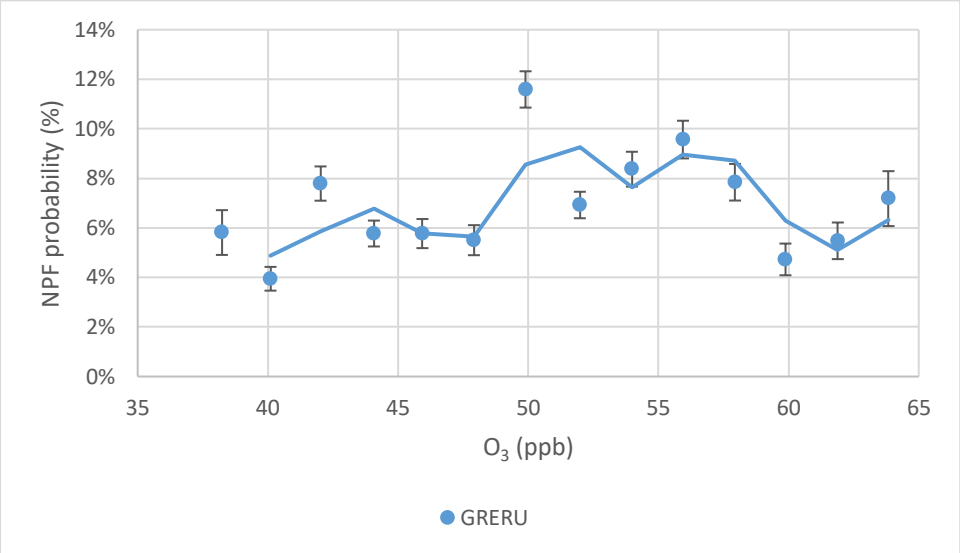


(c)

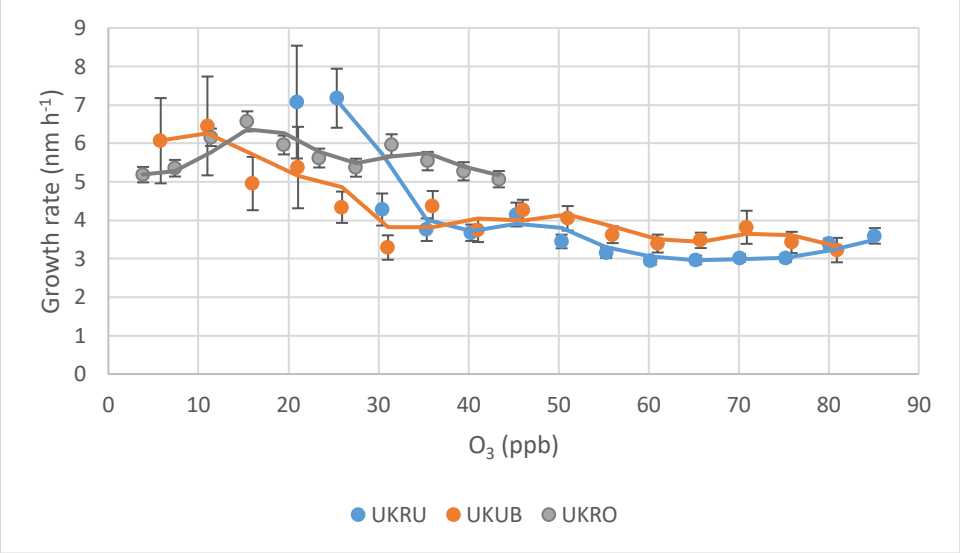
Figure 21 (continued)



(d)

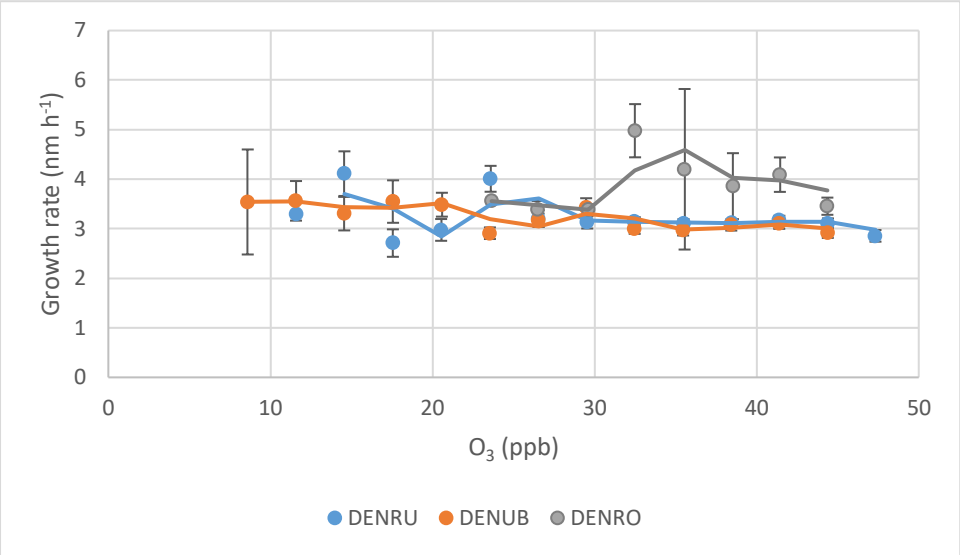


(e)

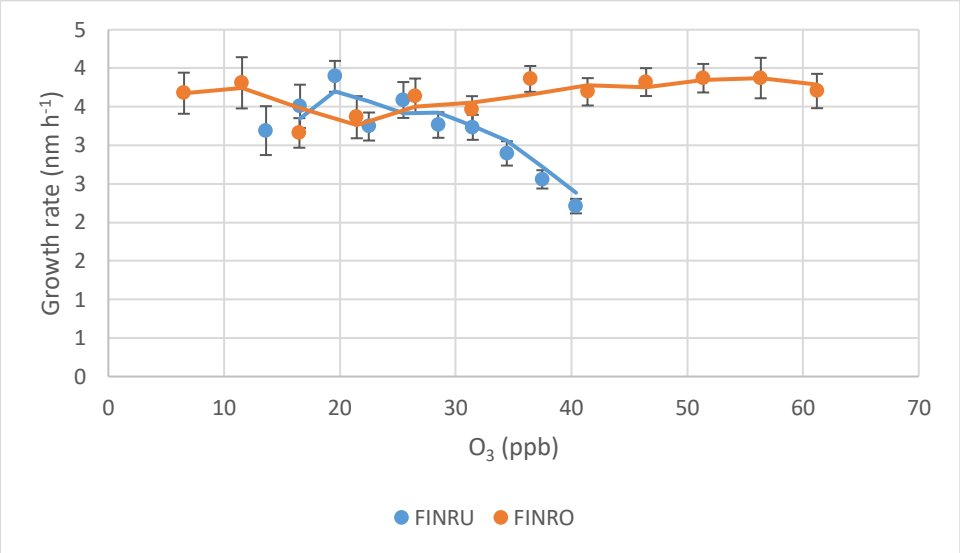


(f)

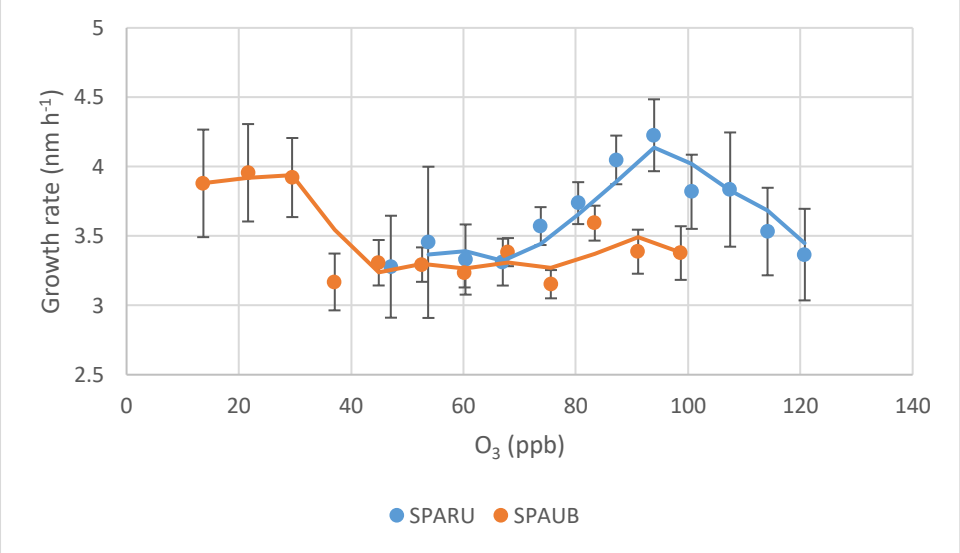
Figure 21 (continued)



(g)

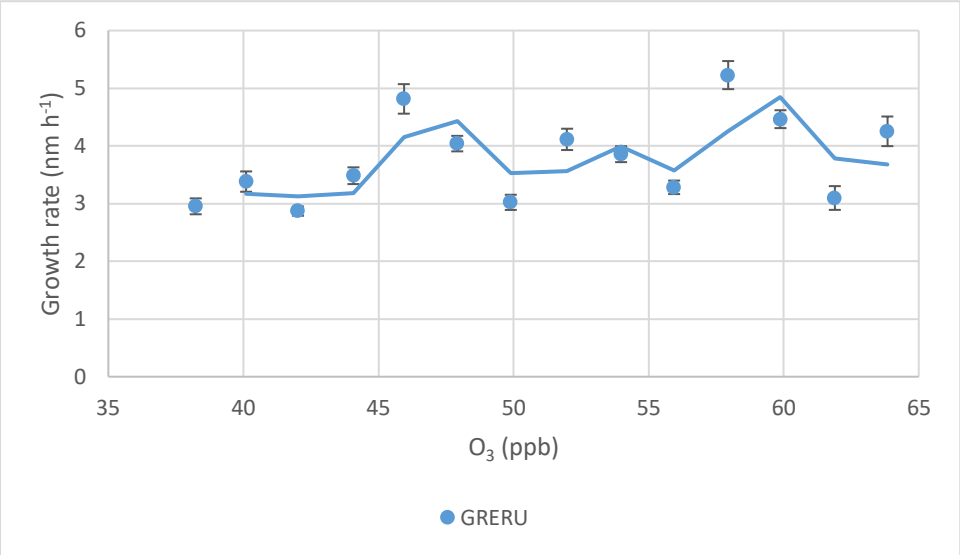


(h)

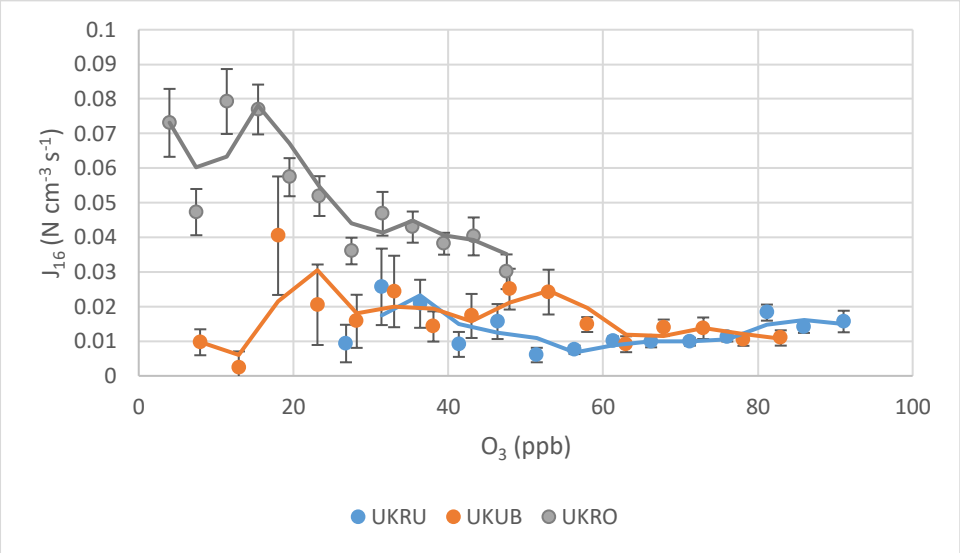


(i)

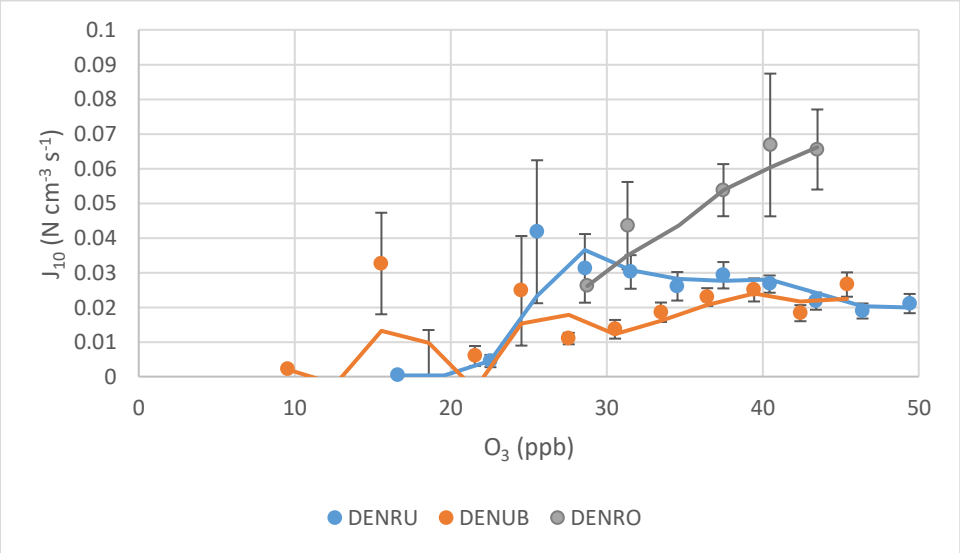
Figure 21 (continued)



(j)

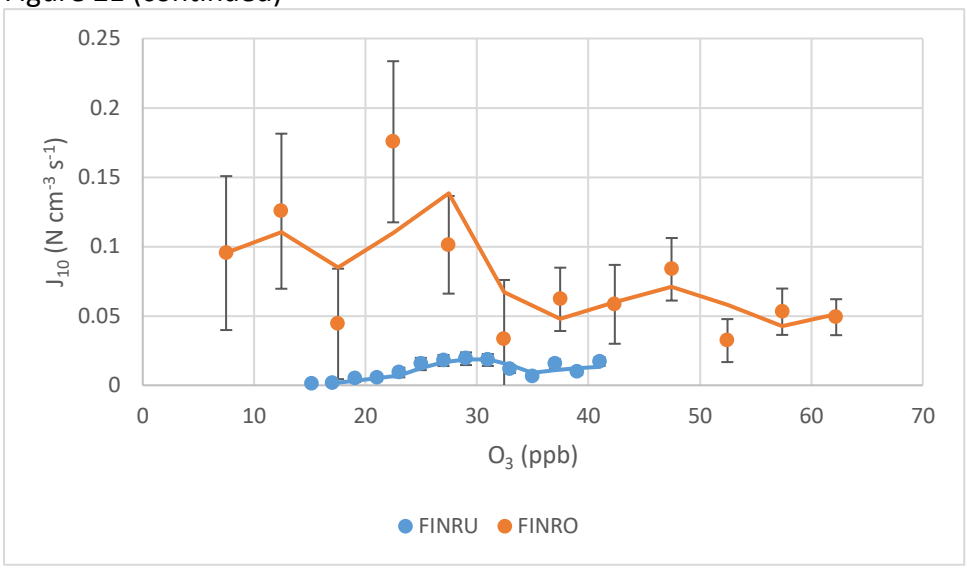


(k)

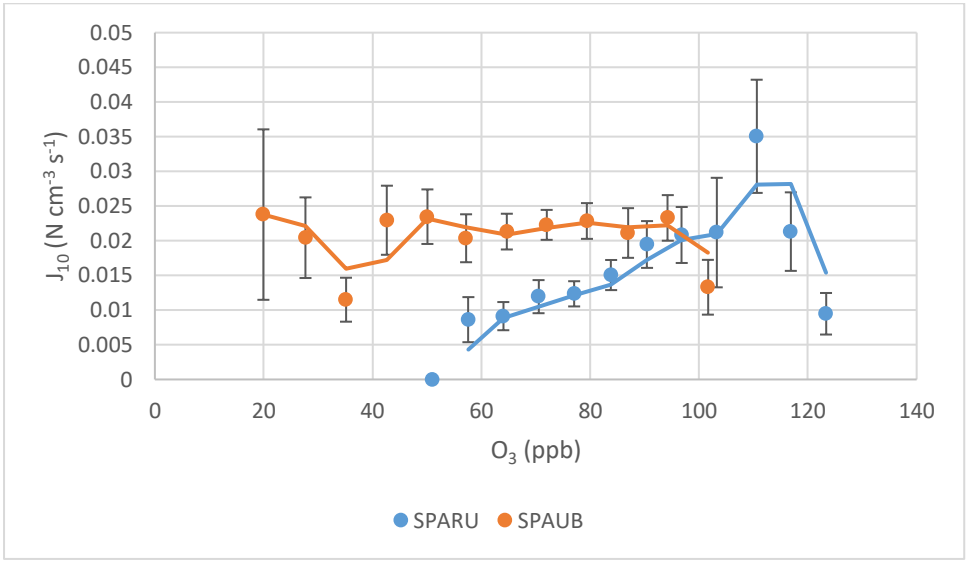


(l)

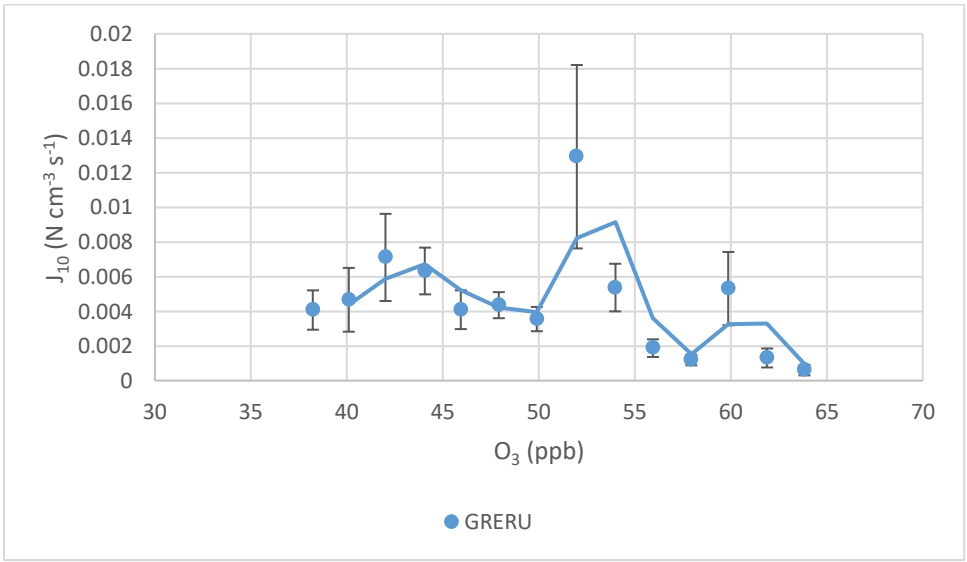
Figure 21 (continued)



(m)



(n)



(o)

Organic compounds

Particulate organic carbon (OC)

Organic carbon (OC) compounds are considered as components with importance in the growth of newly formed particles, with a role that becomes increasingly important as the size of the particles becomes larger (Nieminen et al., 2010; Zhang et al., 2012; Shrivastava et al., 2017). Particulate OC, the data for which is available in the present study (Organic Mass for FINRU), can be associated with pollution, especially in the urban environment. Only a few of the sites of the present study were found to have a strong negative relationship ($R^2 >$ 0.50) of particulate OC with the NPF probability (UKUB, UKRO and DENRU), when linear relationships were considered. Regardless though of the strength of this relationship, all other sites (apart from FINRU) had a negative relationship between these two variables as well, consistent with increased concentrations of particulate OC being associated with increased pollution, which is a suppressing factor in the occurrence of NPF events. Growth rate on the other hand was found to have a slight positive relationship ($R^2 > 0.40$) for most of the sites. This relationship appeared to be stronger (higher R^2) at the roadsides with available data compared to their respective rural background sites. The relationship between particulate OC and the growth rate was positive at all the sites with available data showing that, despite its effect in the occurrence of NPF events, it is still a favourable variable for the growth of the particles. The formation rate was found to have an unclear relationship with particulate OC concentrations. A positive trend was found for the urban sites, which may indicate that its role may be more important at such sites (it should be reminded though that the metric is also associated to an extent with the growth rate). When linear relationships were considered this relationship was significant at half of the sites with available data and was positive in all cases (UKUB, UKRO, DENRU, DENRO).

Volatile organic compounds (VOCs)

Many volatile organic compounds have been found to be associated with the NPF process. Benzene, toluene, ethylbenzene, m+p-xylene, o-xylene and trimethylbenzenes have been reported to be able to form Highly Oxygenated Organic Molecules (HOMs) in flow tubes (Wang et al., 2017a; Molteni et al., 2018), which may act as contributors to particle nucleation and/or growth. Xylenes, and to a lesser extent trimethylbenzenes, are the most efficient at forming HOMs. Benzene and toluene are less efficient and will form more volatile HOMs. These HOMs may all be too volatile to form new particles, though this is not yet confirmed. Chamber studies involving H_2SO_4 and trimethylbenzene oxidation products were associated with high formation rates when measuring $J_{1.5}$ (Metzger et al., 2010). All these HOMs though will be sufficiently involatile to contribute to particle growth. Those with higher oxygen content or carbon number will be classed as LVOC and if they dimerise, they will form ELVOC (Bianchi et al., 2019). Monoterpenes can also form HOMs which drive both the formation (Ehn et al., 2014; Riccobono et al., 2014) and growth (Tröstl et al., 2016), while isoprene can act as a sink for hydroxyl radical (Kiendler-Scharr et al., 2009) and is not as effective in HOM and secondary organic aerosol formation compared to monoterpenes (McFiggans et al., 2019).

Volatile organic compound data was available for three of the sites of this study (Table S3). Two of the sites with VOC data were from the rural background and the roadside in the UK. Most of the compounds are associated with combustion sources and were found to have a negative relationship with NPF event occurrence at both sites, with high R^2 in most cases when linear relationships were considered. Additionally, isoprene, which may have either biogenic or anthropogenic sources (Wagner and Kuttler, 2014) was also found to have a negative relationship with NPF event occurrence at Marylebone Road-UKRO. This result is in

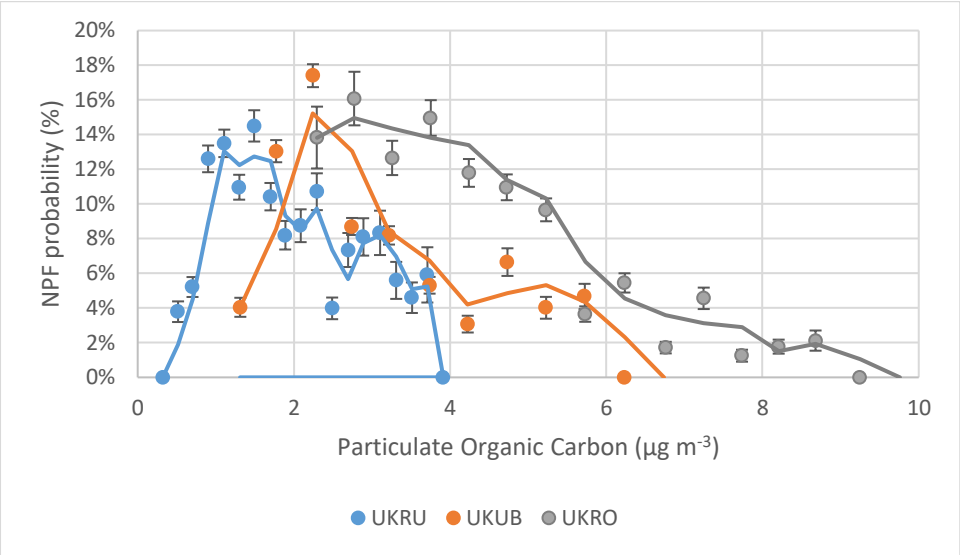
line with the VOCs being strongly correlated with particulate OC (which presented a negative relationship with NPF event probability, as previously discussed), as well as with the CS (which also presented a negative relationship with NPF event probability, as mentioned in the CS analysis), further associating these compounds with combustion emissions.

Growth rate was found to have a positive relationship with VOCs in almost all cases for both UK sites. Few exceptions were found (with only 1,3 butadiene having a relatively high R^2) which presented a negative relationship with the growth rate in rural Harwell-UKRU. Finally, the formation rate presented a different behaviour between the two sites. At Harwell-UKRU, the relationship was unclear in most cases, with a group of VOCs presenting a negative relationship with the formation rate (ethane, ethene, propane, 1,3 butadiene, toluene, ethylbenzene, o-xylene and 1,2,4 trimethylbenzene – with $R^2 > 0.40$), two VOCs presented a rather clear positive relationship with the formation rate (iso-pentane and 2-methylbenzene) and the rest of the VOCs had an unclear relationship. At Marylebone Road-UKRO though, VOCs presented a positive relationship with the formation rate (for particles of diameter 16 nm). This is probably due to the fact that these VOCs are associated with pollution emissions (as mentioned earlier) and though a smaller time window was chosen to avoid including the effect of the morning rush hour traffic, this is very difficult in the traffic polluted environment of Marylebone Road-UKRO.

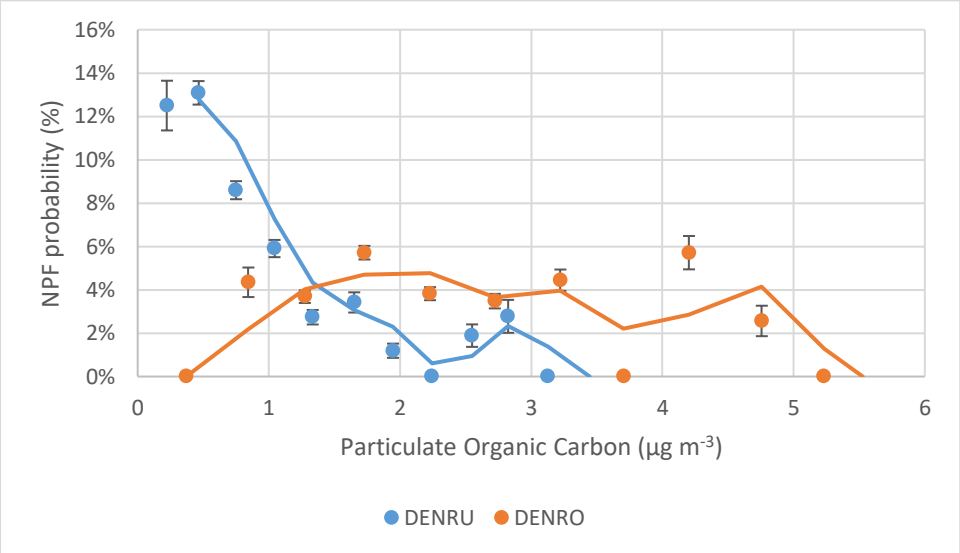
As Hyytiälä (FINRU) is a rural background site far from the direct effect of combustion emissions, different VOCs were measured, which mainly originate from biogenic sources rather than anthropogenic ones. The results were mixed and less clear compared to those from the UK sites (mainly due to the smaller dataset), and three groups were found with specific associations with NPF probability. The first group, including acetonitrile, acetic acid and Methyl Ethyl Ketone (MEK) presented a slight positive relationship. The second group

presented a negative relationship, with the VOCs in this group being MEK, monoterpenes, benzene, isoprene and toluene (only the last two have $R^2 > 0.50$). Finally, the third group included VOCs that presented a peak and then a decline for higher concentrations including methanol, and acetone. Two groups of VOCs were found with respect to their relationship with the growth rate. The one with a positive relationship containing methanol, acetonitrile, acetone, acetic acid, isoprene, MEK, monoterpenes and toluene, while acetaldehyde, MEK and benzene had a negative relationship, with relatively high R^2 in most cases. Finally, the results with the formation rate were unclear with only a handful presenting weak positive (methanol, acetic acid and benzene) or negative (MEK) relationships that do not appear to be significant.

