~~An Analysis~~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, while the growth and formation rates of newly formed particles were higher at roadsides, 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 different countries across Europe and combined with the results from a previous study in the UK, 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 1) are analysed in the size range 3 nm < Dp < 1000 nm. A detailed list of the site locations and the data available for each is found in Table 1 (seasonal data availability is found in Table S1). For site naming the first three letters refer to the country (DEN = Denmark, GER = Germany, FIN = Finland, SPA = Spain, GRE = Greece) while the next two refer to the type of ~~the~~ site (RU = Rural background, UB = Urban background, RO = Roadside). Average meteorological conditions and concentrations of chemical compounds for all sites are found in Tables S2~~1~~ and S3~~2~~ respectively; their seasonal variation is found in Table S4~~3~~.

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):

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

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. Due to the differences in the smallest particle size available between the sites, a discrepancy would exist for the growth rate values presented (sites with lower size cut ~~sh~~would present lower values of growth rate, as the growth rate tends to increase with particle size ~~at~~in this range (Deng et al., 2020)). As a result, a direct comparison of the growth rate values found among sites with significant differences at the smallest particle size available was avoided.

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 (± 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.

As mentioned in the methodology for NPF event selection (chapter 2.2.1) days with particle formation ~~associated with~~resulting directly from traffic emissions were excluded. For those ~~extracted~~identified as NPF event days though, mainly for the roadside sites, such formation still occurs. It is impossible with the data available for this study to remove the traffic related particle formation in the calculations included in this study, by effectively separating it from secondary particle formation or calculate it. Using average conditions for comparison would lead to negative values in most cases since in order for an NPF event to occur other emissions are reduced. This may result~~s~~ in an overestimation of the formation rates at roadside sites presented in this study which, as mentioned earlier, was reduced as far as possible by choosing a time window for which we would have the maximum effect of secondary particle formation and the minimum possible effect from traffic related particle formation.

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:

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

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

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

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.). Finally, it should be mentioned that all the results presented are normalised according the seasonal data availability for each site, based upon the expression:

$$NPF_{probability} = \frac{N_{NPF\ event\ days\ for\ group\ of\ days\ X}}{N_{days\ with\ available\ data\ for\ group\ of\ days\ X}}$$

3. RESULTS ~~AND DISCUSSION~~

The seasonal NPF probability for all sites is found in Table S5. The annual number of NPF events, growth rate and formation rate for all the sites is found in Table S6.

3.1 Frequency and ~~s~~Seasonality of NPF ~~e~~Events

In Denmark, NPF events occurred at all three sites 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%). The seasonal variation favoured summer at DENRU and DENRO, while at DENUB a similar frequency for spring and summer was found (Figure 2). The within-week variation of the events favours weekends compared to weekdays going from the rural background site to the roadside site (Figure 3). Interesting is the increased frequency of NPF events found in all Danish sites on Thursday among the weekdays. This trend though does not have a plausible explanation and is probably coincidental.

A higher frequency of events for all types of environments is found for the German sites compared to all 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. It should be noted though that, due to the lack of spring and summer data for the first two years at the German roadside site, the frequency of events is probably a lot higher, and the seasonal variation should further favour these seasons. No substantial within-week variation was found for any of the sites in this country, 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 in this study.

NPF events at the sites in Finland presented the most diverse seasonal variation, peaking at the background sites in spring and at the roadside site in summer (while the spring data availability is somewhat~~how~~ reduced for the Finnish roadside site, the general trend remains the same if all seasons had the same data availability). The frequency of NPF events at FINRU was higher (8.66%) compared to the urban sites (4.97% at FINUB and 5.20% at FINRO). Strong within-week variation favouring weekends is found for the roadside site, while no clear variation was found for the background sites. This may be due to either the higher condensation sink during weekdays that supresses the events, or the dominant impact of the traffic emissions which could make the detection of NPF events harder.

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 background 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. 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. For both sites a higher NPF probability was found on weekends compared to weekdays, though this trend is stronger at SPAUB. Finally, for Greece data are available for two background sites, though not in close proximity (the distance between the sites is about 350 km). While in Greece meteorological conditions are favourable in general for NPF events, with high solar radiation and low relative humidity, their frequency was only 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 reported in the study by Kalivitis et al. (2019). Most NPF events occurred in spring at both sites, peaking in April. It is interesting that the sites in southern Europe (in Spain and Greece) 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 than at the sites in northern and central Europe, and insolation is higher.

3.2 The Formation and Growth Rates

For the Danish sites the growth rate was found to be higher at the roadside site at 4.45±1.87 nm h^{-1} and it was similar for the other two sites (3.19±1.43 for DENRU and 3.19±1.45 for DENUB) nm h^{-1} (Figure 4), though the peak was found in different seasons (Figure 5), 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 event that occurred in that season). The formation rate (J_{10}) was found to be broadly similar at the rural and urban background sites and higher at DENRO (Figure 6), favoured by different seasons at each site (summer at DENRU, spring at DENUB though with minimal~~um~~ differences and autumn at DENRO) (Figure 7).

Similar to the frequency of NPF events, the German sites also had higher growth rates compared to sites of the same type in other areas of this study, with GERRU having 4.34±1.73 nm h^{-1}, GERUB 4.24±1.69 nm h^{-1} and GERRO 5.17±2.20 nm h^{-1} (Figure 3). While the difference between GERRU and GERUB is not statistically significant, there is a significant difference with GERRO ($p < 0.005$). Higher growth rates were found in summer compared to spring for all sites (Figure 5). 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). Similarly, J_{10} at the German sites was also the highest among the sites of this study, increasing from the GERRU to GERRO. It was found to be higher in summer for the background sites and in autumn for GERRO.

For the Finnish sites, 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 the 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. The formation rate 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.

At the Spanish sites, 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. The formation rate at SPAUB is comparable to the other urban background sites (apart from GERUB) and 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. 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 extent its value. Finally, the growth rate of particles was found to be similar at both Greek 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, having a similar trend with the temperature and particulate organic carbon concentrations in the area. The formation rate presented a unique trend, having high averages in winter for both sites. Interestingly, contrary to most background sites in this study, the lowest average J_{10} was found for summer at both sites.

3.3 Conditions aAffecting NPF eEvents

The average and NPF event days’ conditions are presented in tables S2 and S3 (for meteorological conditions and atmospheric composition respectively). A number of variables present consistent behaviour on NPF days. For all the sites in this study the solar radiation intensity was higher on NPF days compared to the average conditions, while the relative humidity was lower. Additionally, all the chemical compounds with available data present either lower or similar concentrations. This is consistent even for the chemical compounds which are associated with the NPF process (such as the SO_2). This probably pointsindicates that they are in sufficient concentrations for not being a limiting factor in the occurrence of the events, while higher concentrations are associated with increased pollution conditions which may suppress their occurrence. The exceptions found are SPARU and GRERU for NO_2 and FINRU for SO_2. In these sites the concentrations of these gaseous components are very low in general (being rural background sites) and were found to be only marginally higher on NPF event days. These differences pointindicate that the variability of these compounds is not playing a significant role in the occurrence of the events and thus should not be considered as an important factor. The ozone concentration though, was found to be consistently higher on event days compared to the average conditions at all sites regardless of their geographical location and type. As the ozone concentration variability is directly associated with the solar radiation intensity, it is unknown whether it plays a direct role in the occurrence of the events or it is the result of its covariance with the solar radiation intensity.

Following that, differences were found in the variability of some of the meteorological conditions, as well as local conditions (either meteorological or specific pollution sources), which played a significant role in the occurrence and the metrics of NPF events across the sites of this study. These will be further ~~explained~~explored in the following sections.

3.3.1 Denmark

The meteorological conditions that prevailed on NPF event days followed the general trend mentioned earlier, while wind speed and temperature were higher than average (consistently at all sites, meteorological condition~~s’~~ variability was significant for all ($p < 0.001$) ~~but~~except the wind speed). As meteorological data 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 are 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 (Table S7), probably being of marine origin as elemental concentrations showed an increased presence of Na, Cl and Mg (results not included). The wind directions with the highest probability for NPF events presented 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 to 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 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 plant 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~~at both, the occurrence and development of NPF events at DENRO are more pronounced in the data, due to the higher concentrations of condensable compounds.

3.3.2 Germany

Compared to the average conditions, a higher temperature was found on NPF event days, while wind speed was lower at all German sites. The condensation sink was also higher on event days compared to the average, though this may be the result of the high formation rates found for the German sites. The wind profile 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 90º – 270º and although the H/W ratio (surrounding building~~s~~’ height to width ratio) is not high, the effect of a street canyon vortex is observed (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 growth rate are also found for wind directions from the easterly sector, although these are not very frequent for this site. 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, 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 $\mu g\ m^{-3}$ 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.

3.3.3 Finland

At the background sites in Finland, temperature was lower on NPF event days compared to the average conditions, whereas it was 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 explains the lower temperatures found with NPF events at this site. 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 profile for NPF events at FINRO also favours northerly winds with an almost complete absence of NPF in 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. 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 and the limited oxidation processes due to lower incoming solar radiation) 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 to an extent 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~~with ~~released in~~emissions to 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 site 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 ~~rarely~~occasionally found at FINUB. Particle growth at late hours is not a unique feature for the Finnish sites, 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.

3.3.4 Spain

The atmospheric conditions favouring NPF events at both sites are similar to most other sites, though with lower wind speed on event days compared to the average conditions ($p < 0.001$ at SPAUB). The wind profile 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. These incoming wind directions originate from a rather clean area with low concentrations of pollutants and condensation sink. 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.

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 direction profile at SPARU appears to have a daily trend, with almost exclusively stronger southeasterly winds at about midday (Figure S3), ~~which might be~~probably due to a local mesoscale circulation caused by~~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 a higher condensation sink (being affected by the city of Barcelona), ~~which~~and almost consistently bring larger particles at the site during the midday period. 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~~with 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.

3.3.5 Greece

Temperature and wind speed are found to be lower on NPF event days at the Greek sites, though the differences are minimal and are associated with the seasonal variability of the events. The wind rose in 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 a northwesterly wind directions. This may be due to them being associated with the highest solar radiation (probably the result of seasonal and diurnal variation), 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 profile is mainly westerly, and though it coincides with the most important source of pollutants in the area, the city of Herakleio, its effect while observable is not significant due to the topography in the area. The wind profile 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. For the 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 one 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 mainland Crete) 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~~with 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 the Finokalia site, 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 profile, 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.