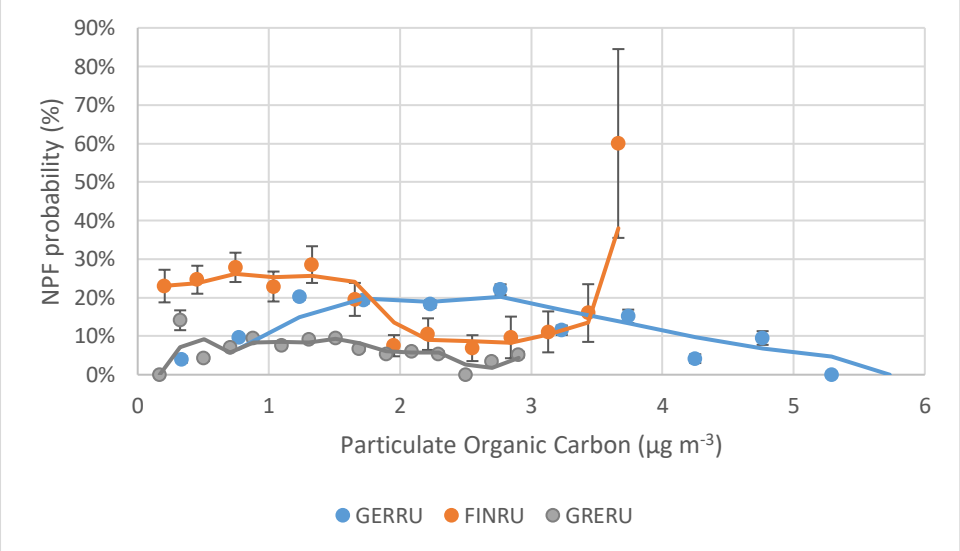
Figure 22: Relationship of particulate organic carbon concentrations with NPF variables.



(a)

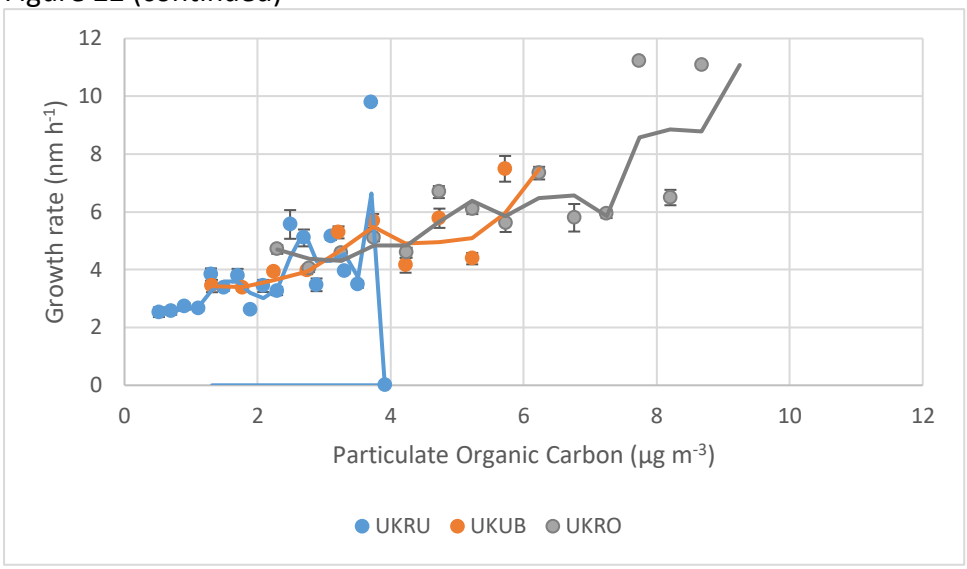


(b)

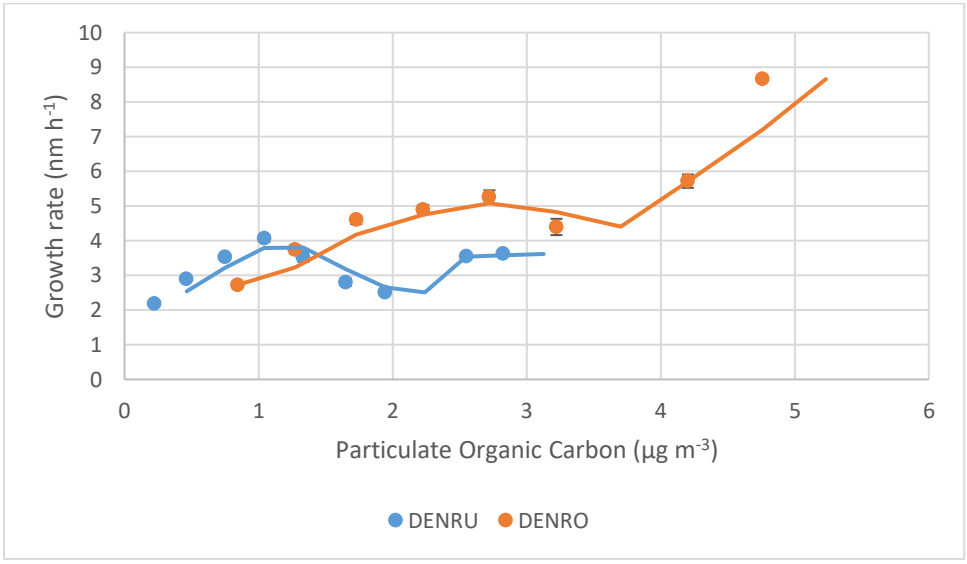


(c)

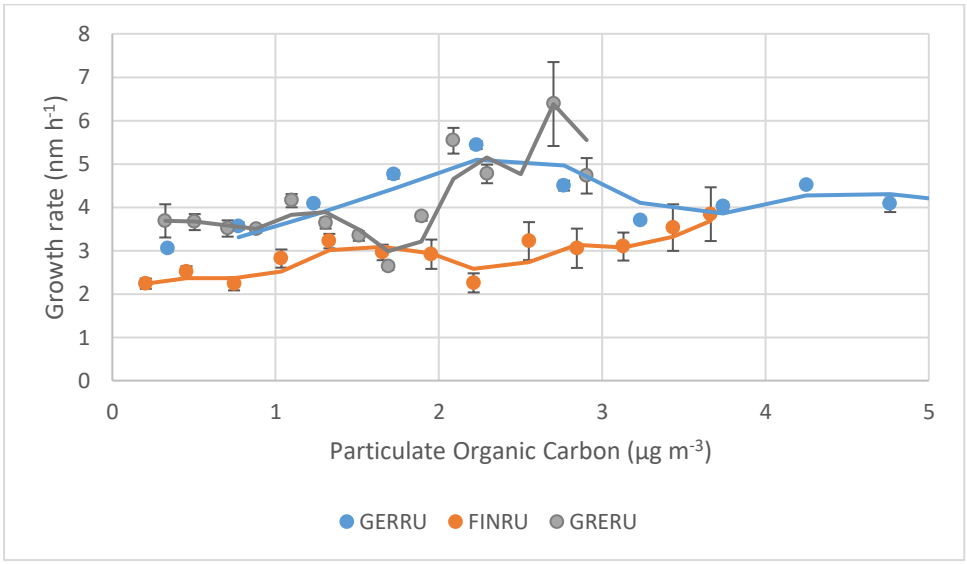
Figure 22 (continued)



(d)

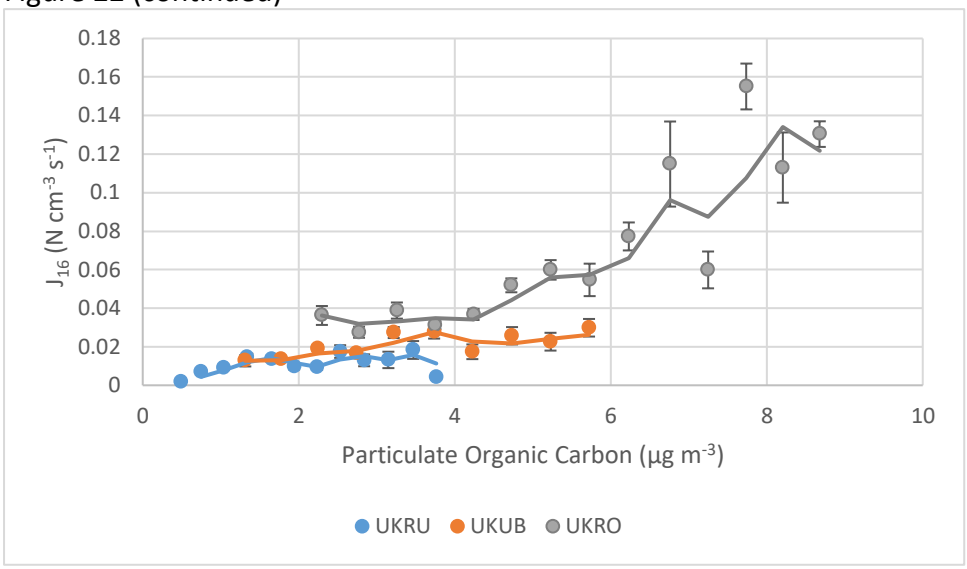


(e)

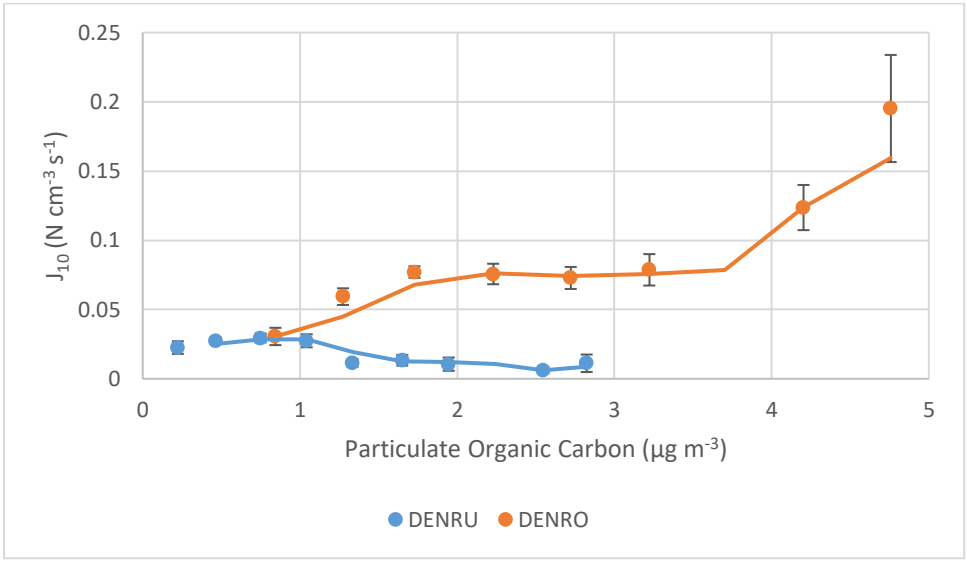


(f)

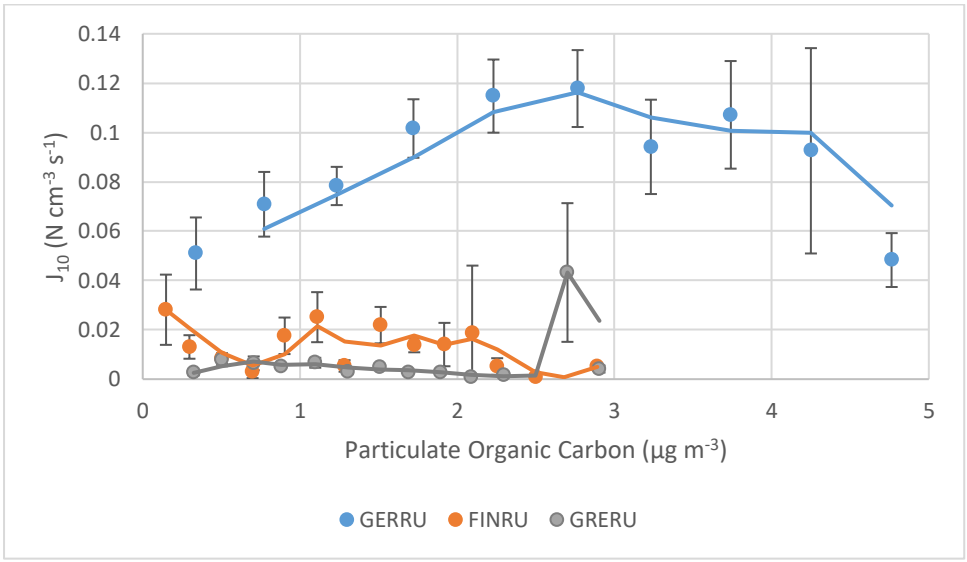
Figure 22 (continued)



(g)



(h)

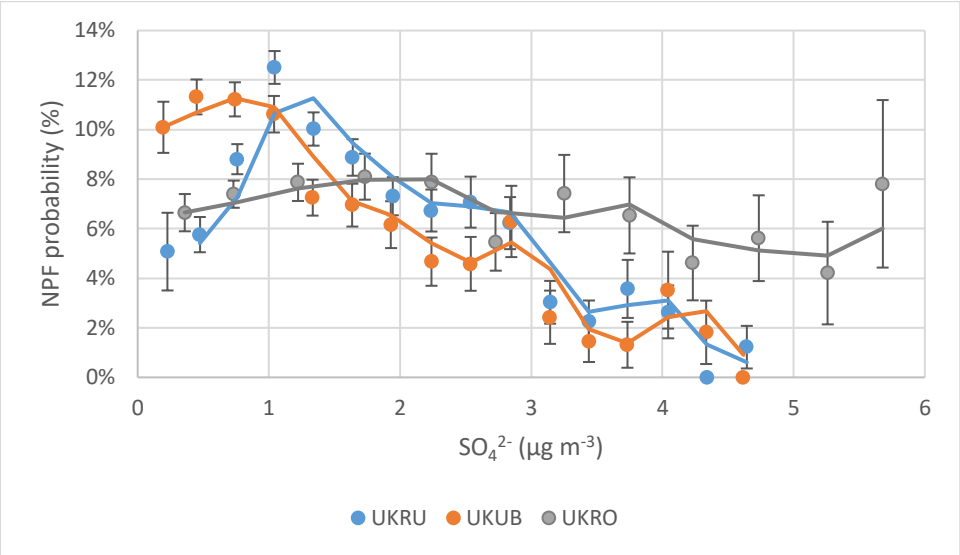


(i)

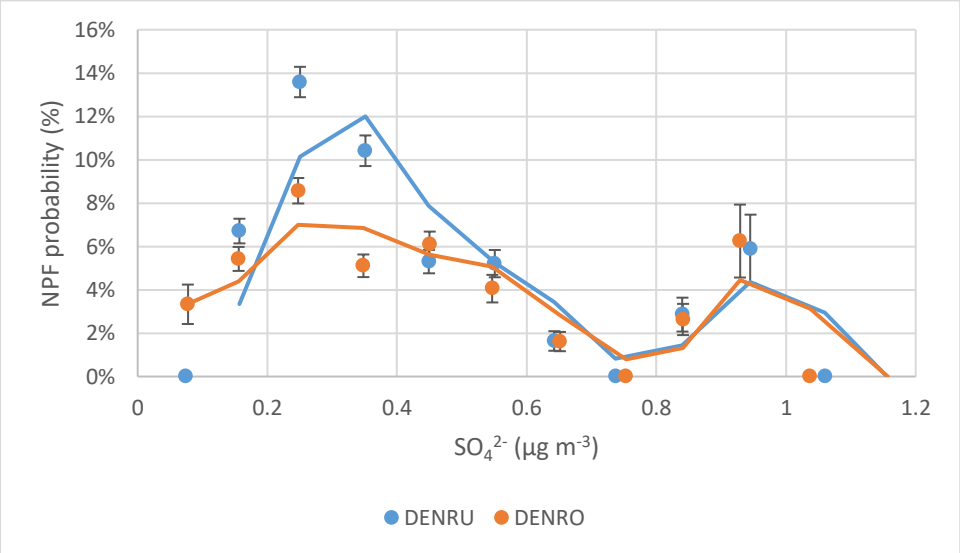
Sulphate (SO_4^{2-})

Sulphate (SO_4^{2-}) is a major secondary constituent of aerosols. Secondary SO_4^{2-} aerosols largely arise from either gas phase reaction between SO_2 and OH, or in the aqueous phase by the reaction of SO_2 and O_3 or H_2O_2, or NO_2 (Hidy et al., 1994). In environments where SO_4^{2-} chemistry is dominant (i.e. remote areas), SO_4^{2-} and ammonium (bi) sulphate ($(NH_4)_2SO_4$ and NH_4HSO_4) particles are a large relative contributor to aerosol mass, while this contribution is lower in environments where other emissions are also significant (i.e. urban areas where the secondary NO_3^- relative contribution is a lot higher). While not well established, a possible association of SO_4^{2-}-containing compounds and variables of NPF events was found in previous studies (Beddows et al., 2015; Minguillón et al., 2015; Wang et al., 2017b). In the present study, only a few sites had SO_4^{2-} data available, for PM_1 (FINRU), $PM_{2.5}$ (Danish sites) or PM_{10} (rest of the sites). While this data cannot be considered as directly associated with the ultrafine particles, for two sites with available AMS (Aerosol Mass Spectrometer) data for ultrafine particles, the direct comparison between SO_4^{2-} aerosol in PM and in the range of particles of about 50 nm, very high correlations were found (results not included). For all the sites with available data the NPF probability presented a negative relationship. The significance of this relationships was found to be relatively high ($R^2 > 0.50$) only for background sites, when linear relationships were considered (apart from GERRU, which has rather low concentrations and probably different mechanisms for the NPF events). Similarly, the growth rate presented a positive relationships at all sites regardless of its significance, when linear relationships were considered. Finally, the formation rate did not present a clear trend as it was found to have both negative and positive relationships for different sites. This relationship was significant only for two rural sites (UKRU and DENRU) and as a result no assumptions can be made.

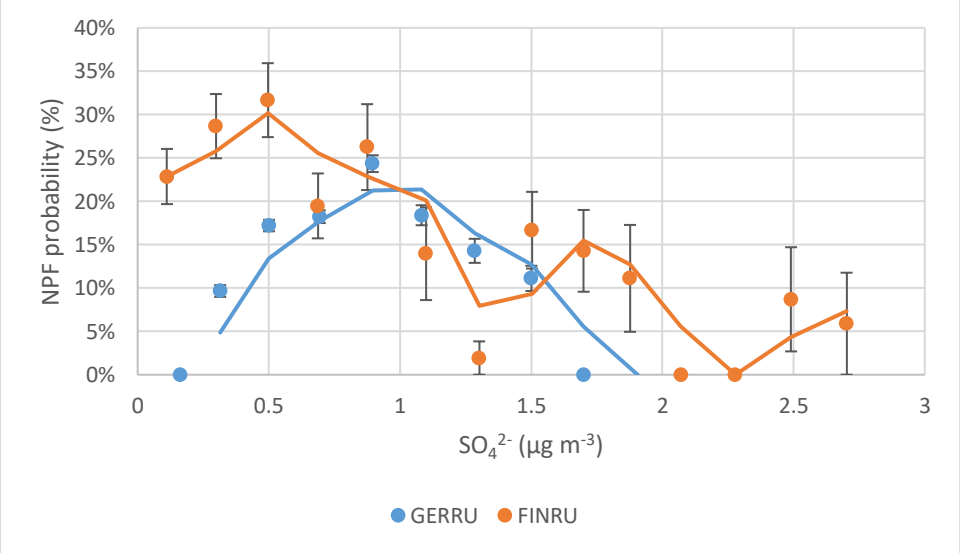
Figure 23: Relationship of SO_4^{2-} concentrations with NPF variables.



(a)



(b)



(c)

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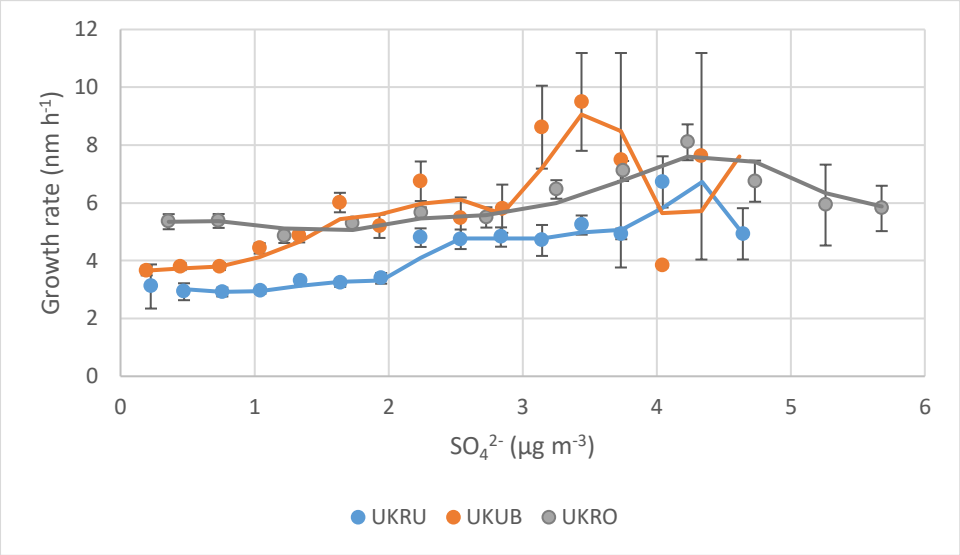
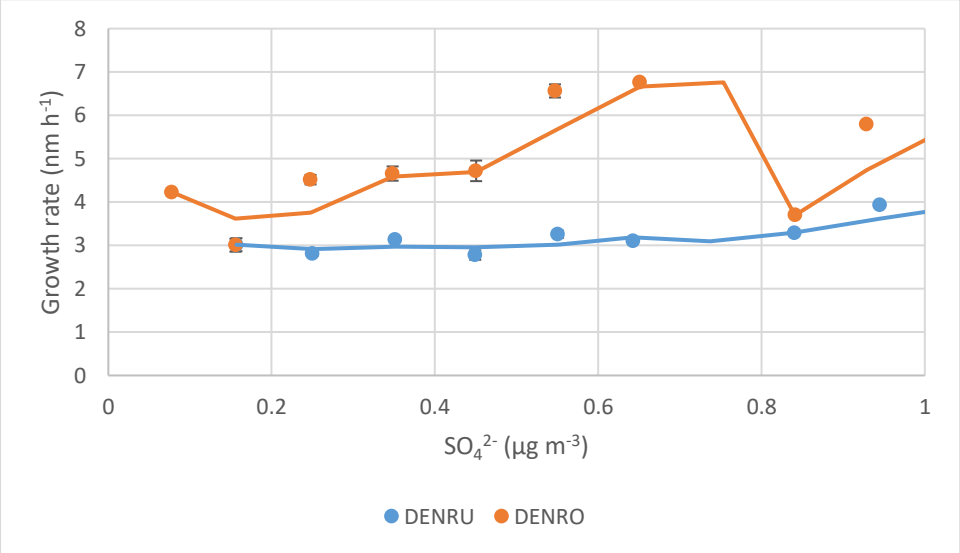
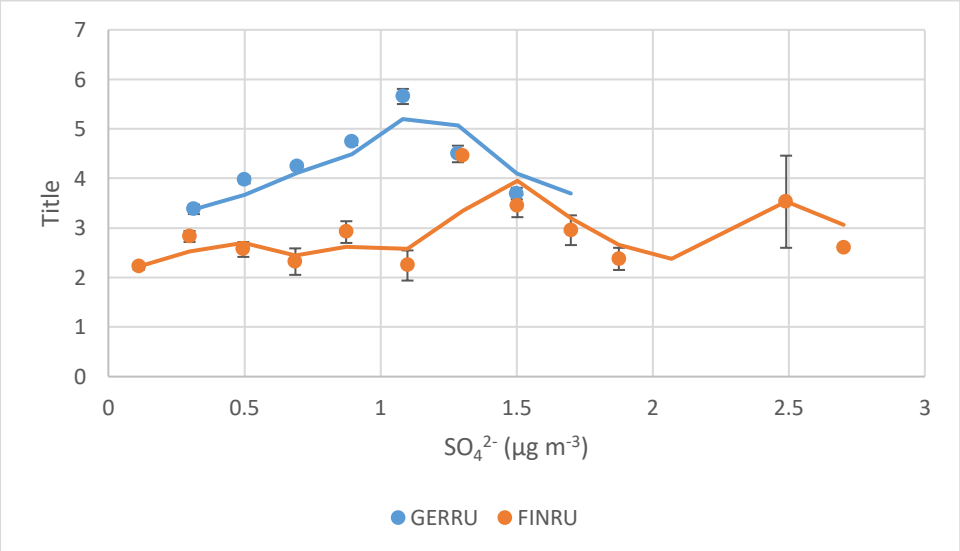


(d)

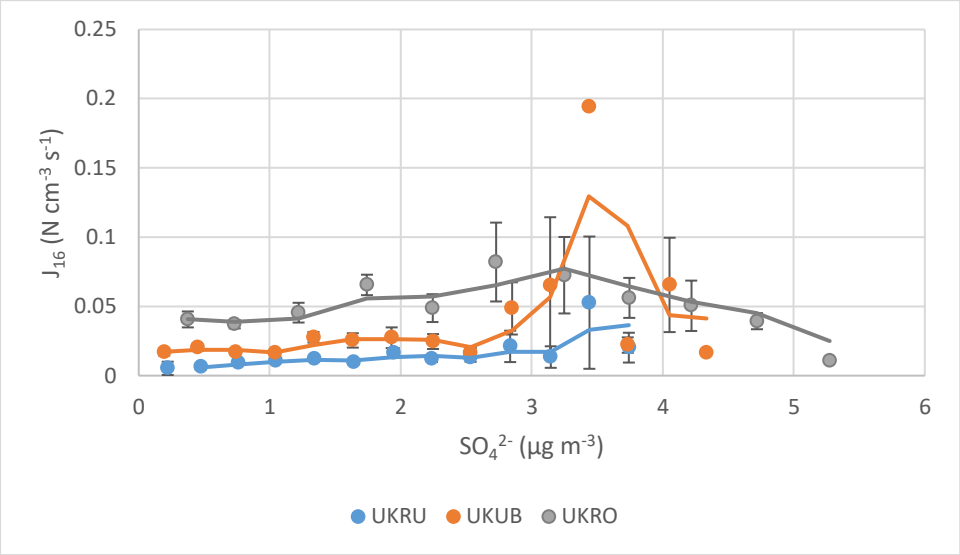


(e)

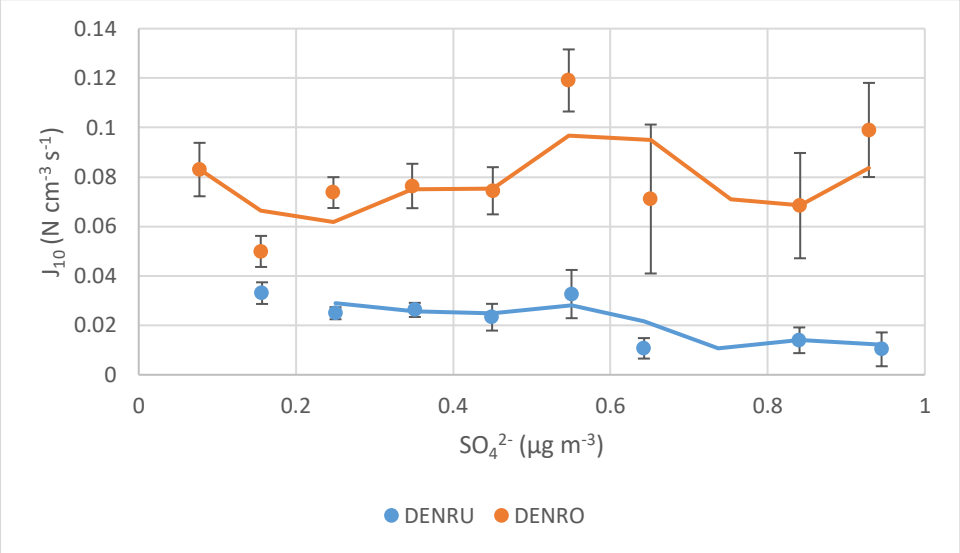


(f)

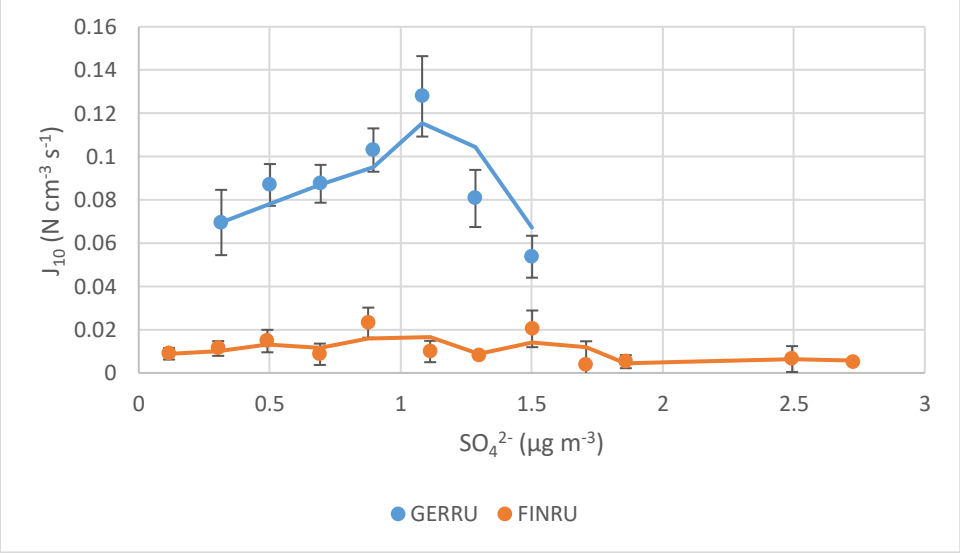
Figure 23 (continued)