3.4 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 the present study, NPF events that took place on the same day at both background sites (urban background and rural) are considered as regional and their conditions are studied (Table S8). 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 8 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.

In Denmark, 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 dependence 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. These cleaner atmospheric conditions are also confirmed by the lower CS ~~on~~associated with region-wide events, which is probably one of the most important factors in the occurrence of these large-scale events. 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 local 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).

In Germany, 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).

In Finland, 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 compared to the averages on total 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, in Spain 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 profile 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.

3.5 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 9. 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 greater than 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 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 an additional 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.

~~3.1 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 nine~~

~~year period of this study (2008 – 2017). For the DENRO and DENRU sites the seasonal variation favoured summer, while at DENUB a higher frequency of events was found for spring (Figure 2). The growth rate was found to be higher at the DENRO site at 4.45±1.87 nm h^{-1} and it was similar for the other two sites (3.19±1.43 for DENRU and 3.19±1.45 for DENUB) nm h^{-1} (Figure 3), though the peak was found in different seasons (Figure 5), 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 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 4). 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 6), favoured by different seasons at each site (summer at DENRU, spring at DENUB though with minimum differences and autumn at DENRO) (Figure 7).~~

~~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 elements like Na, Cl and Mg – 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 are 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 (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 to 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 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 Germany

A higher frequency of NPF events was found for each type of site in Germany compared to the other countries in this study, for the three year period of this study (2008 – 2011). 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 2). 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 having 4.34±1.73 nm h^{-1}, GERUB 4.24±1.69 nm h^{-1} and GERRO 5.17±2.20 nm h^{-1} (Figure 3). 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 5). 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 4), 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. J_{10} at the German sites was also the highest among the sites of this study (Figure 6), increasing from the GERRU to GERRO. It was found to be higher in summer for the background sites and in autumn for GERRO (Figure 7).

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 profile 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 90º – 270º and although the H/W ratio (surrounding buildings' height to width ratio) is not high, the effect of a street canyon vortex is observed (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 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, 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 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 2). The frequency of NPF events at FINRU was higher (8.66%) for the years with available data (2008 – 2011 & 2015 – 2018), while being less at the urban sites (4.97% at FINUB and 5.20% at FINRO) for the three years with available data for each (2008 – 2011 & 2015 - 2018 for FINUB and 2015 – 2018 for 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 3 and 5). Strong within-week variation favouring weekends is found for the roadside, while no clear variation was found for the other two sites (Figure 4). 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 6 and 7).

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 profile 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 and the limited oxidation processes due to lower incoming solar radiation) 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 rarely found at FINUB. 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°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 Spain

For Spain, data was available for an urban and a rural background site in the greater area of Barcelona for the period 2012 - 2015. 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 2). 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 5). 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 6 and 7). For both sites a higher probability for events was found on weekends compared to weekdays, though this trend is stronger at SPAUB (Figure 4). 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 increased growth rate during weekdays may be associated with the increased presence of condensable species due to increased anthropogenic activities, the increased formation rate might be affected by the increased emissions during these days.

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 profile 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 direction profile at SPARU appears to have a daily trend, 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 Greece

Data are available for two background sites in Greece (2012 – 2018 for GRERU and 2015 – 2018 for GREUB), 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 2). 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 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 3 and 5), 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 7).~~

~~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 in 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), 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 profile is mainly westerly, and though it coincides with the most important source of pollutants in the area, the city of Herakleio, its effect while observable is not significant due to the topography in the area. The wind profile 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 the 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 one 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 mainland Crete) 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 the Finokalia site, 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 profile, 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 the present study, NPF events that took place on the same day at both background sites (urban background and rural) 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 8 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.

In Denmark, 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).

In Germany, 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).

In Finland, 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, in Spain 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 profile 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 9. 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 greater than 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 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 ~~f~~Frequency and ~~s~~Seasonality of the ~~e~~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 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. Finally, it should be noted that no clear interannual trend was found in the frequency of the events for any site, even for those with longer datasets.

4.2 Variability and ~~s~~Seasonality of the ~~f~~Formation and ~~g~~Growth ~~r~~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 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 compared to the 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 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. Finally, as with the frequency of the events, no significant interannual trend was found in the variation of the formation or the growth rate across the sites studied.

4.3 Effect of ~~l~~Local ~~c~~Conditions in the ~~o~~Occurrence and ~~d~~Development of NPF ~~e~~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 composition 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.

54. CONCLUSIONS

There are different ways to assess the occurrence 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 significant sources of pollution in close proximity, present more complex trends as the NPF occurrence depends less ~~to~~upon favourable meteorological conditions and more ~~to~~upon the local atmospheric conditions, including composition. As NPF event~~s'~~ occurrence is based on the balance between the rapid growth of the newly formed particles and their loss from processes, such as the evaporation or coagulation of the particles, 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 environment~~s~~ 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. NPF is a complex ~~mechanism~~process, affected by numerous variables, making it extremely difficult to predict any of its metrics without considering ~~all of them~~multiple factors. 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~~are needed to generate greater clarity and predictive capability.

There are different ways to assess occurrences of new particle formation (NPF) events. In this study, the rate of NPF events, the growth rate of the particles and the frequency of NPF events, associated with secondary formation of particles and not primary emissions, at 13 sites from five countries in Europe are considered. 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 associated with anthropogenic emissions, that promotes NPF events more, even compared to the 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. 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 at all types of site in Europe, especially for region-wide events (high solar radiation and low relative humidity and pollutants concentrations). 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, while more polluted tend to have higher growth rates (no consistent trend was found for the formation rate).

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.

Both the growth rate of the newly formed particles and the formation rate of the particles were found to be higher at all the roadsides compared to their respective rural and urban background. While the more polluted urban environment is a limiting factor in the occurrence of NPF events, their development as represented by the number of particles formed and the speed at which they grow is enhanced by the urban environment (which seems to be a prerequisite for NPF events within the more polluted environment), as more condensable compounds, deriving from anthropogenic activities, are available. 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) was 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 though, 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.

While both the formation and growth rates are greater at the roadsides, the relative effect of NPF events on the ultrafine particle concentrations is consistently a lot greater at the rural sites, where in most cases NPF more than doubles (up to 400%) their particle number concentration on the days they occur, as well as in the urban background sites where a substantial increase (up to 240%) is also observed. The effect is considerable at roadside sites as well, increasing the number of ultrafine particles up to 126% on event days (which might be higher as the occurrence of NPF events at roadsides is harder to detect), which is limited compared to background sites due to the stronger effect of local sources influencing the particle number concentration.

~~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 with 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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TABLE LEGENDS:

Table 1: Location and data availability (seasonal data availability is found in Table S4) of the sites in the present ~~study (RU denotes rural site, UB is urban background and RO is roadside)~~. In the studies referenced an~~d~~ extended description of the sites can be found.

FIGURE LEGENDS

Figure 1: Map of the areas of study.

Figure 2: Frequency (a) and seasonal variation (b) of New Particle Formation events (Winter – DJF; Spring – MAM; Summer – JJA; Autumn – SON). ~~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 3: 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.~~Growth rate of particles up to 30 nm (with standard errors of the mean) on New Particle Formation events at all sites.~~

Figure 4: Growth rate of particles up to 30 nm (with standard deviations) during New Particle Formation events at all sites.

~~Ratio of New Particle Formation events probability between weekends to weekdays. The greatest the ratio the more probable is for an event to take place during weekends compared to weekdays.~~

Figure 5: Seasonal variation of growth rate of particles up to 30 nm on New Particle Formation at (a) the rural background, (b) urban background and (c) roadside~~all~~ sites.

Figure 6: Formation rate of 10 nm particles (J_{10}) (with standard ~~errors of the mean~~deviations) from New Particle Formation at all~~all~~ sites.

Figure 7: Seasonal variation of formation rate of 10 nm particles (J_{10}) (with standard deviations) from New Particle Formation events at (a) the rural background, (b) urban background and (c) roadside~~all~~ sites.

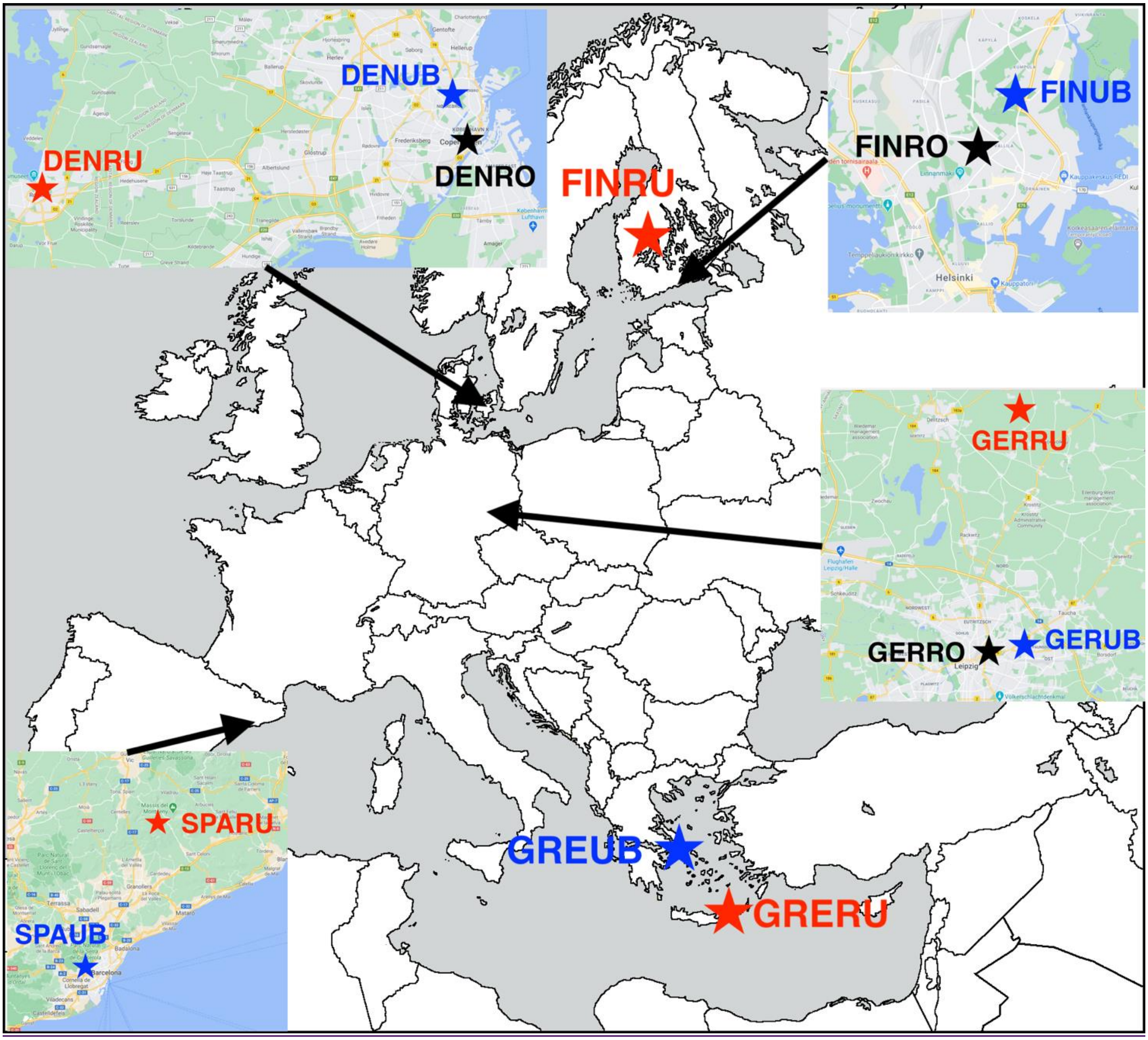
Figure 8: (a) Number of region-wide New Particle Formation events per season (top panel) and (b) fraction of region-wide events to total New Particle Formation events per season for each site. Region-wide events are considered those that occur on the same day on both background sites (Rural and Urban background).

Figure 9: (a) 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 (b) NSF_{GEN} (average annual relative increase of ultrafine particles due to New Particle Formation events) at all sites.

Table 1: Location and data availability (seasonal data availability is found in Table S4) of the sites in the present study (RU denotes rural site, UB is urban background and RO is roadside). In the studies referenced an extended description of the sites can be found.

Site	Location	Available data	Meteorological data location	Data availability	Reference
DENRU	Lille Valby, 25 km W of Copenhagen, (55º 41' 41" N; 12º 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, 65.4% 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 (55º 42' 1" N; 12º 33' 41" E)	DMPS and CPC (5.8 - 700 nm, 59.0% availability), NO, NO_x, O_3, minerals, EC	On site	2008 – 2017	Wang et al., 2010
DENRO	H.C. Andersens Boulevard, Copenhagen, Denmark (55º 40' 28" N; 12º 34' 16" E)	DMPS and CPC (5.8 - 700 nm, 65.0% 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 (51º 31' 31.85" N; 12º 26' 40.30" E)	TDMPS with CPC (4.8 - 800 nm, 87.1% 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 (51º 21' 9.1" N; 12º 26' 5.1" E)	TDMPS with CPC (3 - 800 nm,88.0% availability)	On site	2008 – 2011	Birmili et al., 2016
GERRO	Eisenbahnstraße, Leipzig, Germany (51º 20' 43.80" N; 12º 24' 28.35" E)	TDMPS with CPC (4 - 800 nm, 64.4% availability)	Tropos station	2008 – 2011	Birmili et al., 2016
FINRU	Hyytiälä, 250 km N of Helsinki, Finland (61º 50' 50.70" N; 24º 17' 41.20" E)	TDMPS with CPC (3 – 1000 nm, 98.7% 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 (60º 12' 10.52" N; 24º 57' 40.20" E)	TDMPS with CPC (3.4 - 1000 nm, 94.0% availability)	On site	2008 – 2011 & 2015 – 2018	Järvi et al., 2009
FINRO	Mäkelänkatu street, Helsinki, Finland (60º 11' 47.57" N; 24º 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 (41º 46' 45" N; 2º 21' 29" E)	SMPS (9 – 856 nm, 47.7% 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 (41º 23' 14" N; 2º 6' 56" E)	SMPS (10.9 – 478 nm, 64.2% availability),	On site	2012 – 2015	Dall'Osto et al., 2012

		NO, NO_2, SO_2, O_3, CO, BC, OM, SO_4^{2-}, $PM_{2.5}$, PM_{10}			
GRERU	Finokalia, 70 km E of Heraklion, Greece (35º 20' 16.8" N; 25º 40' 8.4" E)	SMPS (8.77 - 849 nm, 92.4% 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 (37º 59' 41.96" N; 23º 48' 57.56" E)	SMPS (10 – 550 nm, 77.2% availability)	On site	2015 – 2018	Vassilakos et al., 2005


DENUB
DENRO
DENRU
FINRU
FINUB
FINRO
GERRU
GERUB
GERRO
SPARU
SPAUB
GREUB
GRERU

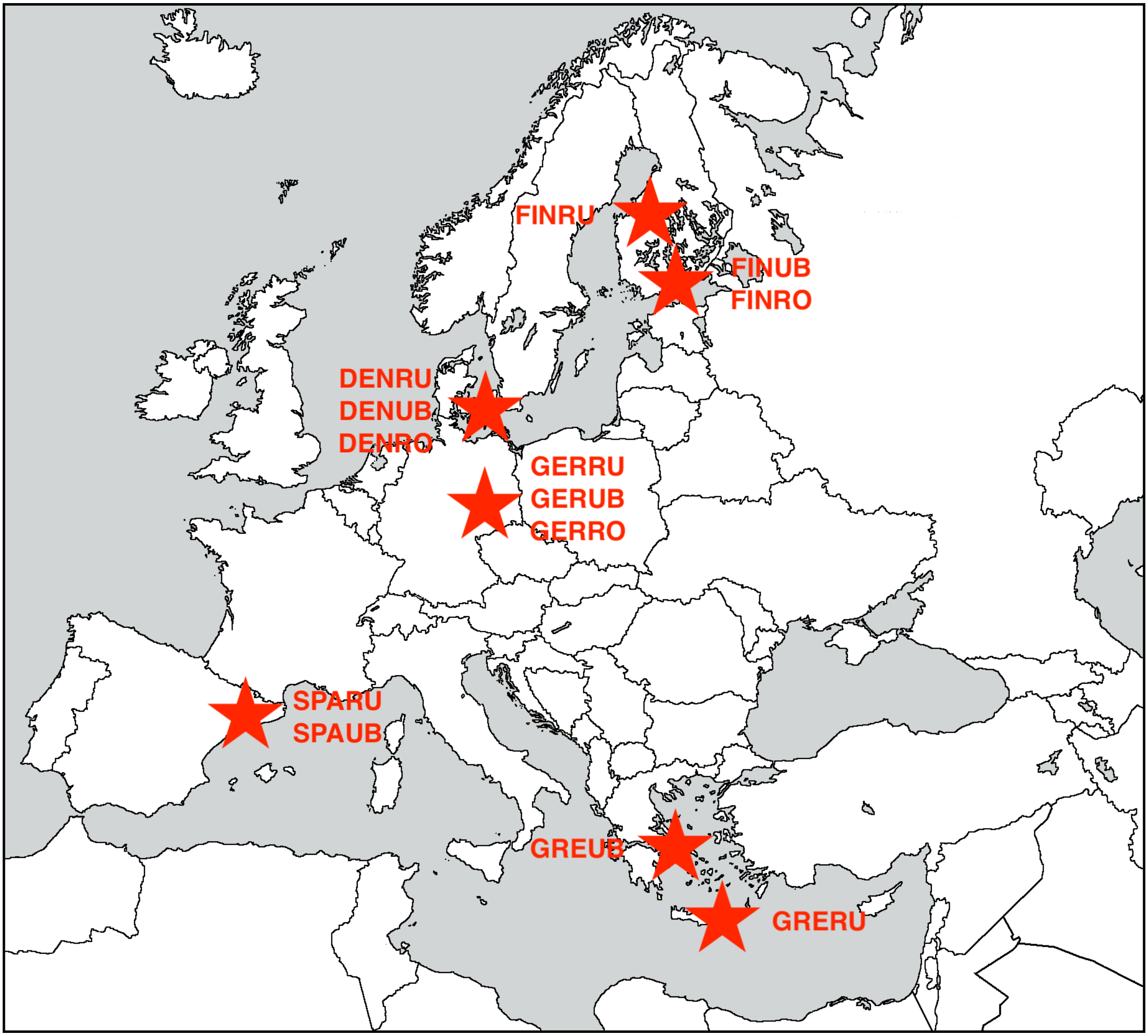
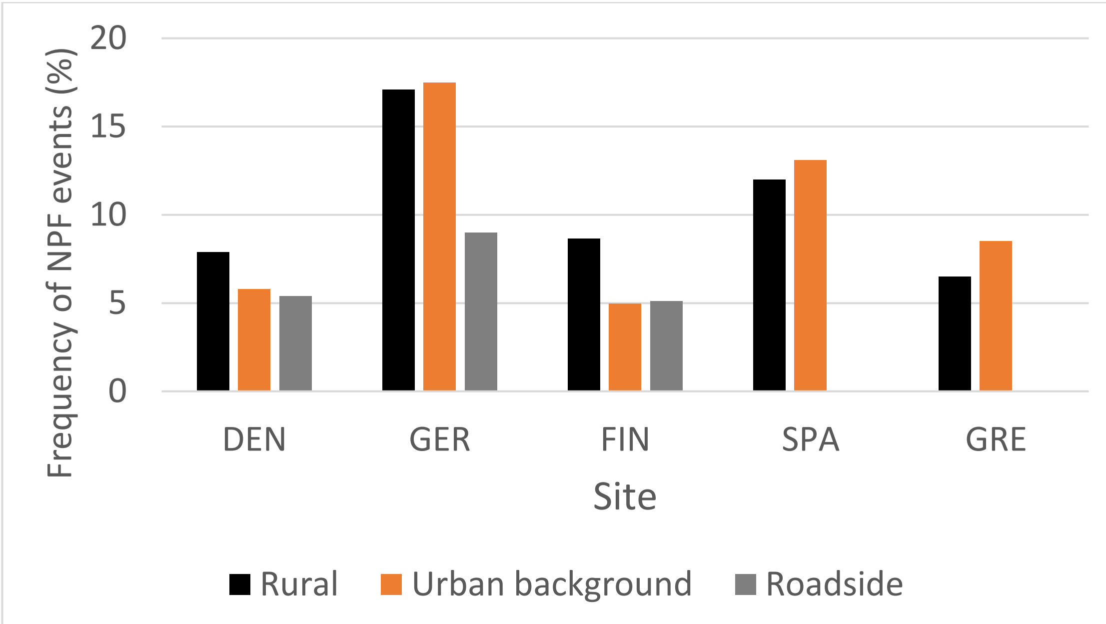
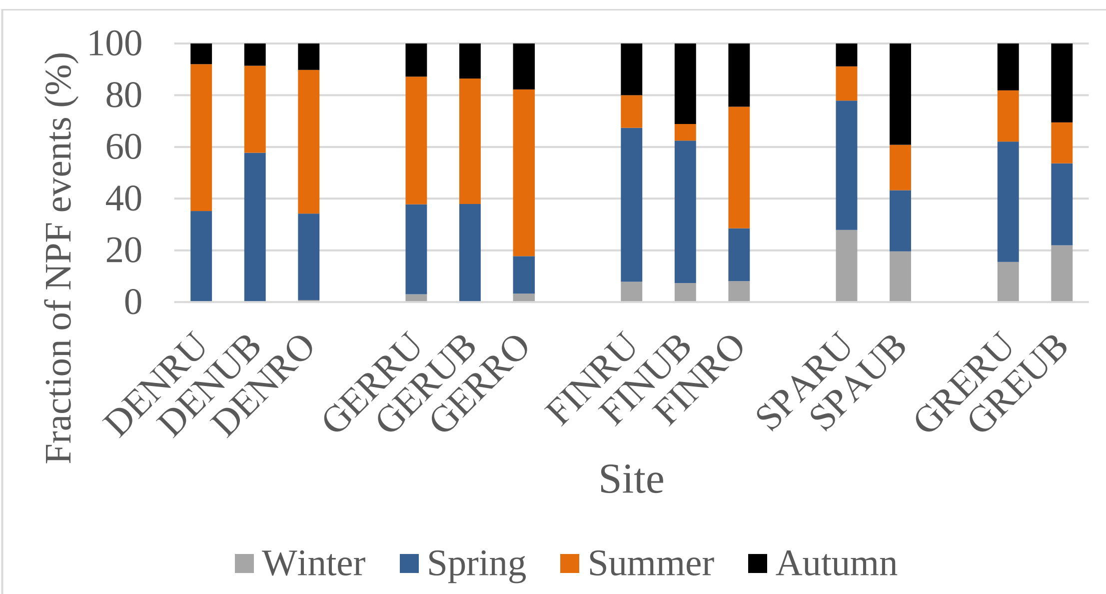