(g)



(h)

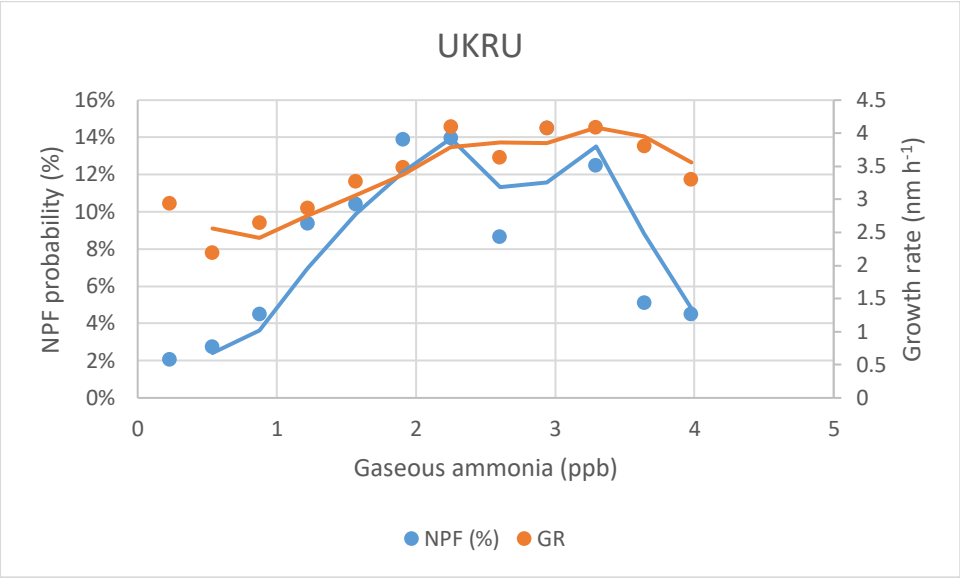


(i)

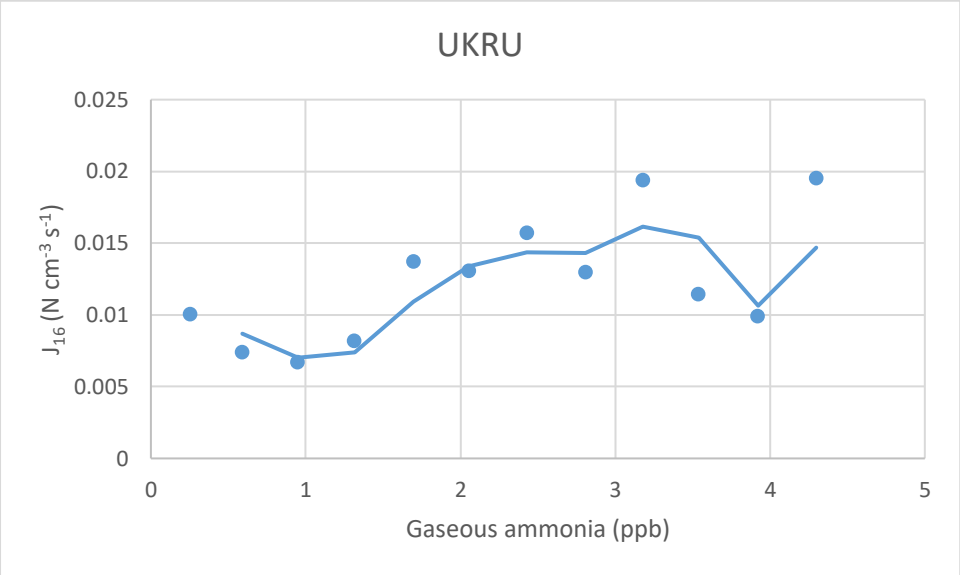
Gaseous ammonia (NH_3)

Ammonia (NH_3) can be an important compound in the nucleation process according to the ternary theory (Napari et al., 2002). It was found that elevations in NH_3 concentrations can lead to elevations to NPF rate (Lehtipalo et al., 2018) and it was also found to be an important factor for NPF event occurrence even when stronger bases are present in high concentrations (Glasoe et al., 2015). No significant variation was found though between event and non-event days in a previous study in Harwell-UKRU (Bousiotis et al., 2019). Data for gaseous ammonia was only available for Harwell-UKRU and presented a positive relationship with NPF probability, until reaching a peak point. Further increase in NH_3 concentrations presented a decline with NPF probability (Figure 24a), which might be due to its association with increased pollution levels. Interesting though is that it presented a clear positive relationship with both the growth rate (though it also appears to decline at high concentrations) and the formation rate.

Figure 24: Relationship of gaseous ammonia concentrations with NPF variables.



(a)



(b)

Condensation sink (CS)

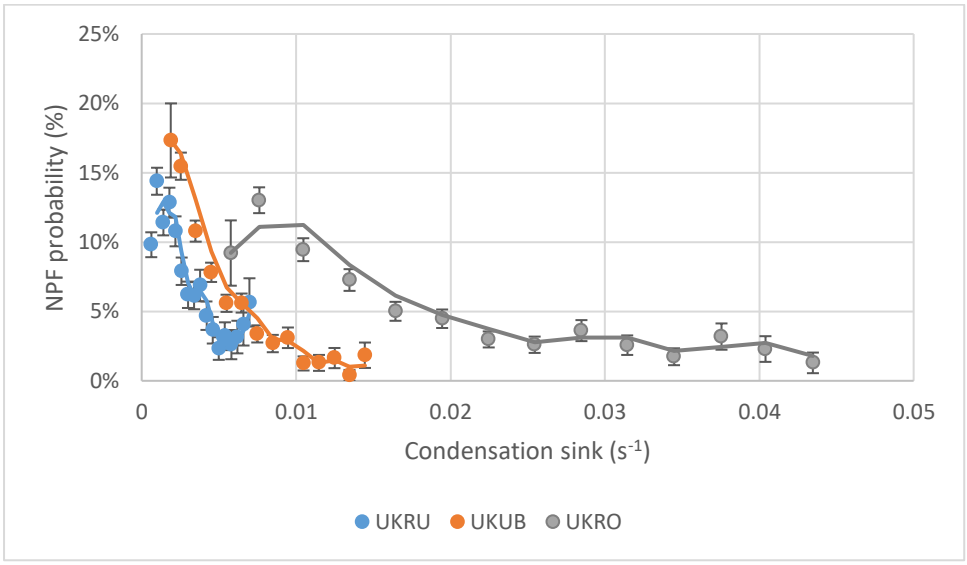
The CS is a measure of the rate at which molecules will condense onto pre-existing aerosols (Lehtinen et al., 2003). It is highly dependent on the number and size of the particles in the atmosphere and as a result it is expected to be affected by both the local emissions within the urban environment as well as the formation and growth of the particles due to NPF events. As a result, for the specific metric a time frame before the events are in full

development was chosen (05:00 to 10:00 LT) to avoid including the effect of the NPF events and provide a picture of the atmospheric conditions that preceded the NPF events. With this data, the NPF probability presented very strong relationships with the condensation sink. Two groups of sites were found though; those which had a positive relationship and those with a negative relationship. In the first group are the sites in Germany and Greece while all others had a negative relationship. This grouping follows the trend between the countries, the sites of which presented a greater (the ones with the positive gradients) or smaller CS on NPF event days, though it is unknown what causes this behaviour (at the German sites and GREUB it may be associated with the very high formation rates on NPF event days). While the gradients from this analysis cannot be used for direct comparisons, a trend was found for which the gradients were more positive or negative at the rural sites compared to their respective roadsides, which might indicate the greater importance of the variability of the CS at the rural sites in the occurrence of NPF events.

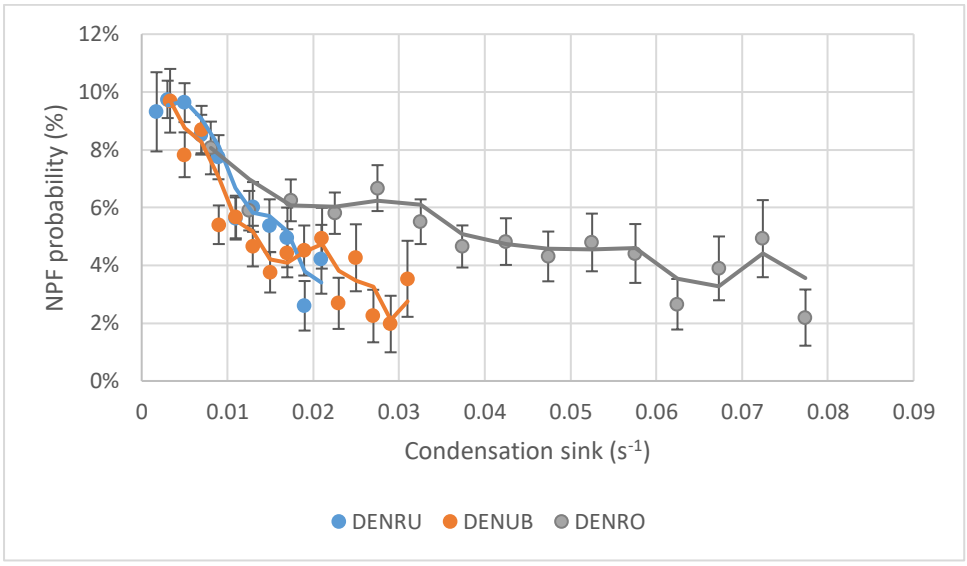
The growth rate was positively correlated with the CS for most of the sites, with strong relationships (high R^2) for about half of them, when linear relationships were considered. As the CS is a metric of pre-existing particles, it is also associated with the level of pollution in a given area. The increased significance and gradient found at the rural sites probably indicates the importance of enhanced presence of condensable compounds in a cleaner environment, which in many cases are associated with the moderate presence of pollution. The formation rate was also found to have a positive relationship with the CS. This relationship was more significant at the roadsides of this study, a result which to some extent is biased by the presence of increased traffic emissions found in the timeframe

chosen. While to an extent, increased presence of condensable compounds can be favourable for greater formation rates, this result should be considered with great caution.

Figure 25: Relationship of the condensation sink with NPF variables.

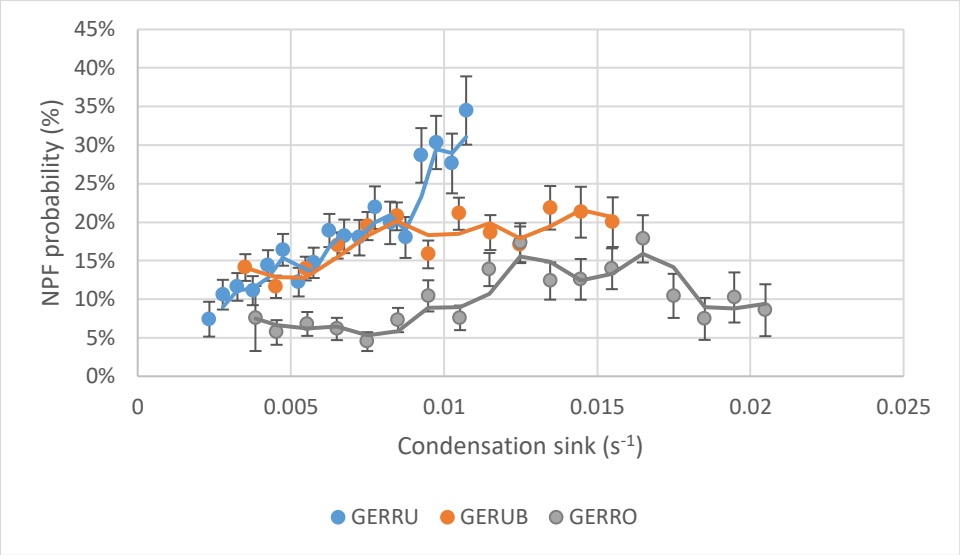


(a)

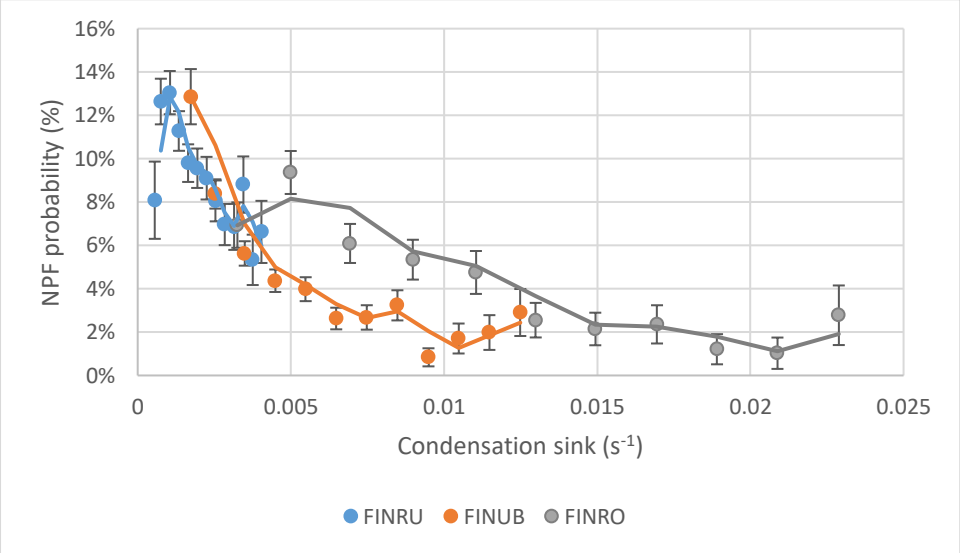


(b)

Figure 25 (continued)



(c)



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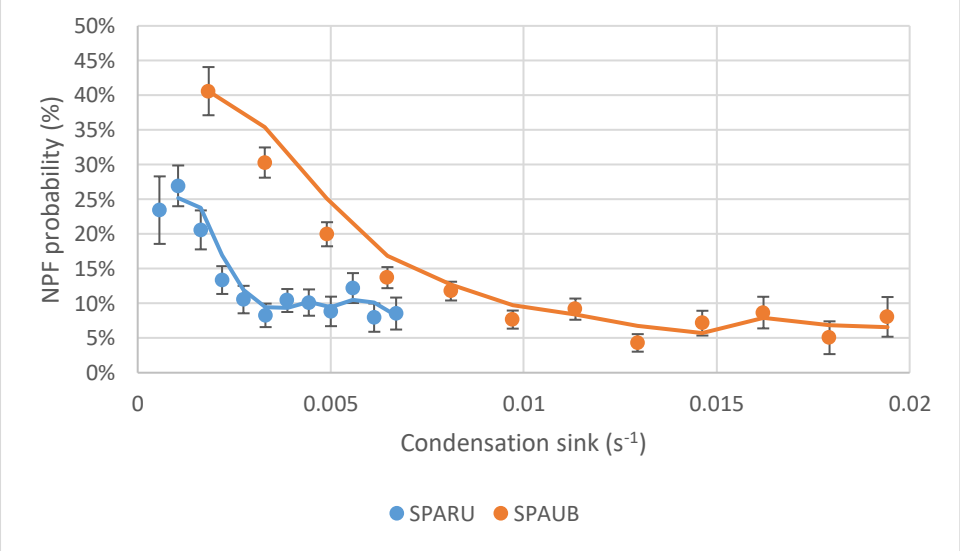
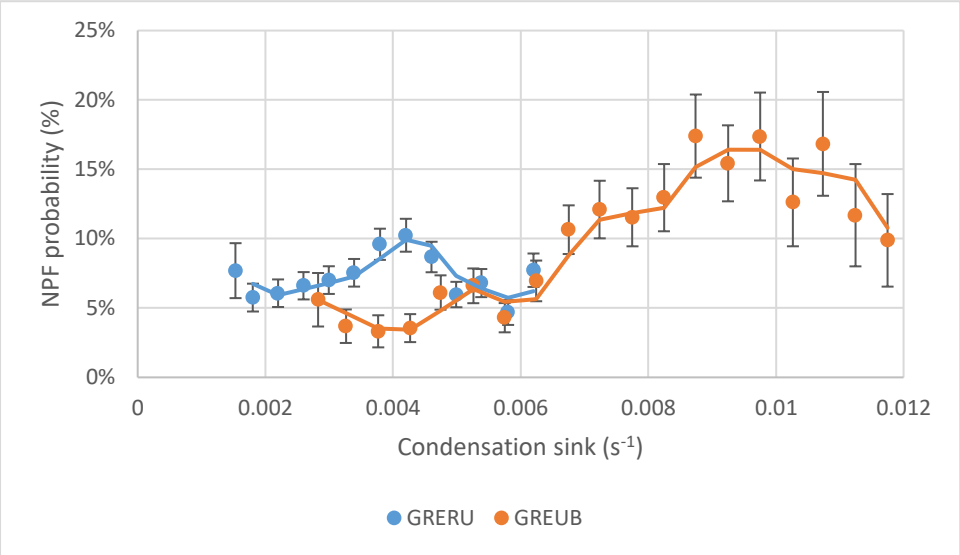


(e)

Figure 25 (continued)



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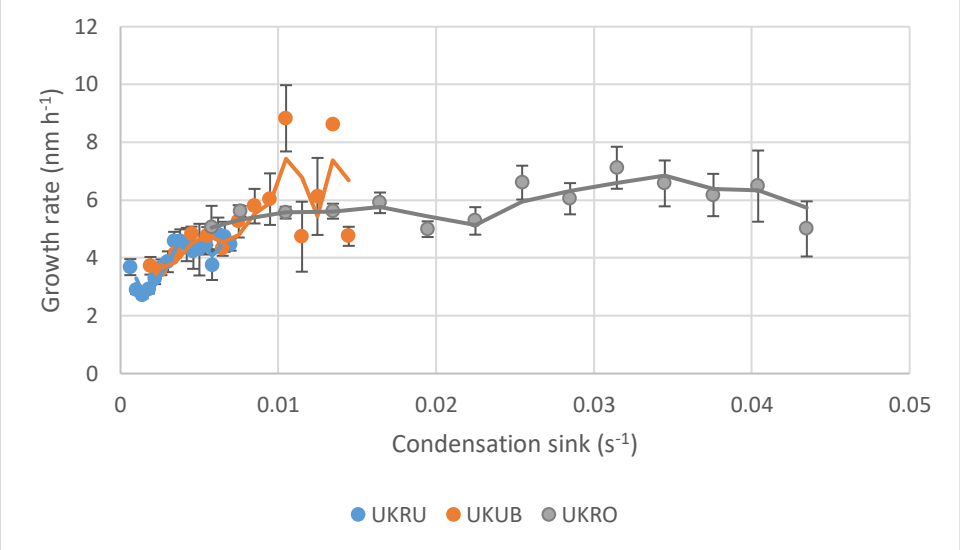

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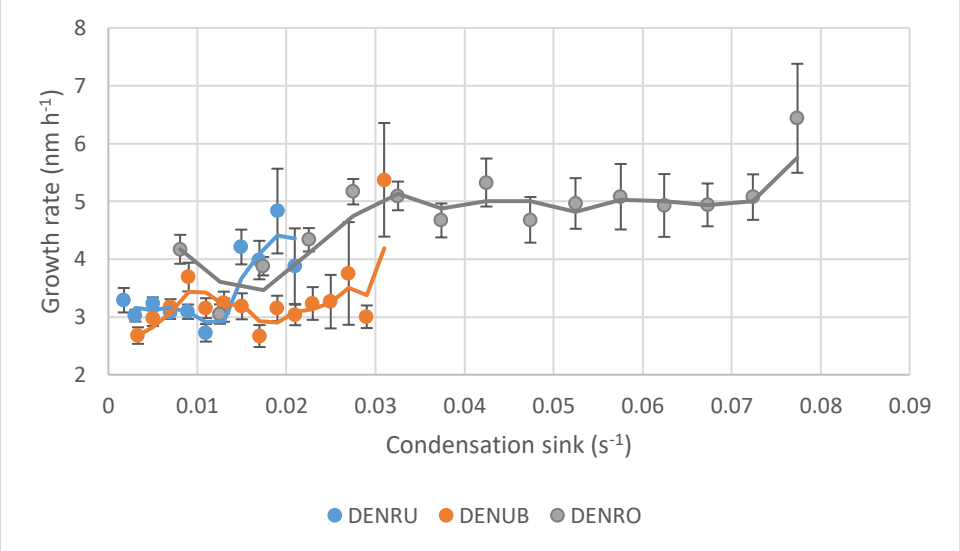
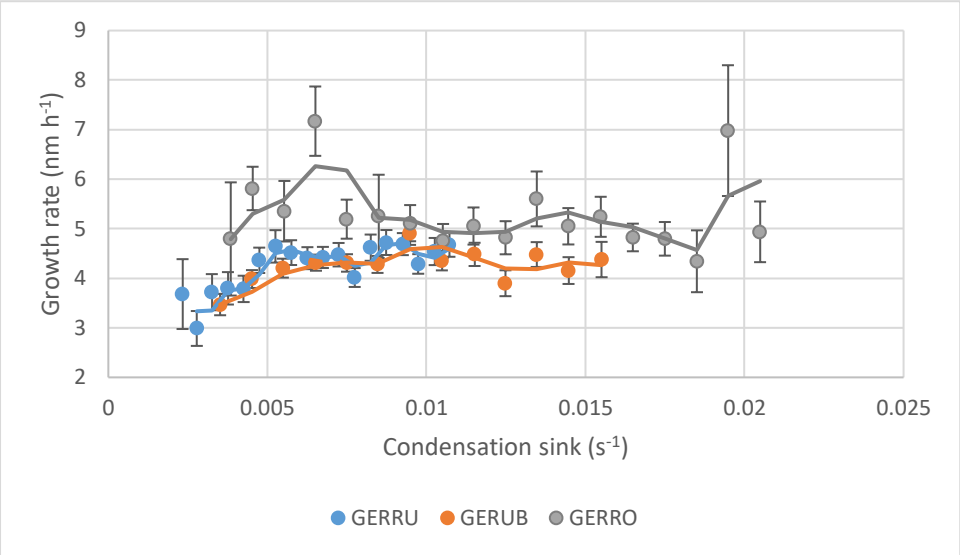
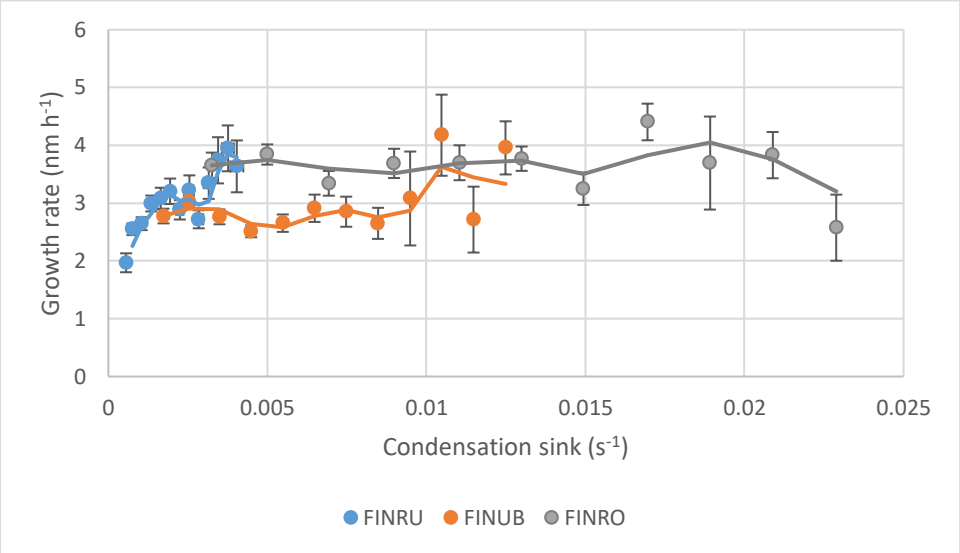


(h)

Figure 25 (continued)



(i)



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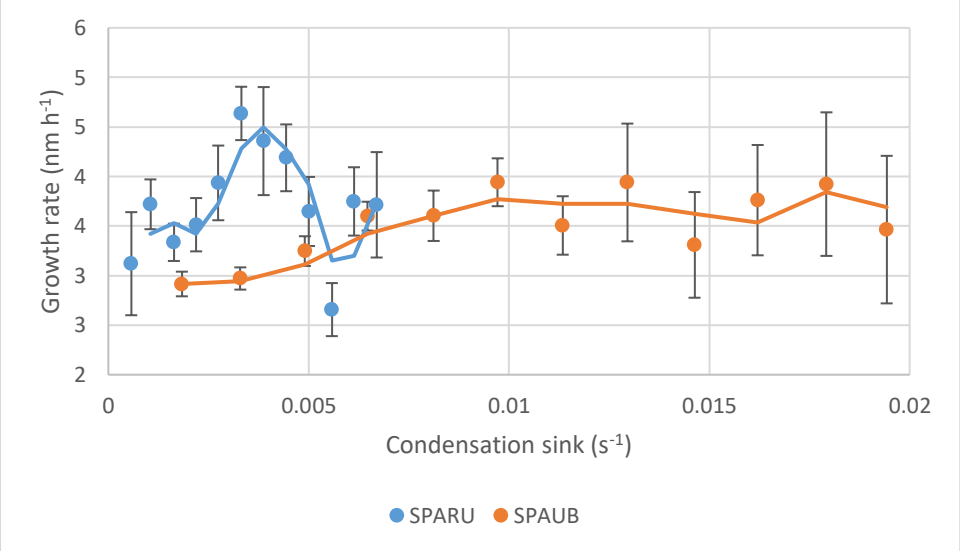
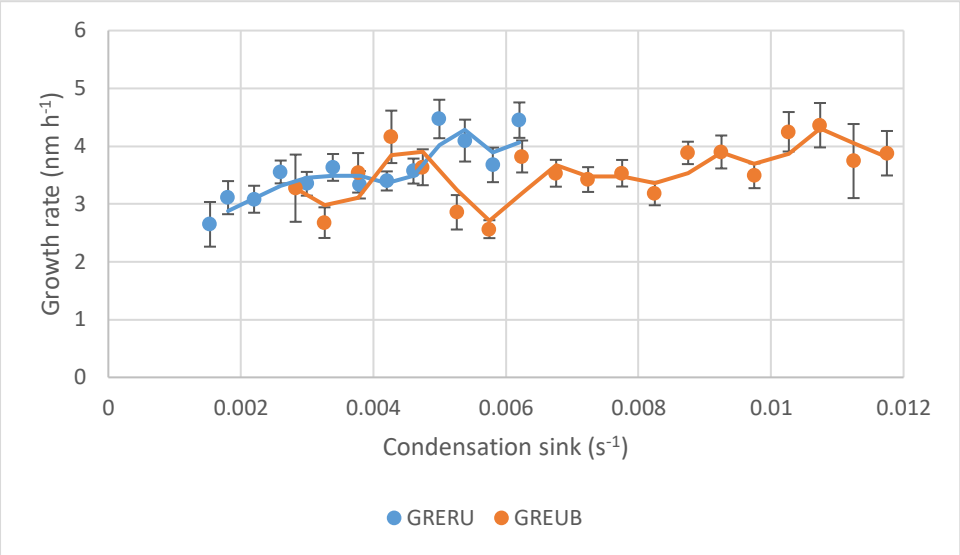


(k)

Figure 25 (continued)



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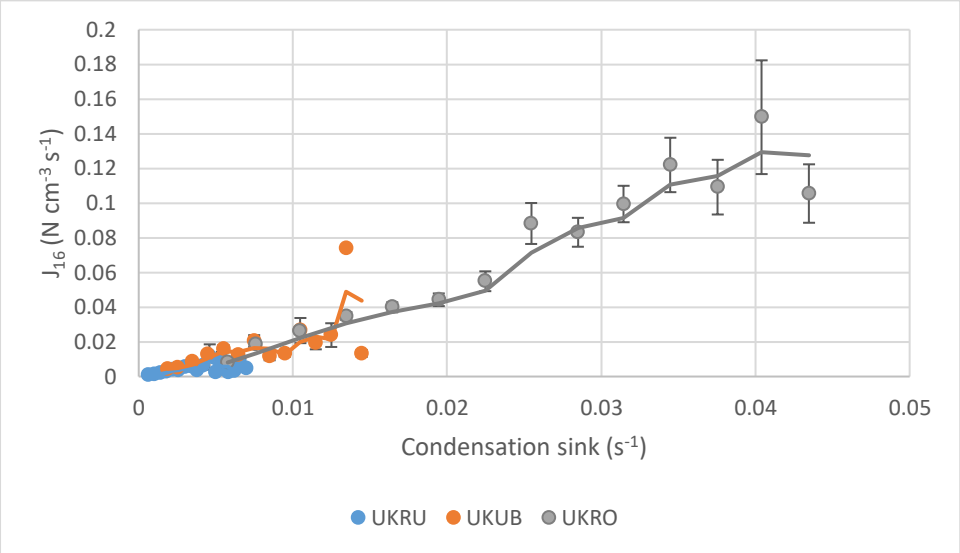

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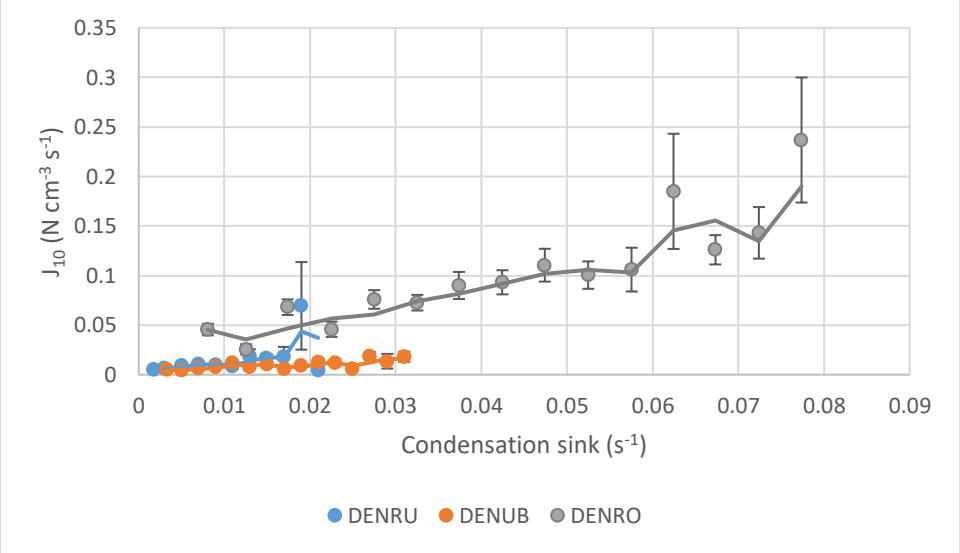
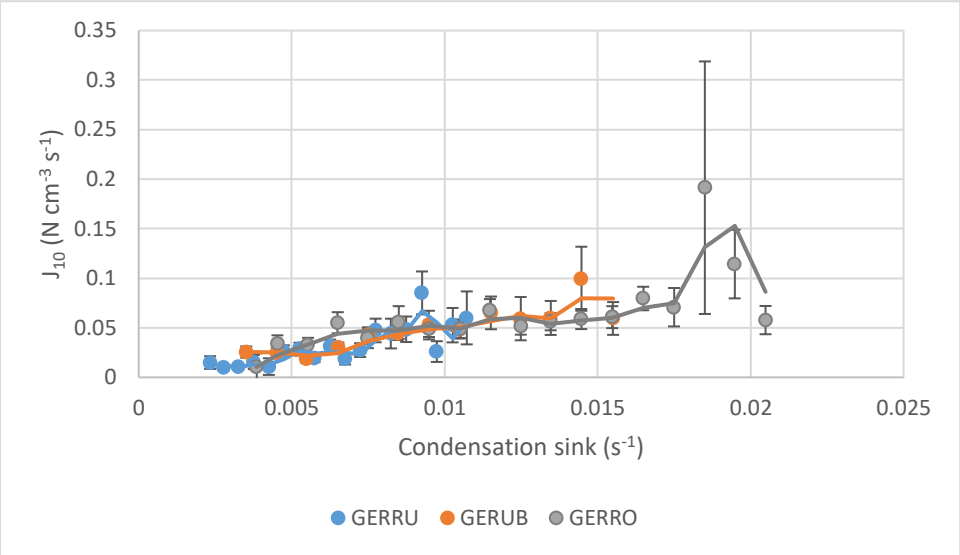


(n)

Figure 25 (continued)



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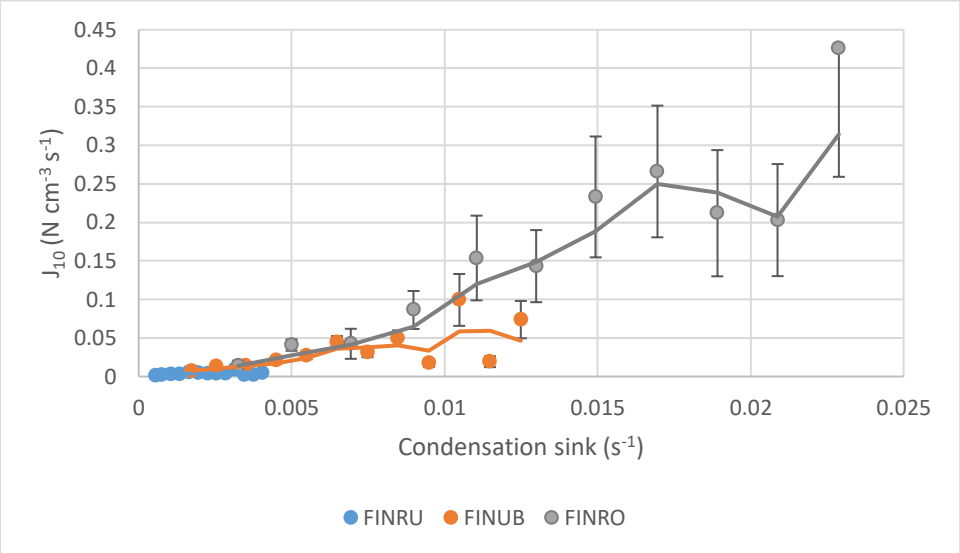

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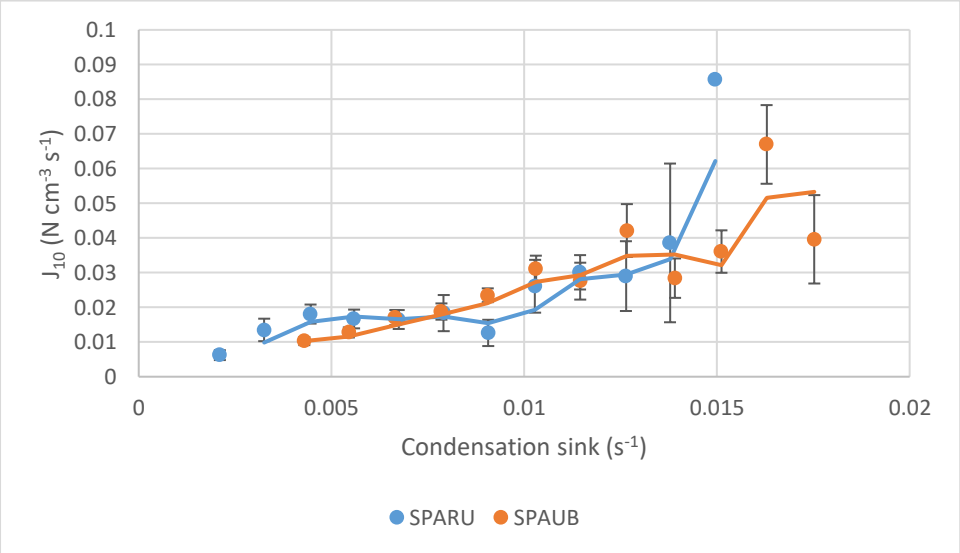
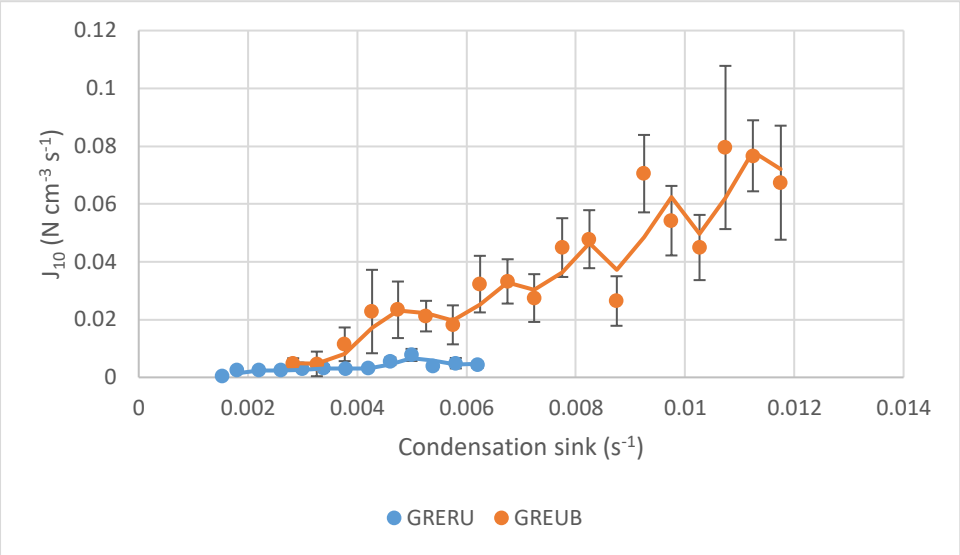


(q)

Figure 25 (continued)



(r)

3.3.3.3 Association of the Effect of the Variables

The Pearson correlation coefficients for the variables studied on each site are found in Table S3. The relatively strong correlation between the solar radiation, temperature and O_3 found, as well as their anticorrelation with the RH may lead to the conclusion that not all these factors play a role in NPF events, but their visible effect is the result of their relationship with each other. There is a similar case with the association of the CS and NO_x (or NO_2), and OC, as well as SO_2, especially at urban sites. However, the factors affect different outcomes differently, as for example the solar radiation intensity does not seem to be as important a factor for the growth rate as temperature, or O_3 does not seem to be strongly associated with either the formation or the growth rate. This is further established by the fact that some of these variables do not correlate well at the southern sites, but still appear to be associated with either the probability of NPF events or the growth or nucleation rate. The effects of all of these factors have been demonstrated in both laboratory and atmospheric studies in the past and were discussed earlier in this paper. By the analysis provided in the

present study, the effect of each of these variables is further established, providing an association of each one of these variables with either the formation or the growth mechanism. However, RH does not seem to be a consistent factor in any mechanism, and it appears that its effect is dependent on location specific conditions, although it was the variable with the most consistent association with NPF event probability at almost all sites.

3.3.3.4 Relationship to a previous multi-station European study

The findings of our study in respect of the background sites show many similarities with the conclusions drawn in the previous multi-station study in Europe by Dall'Osto et al. (2018) despite the two studies using several different sampling stations as well as some in common. Both studies point towards the influence of variables such as solar radiation and CS upon the occurrence of NPF events. The previous study suggested that different compounds participate in the growth of the particles, depending on the area considered. Thus, for northern and southern sites the growth of the particles is suggested to be driven mainly by organic compounds, while for the sites in central Europe sulphate plays a more important role. These findings are confirmed by the present study, as the growth rate was found to correlate better with organic compounds for the rural sites in Finland and Greece, while SO_4^{2-} presented a stronger relationship with the growth rate for the Danish and German sites (the latter presented high gradient values but low R^2 due to a decline at higher SO_4^{2-} concentrations – figure 23i, probably associated with NPF events being suppressed by increased pollution). The growth of the particles at the rural background site in the UK, characterised as "Overlap" in the previous study, was found to be strongly associated with both organic compounds and sulphate, consistent with it being in the central group.

The seasonality of NPF events at northern sites was hard to explain in the previous study, and the possible effect of low temperature was considered. In the present study, the Finnish background sites presented a double-peak relationship of NPF probability with temperature, with one of the peaks being below zero degrees. This might point to the possibility of different compounds driving the events for different temperature ranges, as well as the increased nucleation rate of H_2SO_4 at lower temperatures (Kirkby et al., 2011; Yan et al., 2018), which makes the occurrence of NPF events more probable at lower temperatures in a region with low SO_2 concentrations.

3.3.4 CONCLUSIONS

More than 85 site-years of data from 16 sites from six countries in Europe were analysed for NPF events. A total of 1950 NPF events with consequent growth of the newly formed particles were extracted and with the use of binned linear regression, the relationship between three variables associated with NPF events (NPF event probability, formation and growth rate) with meteorological conditions and atmospheric composition was studied. A summary of the results is presented in Table 6. Among the meteorological conditions, solar radiation, temperature and atmospheric pressure presented a positive relationship with NPF event occurrence, and either promoting the formation or growth rate. Relative humidity presented a negative relationship with NPF event probability which in most cases was associated with it being a limiting factor on particle formation at higher values. Wind speed on the other hand presented variable results, appearing to depend on the location of the sites rather than their type. This shows that while wind speed can be a factor in NPF event occurrence, the origin of the incoming air masses also plays a very important role.

The results for the levels of atmospheric pollutants presented a more interesting picture as most of these, which appear to be either directly or indirectly associated with the NPF process were found to have negative relationships with NPF probability. This is probably due to the fact that increased concentrations of such compounds are associated with more polluted conditions, which are a limiting factor in the occurrence of NPF events, as was found with the negative relationship between the CS and NPF probability in most cases. Thus, SO_2, NO_x (or NO_2), particulate OC and SO_4^{2-} concentrations were negatively correlated with NPF probability in most cases. On the other hand though, these compounds in many cases had a positive relationship (not always though with high significance) with the other variables considered. Thus, particulate OC (and VOCs where data was available) and SO_4^{2-} consistently had a positive relationship with the growth rate, while SO_2 was positively correlated with both the formation and growth rate in most cases. Finally, O_3 was positively correlated with NPF event probability at all sites in this study, though it presented variable results with the other two variables.

The present study attempts to explain the effect of several meteorological and atmospheric variables on the occurrence and development of NPF events, by using a large-scale dataset. It should be noted that the variables considered are in many cases inter-related (e.g. temperature and RH) and this complicates considerably the interpretation in terms of causal factors. Large datasets are very useful in providing more uniform results by removing the possible bias of short period extremities, which may lead to wrong assumptions. Following from this, the importance of a high-resolution measurement network, both site and timewise is underlined, as it can help in elucidating the mechanisms of new particle formation in the real atmosphere.

Table 6: Effect of the variables as found in the study in Chapter 3.3. Upright arrow is a positive effect on the given NPF variable; downright arrow is a negative effect on the given NPF variable. Blue arrow is a relationship between the variable and the NPF variable with $p < 0.05$; red arrow is a relationship between the variable and the NPF variable with $p > 0.05$ (SR refers to solar radiation. WS refers to wind speed).

*The formation rate for the UK sites was calculated for particles of 16nm of diameter

NPF%	SR	RH	T	WS	P	SO_2	NO_x	O_3	OC	SO_4^{2-}	CS
UKRU	↗	↘	↗	↗	↗	↘	↘	↗	↘	↘	↘
UKUB	↗	↘	↗	↗	↗	↘	↘	↗	↘	↘	↘
UKRO	↗	↘	↗	↗	↗	↘	↘	↗	↘	↘	↘
DENRU	↗	↘	↗	↗		↘	↘	↗	↘	↘	↘
DENUB	↗	↘	↗	↗			↘	↗			↘
DENRO	↗	↘	↗	↗		↘	↘	↗	↘	↘	↘
GERRU	↗	↘	↗	↘	↗				↘	↘	↗
GERUB	↗	↘	↗	↘	↗						↗
GERRO	↗	↘	↗	↘	↗						↗
FINRU	↗	↘	↘	↗	↗	↘	↘	↗	↗	↘	↘
FINUB	↗	↘	↘	↘	↗						↘
FINRO	↗	↘	↗	↗	↗		↘	↗			↘
SPARU	↗	↘	↘	↘	↘	↘	↘	↗			↘
SPAUB	↗	↘	↘	↗	↘	↘	↘	↗			↘
GRERU	↗	↘	↗	↗	↗		↗	↗			↗
GREUB	↗	↘	↘	↘	↗				↘		↗

Table 6 (continued)

GR	SR	RH	T	WS	P	SO_2	NO_x	O_3	OC	SO_4^{2-}	CS
UKRU	↗	↗	↗	↘	↗	↘	↗	↘	↗	↗	↗
UKUB	↘	↗	↗	↘	↗	↗	↘	↘	↗	↗	↗
UKRO	↘	↗	↗	↘	↘	↗	↗	↘	↗	↗	↗
DENRU	↗	↗	↗	↘		↗	↘	↘	↗	↗	↗
DENUB	↗	↗	↗	↘			↗	↘			↗
DENRO	↗	↘	↗	↘		↗	↗	↗	↗	↗	↗
GERRU	↗	↘	↗	↘	↗				↗	↗	↗
GERUB	↗	↘	↗	↘	↗						↗
GERRO	↘	↗	↘	↘	↘						↘
FINRU	↗	↘	↗	↘	↗	↘	↘	↘	↗	↗	↗
FINUB	↗	↗	↗	↗	↘						↗
FINRO	↗	↘	↗	↘	↘		↗	↗			↘
SPARU	↗	↘	↗	↗	↗	↘	↗	↗			↗
SPAUB	↗	↗	↗	↗	↗	↗	↗	↘			↗
GRERU	↗	↗	↗	↗	↘		↘	↗			↗
GREUB	↘	↗	↗	↘	↘				↗		↗

Table 6 (continued)

J_{10}	SR	RH	T	WS	P	SO_2	NO_x	O_3	OC	SO_4^{2-}	CS
UKRU*	↗	↘	↗	↗	↗	↗	↘	↘	↗	↗	↗
UKUB*	↗	↘	↗	↗	↗	↗	↘	↘	↗	↗	↗
UKRO*	↗	↘	↗	↗	↘	↘	↗	↘	↗	↘	↗
DENRU	↗	↗	↗	↗		↗	↘	↗	↘	↘	↗
DENUB	↗	↗	↗	↗			↘	↗			↗
DENRO	↗	↘	↗	↗		↗	↗	↗	↗	↗	↗
GERRU	↗	↘	↗	↘	↗				↗	↘	↗
GERUB	↗	↘	↗	↘	↗						↗
GERRO	↗	↘	↗	↘	↘						↗
FINRU	↗	↘	↗	↗	↗	↘	↗	↗	↘	↘	↗
FINUB	↗	↘	↗	↘	↗						↗
FINRO	↗	↗	↘	↘	↗		↗	↘			↗
SPARU	↗	↘	↗	↗	↗	↘	↗	↗			↗
SPAUB	↗	↘	↗	↘	↗	↗	↗	↘			↗
GRERU	↘	↘	↘	↗	↗		↗	↘			↗
GREUB	↗	↗	↘	↘	↗				↗		↗

4. Conclusions

4.1 Conditions of NPF events. Similar but not the same

Many studies have been done to understand the conditions that promote or suppress the events. As found by many other previous studies, increased solar radiation and low relative humidity are the factors that seem to be common at all the areas studied. This result was confirmed for all the sites studied, regardless of the type or geographical location. Additionally, the atmospheric pressure was also found to be higher on event days compared to the average, though it is uncertain whether it is a factor in the occurrence of the events or its variability is the result of its association with weather conditions that promote the events in general. Cloud coverage was only studied for the sites in the UK and was found to be lower on event days compared to the average, though this might be due to its effect on the solar radiation. Greater variability was found for other meteorological conditions, such as the temperature or the wind speed, which have different trends across the countries studied. Interestingly, regional events occurred in more distinct conditions but in most cases following the general trends in each country (when temperature was lower on NPF events compared to the average, it was found to be further lower on regional events).

Regarding the atmospheric composition the conditions were more uniform. Most of the chemical compounds that have any association with NPF (either by promoting or suppressing them) were found to have lower concentrations on event days compared to the average. This holds true even for compounds that are considered to participate in the NPF procedure, either directly or indirectly such as the SO_2, ammonia or organic compounds, which probably points that their concentrations are sufficient for the occurrence of the events and higher concentrations are associated with increased pollution. The only exception was ozone, which

had higher concentrations on event days, which is probably due to it participating in the oxidation of compounds related to the NPF procedure (as well as other compounds, such as hydroxyl radical, since ozone is an indicator of atmospheric photochemistry). Specifically for the U.K., the highest ozone concentrations are associated with southwesterly air masses, which are relatively clean and have low condensation sinks. The variability found for regional events, for which some compounds had higher concentrations probably point to the greater importance of clear meteorological conditions in their occurrence. Finally, the condensation sink, which is a crucial factor in the occurrence of the events was lower at most areas with very few exceptions, which in the case of the German sites might be the result of the very high formation rates found there.

4.2 The role of incoming air masses. Promoting the events in different ways.

The characteristics of the incoming air masses play a very important role in the occurrence of NPF events, as was found in **Papers 1 and 2**. Incoming air masses, depending on their origin as well as their route, can have variable chemical and particle compositions as well as being associated with meteorological conditions which can either suppress or promote NPF events and their development. It should also be noted that the source of incoming air masses may also vary due to the diurnal or seasonal variation in a given area also affecting the events.

While, the origin of the air masses was studied in a different way in the UK compared to the other sites, the results appear to have the same trends. The frequency of the events, both regional and local, tends to be higher with cleaner incoming air masses, while the growth rate of the particles tends to be higher when the air masses originate from more polluted areas.

Air masses that have passed over more polluted areas usually have higher concentrations of pollutants as well as condensation sink. While these pollutants may have a negative effect on the occurrence of the events, if those occur the newly formed particles tend to grow faster due to the greater abundance of condensable compounds that usually comes with greater pollution levels. On the other hand, the opposite conditions are probably the reason for the higher frequency of NPF events and lower growth rates observed with cleaner air masses. Less distinct but similar appears to be the trend for the formation rate J_{10}. This trend though was found to be different for the German sites as well as the very clean Greek rural background, for which both the occurrence of NPF events as well as the growth rate of the particles were enhanced for incoming air masses from the sectors with higher condensation sinks. This can also be due to the increased concentrations of organic carbon also found from that side, originating from the polluted areas found there.

Apart from the level of pollution, meteorological characteristics of the air masses from different origins can also affect NPF events. These followed the general trends of NPF events, with air masses from areas with higher relative humidity having lower frequency of NPF events. The temperature variation also appeared to play a role, with cooler air masses from the north, in both UK and Finland presenting higher frequency of NPF events, while warmer ones having higher growth rates. The latter though can be the result of the seasonal variability of the wind sector frequency distribution in these areas.

Apart from characteristics due to long range transport, features gained from local transport also affect the characteristics of NPF events. At both roadsides in the UK and Germany, the origin of the incoming air masses, when coincided with the street canyon formed, presented lower frequency of NPF events though with different patterns of growth rate of the particles.

4.3 Variation of events with location. Not every house is the same in Europe.

The variability of conditions found throughout Europe resulted in a great variation in the characteristics of the events. While the growth rate peaked in most areas in the summer, probably associated with the increased biogenic activity during that season, the frequency of events as well as the formation rate varied more across Europe, with different areas presenting different seasonality. The reasons for the differences in the seasonality found varied from the chemical composition and meteorological conditions in each geographical region, to very local conditions that usually suppressed the events. These local conditions were either local sources of pollution or specific synoptic conditions, usually affecting the particle composition in the given area by increasing the condensation sink or altering the chemical composition of the given atmosphere. While, as discussed in section 4.1, NPF events occurred on days with higher solar radiation and lower relative humidity, the sites in southern Europe where these conditions are more common did not present greater frequency of events or seasonality that followed these trends.

The effect of the type of the environment was thoroughly discussed on **Papers 1 and 2**. It was pointed out that for all the countries with more than one site with available data, the growth and formation rate was almost always greater moving from the rural background site to the roadside, while the frequency of events had the opposite trend. The former is explained either by the greater presence of condensable compounds in urban environments which enhances the nucleation procedure, or it can be a prerequisite for NPF events to occur in more polluted areas, as particles should be formed and grow in higher rates to overcome the increased

condensation sink found in such areas. The latter is probably explained by the cleaner conditions found at the rural sites, which are more favourable for the occurrence of NPF events. Specifically for the UK, in **Paper 1** using the Urban Increment (U.I), a metric proposed to calculate the effect of the urban environment on the concentrations of particles between 16 and 20 nm during NPF events, it was found that the urban environment enhanced the formation and survival of particles in this range, increasing the particle concentration in that size range about 20% more compared to the rural background site, located about 80 km away (the meteorological conditions can be considered as similar and thus are not a factor).

What can be pointed out as a bottom line is that, while there is a general trend in the conditions that affect the events, local conditions have a decisive effect on the frequency and characteristics of the events, which is the factor that makes the understanding of the effect of each variable clear.

4.4 The variability of the effect of conditions. Same variables, different results.

Many studies, both in laboratories and in real atmospheric conditions were done to elucidate the effect of meteorological conditions and chemical compounds on the development of NPF events. In many cases though the results between them were not in agreement. In **Chapter 3.3** not only the behaviour of many conditions and compounds was studied on their effect in the occurrence of the events, but the effect and its extent in the metrics of NPF events (formation and growth rates) was studied across different types of environments and geographical areas. As mentioned earlier, NPF events are the formation and growth of new particles in the atmosphere; these two procedures appear to have different mechanics and

compounds that participate on them. Table 6 summarises the results of the study in **Chapter 3.3.** Solar radiation, relative humidity and temperature appear to be important factors in the occurrence of NPF events, though affecting different stages of the process, either the formation or the growth of the particles. Other meteorological factors, such as the wind speed and pressure appeared to have different effects at different European sites and probably are either not a factor in the development of the events or their apparent effect is the result of their association with other factors which have a more distinct effect. In general, meteorological conditions appeared to be a more important factor for the events at the rural sites compared to the urban ones.

The chemical compounds studied were found to be a more important factor within the urban environment. Sulphur dioxide which indirectly participates in both the formation and growth of the particles was found to negatively correlate with the occurrence of the events. Interestingly though, an intercomparison between the sites (mainly the background ones) of the study showed that while it has a negative effect in the occurrence of events within the sites, those with higher concentrations presented higher probability of NPF events, having a positive relationship with the growth rate of the particles. Similar results were found for other chemical compounds such as the organic carbon and sulphates, which might associate the greater abundance of these compounds with an increased presence of condensable species that participate in the growth procedure. The result for ozone presented a positive relationship with the probability of the events, especially with the sites in northern Europe, that seems to be associated with the formation of the particles. The condensation sink, as well as NO_x/NO_2 presented a negative relationship with the events in most cases, as increased values of these are associated with increased pollution levels. Interestingly, the variation found for some of these variables around the sites of the study showed that, while some of

them have a clear effect on the events, when in great abundance they may become less important as factors.

4.5 The effect of NPF events on the ultrafine particle number concentrations.

New particle formation events are a significant source of ultrafine particles in all types of environments. To quantify this effect the Nucleation Strength Factors were used. The importance of NPF was found to be variable not only for the different types of areas, but also between similar sites in different regions, depending on both the frequency as well as the survivability of the newly formed particles.

The greatest relative increase in the ultrafine particle concentrations was found at the rural background sites. This type of sites usually has NPF events as their main source of smaller particles (especially for particles < 30 nm) and as a result the increase found on the ultrafine particles is mainly due to them. The increase found ranged from a bit less than 100% in Germany, up to more than 4 times in the Finnish rural background. It should be noted that, as the events extracted were only the stronger and clearer ones, this effect is probably greater if we consider the weaker or less clear ones as well. This makes NPF events a very important factor for the physical properties in these types of environment, which can also affect their surrounding areas.

A significant increase was also found at the urban background sites of the study. While not reaching the level of the rural background sites, a significant increase in the number concentration of ultrafine particles on event days was found, reaching up to more than 200% in average at the Finnish site. Since this type of site consists of residential areas, NPF events

doubling the number of ultrafine particles are a factor that should be considered for public health studies and regulations. Also, considering the increased particle concentrations in the urban environments, such an increase can be even greater in absolute values compared to the rural sites, as found in **Paper 1**.

Finally, at the roadside sites the relative effect of NPF events was found to be a lot smaller, varying between no difference in ultrafine particle concentrations on event days up to an increase of 20%. Specifically for the roadside sites, there are many factors that should be considered when trying to explain the result with the metric used. Initially, the result found shows the limited effect of NPF events in an environment where there are other strong sources of ultrafine particles. As traffic is a major contributor of ultrafine particles in these sites the effect of NPF events appears to be smaller. It should be noted though that due to the conditions at the roadsides, it was found that the condensation sink as well as the number of particles of all sizes was greatly reduced when the events took place, which probably is a prerequisite for them to occur in such areas. This means that an event day on such environments is very different compared to the average conditions and a direct comparison between them will provide with untrustworthy results. Furthermore, as was proven by recent studies, traffic can be a source of particles even smaller than 3 nm. This makes it harder to distinguish whether the particles observed are from secondary formation or direct emissions.

5. Final thoughts and future challenges

The present study explored the conditions of NPF events in a number of different sites in Europe, as well as tried to quantify the exact effect of the variables associated with the events. While it managed to present a more in-depth picture of the events around Europe, many aspects of the events and the mechanisms that drive them are yet to be elucidated. New particle formation is a complex mechanism, which depends on the combination of a wide range of different variables. As it is found to occur everywhere, both horizontally and vertically in the atmosphere, the mix of conditions and compounds needed varies as much as all the different environments and conditions that exist. In general, there are factors with a constant behaviour everywhere and others that vary depending on the mix. And even the factors that seem to have a uniform behaviour also vary in the intensity of their effect. This study, apart from pointing on this variability, further confirmed the general consent: formation and growth of particles are two separate processes, governed by specific general rules but vary according to the intensity of the factors that drive them and the environment in which they occur. While in controlled laboratory studies specific conditions can be tested, the atmospheric "laboratory" presents such a variability that it is almost impossible, with our present knowledge and measuring ability to know the outcome of the mix of the conditions found in each case.