Figure 1: Map of the areas of study.



(a)



(b)

Figure 2: Frequency (a) and seasonal variation (b) of New Particle Formation events (Winter – DJF; Spring – MAM; Summer – JJA; Autumn – SON).

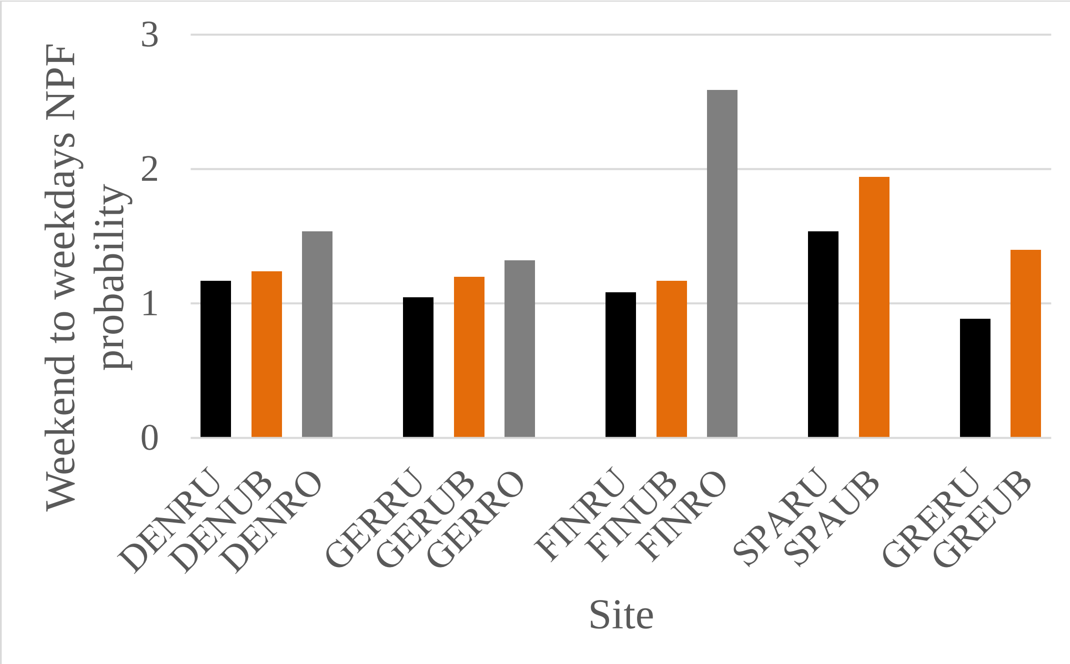


Figure 3: Ratio of New Particle Formation event probability between weekends to weekdays. The greater the ratio the more probable it is for an event to take place during weekends compared to weekdays.~~For site naming first three letters refer to the country (DEN = Denmark, GER = Germany, FIN = Finland, SPA = Spain, GRE = Greece) while next two to the type of the site (RU = Rural background, UB = Urban background, RO = Roadside)~~