Continuing from the work started with this study, while the effect of individual variables was studied, the combined study of these variables will provide information not only of those that may not have an effect but present a relation due to their association with other factors, but also explore their effect when combined, thus finding the real mixes of conditions that lead to

the events, as well as quantifying the characteristics of the events depending on the initial conditions.

Additionally, though studying the conditions that lead to NPF events at all kinds of environments is important, it is crucial to understand the mechanisms of NPF events on a molecular level. Studies that focus on the chemical compounds that lead to the formation of new particles while already conducted, should continue, to provide insight on the role of each one of the compounds participating. Fully understanding the initial formation and growth of particles down to the very start of the procedure will help in fully elucidating the role and behaviour of the chemical compounds that participate in it and thus explain the variation found in real atmospheric conditions by understanding how NPF events are either promoted or suppressed. In this direction, the CLOUD experiment at the Conseil Europèen pour la Recherche Nuclèaire (CERN) in Switzerland, for almost a decade aims to understand the mechanics of both aerosol nucleation and subsequent particle growth with the use of advanced instrumentation in an environment with extremely low concentrations of impurities (Lee et al., 2019).

Not many years ago, NPF events were considered as unlikely to occur within the more polluted urban environments. As a result, the study of the secondary formation of particles in urban areas was limited. This is a new field that needs to be focused on the future as the extent of the effect of secondary formation is probably greater than anticipated. Due to the more complex chemical composition found in the urban atmosphere, many different compounds that result from the activities within urban areas can participate in the NPF procedure and many pathways, such as those found at the CLOUD experiment involving the cluster – cluster collision growth mechanism (Lehtipalo et al., 2016) or growth by condensation of nitric acid and ammonia on fresh particles (Wang et al., 2020), should be explored. A study like this will

be useful not only to explain the mechanisms of nucleation in urban areas, but to also assess future regulations associated with pollution from ultrafine particles. Furthermore, the role of anthropogenic VOCs, which are in great abundance in urban areas should be studied. This will also help in defining the sources of the particles in the more polluted cities and help in exploring the possible health effects of the ultrafine particles in them.

Apart from the secondary formation occurring in the atmosphere, evidence exist that it also takes place indoors (Vu et al., 2019). Most people spend the majority of their time indoors, which makes the importance of air quality within buildings a very important factor. Along with other pollutants found, secondary formation may play an important role on the indoor air quality as there are the conditions for it to be a significant source of ultrafine particles.

Additionally, special cases of NPF event's development should be studied, such as night-time events or particle shrinkage. These mechanisms were found to occur in several areas where NPF events were studied and can be a factor in public health, as they can affect the particle composition with mechanisms that are not yet explained.

Finally, while ultrafine particles have not been regulated yet, many compounds that are associated with the secondary formation have been. Both regulations as well as technologic advancements aim in the improvement of air quality by reducing the concentrations of particulate matter and harmful chemical compounds. Among them the emissions of sulphur dioxide have been restricted during the last couple of decades and its concentrations in the atmosphere decline rapidly. As it is one of the major components associated with secondary particle formation, the effects of this change may have a great impact on many atmospheric variables. Similar is the case with other compounds which may be associated with anthropogenic activities and participate in the secondary formation process. The effect of these changes is hard to predict. For example, while the reduction of the concentrations of

sulphuric species may lead to a limitation of NPF, the associated reduction of other anthropogenic emissions will also reduce the condensation sink which will have a positive effect on NPF. Unknown are also the changes that may occur in the dominant mechanisms of NPF, as well as in the importance of the chemical compounds that participate in it. Depending on the range of these changes one can only speculate on the effect they will have in both the particle concentrations as well as their composition in different environments, favouring different size ranges and suppressing others.

To achieve the aforementioned, new methods and instruments that will help in accurately measuring both atmospheric particles and their chemical composition in the lower end of the particle size spectra should be developed. This is crucial for the understanding of the mechanisms that occur in the first stages of particle formation in atmospheric conditions. Along with the development of such instruments, effort should be made in making them as cost efficient as possible. The development of low-cost instruments, which has already started, should be further focused and funded, as these will not only help in understanding the mechanisms of NPF, but will also help in monitoring the air quality better through a denser measuring network.

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7. Appendix

7.1 Supplement of "Analysis of new particle formation (NPF) events at nearby rural, urban background and urban roadside sites"

Authors: Dimitrios Bousiotis, Manuel Dall'Osto, David C.S. Beddows, Francis D. Pope, Roy M. Harrison

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Atmospheric Chemistry and Physics
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EGU

Supplement of

Analysis of new particle formation (NPF) events at nearby rural, urban background and urban roadside sites

Dimitrios Bousiotis et al.

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Table S1: Data availability per season (all numbers are percentages of available data)

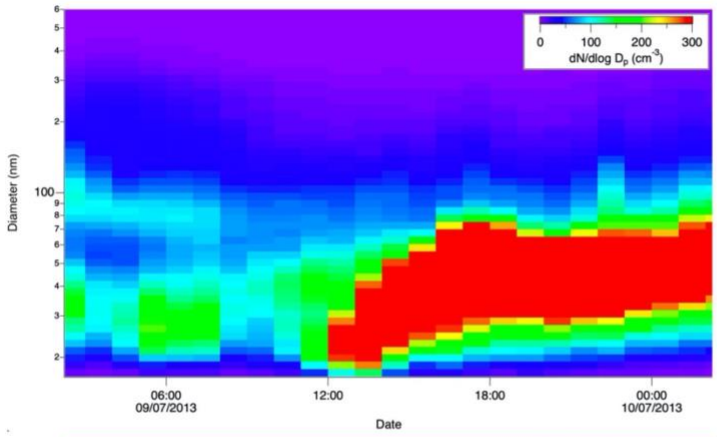
	Harwell				N. Kensington				Marylebone Road			
	Winter	Spring	Summer	Autumn	Winter	Spring	Summer	Autumn	Winter	Spring	Summer	Autumn
2009	15	97	10	80	57	97	100	100	100	65	86	68
2010	37	53	100	95	58	87	93	100	46	100	87	86
2011	72	75	99	73	89	87	73	89	79	99	100	67
2012	82	86	100	95	56	88	99	86	0	0	87	66
2013	91	70	99	100	84	92	98	98	57	92	84	100
2014	97	62	99	99	84	78	97	98	89	79	76	99
2015	77	100	61	70	80	99	65	100	74	100	98	100

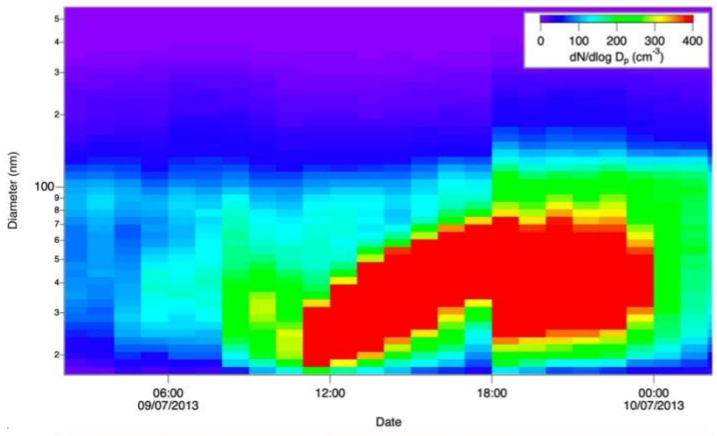
Table S2: Conditions per air mass origin for NPF event days (April – October average in parenthesis) for all areas of study.

Harwell				
	Continental	Arctic	Polar	Tropical
Condensation sink (s^{-1})	5.05E-03 (5E-03)	2.71E-03 (3.32E-03)	2.57E-03 (2.87E-03)	3.19E-03 (2.87E-03)
Wind speed (m s^{-1})	3.52 (3.63)	3.87 (3.47)	3.64 (3.69)	3.74 (4.17)
Temperature (°C)	15.5 (13.6)	12.2 (11.5)	13.6 (13.1)	16.3 (15)
SO_2 (μg m^{-3})	1.87 (1.81)	1.11 (1.82)	1.11 (1.27)	1.22 (1.36)
NO_x (μg m^{-3})	9.58 (13.9)	5.49 (8.01)	4.66 (7.2)	5.81 (7.69)
SO_4^{2-} (μg m^{-3})	2.70 (3.3)	1.37 (2.05)	1.44 (1.64)	1.37 (1.57)
Particulate OC (μg m^{-3})	2.85 (2.88)	1.35 (1.59)	1.52 (1.63)	1.98 (1.76)

North Kensington				
	Continental	Arctic	Polar	Tropical
Condensation sink (s^{-1})	7.20E-03 (9.35E-03)	5.20E-03 (6.37E-03)	5.40E-03 (6.38E-03)	4.89E-03 (6.32E-03)
Wind speed (m s^{-1})	3.89 (3.44)	3.92 (3.65)	4.46 (4.2)	4.74 (4.44)
Temperature (°C)	18.4 (15)	12.7 (13.1)	15.5 (14.6)	17 (16.4)
SO_2 (μg m^{-3})	1.68 (2.23)	1.33 (1.89)	1.73 (1.75)	1.74 (1.72)
NO_x (μg m^{-3})	33.5 (55)	28.5 (39.2)	30.3 (39.4)	24 (34.9)
SO_4^{2-} (μg m^{-3})	1.93 (2.23)	0.95 (1.36)	0.98 (1.13)	1.30 (1.47)
Particulate OC (μg m^{-3})	3.84 (4.90)	2.24 (2.95)	2.81 (2.96)	2.43 (3.03)

Marylebone				
	Continental	Arctic	Polar	Tropical
Condensation sink (s^{-1})	1.65E-02 (1.96E-02)	1.16E-02 (1.57E-02)	1.4E-02 (2.14E-02)	1.82E-02 (2.39E-02)
Wind speed (m s^{-1})	3.92 (3.41)	3.50 (3.64)	3.84 (4.13)	4.77 (4.4)
Temperature (°C)	17.4 (15.2)	13.4 (13.4)	15.3 (14.8)	16.9 (16.3)
SO_2 (μg m^{-3})	4.99 (6.39)	4.31 (5.63)	5.38 (7.43)	6.95 (8.17)
NO_x (μg m^{-3})	172 (250)	139 (214)	191 (303)	269 (336)
SO_4^{2-} (μg m^{-3})	3.24 (3.35)	1.47 (1.6)	1.52 (1.61)	1.24 (1.8)
Particulate OC (μg m^{-3})	6.03 (6.91)	3.81 (4.73)	4.67 (5.97)	5.31 (6.6)





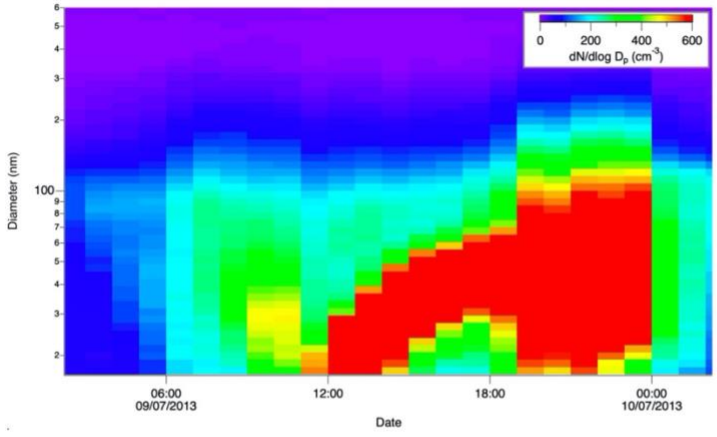
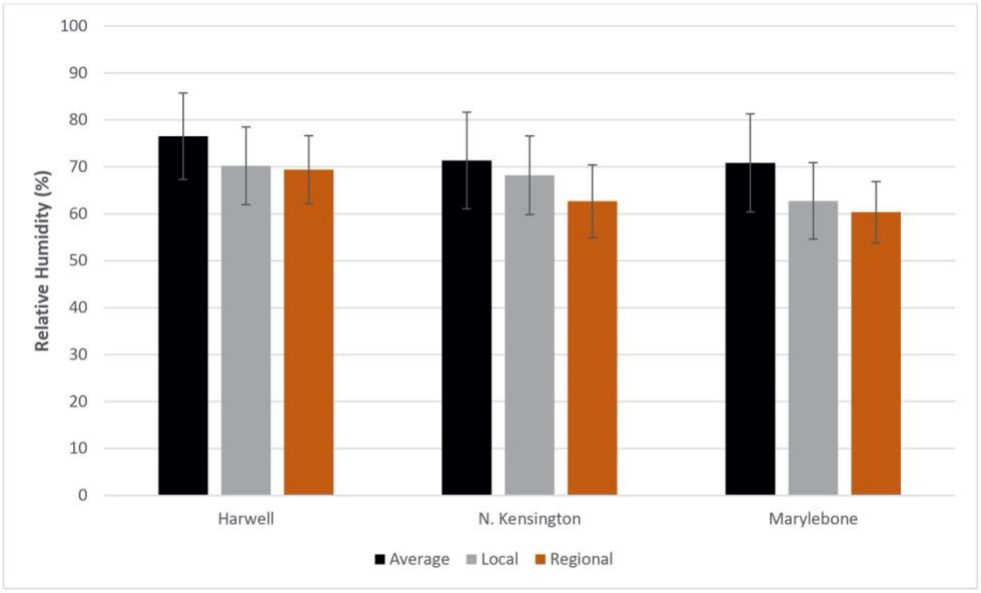
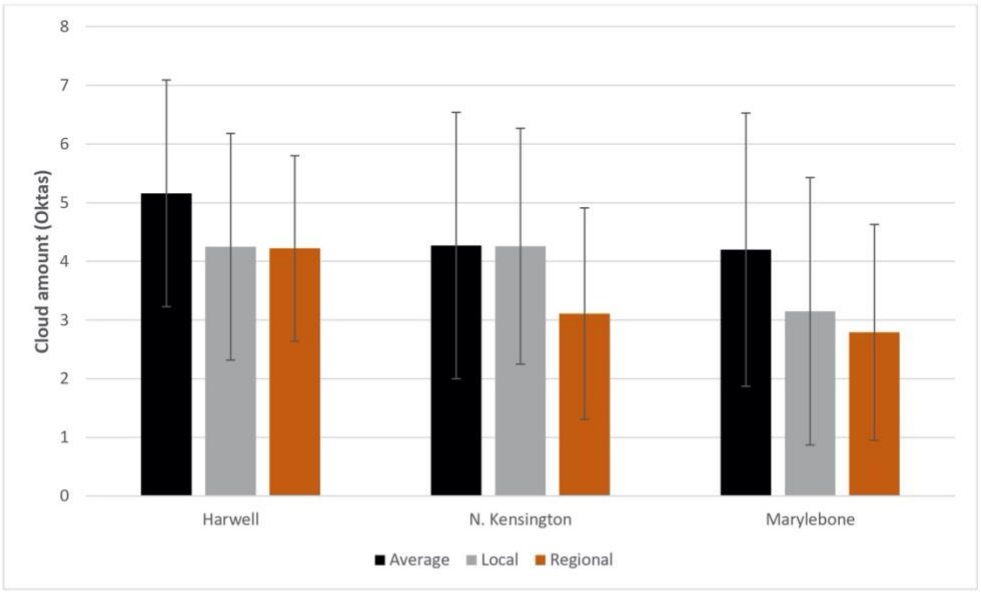


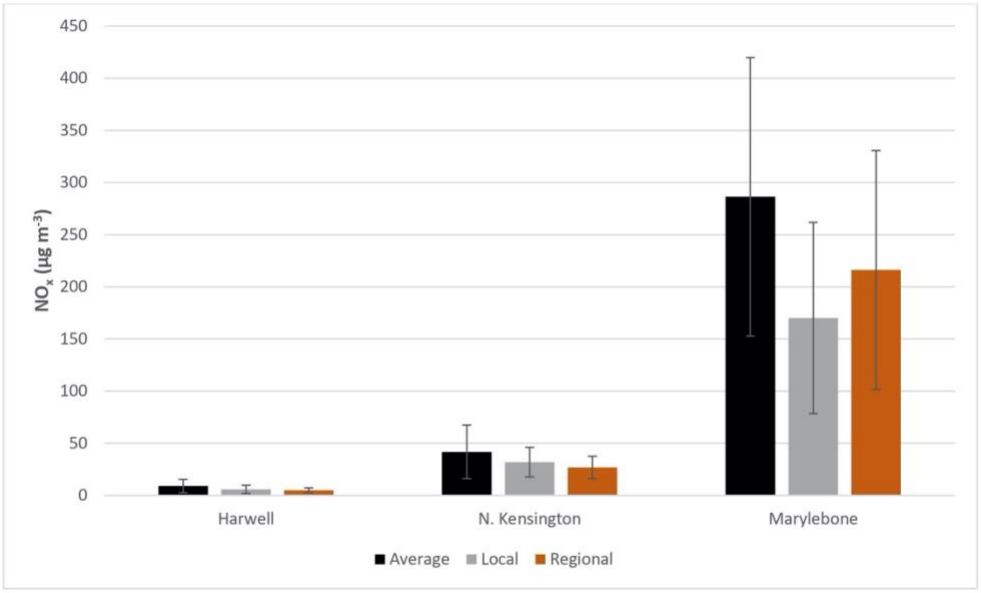
Figure S1: Example of a regional NPF event for all the sites of the present study (from top to bottom is HW, NK, MR)


100
90
80
70
60
50
40
30
20
10
0
Relative Humidity (%)
Harwell
N. Kensington
Marylebone
Average
Local
Regional

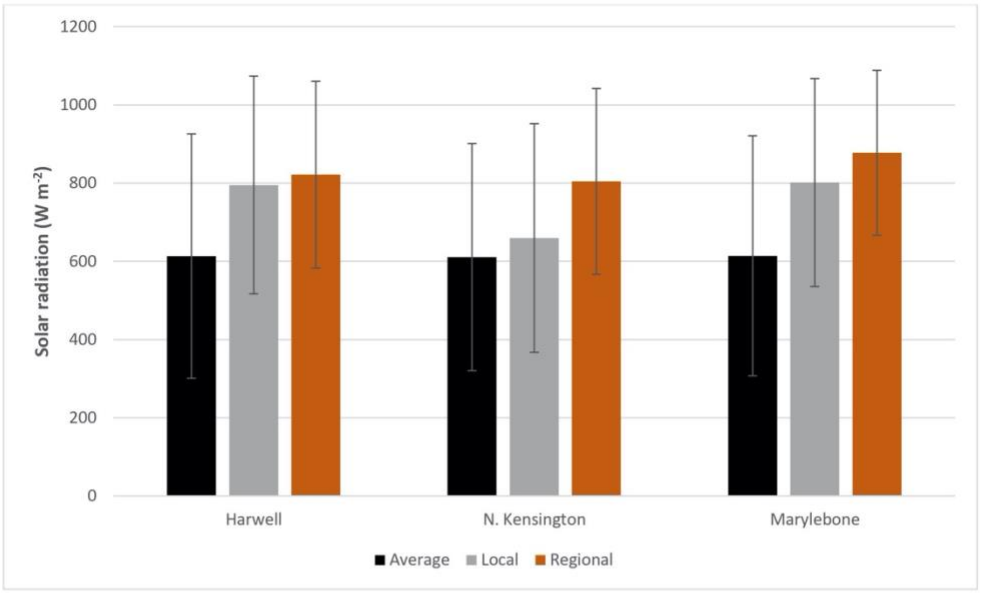
26


8
7
6
5
4
3
2
1
0
Cloud amount (Oktas)
Harwell
N. Kensington
Marylebone
Average
Local
Regional

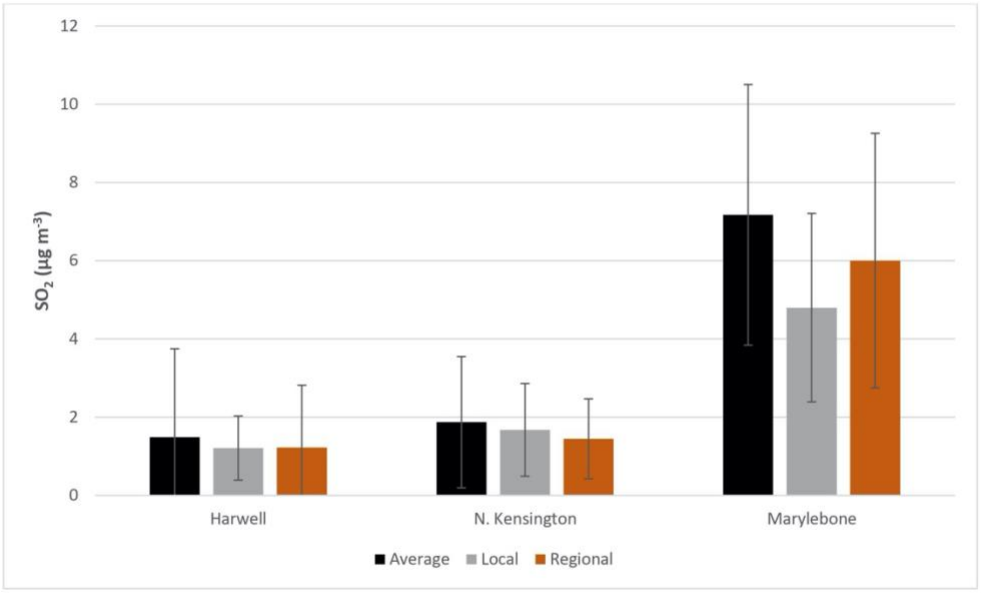
27


450
400
350
300
250
200
150
100
50
0
NO_x (µg m^{-3})
Harwell
N. Kensington
Marylebone
Average
Local
Regional

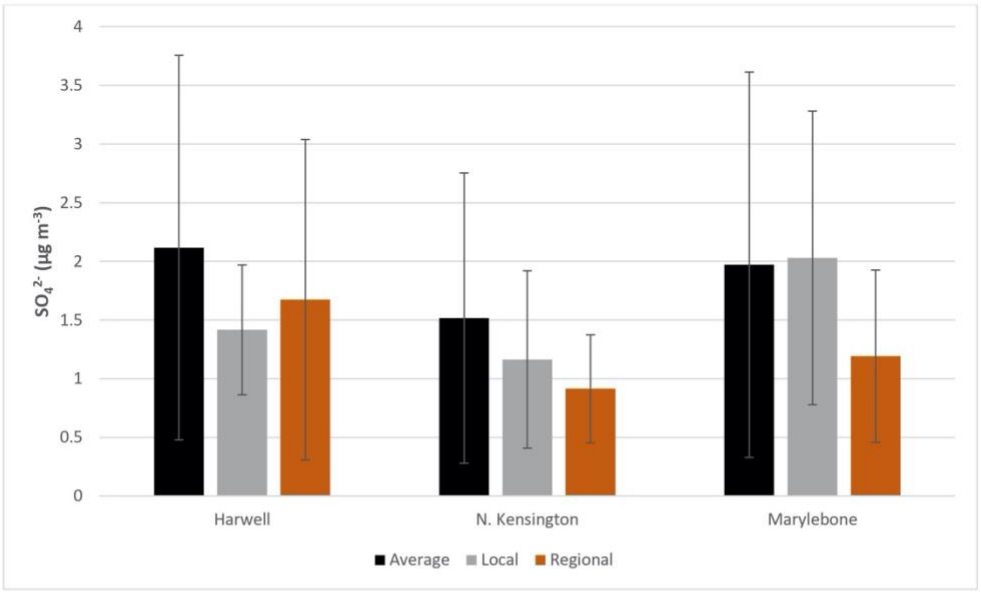
28


1200
1000
800
600
400
200
0
Solar radiation (W m^{-2})
Harwell
N. Kensington
Marylebone
Average
Local
Regional

29

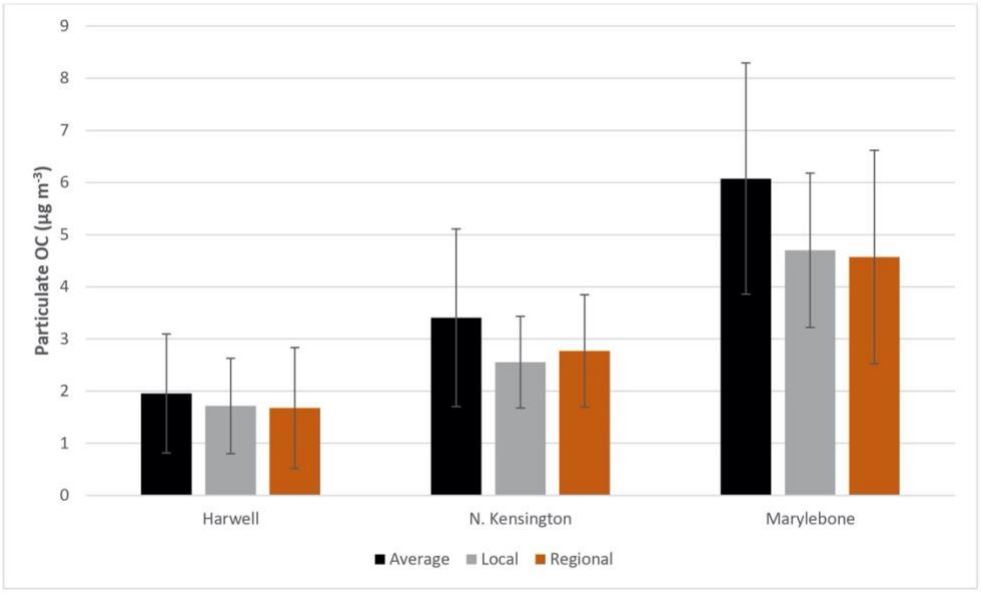

12
10
8
6
4
2
0
SO_2 (µg m^{-3})
Harwell
N. Kensington
Marylebone
Average
Local
Regional

30


4
3.5
3
2.5
2
1.5
1
0.5
0
SO_4^{2-} (µg m^{-3})
Harwell
N. Kensington
Marylebone
Average
Local
Regional

31

32



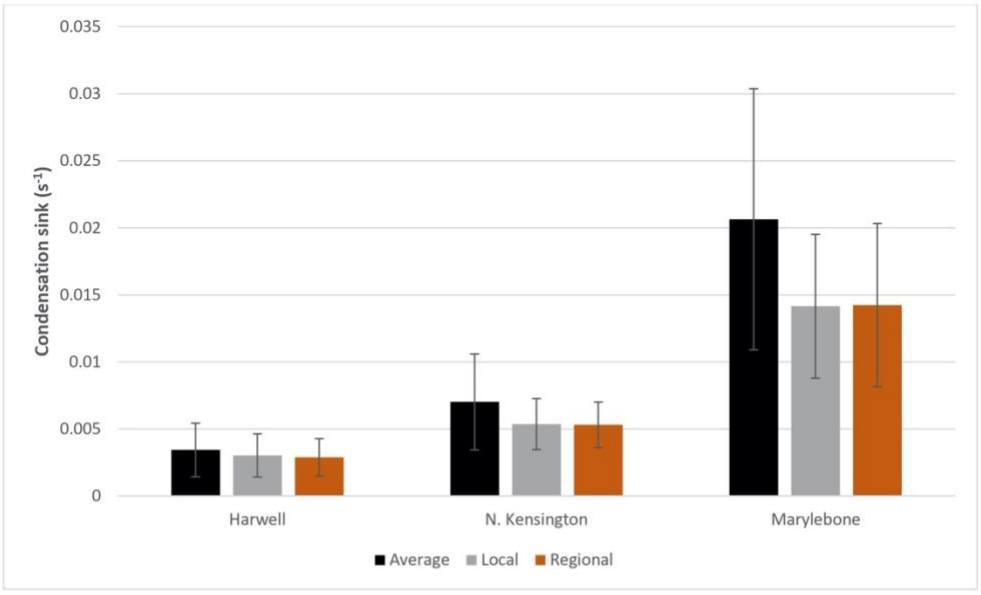


Figure S2: Average and NPF event conditions for Harwell and N. Kensington and Marylebone Road. On Marylebone Road, Regional events' conditions refer to the Regional event days for all 3 sites.