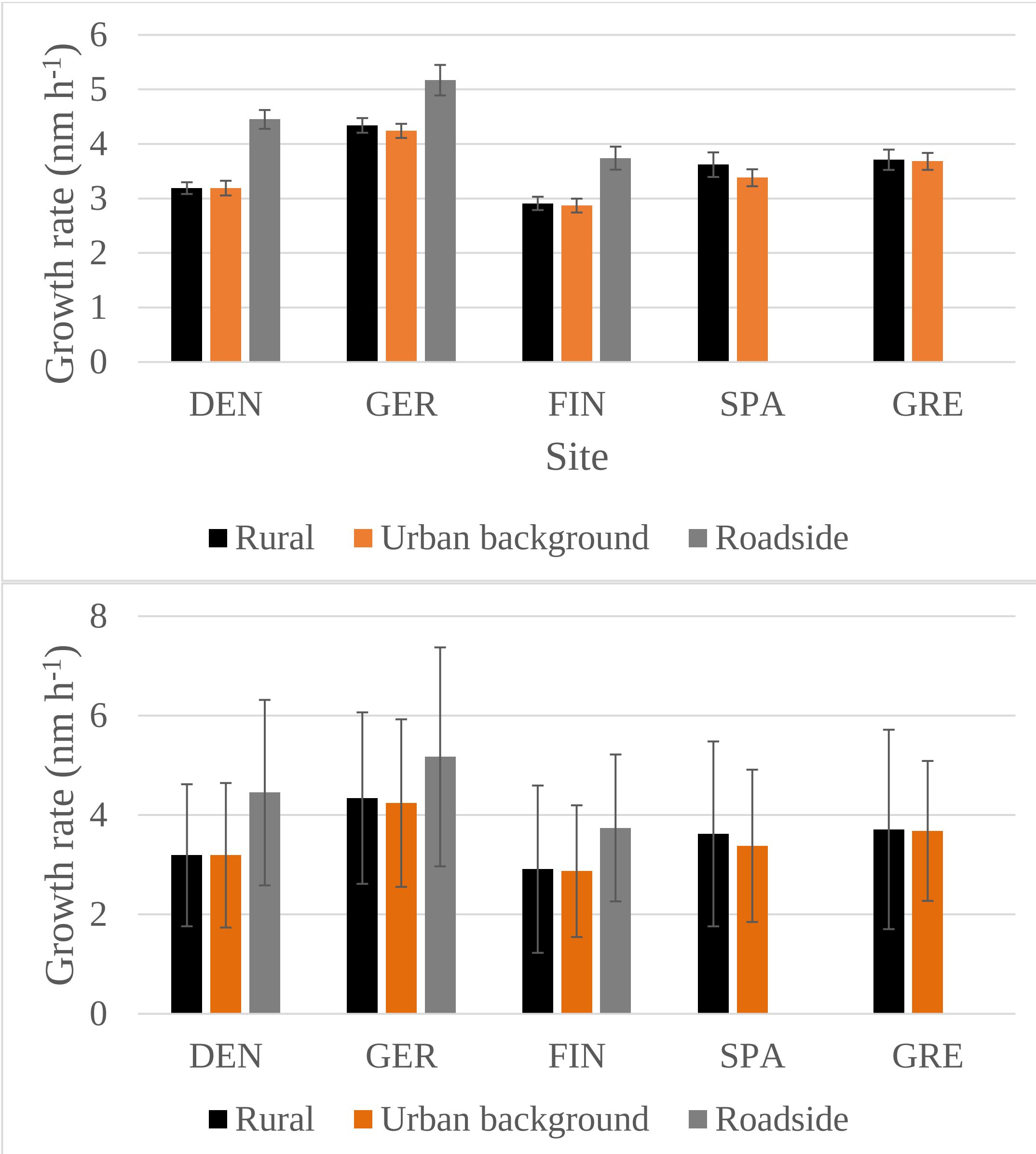
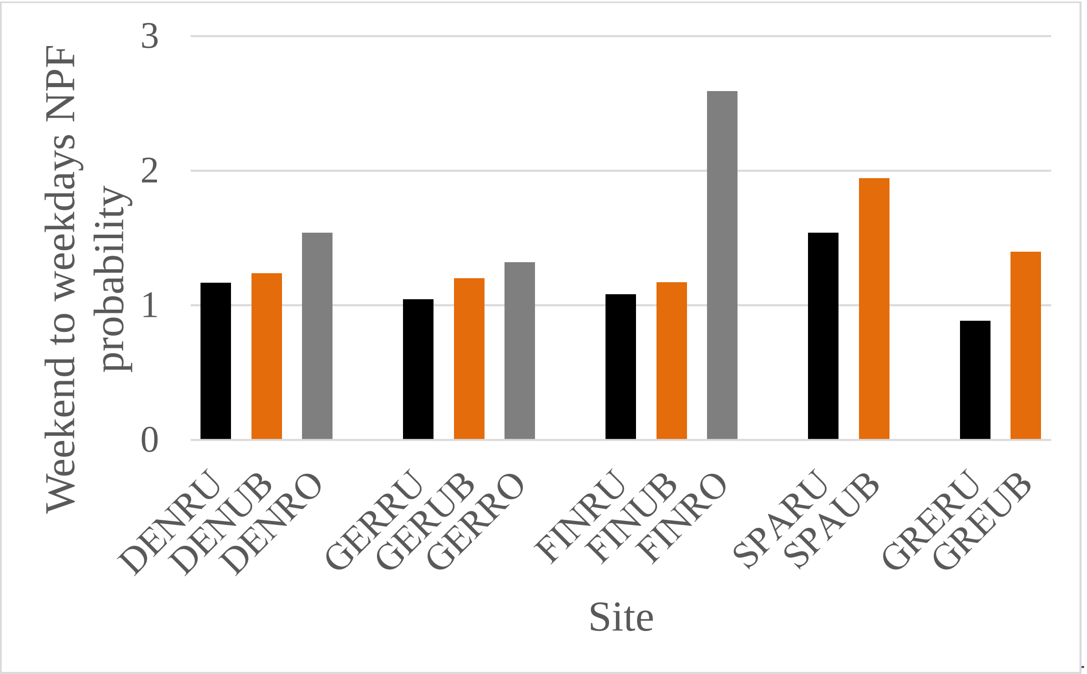
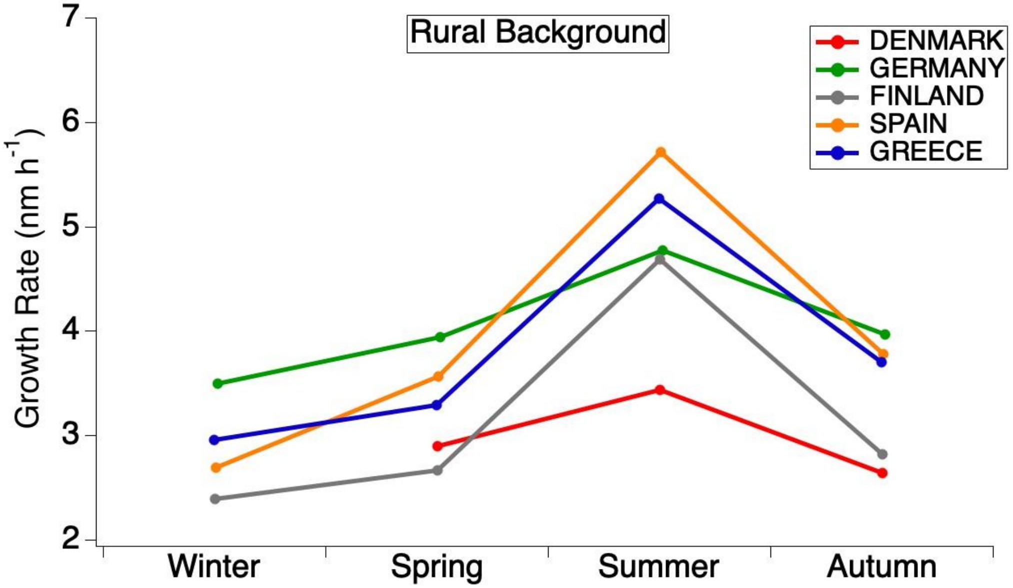
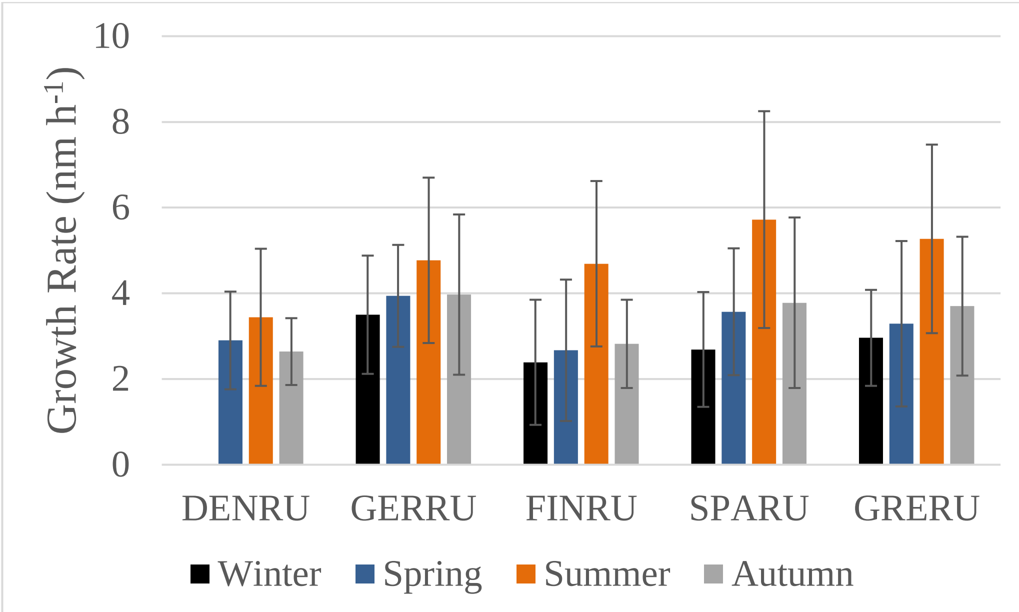


Figure 43: Growth rate of particles up to 30 nm (with standard deviations) during New Particle Formation events at all sites.

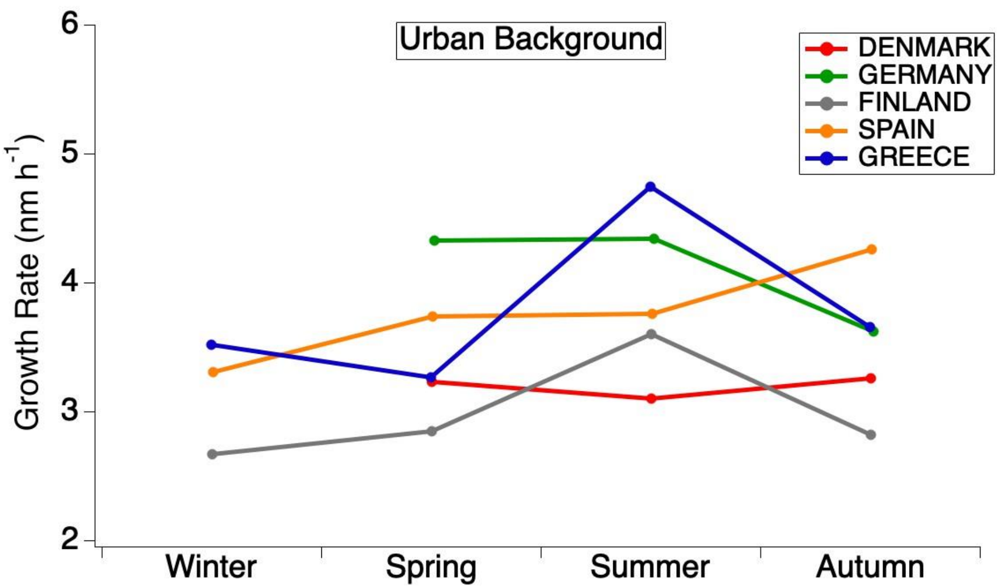


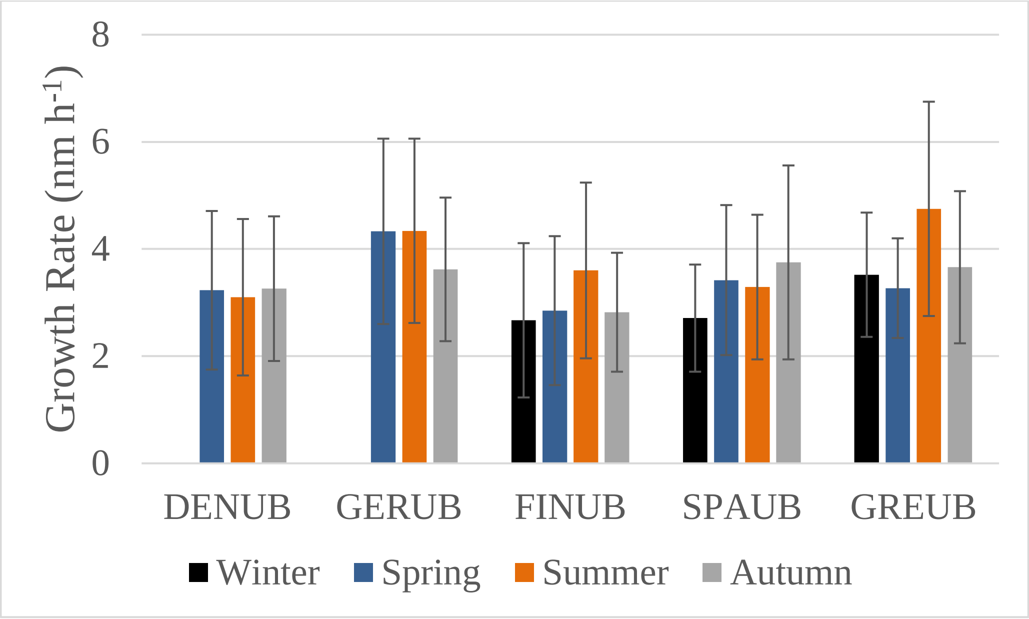
~~**Figure 4:** Ratio of New Particle Formation event probability between weekends to weekdays. The greater the ratio the more probable it is for an event to take place during weekends compared to weekdays.~~


Rural Background
DENMARK
GERMANY
FINLAND
SPAIN
GREECE
Growth Rate (nm h^{-1})
7
6
5
4
3
2
Winter
Spring
Summer
Autumn

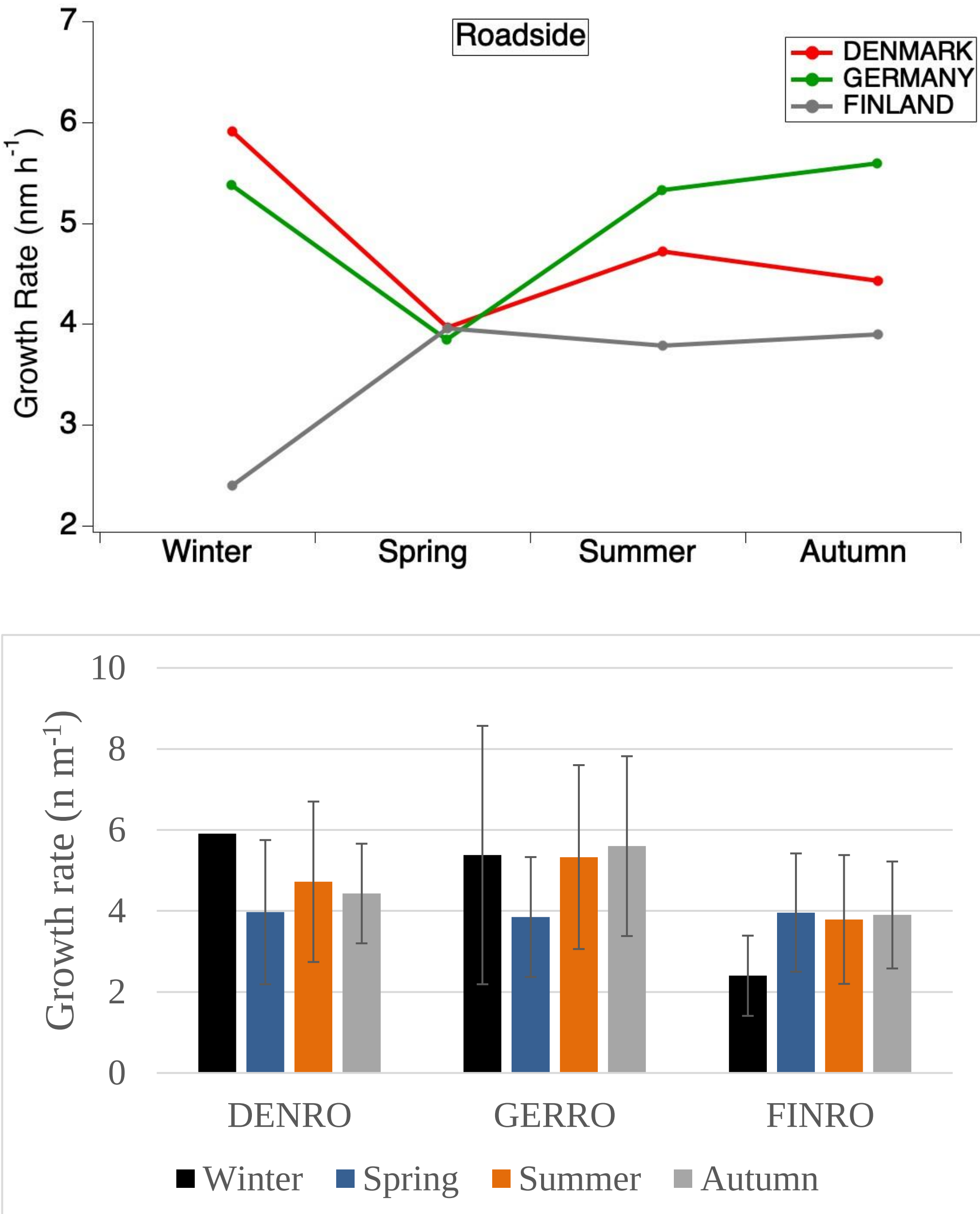

10
8
6
4
2
0
Growth Rate (nm h-1)
DENRU
GERRU
FINRU
SPARU
GRERU
Winter
Spring
Summer
Autumn

(a)


Urban Background
DENMARK
GERMANY
FINLAND
SPAIN
GREECE
6
5
4
3
2
Growth Rate (nm h-1)
Winter
Spring
Summer
Autumn


Growth Rate (nm h-1)
0
2
4
6
8
DENUB
GERUB
FINUB
SPAUB
GREUB
Winter
Spring
Summer
Autumn

(b)



(c)

Figure 5: Seasonal variation of growth rate of particles up to 30 nm on New Particle Formation at (a) the rural background, (b) urban background and (c) roadside~~all~~ sites.

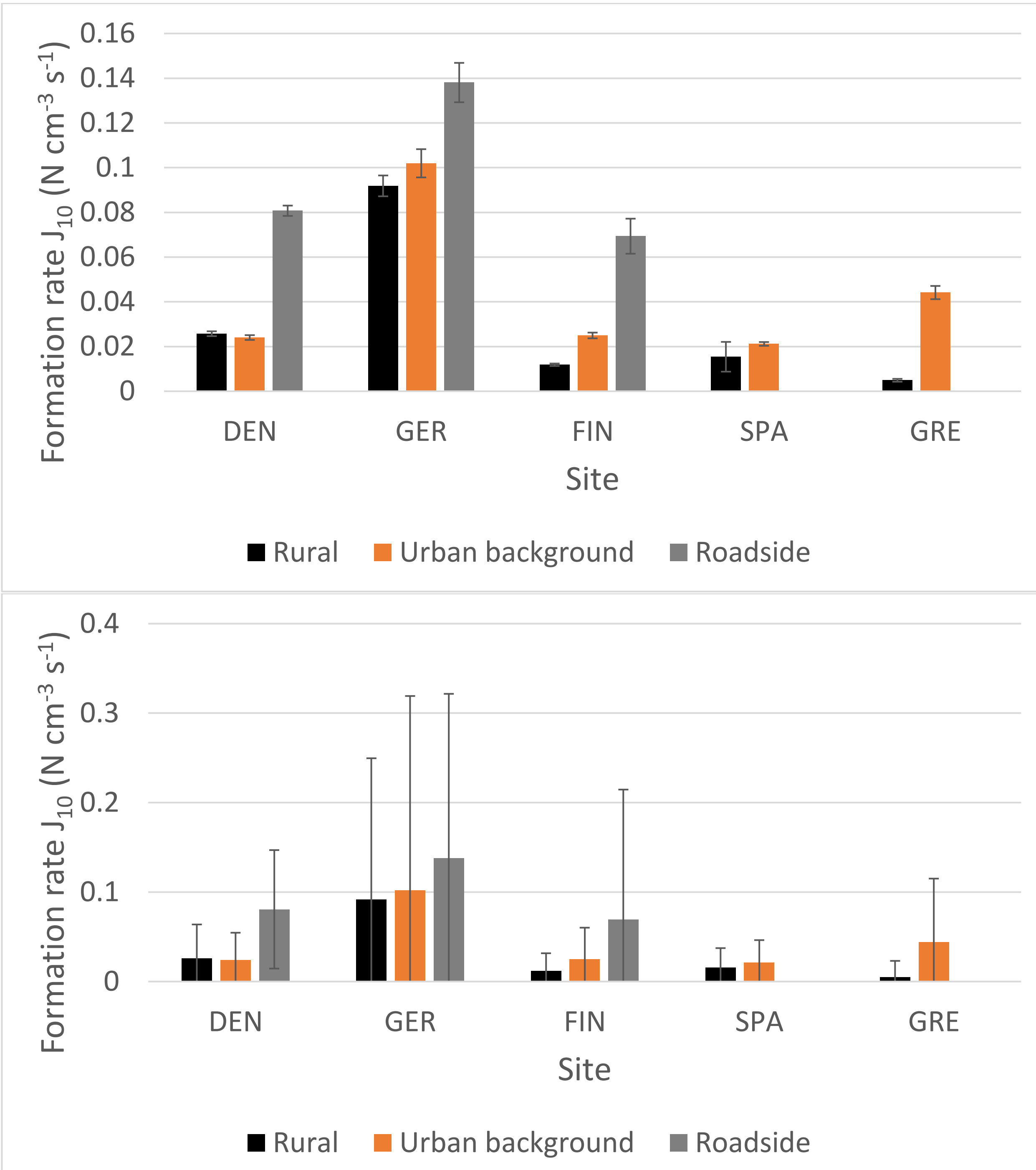
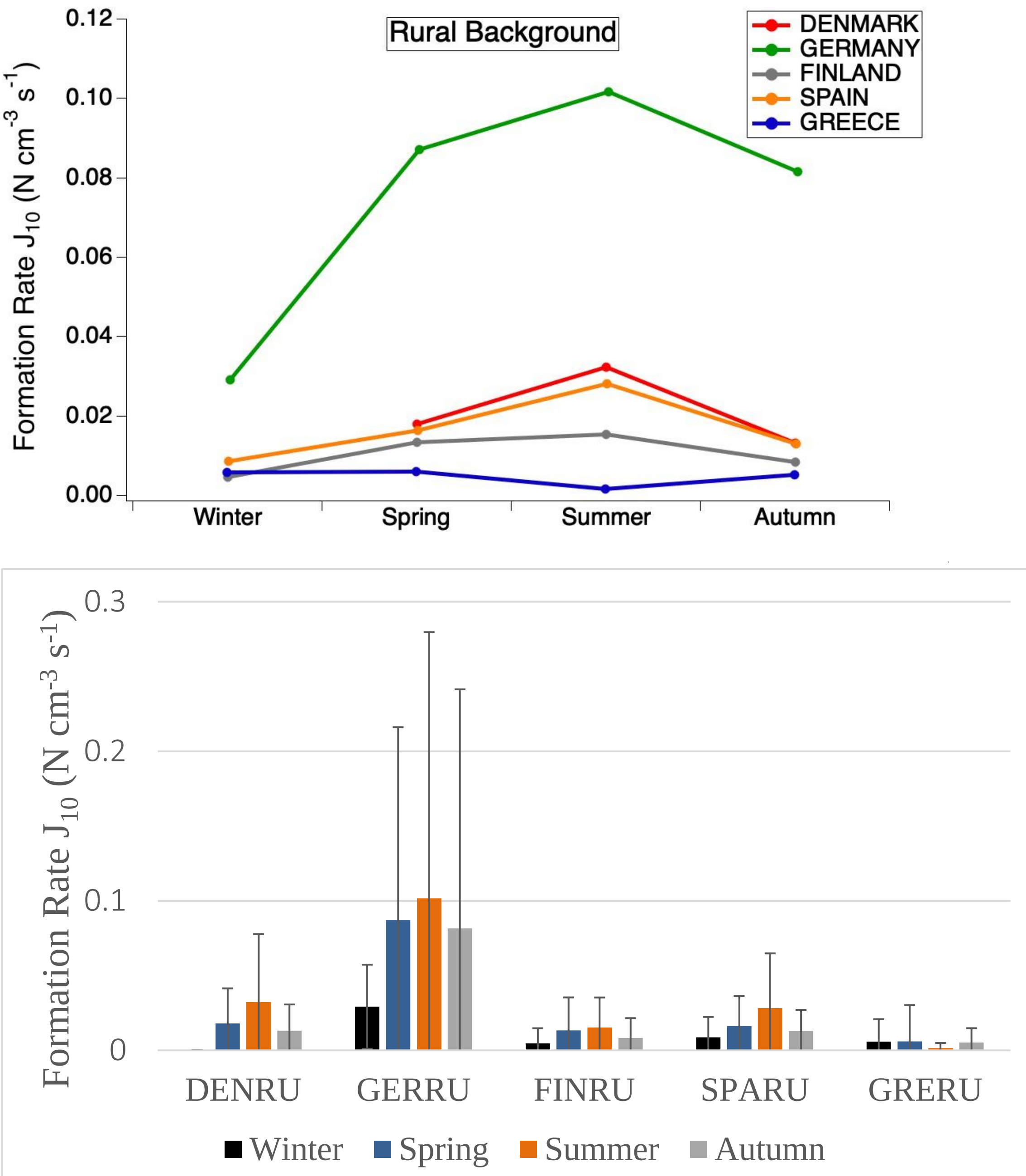
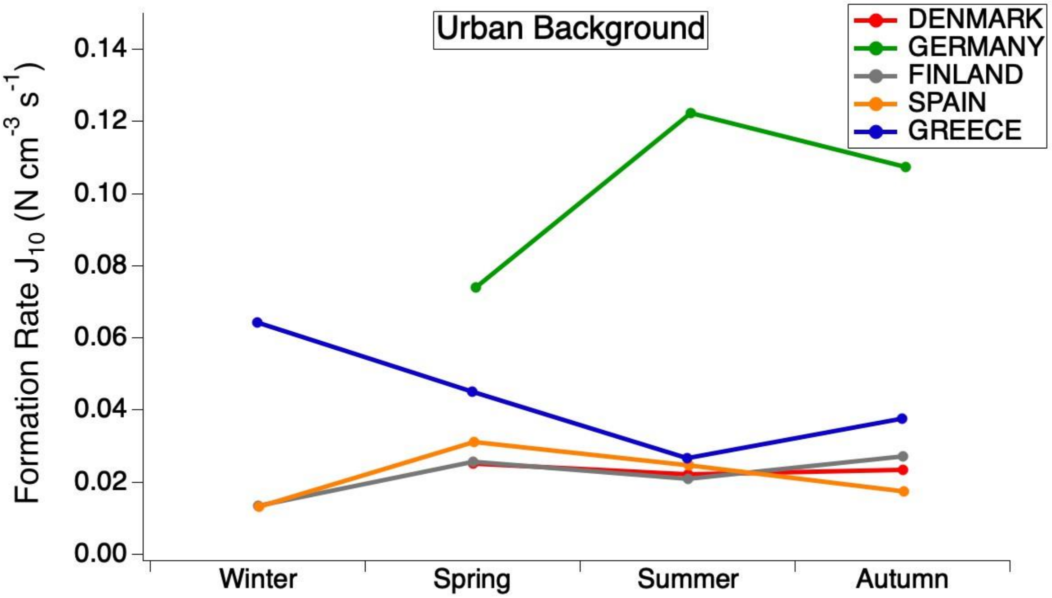
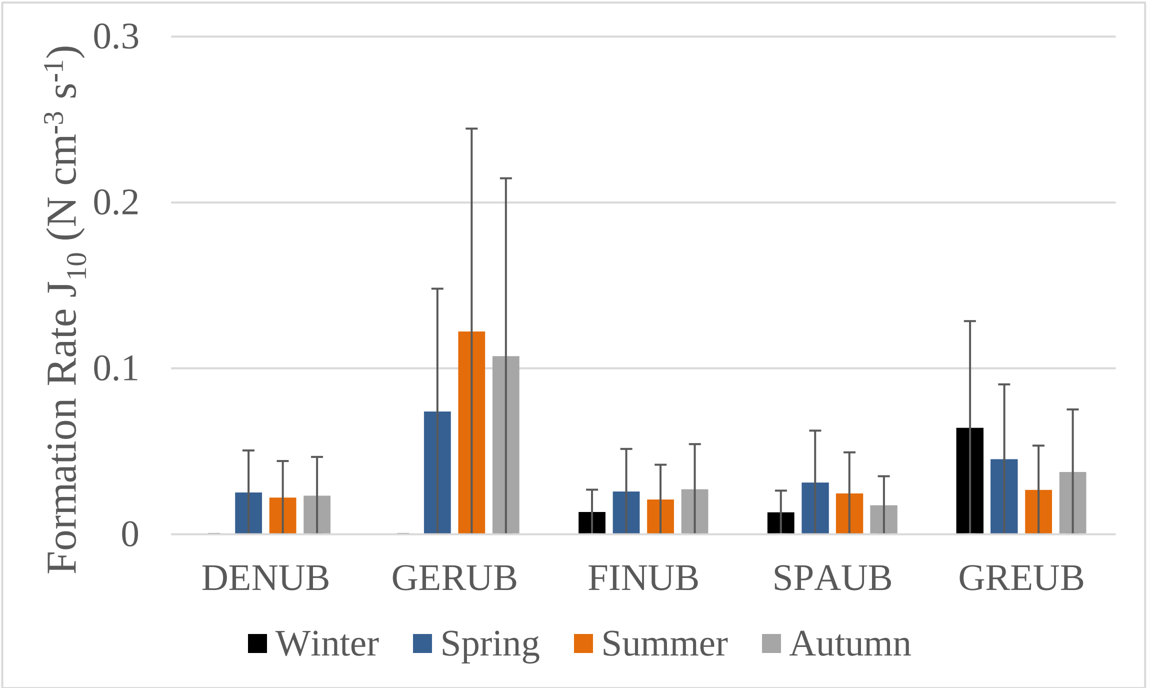