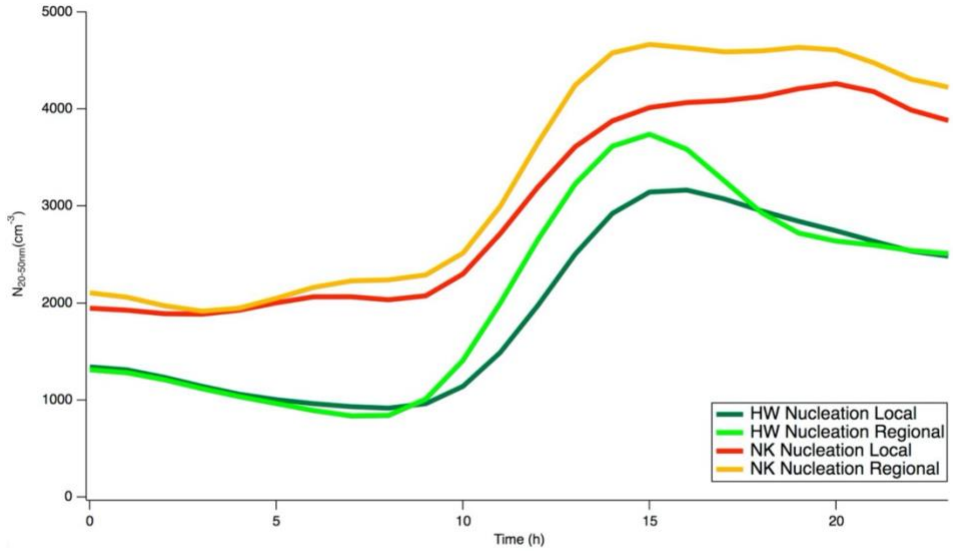


Figure S3: Diurnal variation of $N_{20\text{-}50nm}$ for Harwell and N. Kensington during local and regional NPF events.

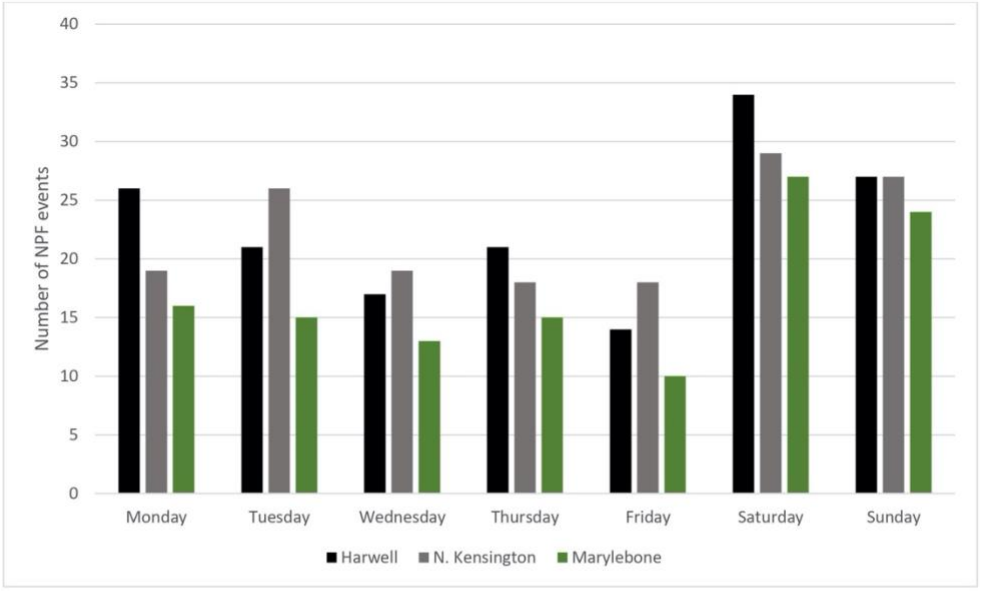
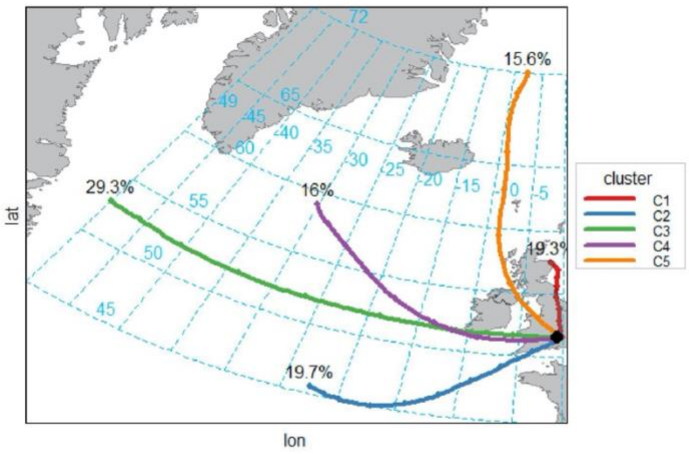
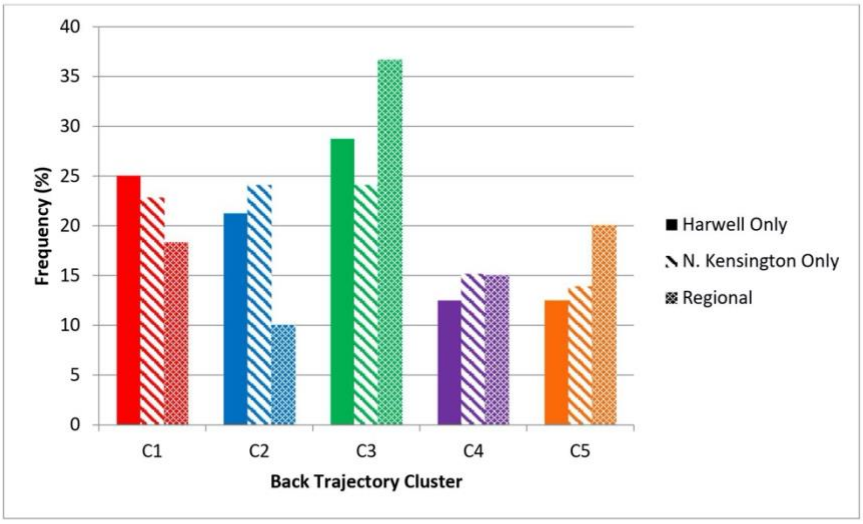


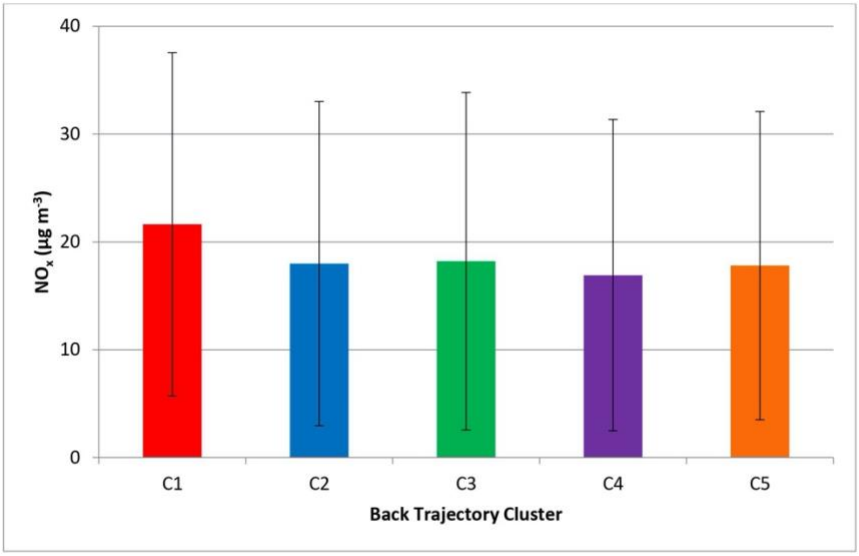
Figure S4: Weekly variation of NPF events in Marylebone Road.


15.6%
29.3%
16%
19.3%
19.7%
cluster
C1
C2
C3
C4
C5
lat
lon

47

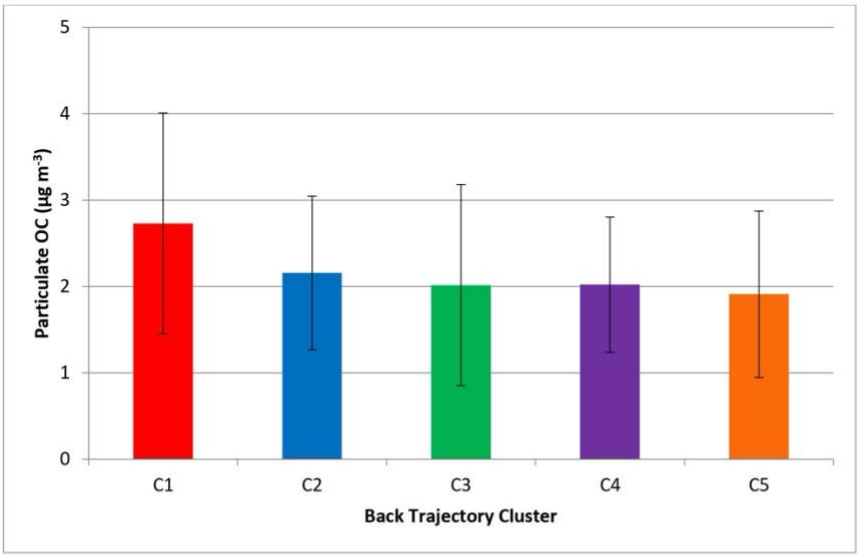

Frequency (%)
Back Trajectory Cluster
C1
C2
C3
C4
C5
Harwell Only
N. Kensington Only
Regional

48


40
30
20
10
0
NOx (µg m-3)
C1
C2
C3
C4
C5
Back Trajectory Cluster

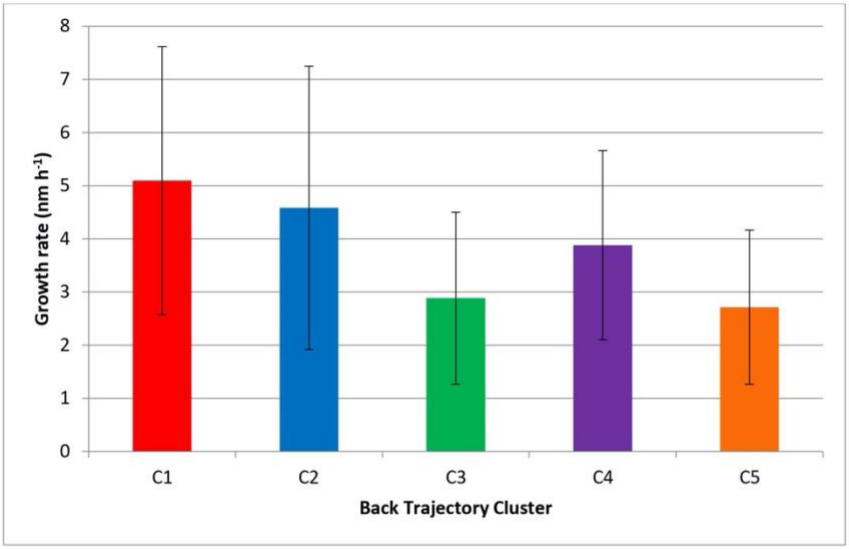
49

50

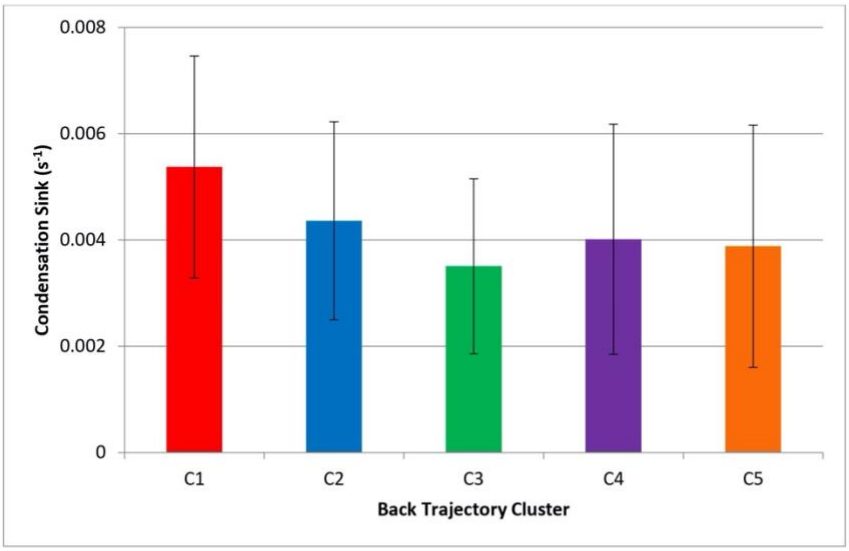

5
4
3
2
1
0
Particulate OC (µg m-3)
C1
C2
C3
C4
C5
Back Trajectory Cluster

51

52

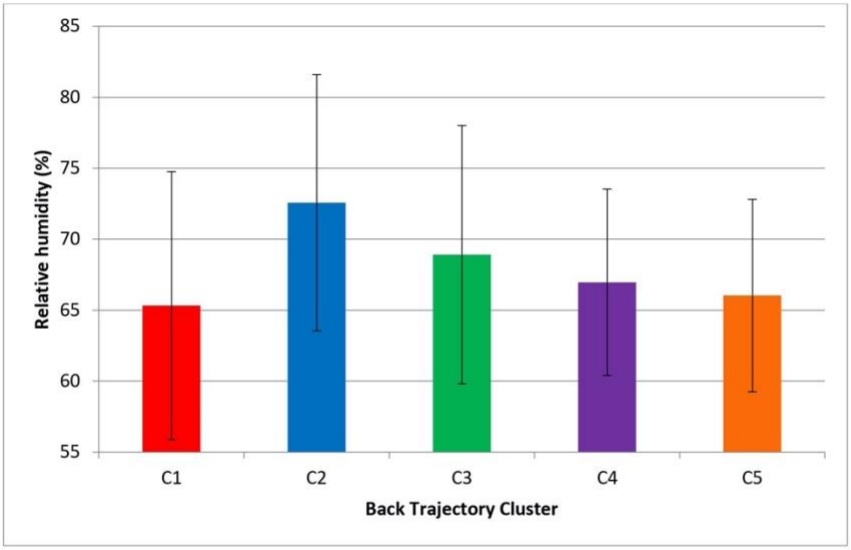

Growth rate (nm h-1)
8
7
6
5
4
3
2
1
0
C1
C2
C3
C4
C5
Back Trajectory Cluster

53

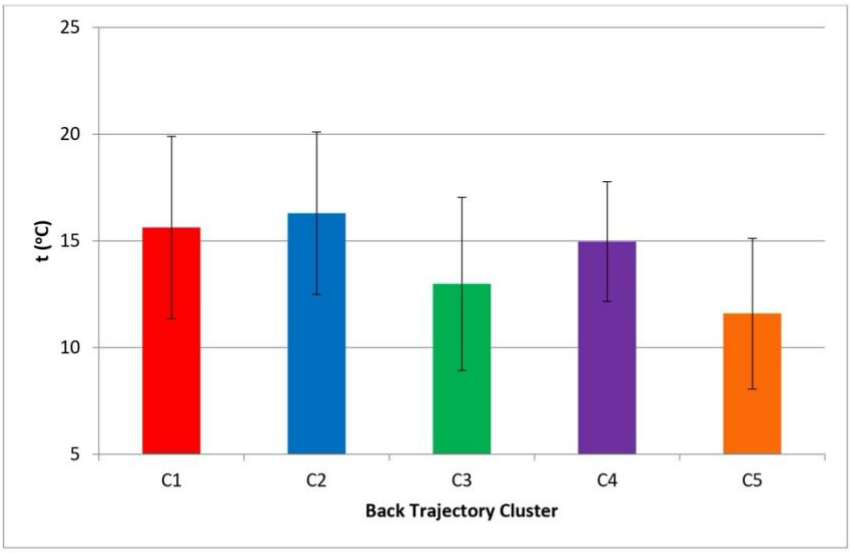

Condensation Sink (s-1)
0.008
0.006
0.004
0.002
0
C1
C2
C3
C4
C5
Back Trajectory Cluster

54

55


85
80
75
70
65
60
55
Relative humidity (%)
C1
C2
C3
C4
C5
Back Trajectory Cluster

56


25
20
15
10
5
t (°C)
C1
C2
C3
C4
C5
Back Trajectory Cluster

57

58

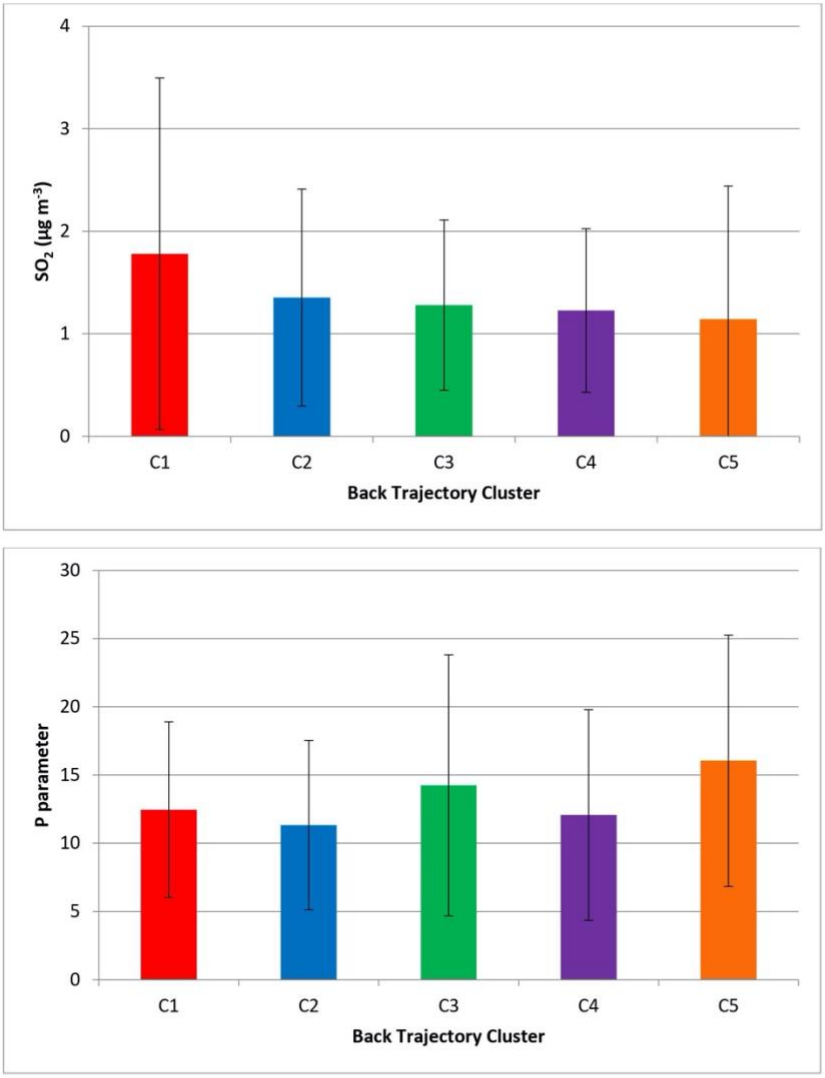
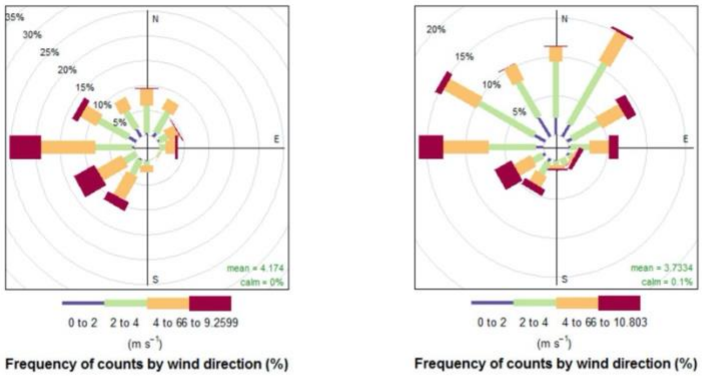


Figure S5: Air mass origin frequency and conditions for NPF events in Harwell and N. Kensington.

65



66

67

68 **Figure S6:** Wind profile for Local (left) and Regional (right) NPF events in Marylebone Road.

69

7.2 Supplement of "An Analysis of New Particle Formation (NPF) at Several European Sites"

Authors: Dimitrios Bousiotis, Francis D. Pope, Manuel Dall'Osto, Andreas Massling, Jacob Klenø Nøjgaard, Claus Nørdstrom, Jarrko V. Niemi, Harri Portin, Tuuka Petäjä, Noemi Perez, Andrés Alastuey, Xavier Querol, Giorgos Kouvarakis, Stergios Vratolis, Konstantinos Eleftheriadis, and Roy M. Harrison

Submitted to: Atmospheric Chemistry and Physics (acp-2020-414)

SUPPLEMENTARY INFORMATION

An Analysis of New Particle Formation (NPF) at Thirteen European Sites

Dimitrios Bousiotis, Francis D. Pope, David C. Beddows, Manuel Dall'Osto, Andreas Massling, Jacob Klenø Nøjgaard, Claus Nørdstrom, Jarkko V. Niemi, Harri Portin, Tuukka Petäjä, Noemi Perez, Andrés Alastuey, Xavier Querol, Giorgos Kouvarakis, Stergios Vratolis, Konstantinos Eleftheriadis, Alfred Wiedensohler, Kay Weinhold, Maik Merkel, Thomas Tuch and Roy M. Harrison

Table S1: Meteorological conditions and condensation sink on average (upper) and NPF event days' (lower).

Site	Temperature (°C)	Solar radiation (W m^{-2})	Wind speed (m s^{-1})	Relative humidity (%)	Condensation sink (s^{-1})
DENRU	9.80 ± 6.75	115 ± 98.0	4.17 ± 1.49	75.7 ± 11.7	9.46E-03 ± 6.56E-03
DENUB	9.82 ± 6.76	115 ± 98.0	4.17 ± 1.49	75.7 ± 11.7	1.42E-02 ± 8.80E-03
DENRO	10.0 ± 6.68	117 ± 98.5	4.16 ± 1.48	75.7 ± 11.4	3.10E-02 ± 1.79E-02
GERRU	10.3 ± 7.83	130 ± 94.0	2.58 ± 1.32	81.9 ± 10.4	7.02E-03 ± 3.54E-03
GERUB	11.1 ± 8.37	114 ± 86.3	2.33 ± 0.84	78.7 ± 14.7	9.11E-03 ± 4.48E-03
GERRO	11.1 ± 8.37	114 ± 86.3	2.33 ± 0.84	78.7 ± 14.7	1.20E-02 ± 5.58E-03
FINRU	4.79 ± 8.79	91 ± 89.0	1.31 ± 0.86	80.1 ± 15.7	2.32E-03 ± 1.25E-03
FINUB	6.52 ± 8.34	111 ± 110	3.43 ± 1.53	76.5 ± 13.9	6.34E-03 ± 3.20E-02
FINRO	7.72 ± 7.55	114 ± 103	4.26 ± 1.44	71.1 ± 11.4	8.96E-03 ± 3.70E-02
SPARU	13.9 ± 6.27	162 ± 82.3	0.94 ± 0.56	66.4 ± 15.6	5.49E-03 ± 2.70E-03
SPAUB	18.2 ± 5.68	180 ± 93.3	2.05 ± 0.99	69.2 ± 11.7	1.00E-02 ± 4.00E-03
GRERU	18.2 ± 6.01	201 ± 104	6.06 ± 3.38	70.0 ± 8.59	4.66E-03 ± 2.08E-03
GREUB	17.6 ± 7.37	183 ± 92.3	1.87 ± 0.74	60.5 ± 15.5	7.55E-03 ± 3.23E-03

Site	Temperature (°C)	Solar radiation (W m^{-2})	Wind speed (m s^{-1})	Relative humidity (%)	Condensation sink (s^{-1})
DENRU	13.5 ± 4.61	218 ± 71.0	4.29 ± 1.31	65.1 ± 9.38	8.67E-03 ± 5.46E-03
DENUB	13.4 ± 5.17	206 ± 76.4	4.59 ± 1.43	64.6 ± 9.96	1.12E-02 ± 5.80E-03
DENRO	13.6 ± 4.82	226 ± 78.5	4.49 ± 1.33	66.2 ± 8.48	2.66E-02 ± 1.41E-02
GERRU	15.4 ± 5.90	229 ± 68.3	2.05 ± 0.98	71.2 ± 7.75	8.61E-03 ± 2.98E-03
GERUB	17.6 ± 5.66	215 ± 60.6	2.08 ± 0.71	62.0 ± 10.5	9.96E-03 ± 3.86E-03
GERRO	18.7 ± 6.44	217 ± 65.70	1.96 ± 0.62	60.6 ± 11.1	1.42E-02 ± 4.51E-03
FINRU	3.48 ± 6.74	149 ± 80.8	1.44 ± 0.93	64.8 ± 14.6	2.15E-03 ± 9.52E-03
FINUB	5.32 ± 5.44	150 ± 85.5	3.28 ± 1.42	62.0 ± 13.3	4.33E-03 ± 2.01E-03
FINRO	11.6 ± 6.28	169 ± 91.9	4.47 ± 1.46	64.2 ± 14.0	6.89E-03 ± 2.62E-03
SPARU	12.0 ± 4.71	169 ± 67.3	1.00 ± 0.47	59.2 ± 13.4	4.68E-03 ± 2.34E-03
SPAUB	17.7 ± 5.29	196 ± 76.2	2.58 ± 1.04	57.9 ± 10.3	8.45E-03 ± 3.57E-03
GRERU	18.1 ± 4.99	233 ± 91.5	6.49 ± 3.33	67.6 ± 7.98	4.80E-03 ± 1.73E-03
GREUB	16.8 ± 7.50	192 ± 74.9	1.79 ± 0.72	54.6 ± 12.0	9.31E-03 ± 3.49E-03

Table S2: Average (top panel) and NPF event day (lower panel) concentrations of chemical components (*refers to NO_2, $^+$refers to Organic Mass from AMS measurements in PM_1, $^\bullet$refers to measurements at the Kalio site).