Formation rate J10 (N cm-3 s-1)
0.16
0.14
0.12
0.1
0.08
0.06
0.04
0.02
0
DEN
GER
FIN
SPA
GRE
Site
Rural
Urban background
Roadside
Formation rate J10 (N cm-3 s-1)
0.4
0.3
0.2
0.1
0
DEN
GER
FIN
SPA
GRE
Site
Rural
Urban background
Roadside

Figure 6: Formation rate of 10 nm particles (J_{10}) (with standard ~~errors of the mean~~deviations) during New Particle Formation events at all sites.

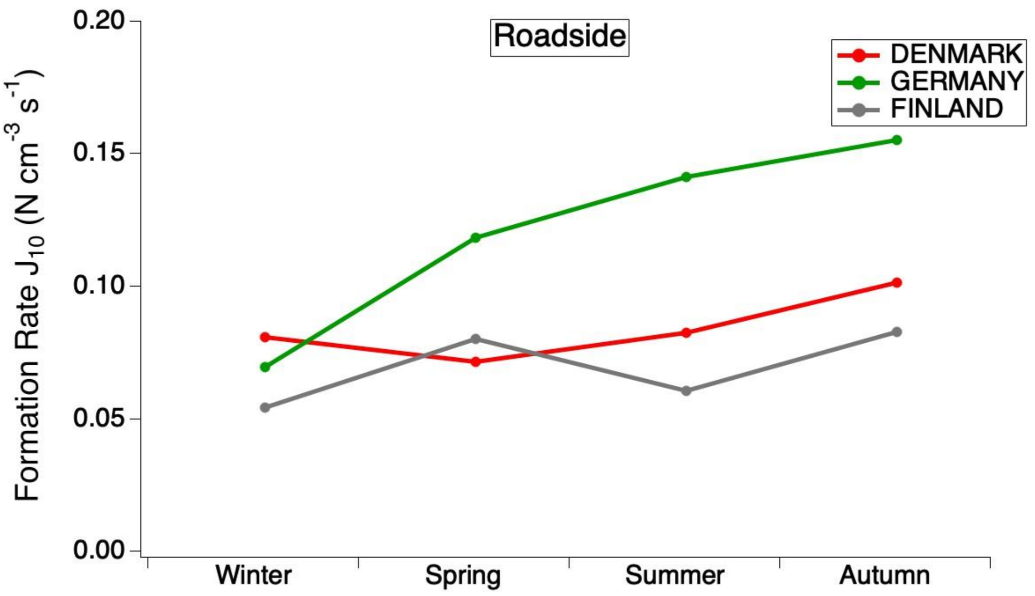

Rural Background
DENMARK
GERMANY
FINLAND
SPAIN
GREECE
Formation Rate J_{10} (N cm^{-3} s^{-1})
0.12
0.10
0.08
0.06
0.04
0.02
0.00
Winter
Spring
Summer
Autumn
Formation Rate J_{10} (N cm^{-3} s^{-1})
0.3
0.2
0.1
0
DENRU
GERRU
FINRU
SPARU
GRERU
Winter
Spring
Summer
Autumn

(a)

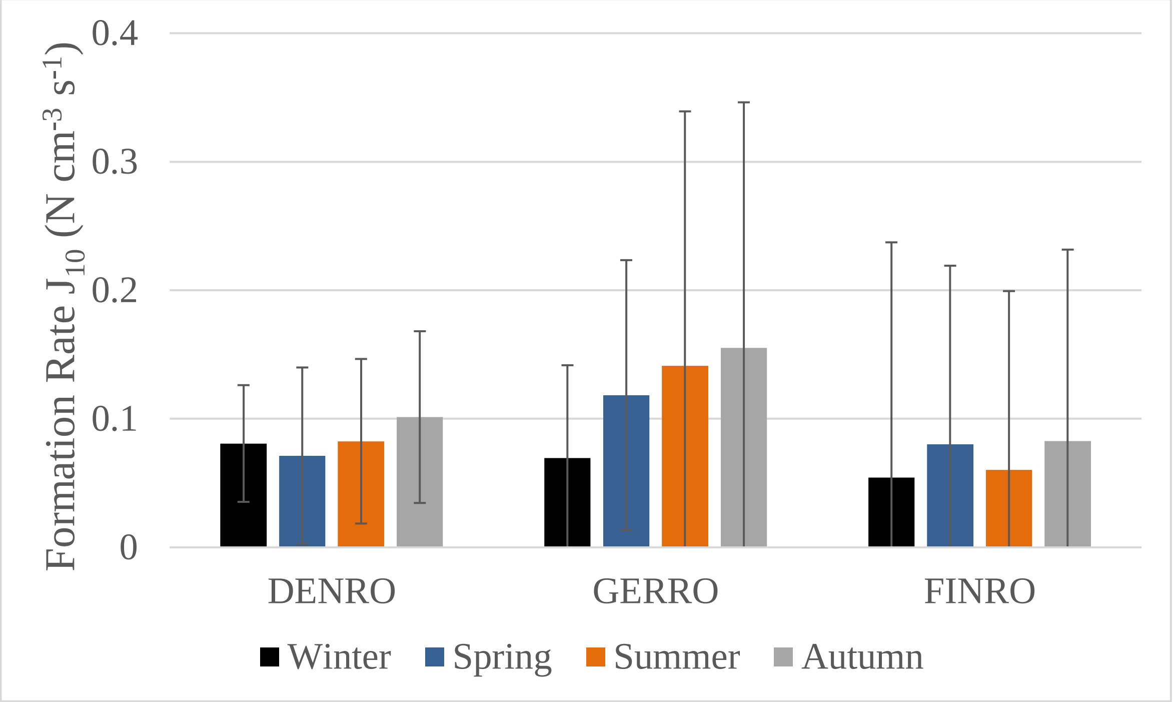

Urban Background
DENMARK
GERMANY
FINLAND
SPAIN
GREECE
Formation Rate J_{10} (N cm^{-3} s^{-1})
0.14
0.12
0.10
0.08
0.06
0.04
0.02
0.00
Winter
Spring
Summer
Autumn


Formation Rate J_{10} (N cm^{-3} s^{-1})
0.3
0.2
0.1
0
DENUB
GERUB
FINUB
SPAUB
GREUB
Winter
Spring
Summer
Autumn

(b)



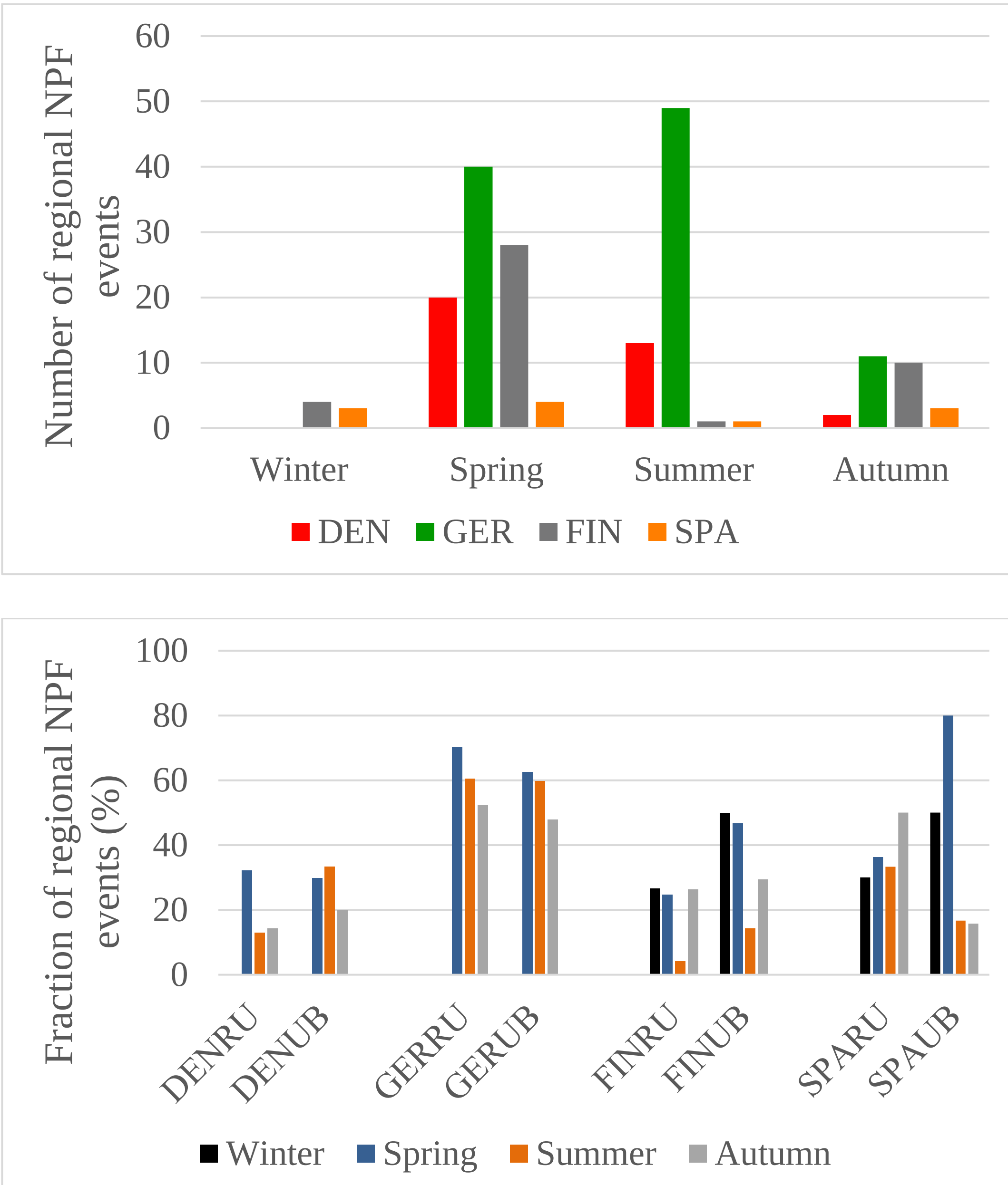
Roadside
DENMARK
GERMANY
FINLAND
Formation Rate J10 (N cm-3 s-1)
0.20
0.15
0.10
0.05
0.00
Winter
Spring
Summer
Autumn



Formation Rate J10 (N cm-3 s-1)
0.4
0.3
0.2
0.1
0
DENRO
GERRO
FINRO
Winter
Spring
Summer
Autumn

(c)

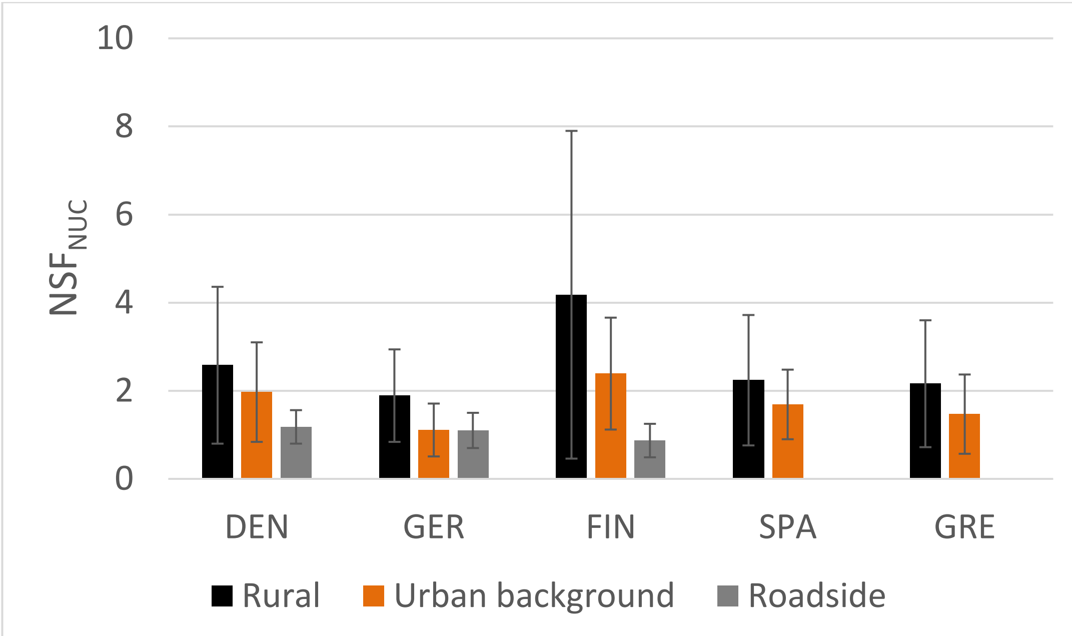
Figure 7: Seasonal variation of formation rate of 10 nm particles (J_{10}) (with standard deviations) from New Particle Formation events at (a) the rural background, (b) urban background and (c) roadside~~all~~ sites.



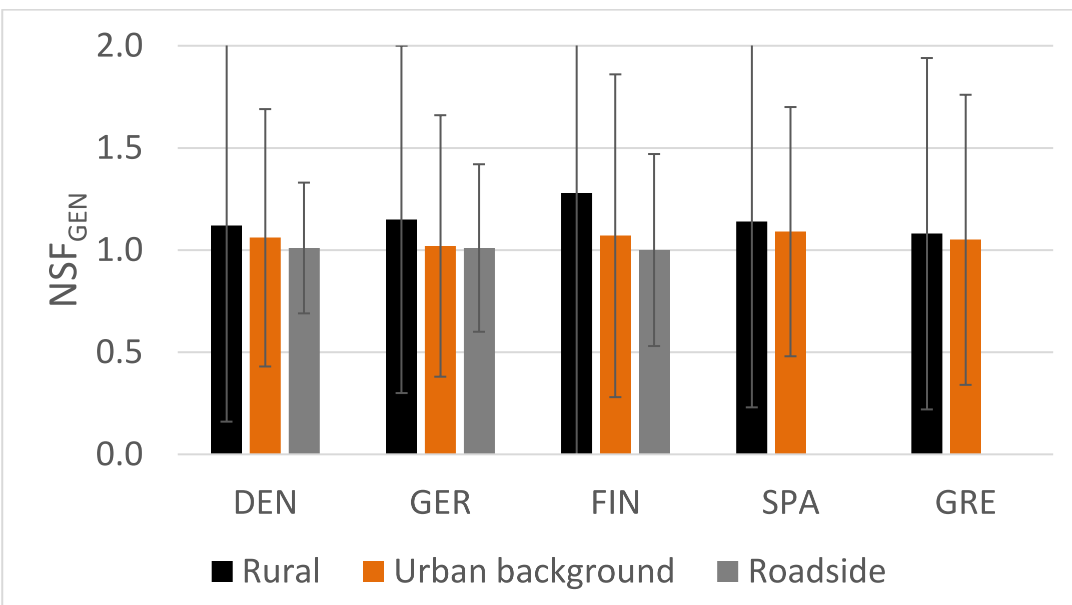
(a)

(b)

Figure 8: (a) Number of region-wide New Particle Formation events per season ~~(top panel)~~ 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)

Figure 9: (a) 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 (b) NSF$_{GEN}$ (average annual relative increase of ultrafine particles due to New Particle Formation events) at all sites.