Site	NO_x/NO_2 (μg m^{-3})	SO_2 (μg m^{-3})	O_3 (μg m^{-3})	Organic carbon ($PM_{2.5}$) (μg m^{-3})	Sulphate ($PM_{2.5}$) (μg m^{-3})
DENRU	5.42 ± 4.55	0.26 ± 0.37	30.1 ± 9.32	1.48 ± 1.37	0.52 ± 0.46
DENUB	10.5 ± 6.24	-	28.2 ± 9.36	-	-
DENRO	68.5 ± 30.4	0.97 ± 0.78	31.1 ± 7.13	2.59 ± 1.81	0.55 ± 0.50
GERRU	-	-	-	2.18 ± 2.08	0.83 ± 0.12
GERUB	-	-	-	-	-
GERRO	-	-	-	-	-
FINRU	0.72 ± 0.55	0.09 ± 0.16	27.4 ± 8.07	1.78 ± 1.24$^+$	1.02 ± 0.82
FINUB	-	-	-	-	-
FINRO	88.1 ± 54.4	0.93 ± 1.27$^\bullet$	37.1 ± 14.0	-	-
SPARU	3.26 ± 2.46*	0.95 ± 0.57	75.9 ± 20.0	2.69 ± 1.34	1.21 ± 0.97
SPAUB	31.4 ± 14.2*	1.99 ± 0.88	54.9 ± 19.0	-	-
GRERU	0.52 ± 0.22*	-	49.5 ± 8.61	1.58 ± 0.97	-
GREUB	-	-	-	-	-

Site	NO_x/NO_2 (μg m^{-3})	SO_2 (μg m^{-3})	O_3 (μg m^{-3})	Organic carbon ($PM_{2.5}$) (μg m^{-3})	Sulphate ($PM_{2.5}$) (μg m^{-3})
DENRU	2.59 ± 1.64	0.18 ± 0.19	37.0 ± 6.10	0.74 ± 0.45	0.33 ± 0.15
DENUB	6.55 ± 2.77	-	35.5 ± 5.54	-	-
DENRO	53.9 ± 22.7	0.66 ± 0.54	36.3 ± 4.09	2.13 ± 0.87	0.34 ± 0.18
GERRU	-	-	-	1.83 ± 0.98	0.69 ± 0.28
GERUB	-	-	-	-	-
GERRO	-	-	-	-	-
FINRU	0.50 ± 0.42	0.13 ± 0.20	33.8 ± 7.12	1.21 ± 0.63$^+$	0.66 ± 0.45
FINUB	-	-	-	-	-
FINRO	52.9 ± 31.6	0.66 ± 0.60$^\bullet$	45.1 ± 16.0	-	-
SPARU	3.27 ± 3.39*	0.96 ± 0.71	78.1 ± 15.0	1.47 ± 0.76	0.53 ± 0.68
SPAUB	25.4 ± 12.7*	1.95 ± 0.81	59.6 ± 16.2	-	-
GRERU	0.56 ± 0.28*	-	50.8 ± 7.48	1.46 ± 0.74	-
GREUB	-	-	-	-	-

Table S3: Seasonal meteorological conditions and chemical compounds' concentrations (Winter – DJF; Spring – MAM; Summer – JJA; Autumn – SON)

DENRU	Solar radiation ($W\ m^{-2}$)	Wind speed ($m\ s^{-1}$)	Temperature (ºC)	Relative humidity (%)	NO_x ($\mu g\ m^{-3}$)	O_3 ($\mu g\ m^{-3}$)	Organic carbon ($PM_{2.5}$) ($\mu g\ m^{-3}$)	Condensation sink (s^{-1})
Winter	24	4.60	2.26	82.2	7.29	25.5	1.99	8.34E-03
Spring	163	4.06	8.69	69.4	5.07	36.3	1.43	1.09E-02
Summer	210	3.80	17.8	70.7	3.16	34.9	1.10	1.07E-02
Autumn	68	4.21	10.8	80.0	6.09	24.2	1.43	8.03E-03

DENUB	Solar radiation ($W\ m^{-2}$)	Wind speed ($m\ s^{-1}$)	Temperature (ºC)	Relative humidity (%)	NO_x ($\mu g\ m^{-3}$)	O_3 ($\mu g\ m^{-3}$)	Condensation sink (s^{-1})
Winter	24	4.62	2.19	82.2	12.08	22.7	1.37E-02
Spring	163	4.06	8.69	69.4	10.12	34.1	1.45E-02
Summer	210	3.80	17.8	70.7	8.25	33.5	1.61E-02
Autumn	68	4.21	10.8	80.0	11.71	22.2	1.25E-02

DENRO	Solar radiation ($W\ m^{-2}$)	Wind speed ($m\ s^{-1}$)	Temperature (ºC)	Relative humidity (%)	SO_2 ($\mu g\ m^{-3}$)	NO_x ($\mu g\ m^{-3}$)	O_3 ($\mu g\ m^{-3}$)	Organic carbon ($PM_{2.5}$) ($\mu g\ m^{-3}$)	Condensation sink (s^{-1})
Winter	23	4.61	2.40	82.6	0.90	68.99	32.4	2.84	3.19E-02
Spring	166	4.04	8.91	69.4	0.79	65.07	28.8	2.44	2.96E-02
Summer	210	3.80	17.8	70.7	0.94	61.40	31.2	2.19	3.08E-02
Autumn	68	4.21	10.8	80.0	1.24	78.45	32.0	2.89	3.21E-02

GERRU	Solar radiation ($W\ m^{-2}$)	Wind speed ($m\ s^{-1}$)	Temperature (ºC)	Relative humidity (%)	Organic carbon ($PM_{2.5}$) ($\mu g\ m^{-3}$)	Condensation sink (s^{-1})
Winter	39	2.94	0.69	89.6	3.37	7.94E-03
Spring	164	2.68	9.83	79.1	2.01	7.03E-03
Summer	215	2.12	19.0	74.8	1.44	6.92E-03
Autumn	85	2.62	10.3	85.3	1.81	6.28E-03

GERUB	Solar radiation ($W\ m^{-2}$)	Wind speed ($m\ s^{-1}$)	Temperature (°C)	Relative humidity (%)	Condensation sink (s^{-1})
Winter	39	2.94	0.69	89.6	1.12E-02
Spring	164	2.68	9.83	79.1	7.63E-03
Summer	215	2.12	19.0	74.8	8.58E-03
Autumn	85	2.62	10.3	85.3	9.19E-03

GERRO	Solar radiation ($W\ m^{-2}$)	Wind speed ($m\ s^{-1}$)	Temperature (°C)	Relative humidity (%)	Condensation sink (s^{-1})
Winter	30	2.45	0.52	92.3	1.42E-02
Spring	150	2.42	11.1	74.2	1.16E-02
Summer	194	2.19	20.2	66.1	1.16E-02
Autumn	70	2.27	11.0	83.5	1.11E-02

FINRU	Solar radiation ($W\ m^{-2}$)	Wind speed ($m\ s^{-1}$)	Temperature (°C)	Relative humidity (%)	SO_2 ($\mu g\ m^{-3}$)	NO_x ($\mu g\ m^{-3}$)	O_3 ($\mu g\ m^{-3}$)	Organic carbon (PM_1) ($\mu g\ m^{-3}$)	Condensation sink (s^{-1})
Winter	12	1.34	-5.56	93.1	0.14	1.14	27.3	NA	1.84E-03
Spring	131	1.34	3.74	72.0	0.12	0.69	35.5	1.52	2.52E-03
Summer	179	1.24	14.5	71.4	0.05	0.37	26.0	4.69	2.94E-03
Autumn	43	1.31	4.36	86.8	0.05	0.81	20.7	1.81	1.85E-03

FINUB	Solar radiation ($W\ m^{-2}$)	Wind speed ($m\ s^{-1}$)	Temperature (°C)	Relative humidity (%)	Condensation sink (s^{-1})
Winter	17	3.76	-2.54	84.5	5.97E-03
Spring	158	3.25	5.35	70.0	8.59E-03
Summer	213	3.18	16.2	70.0	5.79E-03
Autumn	55	3.52	6.94	81.7	4.84E-03

FINRO	Solar radiation ($W\ m^{-2}$)	Wind speed ($m\ s^{-1}$)	Temperature (ºC)	Relative humidity (%)	SO_2* ($\mu g\ m^{-3}$)	NO_x ($\mu g\ m^{-3}$)	O_3 ($\mu g\ m^{-3}$)	Condensation sink (s^{-1})
Winter	17	4.65	-0.68	77.2	1.35	102.34	32.9	8.67E-03
Spring	156	4.10	6.16	65.1	1.11	85.28	46.6	8.94E-03
Summer	212	3.98	16.3	67.2	0.69	77.13	39.6	9.39E-03
Autumn	56	4.37	7.53	76.0	0.64	89.79	28.4	8.76E-03

*SO_2 data are from the nearby Kalio station

SPARU	Solar radiation ($W\ m^{-2}$)	Wind speed ($m\ s^{-1}$)	Temperature (ºC)	Relative humidity (%)	SO_2 ($\mu g\ m^{-3}$)	NO_x ($\mu g\ m^{-3}$)	O_3 ($\mu g\ m^{-3}$)	Condensation sink (s^{-1})
Winter	94	0.83	6.87	62.3	0.86	3.89	61.5	3.60E-03
Spring	179	1.03	11.6	68.6	1.06	3.21	83.1	5.04E-03
Summer	234	0.97	20.6	63.2	0.89	2.86	91.2	7.67E-03
Autumn	129	0.90	15.3	71.4	1.02	2.99	66.9	5.00E-03

SPAUB	Solar radiation ($W\ m^{-2}$)	Wind speed ($m\ s^{-1}$)	Temperature (ºC)	Relative humidity (%)	SO_2 ($\mu g\ m^{-3}$)	NO_x ($\mu g\ m^{-3}$)	O_3 ($\mu g\ m^{-3}$)	Organic carbon ($PM_{2.5}$) ($\mu g\ m^{-3}$)	Condensation sink (s^{-1})
Winter	96	2.32	11.6	65.7	1.50	35.05	39.9	4.23	9.29E-03
Spring	220	2.22	15.9	69.5	1.86	30.73	63.0	3.67	1.00E-02
Summer	277	1.81	24.8	68.6	2.67	26.07	70.6	4.05	9.91E-03
Autumn	143	1.93	20.0	72.3	1.91	33.27	48.4	3.96	1.06E-02

GRERU	Solar radiation ($W\ m^{-2}$)	Wind speed ($m\ s^{-1}$)	Temperature (ºC)	Relative humidity (%)	NO_x ($\mu g\ m^{-3}$)	O_3 ($\mu g\ m^{-3}$)	Organic carbon ($PM_{2.5}$) ($\mu g\ m^{-3}$)	Condensation sink (s^{-1})
Winter	100	6.29	13.2	71.9	0.59	40.8	1.36	2.95E-03
Spring	239	5.21	17.1	69.5	0.58	51.6	1.53	4.04E-03
Summer	301	7.33	23.9	67.4	0.48	58.0	2.16	6.38E-03
Autumn	161	5.67	19.7	71.4	0.50	46.5	1.51	4.33E-03

GREUB	Solar radiation ($W\ m^{-2}$)	Wind speed ($m\ s^{-1}$)	Temperature (^oC)	Relative humidity (%)	Condensation sink (s^{-1})
Winter	88	1.86	9.30	71.9	8.81E-03
Spring	215	1.96	15.9	59.2	8.02E-03
Summer	282	2.00	26.5	46.0	6.93E-03
Autumn	144	1.68	18.5	65.2	6.73E-03

Table S4: Average conditions as a function of incoming wind direction at each site. (Wind directions in degrees, N – 337.5° – 22.5°; NE – 22.5° – 67.5°; E – 67.5° – 112.5°; SE – 112.5° – 157.5°; S – 157.5° – 202.5°; SW – 202.5° – 247.5°; W – 247.5° – 292.5°; NW – 292.5° – 337.5°)

DENRU	WS (m s^{-1})	RH (%)	T (°C)	CS (s^{-1})	OC (µg m^{-3})	NPF probability (%)	GR (nm h^{-1})	J_{10} (N cm^{-3} s^{-1})
N	3.45	72.8	7.85	6.62E-03	1.35	2.88%	3.47	9.58E-03
NE	3.92	74.2	8.35	9.57E-03	1.81	3.19%	3.30	2.21E-02
E	3.93	74.2	9.19	1.20E-02	2.41	2.33%	3.27	2.24E-02
SE	3.92	74.1	10.5	1.35E-02	2.39	1.36%	3.52	1.69E-02
S	4.15	77.9	10.5	1.16E-02	1.40	3.54%	3.47	2.50E-02
SW	4.34	80.1	9.64	7.61E-03	0.96	6.54%	3.07	3.19E-02
W	4.87	75.1	10.1	6.21E-03	0.94	10.4%	3.05	2.97E-02
NW	3.71	70.2	9.06	6.65E-03	1.06	8.01%	3.26	1.84E-02

DENUB	WS (m s^{-1})	RH (%)	T (°C)	CS (s^{-1})	NPF probability (%)	GR (nm h^{-1})	J_{10} (N cm^{-3} s^{-1})
N	3.45	72.8	7.86	1.07E-02	3.14%	3.28	1.95E-02
NE	3.92	74.2	8.36	1.32E-02	1.81%	3.36	2.74E-02
E	3.93	74.2	9.19	1.67E-02	1.85%	3.21	2.63E-02
SE	3.93	74.1	10.5	2.06E-02	2.05%	3.31	2.52E-02
S	4.16	77.9	10.6	1.70E-02	2.01%	3.22	2.45E-02
SW	4.34	80.0	9.69	1.27E-02	3.04%	3.02	1.69E-02
W	4.87	75.0	10.1	1.04E-02	7.76%	2.94	2.27E-02
NW	3.71	70.2	9.10	1.04E-02	5.20%	3.47	2.90E-02

DENRO	WS (m s^{-1})	RH (%)	T (°C)	CS (s^{-1})	OC (µg m^{-3})	NPF probability (%)	GR (nm h^{-1})	J_{10} (N cm^{-3} s^{-1})
N	3.47	72.7	8.02	2.46E-02	2.42	1.92%	4.33	5.46E-02
NE	3.92	74.3	8.56	3.62E-02	2.91	2.41%	5.27	1.04E-01
E	3.92	74.4	9.50	3.46E-02	3.43	2.95%	5.02	1.07E-01
SE	3.90	74.3	10.9	3.53E-02	3.30	2.16%	5.16	1.23E-01
S	4.15	77.9	10.7	3.18E-02	2.51	2.57%	4.56	7.35E-02
SW	4.34	80.2	9.91	3.08E-02	2.17	2.81%	4.00	5.76E-02
W	4.84	75.0	10.3	2.36E-02	2.12	7.44%	3.95	7.09E-02
NW	3.72	70.2	9.36	2.79E-02	2.20	3.49%	5.02	8.74E-02

GERRU	WS (m s^{-1})	RH (%)	T (°C)	CS (s^{-1})	OC (µg m^{-3})	NPF probability (%)	GR (nm h^{-1})	J_{10} (N cm^{-3} s^{-1})
N	2.00	84.5	9.08	7.47E-03	2.68	13.0%	4.40	7.21E-02
NE	2.19	81.9	8.86	9.30E-03	3.47	21.6%	4.47	8.48E-02
E	2.08	77.5	10.3	9.30E-03	3.34	25.0%	4.61	1.17E-01
SE	1.64	81.3	11.2	7.90E-03	2.18	21.9%	4.41	1.08E-01
S	2.45	81.6	10.8	6.48E-03	1.89	13.8%	4.33	1.23E-01
SW	3.24	82.7	10.3	6.49E-03	1.66	10.9%	4.29	1.02E-01
W	3.23	81.7	10.5	5.05E-03	1.38	12.5%	3.81	4.81E-02
NW	2.35	79.7	11.8	5.28E-03	1.55	10.3%	4.27	3.53E-02

GERUB	WS (m s^{-1})	RH (%)	T (ºC)	CS (s^{-1})	NPF probability (%)	GR (nm h^{-1})	J_{10} (N cm^{-3} s^{-1})
N	1.99	84.3	9.18	8.67E-03	13.8%	4.21	7.86E-02
NE	2.19	82.1	8.73	1.00E-02	26.7%	4.58	6.74E-02
E	2.07	77.3	10.4	1.14E-02	28.4%	4.67	1.05E-01
SE	1.64	81.1	11.3	1.12E-02	22.9%	4.04	1.31E-01
S	2.45	81.4	10.9	9.99E-03	11.4%	3.96	1.87E-01
SW	3.22	82.8	10.3	8.64E-03	8.7%	3.97	1.35E-01
W	3.21	81.6	10.6	6.82E-03	12.5%	3.66	6.84E-02
NW	2.38	79.8	11.7	6.72E-03	11.1%	4.08	3.76E-02

GERRO	WS (m s^{-1})	RH (%)	T (ºC)	CS (s^{-1})	NPF probability (%)	GR (nm h^{-1})	J_{10} (N cm^{-3} s^{-1})
N	2.10	81.6	9.84	1.55E-02	2.69%	4.94	6.71E-02
NE	2.09	79.8	9.10	1.59E-02	7.32%	4.97	1.30E-01
E	1.75	72.6	12.4	1.54E-02	15.5%	4.92	1.23E-01
SE	1.62	77.1	11.9	1.40E-02	10.4%	5.05	1.73E-01
S	2.05	80.1	10.9	8.95E-03	3.36%	5.36	1.90E-01
SW	2.99	78.7	11.3	8.41E-03	3.17%	5.53	1.24E-01
W	2.85	77.5	12.0	1.17E-02	5.72%	5.31	1.04E-01
NW	2.70	79.9	11.5	1.32E-02	4.50%	5.12	1.25E-01

FINRU	WS (m s^{-1})	RH (%)	T (ºC)	CS (s^{-1})	NPF probability (%)	GR (nm h^{-1})	J_{10} (N cm^{-3} s^{-1})
N	1.96	73.7	3.20	1.82E-03	14.6%	2.98	7.20E-03
NE	1.37	80.0	1.84	1.95E-03	10.9%	3.21	1.54E-02
E	0.98	81.8	3.21	2.49E-03	7.10%	3.09	1.41E-02
SE	1.19	83.4	4.66	2.97E-03	4.49%	3.08	1.47E-02
S	1.19	82.8	6.11	2.60E-03	5.20%	2.93	9.35E-03
SW	1.39	78.5	6.99	2.23E-03	8.42%	2.97	1.39E-02
W	1.26	78.0	4.75	1.75E-03	12.3%	2.61	1.23E-02
NW	1.88	73.9	3.41	1.78E-03	18.5%	2.91	9.58E-03

FINUB	WS (m s^{-1})	RH (%)	T (ºC)	CS (s^{-1})	NPF probability (%)	GR (nm h^{-1})	J_{10} (N cm^{-3} s^{-1})
N	2.95	72.1	4.31	5.49E-03	8.06%	2.61	2.85E-02
NE	2.86	77.5	3.17	6.82E-03	5.11%	2.67	3.66E-02
E	3.26	77.2	7.09	7.48E-03	1.57%	3.66	2.60E-02
SE	3.29	76.9	7.08	6.82E-03	1.73%	4.18	2.84E-02
S	3.77	81.0	6.98	6.67E-03	1.44%	4.14	3.55E-02
SW	4.14	79.0	9.34	4.86E-03	1.92%	3.21	1.39E-02
W	3.59	77.5	6.72	7.96E-03	7.53%	2.67	2.38E-02
NW	3.13	70.2	5.52	4.51E-03	13.3%	2.78	2.17E-02

FINRO	WS (m s^{-1})	RH (%)	T (ºC)	CS (s^{-1})	NPF probability (%)	GR (nm h^{-1})	J_{10} (N cm^{-3} s^{-1})
N	3.89	66.8	5.54	7.38E-03	9.31%	4.06	6.21E-02
NE	3.74	71.2	2.75	6.64E-03	8.52%	3.44	5.52E-02
E	4.21	70.3	9.30	7.87E-03	6.22%	3.75	4.29E-02
SE	4.23	72.3	7.94	9.41E-03	3.69%	4.00	6.76E-02
S	4.62	74.8	7.71	1.04E-02	1.81%	3.47	1.23E-01
SW	4.64	72.1	10.1	9.90E-03	3.22%	3.48	8.51E-02
W	4.12	72.4	8.07	9.64E-03	3.27%	3.83	8.64E-02
NW	4.23	67.0	6.61	8.44E-03	4.47%	3.99	7.07E-02

SPARU	WS (m s^{-1})	RH (%)	T (ºC)	CS (s^{-1})	NPF probability (%)	GR (nm h^{-1})	J_{10} (N cm^{-3} s^{-1})
N	0.46	72.3	13.0	4.64E-03	12.8%	3.57	2.54E-02
NE	0.26	72.1	13.1	5.00E-03	10.9%	3.67	8.75E-03
E	0.37	72.2	13.7	5.26E-03	10.8%	3.91	1.74E-02
SE	1.47	64.8	18.0	7.43E-03	9.28%	3.97	1.48E-02
S	0.93	58.7	17.1	6.46E-03	10.7%	3.68	1.67E-02
SW	0.45	60.9	13.7	4.45E-03	14.0%	3.23	2.07E-02
W	0.59	59.0	13.6	4.33E-03	17.5%	3.62	1.94E-02
NW	0.99	64.5	12.8	4.19E-03	16.7%	3.52	1.23E-02

SPAUB	WS (m s^{-1})	RH (%)	T (ºC)	CS (s^{-1})	NPF probability (%)	GR (nm h^{-1})	J_{10} (N cm^{-3} s^{-1})
N	1.24	73.8	15.9	1.14E-02	9.22%	3.02	8.62E-03
NE	2.56	79.0	17.1	9.39E-03	5.49%	3.30	2.63E-02
E	2.80	74.2	20.1	9.71E-03	6.83%	3.25	1.40E-02
SE	1.94	68.5	21.2	1.19E-02	8.42%	2.79	2.13E-02
S	1.63	63.6	22.3	1.22E-02	12.4%	3.32	3.35E-02
SW	1.60	65.9	21.6	1.13E-02	12.3%	3.91	2.59E-02
W	2.37	65.9	16.3	9.12E-03	21.5%	3.30	2.12E-02
NW	2.30	64.1	14.4	7.52E-03	22.0%	3.50	9.87E-03

GRERU	WS (m s^{-1})	RH (%)	T (ºC)	CS (s^{-1})	OC (µg m^{-3})	NPF probability (%)	GR (nm h^{-1})	J_{10} (N cm^{-3} s^{-1})
N	6.23	72.3	17.0	3.75E-03	1.39	5.51%	2.99	4.65E-03
NE	2.52	68.8	19.6	3.78E-03	1.42	5.87%	2.34	1.71E-03
E	3.36	70.6	18.7	3.82E-03	1.47	3.37%	2.68	5.41E-03
SE	8.44	65.5	18.1	3.28E-03	1.58	5.15%	3.60	2.38E-03
S	4.44	69.9	17.6	3.10E-03	1.53	5.25%	3.54	3.84E-02
SW	5.20	72.7	19.0	4.43E-03	1.79	5.56%	3.63	1.23E-02
W	6.39	69.5	21.1	5.41E-03	1.86	6.68%	3.97	2.69E-03
NW	7.08	67.9	17.7	4.51E-03	1.53	10.3%	3.42	5.25E-03

GREUB	WS (m s^{-1})	RH (%)	T (ºC)	CS (s^{-1})	NPF probability (%)	GR (nm h^{-1})	J_{10} (N cm^{-3} s^{-1})
N	1.90	48.8	7.28	7.57E-03	9.35%	3.20	3.62E-02
NE	2.54	55.2	17.6	6.29E-03	4.82%	3.30	3.86E-02
E	1.56	60.7	22.3	7.12E-03	4.73%	3.69	3.33E-02
SE	1.99	64.6	21.1	7.61E-03	7.44%	3.57	1.38E-02
S	0.88	68.5	17.3	8.23E-03	7.78%	3.62	4.53E-02
SW	1.55	65.0	17.4	7.53E-03	8.61%	3.87	4.44E-02
W	2.05	53.5	16.4	9.01E-03	10.6%	3.98	5.07E-02
NW	1.30	51.8	20.6	1.01E-02	11.8%	3.99	6.56E-02

Table S5: Average conditions for local and region-wide events (*refers to NO_2).

Site		WS (m s^{-1})		T (ºC)		RH (%)		SR (W m^{-2})	
		Local	Regional	Local	Regional	Local	Regional	Local	Regional
DEN	RU	4.20 ± 1.29	4.72 ± 1.34	14.1 ± 4.51	10.6 ± 4.05	65 ± 9.51	65.6 ± 8.90	223 ± 69.3	197 ± 76.1
	UB	4.54 ± 1.47	4.72 ± 1.34	12.2 ± 5.49	10.6 ± 4.05	64.3 ± 10.4	65.6 ± 8.90	210 ± 76.8	197 ± 76.1
GER	RU	2.18 ± 1.59	1.97 ± 1.50	14.6 ± 8.44	15.9 ± 7.64	74.4 ± 20.2	69.2 ± 21.8	196 ± 247	251 ± 287
	UB	2.12 ± 1.55	1.97 ± 1.50	15.6 ± 8.14	15.9 ± 7.64	72.5 ± 21.3	69.2 ± 21.8	225 ± 269	251 ± 287
FIN	RU	1.37 ± 1.02	1.67 ± 1.12	4.3 ± 7.69	0.87 ± 5.93	64.7 ± 21.5	64.3 ± 19.8	153 ± 202	148 ± 197
	UB	3.38 ± 1.89	3.14 ± 1.54	6.25 ± 6.15	3.75 ± 5.24	65.4 ± 17.6	56.4 ± 17.5	142 ± 206	168 ± 224
SPA	RU	1.16 ± 0.51	1.07 ± 0.25	11.8 ± 4.76	13.6 ± 5.41	63.2 ± 12.2	55.2 ± 13.1	149 ± 66.3	167 ± 65.1
	UB	2.49 ± 1.06	3.13 ± 0.80	17.9 ± 5.41	17.5 ± 4.99	58.6 ± 9.44	54.2 ± 10.2	199 ± 77.3	194 ± 69.7

Site		CS (s^{-1})		GR (nm h^{-1})		J_{10} (N cm^{-3} s^{-1})	
		Local	Regional	Local	Regional	Local	Regional
DEN	RU	9.03E-03 ± 5.69E-03	6.94E-03 ± 3.84E-03	3.26 ± 1.49	2.89 ± 1.12	2.39E-02 ± 3.27E-02	2.00E-02 ± 2.45E-02
	UB	1.17E-02 ± 6.35E-03	1.01E-02 ± 3.86E-03	3.36 ± 1.59	2.78 ± 0.92	2.67E-02 ± 4.01E-02	2.16E-02 ± 2.76E-02
GER	RU	7.36E-03 ± 3.42E-03	9.42E-03 ± 4.22E-03	4.11 ± 1.71	4.48 ± 1.71	7.58E-02 ± 3.76E-02	1.02E-01 ± 1.65E-01
	UB	9.63E-03 ± 1.03E-02	1.01E-02 ± 5.24E-03	4.19 ± 1.71	4.27 ± 1.66	1.00E-01 ± 2.02E-01	1.03E-01 ± 2.28E-01
FIN	RU	2.26E-03 ± 1.33E-03	1.85E-03 ± 1.17E-03	2.96 ± 1.76	2.75 ± 1.37	1.03E-02 ± 1.82E-02	1.29E-02 ± 2.34E-02
	UB	4.46E-03 ± 3.16E-03	4.04E-03 ± 2.70E-03	3.06 ± 1.53	2.60 ± 0.87	2.19E-02 ± 3.33E-02	2.45E-02 ± 3.67E-02
SPA	RU	4.00E-03 ± 2.13E-03	4.30E-03 ± 1.59E-03	3.98 ± 2.22	3.38 ± 1.32	1.32E-02 ± 1.74E-02	1.32E-02 ± 1.68E-02
	UB	8.92E-03 ± 3.60E-03	6.66E-03 ± 2.12E-03	3.37 ± 1.56	3.31 ± 0.81	2.28E-02 ± 2.67E-02	1.44E-02 ± 1.42E-02

Site		NOx / NO_2(µg m^{-3})		O_3 (µg m^{-3})		SO_2 (µg m^{-3})	
		Local	Regional	Local	Regional	Local	Regional
DEN	RU	2.65 ± 1.73	2.35 ± 1.23	36.6 ± 6.37	38.3 ± 4.91	0.19 ± 0.21	0.14 ± 0.07
	UB	6.89 ± 2.96	5.89 ± 2.24	35.3 ± 5.78	35.7 ± 5.09	-	-
GER	RU	-	-	-	-	-	-
	UB	-	-	-	-	-	-
FIN	RU	0.51 ± 0.54	0.51 ± 0.59	33.4 ± 8.59	35.6 ± 6.63	0.13 ± 0.27	0.13 ± 0.19
	UB	-	-	-	-	-	-
SPA	RU	2.38 ± 1.01*	2.74 ± 0.66*	72.2 ± 8.54	81.4 ± 14.2	0.95 ± 0.89	0.90 ± 0.66
	UB	26.5 ± 13.1*	19.6 ± 7.21*	59.2 ± 15.8	67.6 ± 15.4	1.94 ± 0.81	1.94 ± 0.93

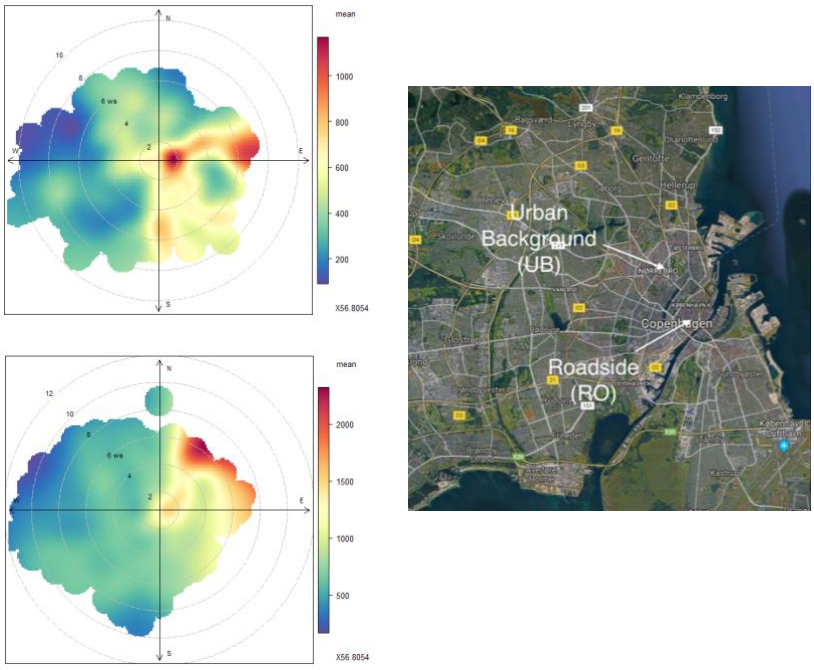


Figure S1: Sources of 56 nm particles at DENUB and DENRO (map provided by ©Google Maps)

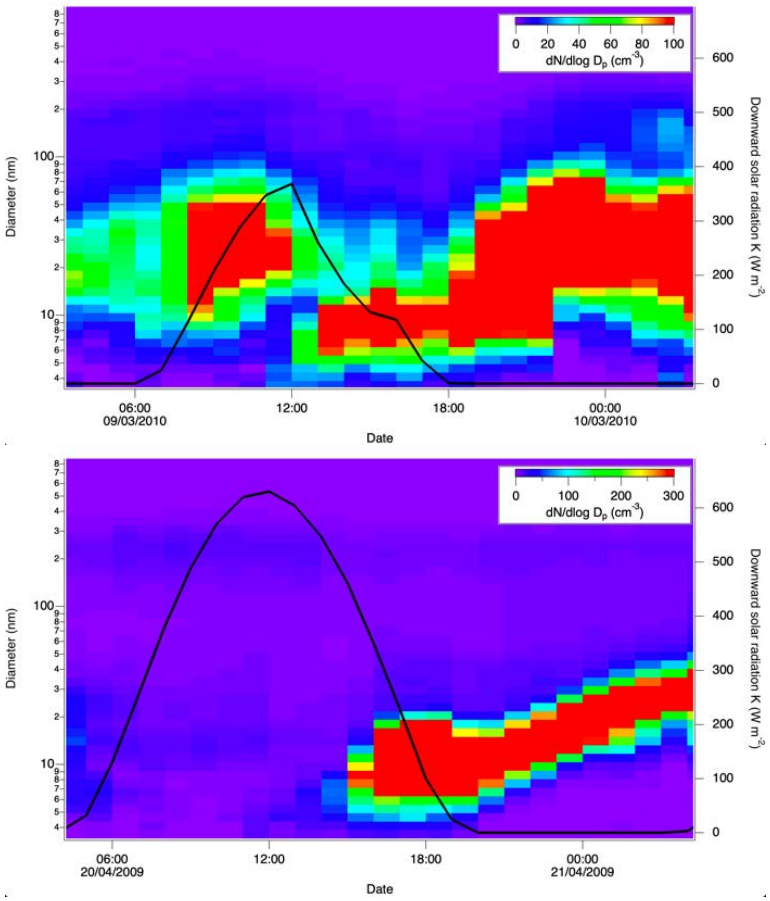


Figure S2: Examples of NPF events with late growth of the particles at FINRU.

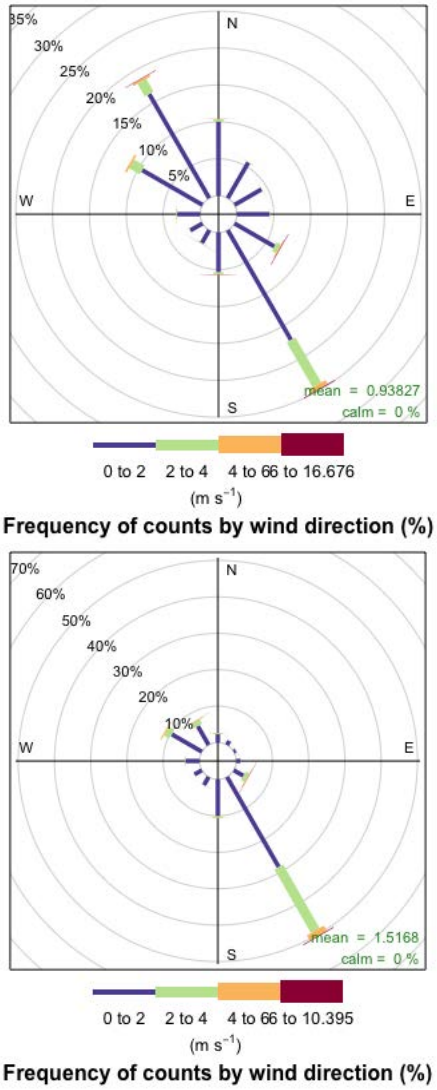


Figure S3: Average wind rose (top panel) and wind rose for the time window 9:00 to 15:00 (bottom panel) for SPARU.

7.3 Supplement of "The effect of meteorological conditions and atmospheric composition in the occurrence and development of New Particle Formation (NPF) events in Europe"

Table S1: Number of NPF events per day of the week and season of the year

Site	Monday	Tuesday	Wednesday	Thursday	Friday	Saturday	Sunday	**Total**
UKRU	26	21	17	21	14	34	27	**160**
UKUB	19	26	19	18	18	29	27	**156**
UKRO	16	15	13	15	10	27	24	**120**
DENRU	15	25	24	30	27	24	31	**176**
DENUB	17	19	7	18	17	16	22	**116**
DENRO	12	11	11	24	14	21	24	**117**
GERRU	25	19	26	24	24	23	23	**164**
GERUB	21	23	28	21	22	24	30	**169**
GERRO	5	10	12	5	9	9	12	**62**
FINRU	20	29	27	29	28	27	30	**190**
FINUB	19	10	18	17	11	20	15	**110**
FINRO	4	4	4	8	4	13	12	**49**
SPARU	11	10	7	9	5	15	11	**68**
SPAUB	9	11	10	15	11	16	25	**97**
GRERU	18	13	17	19	19	15	15	**116**
GREUB	12	11	6	17	8	17	11	**82**

	UKRU	UKUB	UKRO	DENRU	DENUB	DENRO	GERRU	GERUB	GERRO	FINRU	FINUB	FINRO	SPARU	SPAUB	GRERU	GREUB
Winter	2	7	0	0	0	1	5	0	2	15	8	4	19	19	18	18
Spring	47	40	39	62	67	39	57	64	9	113	60	10	34	23	54	26
Summer	93	86	63	100	39	65	81	82	40	24	7	23	9	17	23	13
Autumn	18	23	17	14	10	12	21	23	11	38	34	12	6	38	21	25
Total	**160**	**156**	**119**	**176**	**116**	**117**	**164**	**169**	**62**	**190**	**109**	**49**	**68**	**97**	**116**	**82**

Table S2: Correlation matrices of the meteorological and atmospheric variables. Correlations greater than 0.50 or less than -0.50 are marked bold.

UKRU	SR	RH	T	WS	P	SO_2	NOx	O_3	OC	SO_4^{2-}	CS
SR	**1.00**	**-0.70**	**0.55**	0.12	0.13	0.05	-0.12	0.45	0.07	0.05	0.00
RH	**-0.70**	**1.00**	**-0.55**	-0.29	-0.10	-0.10	0.20	**-0.59**	0.01	-0.04	0.01
T	**0.55**	**-0.55**	**1.00**	0.12	0.09	-0.01	-0.26	0.37	0.15	0.11	-0.03
WS	0.12	-0.29	0.12	**1.00**	-0.42	0.04	-0.19	0.41	-0.29	-0.12	-0.32
P	0.13	-0.10	0.09	-0.42	**1.00**	-0.07	0.03	-0.09	0.13	0.15	0.23
SO_2	0.05	-0.10	-0.01	0.04	-0.07	**1.00**	0.05	0.06	0.03	0.37	0.31
NOx	-0.12	0.20	-0.26	-0.19	0.03	0.05	**1.00**	**-0.58**	0.48	0.16	**0.54**
O_3	0.45	**-0.59**	0.37	0.41	-0.09	0.06	**-0.58**	**1.00**	-0.30	-0.07	-0.34
OC	0.07	0.01	0.15	-0.29	0.13	0.03	0.48	-0.30	**1.00**	0.37	**0.59**
SO_4^{2-}	0.05	-0.04	0.11	-0.12	0.15	0.37	0.16	-0.07	0.37	**1.00**	0.44
CS	0.00	0.01	-0.03	-0.32	0.23	0.31	**0.54**	-0.34	**0.59**	0.44	**1.00**

UKUB	SR	RH	T	WS	P	SO_2	NOx	O_3	OC	SO_4^{2-}	CS
SR	**1.00**	**-0.70**	**0.53**	0.22	0.07	0.08	-0.15	0.47	0.01	0.02	-0.14
RH	**-0.70**	**1.00**	**-0.56**	-0.22	-0.19	-0.08	0.20	**-0.66**	-0.10	-0.01	0.08
T	**0.53**	**-0.56**	**1.00**	0.21	-0.05	-0.05	-0.38	**0.52**	-0.12	0.01	-0.18
WS	0.22	-0.22	0.21	**1.00**	-0.33	-0.16	-0.44	0.41	-0.43	-0.25	**-0.50**
P	0.07	-0.19	-0.05	-0.33	**1.00**	0.18	0.22	-0.06	0.31	0.25	0.26
SO_2	0.08	-0.08	-0.05	-0.16	0.18	**1.00**	0.44	-0.16	0.29	0.40	0.39
NOx	-0.15	0.20	-0.38	-0.44	0.22	0.44	**1.00**	**-0.56**	**0.57**	0.29	**0.79**
O_3	0.47	**-0.66**	**0.52**	0.41	-0.06	-0.16	**-0.56**	**1.00**	-0.14	-0.14	-0.40
OC	0.01	-0.10	-0.12	-0.43	0.31	0.29	**0.57**	-0.14	**1.00**	0.46	**0.63**
SO_4^{2-}	0.02	-0.01	0.01	-0.25	0.25	0.40	0.29	-0.14	0.46	**1.00**	0.36
CS	-0.14	0.08	-0.18	**-0.50**	0.26	0.39	**0.79**	-0.40	0.63	0.36	**1.00**

UKRO	SR	RH	T	WS	P	SO_2	NOx	O_3	OC	SO_4^{2-}	CS
SR	**1.00**	**-0.68**	**0.51**	0.11	0.15	0.14	0.17	0.16	-0.03	0.03	0.06
RH	**-0.68**	**1.00**	-0.49	-0.14	-0.24	0.01	-0.01	-0.35	0.09	-0.01	0.06
T	**0.51**	-0.49	**1.00**	0.16	0.21	0.18	0.15	-0.02	0.02	0.01	0.15
WS	0.11	-0.14	0.16	**1.00**	-0.34	0.17	0.17	0.08	-0.16	-0.19	-0.05
P	0.15	-0.24	0.21	-0.34	**1.00**	-0.10	-0.05	0.04	0.15	0.08	0.01
SO_2	0.14	0.01	0.18	0.17	-0.10	**1.00**	**0.91**	**-0.65**	0.36	-0.13	**0.72**
NOx	0.17	-0.01	0.15	0.17	-0.05	**0.91**	**1.00**	-0.63	0.34	-0.04	**0.81**
O_3	0.16	-0.35	-0.02	0.08	0.04	**-0.65**	-0.63	**1.00**	-0.43	0.02	**-0.64**
OC	-0.03	0.09	0.02	-0.16	0.15	0.36	0.34	-0.43	**1.00**	0.24	0.47
SO_4^{2-}	0.03	-0.01	0.01	-0.19	0.08	-0.13	-0.04	0.02	0.24	**1.00**	0.18
CS	0.06	0.06	0.15	-0.05	0.01	**0.72**	**0.81**	**-0.64**	0.47	0.18	**1.00**

DENRU	SR	RH	T	WS	SO_2	NOx	O_3	OC	SO_4^{2-}	CS
SR	**1.00**	**-0.56**	0.44	0.07	-0.05	-0.12	0.43	-0.04	-0.09	0.05
RH	**-0.56**	**1.00**	-0.39	0.02	0.02	0.17	**-0.54**	0.01	0.18	-0.08
T	0.44	-0.39	**1.00**	-0.18	-0.09	-0.19	0.37	-0.13	-0.06	0.22
WS	0.07	0.02	-0.18	**1.00**	0.02	-0.28	0.22	-0.09	0.02	-0.32
SO_2	-0.05	0.02	-0.09	0.02	**1.00**	0.18	-0.06	0.48	**0.51**	0.34
NOx	-0.12	0.17	-0.19	-0.28	0.18	**1.00**	**-0.58**	0.34	0.22	**0.54**
O_3	0.43	**-0.54**	0.37	0.22	-0.06	-0.58	**1.00**	-0.17	-0.18	-0.17
OC	-0.04	0.01	-0.13	-0.09	0.48	0.34	-0.17	**1.00**	0.65	**0.58**
SO_4^{2-}	-0.09	0.18	-0.06	0.02	**0.51**	0.22	-0.18	**0.65**	**1.00**	0.41
CS	0.05	-0.08	0.22	-0.32	0.34	**0.54**	-0.17	**0.58**	0.41	**1.00**

DENUB	SR	RH	T	WS	NOx	O_3	CS
SR	**1.00**	**-0.55**	0.45	0.06	-0.02	0.39	0.04
RH	**-0.55**	**1.00**	-0.40	-0.02	0.15	**-0.58**	-0.04
T	0.45	-0.40	**1.00**	-0.13	-0.11	0.40	0.18
WS	0.06	-0.02	-0.13	**1.00**	-0.37	0.26	-0.35
NOx	-0.02	0.15	-0.11	-0.37	**1.00**	-0.59	**0.55**
O_3	0.39	**-0.58**	0.40	0.26	-0.59	**1.00**	-0.23
CS	0.04	-0.04	0.18	-0.35	**0.55**	-0.23	**1.00**

DENRO	SR	RH	T	WS	SO_2	NOx	O_3	OC	CS
SR	**1.00**	**-0.55**	0.30	0.21	0.37	0.29	0.41	0.00	0.26
RH	**-0.55**	**1.00**	-0.45	-0.09	-0.26	-0.17	-0.42	-0.20	-0.29
T	0.30	-0.45	**1.00**	0.04	0.22	0.12	0.25	0.39	0.41
WS	0.21	-0.09	0.04	**1.00**	-0.16	-0.12	0.53	-0.19	-0.12
SO_2	0.37	-0.26	0.22	-0.16	**1.00**	**0.80**	0.01	0.31	**0.62**
NOx	0.29	-0.17	0.12	-0.12	**0.80**	**1.00**	-0.02	0.20	**0.67**
O_3	0.41	-0.42	0.25	0.53	0.01	-0.02	**1.00**	-0.01	0.05
OC	0.00	-0.20	0.39	-0.19	0.31	0.20	-0.01	**1.00**	0.36
CS	0.26	-0.29	0.41	-0.12	**0.62**	**0.67**	0.05	0.36	**1.00**

GERRU	SR	RH	T	WS	P	OC	SO_4^{2-}	CS
SR	**1.00**	**-0.70**	**0.55**	0.20	0.13	-0.07	-0.07	0.02
RH	**-0.70**	**1.00**	**-0.61**	-0.31	-0.12	0.08	0.10	-0.01
T	**0.55**	**-0.61**	**1.00**	0.01	0.11	-0.34	-0.29	-0.11
WS	0.20	-0.31	0.01	**1.00**	-0.24	-0.14	-0.09	-0.35
P	0.13	-0.12	0.11	-0.24	**1.00**	0.11	0.13	0.23
OC	-0.07	0.08	-0.34	-0.14	0.11	**1.00**	0.83	**0.65**
SO_4^{2-}	-0.07	0.10	-0.29	-0.09	0.13	0.83	**1.00**	**0.52**
CS	0.02	-0.01	-0.11	-0.35	0.23	**0.65**	**0.52**	**1.00**

GERUB	SR	RH	T	WS	P	CS
SR	**1.00**	**-0.72**	**0.55**	0.25	0.16	-0.06
RH	**-0.72**	**1.00**	**-0.61**	-0.32	-0.17	0.10
T	**0.55**	**-0.61**	**1.00**	0.05	0.11	-0.20
WS	0.25	-0.32	0.05	**1.00**	-0.21	-0.31
P	0.16	-0.17	0.11	-0.21	**1.00**	0.21
CS	-0.06	0.10	-0.20	-0.31	0.21	**1.00**

GERRO	SR	RH	T	WS	P	CS
SR	**1.00**	**-0.65**	**0.50**	0.19	0.14	0.05
RH	**-0.65**	**1.00**	**-0.72**	-0.14	-0.16	0.03
T	**0.50**	**-0.72**	**1.00**	-0.03	0.16	-0.14
WS	0.19	-0.14	-0.03	**1.00**	-0.15	-0.34
P	0.14	-0.16	0.16	-0.15	**1.00**	0.19
CS	0.05	0.03	-0.14	-0.34	0.19	**1.00**

FINRU	SR	RH	T	WS	P	SO_2	NOx	O_3	OM	SO_4^{2-}	CS
SR	**1.00**	**-0.67**	**0.50**	0.11	0.11	0.00	-0.24	0.30	-0.05	-0.14	0.09
RH	**-0.67**	**1.00**	**-0.56**	-0.21	-0.27	-0.12	0.31	**-0.55**	0.00	0.17	-0.20
T	**0.50**	**-0.56**	**1.00**	0.01	0.03	-0.20	-0.28	-0.14	0.27	-0.20	0.28
WS	0.11	-0.21	0.01	**1.00**	0.17	0.11	0.13	0.35	-0.20	-0.20	-0.07
P	0.11	-0.27	0.03	0.17	**1.00**	0.00	-0.08	-0.08	0.34	0.12	0.19
SO_2	0.00	-0.12	-0.20	0.11	0.00	**1.00**	0.18	0.09	NA	NA	0.21
NOx	-0.24	0.31	-0.28	0.13	-0.08	0.18	**1.00**	-0.24	NA	NA	0.12
O_3	0.30	**-0.55**	-0.14	0.35	-0.08	0.09	-0.24	**1.00**	NA	NA	0.02
OM	-0.05	0.00	0.27	-0.20	0.34	NA	NA	NA	**1.00**	0.43	0.61
SO_4^{2-}	-0.14	0.17	-0.20	-0.20	0.12	NA	NA	NA	0.43	**1.00**	0.18
CS	0.09	-0.20	0.28	-0.07	0.19	0.21	0.12	0.02	0.61	0.18	**1.00**

FINUB	SR	RH	T	WS	P	CS
SR	**1.00**	**-0.54**	0.45	0.05	0.09	0.00
RH	**-0.54**	**1.00**	-0.35	0.04	-0.23	-0.01
T	0.45	-0.35	**1.00**	-0.02	-0.01	0.00
WS	0.05	0.04	-0.02	**1.00**	-0.26	0.00
P	0.09	-0.23	-0.01	-0.26	**1.00**	0.00
CS	0.00	-0.01	0.00	0.00	0.00	**1.00**

FINRO	SR	RH	T	WS	P	NOx	O_3	CS
SR	**1.00**	**-0.58**	0.47	0.03	0.08	0.05	0.20	0.09
RH	**-0.58**	**1.00**	-0.29	-0.05	-0.24	0.02	-0.34	0.01
T	0.47	-0.29	**1.00**	-0.07	-0.02	-0.08	0.18	0.05
WS	0.03	-0.05	-0.07	**1.00**	-0.25	-0.29	0.41	-0.32
P	0.08	-0.24	-0.02	-0.25	**1.00**	0.10	-0.09	0.13
NOx	0.05	0.02	-0.08	-0.29	0.10	**1.00**	-0.61	**0.75**
O_3	0.20	-0.34	0.18	0.41	-0.09	**-0.61**	**1.00**	-0.51
CS	0.09	0.01	0.05	-0.32	0.13	**0.75**	-0.51	**1.00**

SPARU	SR	RH	T	WS	P	SO_2	NO2	O_3	CS
SR	**1.00**	**-0.45**	**0.50**	0.38	0.09	0.10	-0.02	0.34	0.34
RH	**-0.45**	**1.00**	-0.29	-0.20	-0.24	-0.08	0.05	-0.48	-0.06
T	**0.50**	-0.29	**1.00**	0.16	0.24	0.07	-0.05	0.54	0.47
WS	0.38	-0.20	0.16	**1.00**	-0.16	0.13	-0.02	0.25	0.10
P	0.09	-0.24	0.24	-0.16	**1.00**	-0.15	0.12	0.09	0.14
SO_2	0.10	-0.08	0.07	0.13	-0.15	**1.00**	0.14	0.19	0.25
NO_2	-0.02	0.05	-0.05	-0.02	0.12	0.14	**1.00**	-0.02	0.42
O_3	0.34	-0.48	0.54	0.25	0.09	0.19	-0.02	**1.00**	0.44
CS	0.34	-0.06	0.47	0.10	0.14	0.25	0.42	0.44	**1.00**

SPAUB	SR	RH	T	WS	P	SO_2	NO2	O_3	CS
SR	**1.00**	-0.43	0.44	0.18	0.03	0.25	-0.09	0.32	0.00
RH	-0.43	**1.00**	-0.04	-0.23	-0.16	-0.12	0.10	-0.23	0.16
T	0.44	-0.04	**1.00**	-0.14	0.11	0.35	-0.07	0.38	0.11
WS	0.18	-0.23	-0.14	**1.00**	-0.26	-0.08	-0.34	0.32	-0.43
P	0.03	-0.16	0.11	-0.26	**1.00**	0.13	0.15	-0.10	0.10
SO_2	0.25	-0.12	0.35	-0.08	0.13	**1.00**	0.20	0.13	0.16
NO_2	-0.09	0.10	-0.07	-0.34	0.15	0.20	**1.00**	**-0.66**	**0.59**
O_3	0.32	-0.23	0.38	0.32	-0.10	0.13	**-0.66**	**1.00**	-0.35
CS	0.00	0.16	0.11	-0.43	0.10	0.16	**0.59**	-0.35	**1.00**

GRERU	SR	RH	T	WS	P	NO_2	O_3	OC	CS
SR	**1.00**	-0.30	0.33	0.02	-0.11	0.36	0.19	0.09	0.18
RH	-0.30	**1.00**	-0.25	-0.27	0.20	-0.20	-0.12	-0.06	0.08
T	0.33	-0.25	**1.00**	0.00	**-0.53**	0.02	**0.54**	0.35	0.46
WS	0.02	-0.27	0.00	**1.00**	-0.21	-0.03	0.15	0.14	0.11
P	-0.11	0.20	**-0.53**	-0.21	**1.00**	-0.10	-0.35	-0.24	-0.09
NO_2	0.36	-0.20	0.02	-0.03	-0.10	**1.00**	0.00	0.01	-0.02
O_3	0.19	-0.12	**0.54**	0.15	-0.35	0.00	**1.00**	**0.50**	**0.62**
OC	0.09	-0.06	0.35	0.14	-0.24	0.01	**0.50**	**1.00**	0.47
CS	0.18	0.08	0.46	0.11	-0.09	-0.02	**0.62**	0.47	**1.00**

GREUB	SR	RH	T	WS	P	CS
SR	**1.00**	**-0.55**	0.48	0.47	-0.15	0.04
RH	**-0.55**	**1.00**	**-0.67**	-0.30	0.18	-0.07
T	0.48	**-0.67**	**1.00**	0.20	**-0.51**	-0.06
WS	0.47	-0.30	0.20	**1.00**	-0.15	-0.21
P	-0.15	0.18	**-0.51**	-0.15	**1.00**	0.16
CS	0.04	-0.07	-0.06	-0.21	0.16	**1.00**

Table S3: Gradients and R^2 for the relation between VOCs and NPF event variables. Gradients of $R^2 > 0.50$ are in bold

UKRU	a_N	R^2	a_{GR}	R^2	a_J	R^2
Benzene	**-3.37E-01**	0.88	1.24E+00	0.16	-5.99E-03	0.07
Ethane	**-5.42E-02**	0.88	-4.79E-01	0.26	**-4.61E-03**	0.77
Ethene	**-1.65E-01**	0.83	**2.64E+00**	0.60	**-1.70E-02**	0.57
Ethylbenzene	**-7.01E-01**	0.79	6.78E+00	0.41	**-5.77E-02**	0.63
Isobutane	**-2.06E-01**	0.75	**1.41E+00**	0.70	-5.62E-03	0.11
Isooctane	-5.23E-01	0.45	**1.09E+01**	0.80	9.32E-03	0.11
Isopentane	**-1.96E-01**	0.74	**2.36E+00**	0.58	**2.36E-02**	0.72
m-p-xylene	**-2.92E-01**	0.86	**3.21E+00**	0.68	-1.98E-02	0.35
n-butane	**-1.67E-01**	0.79	1.04E+00	0.44	1.43E-02	0.11
n-heptane	**-9.63E-01**	0.80	**1.36E+01**	0.73	-1.46E-02	0.13
n-hexane	**-1.21E+00**	0.84	**6.82E+00**	0.67	1.33E-02	0.11
n-pentane	**-3.71E-01**	0.67	**3.49E+00**	0.64	-8.97E-03	0.06
o-xylene	**-5.34E-01**	0.71	**8.59E+00**	0.86	-1.81E-02	0.42
Propane	**-7.77E-02**	0.76	1.97E-01	0.24	-4.28E-03	0.49
Propene	**-1.50E-01**	0.67	-4.01E-01	0.02	6.20E-03	0.08
Toluene	**-1.48E-01**	0.79	**1.88E+00**	0.81	-9.26E-03	0.43
1-2-4 trimethylbenzene	-4.36E-01	0.46	5.38E+00	0.29	**-4.78E-02**	0.68
1-3 butadiene	-1.17E+00	0.40	**-1.68E+01**	0.71	**-7.55E-02**	0.66
1-butene	-9.39E-02	0.03	-4.77E+00	0.25	-1.99E-02	0.07
2-methylpentane	**-7.66E-01**	0.77	**8.49E+00**	0.57	**4.56E-02**	0.64

FINRU	a_N	R^2	a_{GR}	R^2	a_J	R^2
Acetaldehyde	-1.04E-01	0.05	**-2.16E+00**	0.69	1.23E-02	0.07
Aceticacid	1.19E-01	0.13	**5.88E+00**	0.77	3.33E-02	0.21
Acetolnitrite	-1.02E+00	0.13	**1.33E+01**	0.59	6.62E-02	0.18
Acetone	-4.63E-02	0.08	**3.38E+00**	0.74	5.85E-03	0.19
Benzene	-4.46E-01	0.11	**-2.02E+01**	0.83	-4.13E-02	0.02
Ethanol formic acid	4.04E-02	0.06	1.31E+00	0.10	4.77E-03	0.10
Isoprene	**-3.17E+00**	0.51	**1.59E+01**	0.87	-1.50E+00	0.31
MEK	6.45E-01	0.34	-8.03E+00	0.36	2.95E-02	0.03
Methacrolein MVK	-5.15E+00	0.45	**3.75E+01**	0.66	2.92E-02	0.02
Methanol	1.68E-02	0.05	**1.48E+00**	0.75	3.48E-03	0.12
Monoterpenes	-1.17E-01	0.38	**2.84E+00**	0.56	1.11E-03	0.00
Toluene	**-4.25E+00**	0.59	**2.88E+01**	0.80	-5.55E-02	0.13

UKRO	a_N	R^2	a_{GR}	R^2	a_J	R^2
Benzene	**-1.03E-01**	0.68	**1.36E+00**	0.80	**4.42E-02**	0.78
Cis-2-butene	**-1.93E-01**	0.59	8.33E-01	0.02	1.70E-01	0.48
Ethane	**-2.45E-02**	0.53	2.99E-02	0.06	2.28E-03	0.14
Ethene	**-4.59E-02**	0.69	**5.74E-01**	0.83	**2.50E-02**	0.97
Ethylbenzene	**-7.13E-02**	0.87	**1.22E+00**	0.77	3.59E-02	0.41
Ethyne	**-8.43E-02**	0.74	**1.23E+00**	0.75	**4.22E-02**	0.64
Isobutane	**-4.70E-02**	0.55	**6.07E-01**	0.78	**1.79E-02**	0.92
Isooctane	**-7.53E-02**	0.80	**2.14E+00**	0.78	**7.35E-02**	0.67
Isopentane	**-1.10E-02**	0.70	**2.64E-01**	0.72	**1.00E-02**	0.82
Isoprene	-2.75E-02	0.07	4.34E-01	0.01	2.24E-03	0.00
m-p-xylene	**-1.99E-02**	0.91	**3.81E-01**	0.56	**1.47E-02**	0.64
n-butane	**-2.17E-02**	0.61	**2.58E-01**	0.78	4.07E-03	0.17
n-heptane	**-1.53E-01**	0.75	**2.51E+00**	0.80	**1.15E-01**	0.82
n-hexane	**-1.10E-01**	0.63	**2.86E+00**	0.75	**8.28E-02**	0.74
n-octane	**-2.64E-01**	0.55	**7.06E+00**	0.72	**2.73E-01**	0.98
n-pentane	**-5.44E-02**	0.53	**1.03E+00**	0.80	**2.99E-02**	0.86
o-xylene	**-4.69E-02**	0.88	**9.58E-01**	0.65	**4.37E-02**	0.86
Propane	**-3.16E-02**	0.68	1.95E-01	0.32	**1.01E-02**	0.90
Propene	**-6.69E-02**	0.87	**1.15E+00**	0.85	**3.55E-02**	0.78
Toluene	**-1.22E-02**	0.84	**2.76E-01**	0.74	**1.15E-02**	0.85
Trans-2-butene	**-2.63E-01**	0.72	3.16E+00	0.35	**1.41E-01**	0.60
Trans-2-pentene	**-1.67E-01**	0.73	2.69E+00	0.31	**1.16E-01**	0.52
1-2-3 trimethylbenzene	**-1.45E-01**	0.78	**3.31E+00**	0.66	**1.28E-01**	0.81
1-2-4 trimethylbenzene	**-4.89E-02**	0.85	7.64E-01	0.43	3.26E-02	0.46
1-3-5 trimethylbenzene	**-8.62E-02**	0.77	**1.56E+00**	0.67	**6.65E-02**	0.64
1-3 butadiene	**-1.78E-01**	0.81	2.99E+00	0.44	9.04E-02	0.26
1-butene	-2.18E-01	0.38	2.51E+00	0.25	**1.24E-01**	0.64
1-pentene	**-2.43E-01**	0.52	6.92E+00	0.37	**3.00E-01**	0.82
2-methylpentane	**-3.73E-02**	0.68	**8.57E-01**	0.67	**2.83E-02**	0.80

Abbreviations

AMS: Aerosol Mass Spectrometer

BVOC: Biogenic Volatile Organic Compound

CCN: Cloud Condensation Nuclei

CoagS: Coagulation Sink

CPC: Condensation Particle Counter

CS: Condensation Sink

GMD: Geometrical Mean Diameter

GR: Growth Rate

HOM: Highly Oxygenated Organic Molecules

NPF: New Particle Formation

NSF: Nucleation Strength Factor

PM: Particulate Matter

RH: Relative Humidity

SMPS: Scanning Mobility Particle Sizer

U.I.: Urban Increment

VOC: Volatile organic compound