The Effect of Meteorological Conditions and Atmospheric Composition in the Occurrence and Development of New Particle Formation (NPF) Events in Europe

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ABSTRACT

Although new particle formation (NPF) events have been studied extensively for some decades, the mechanisms that drive their occurrence and development are yet to be fully elucidated. Laboratory studies have done much to elucidate the molecular processes involved in nucleation, but this knowledge has yet to be conclusively linked to NPF events in the atmosphere. There is great difficulty in successful application of the results from laboratory studies to real atmospheric conditions, due to the diversity of atmospheric conditions and observations found, as NPF events occur almost everywhere in the world without always following a clearly defined trend of frequency, seasonality, atmospheric conditions or event development. The present study seeks common features in nucleation events by applying a binned linear regression over an extensive dataset from 16 sites of various types (combined dataset of 85 years from rural and urban backgrounds as well as roadside sites) in Europe. At most sites, a clear positive relation is found between the solar radiation intensity (up to $R^2 = 0.98$), temperature (up to $R^2 = 0.98$) and atmospheric pressure (up to $R^2 = 0.97$) with the probability of NPF events, while relative humidity (RH) presents a negative relation (up to $R^2 = 0.95$) with NPF event probability. Wind speed presents a less consistent relationship which appears to be heavily affected by local conditions. While some meteorological variables (such as the solar radiation intensity and RH) appear to have a crucial effect on the occurrence and characteristics of NPF events, especially at rural sites, it appears that their role becomes less marked when at higher average values.

The analysis of chemical composition data presents interesting results. Concentrations of almost all chemical compounds studied (apart from O_3) and the Condensation Sink (CS) have a negative relationship with NPF event probability, though areas with higher average concentrations of SO_2 had higher NPF event probability. Particulate Organic Carbon (OC), Volatile Organic Compounds (VOCs) and particulate phase sulphate consistently had a positive relation with the growth rate of the newly formed particles. As with some meteorological variables, it appears that at increased concentrations of pollutants or the CS, their influence upon NPF probability is reduced.

1. INTRODUCTION

New Particle Formation (NPF) events are an important source of particles in the atmosphere (Merikanto et al., 2009; Spracklen et al., 2010), which are known to have adverse effects on human health (Schwartz et al., 1996; Politis et al., 2008; Kim, et al., 2015) as well as affecting the optical and physical properties of the atmosphere (Makkonen et al., 2012; Seinfeld and Pandis, 2012). While they occur almost everywhere in the world (Dall'Osto et al., 2018; Kulmala et al., 2017; O'Dowd et al., 2002; Wiedensohler et al., 2019; Chu et al., 2019; Kerminen et al., 2018), with some exceptions mentioned in the literature in forest (Lee et al., 2016; Pillai et al., 2013; Rizzo et al., 2010) or high-elevation sites (Bae et al., 2010; Hallar et al., 2016), great diversity is found in the atmospheric conditions within which they take place. Many studies have been done in a large number of different types of locations (urban, traffic, regional background) around the world and differences were found in both the seasonality and intensity of NPF events. To an extent this variability is due to the mix of conditions that are specific to each location, which blurs the general understanding of the conditions that are favourable for the occurrence of NPF events (Berland et al., 2017; Bousiotis et al., 2020). For example, solar radiation is considered as one of the most important factors in the occurrence of NPF events (Kulmala and Kerminen, 2008; Kürten et al., 2016; Pikridas et al., 2015; Salma et al., 2011), as it is needed for the photochemical reactions that lead to the formation of sulphuric acid (Petäjä et al., 2009; Cheung et al., 2013). Sulphuric acid is considered as the main component of the formation and growth of the initial clusters (Iida et al., 2008; Stolzenburg et al., 2020; Weber et al., 1995). Nevertheless, in many cases NPF events did not occur in the seasons with the highest insolation (Park et al., 2015; Vratolis et al., 2019). Similarly, uncertainty exists over the effect of temperature (Yli-Juuti et al., 2020; Stolzenburg et al., 2018). Higher temperatures are considered favourable for the growth of the newly formed particles as increased concentrations of both Biogenic Volatile Organic Compounds (BVOCs) and Anthropogenic Volatile Organic Compounds (AVOCs) (Yamada, 2013; Paasonen et al., 2013) and their oxidation products (Ehn et al., 2014) are associated to the growth of the particles. The negative effect of increasing temperatures in increasing the energy barriers the clusters have to overcome to become stable and grow in size though should not be overlooked (Kürten et al, 2018; Zhang et al., 2012). This appears to be true in most cases, as higher growth rates are found in most cases in the local summer (Nieminen et al., 2018), although the actual importance of those VOCs in the occurrence of NPF events is still not fully elucidated, with oxidation mechanisms still under intense research (Tröstl et al., 2016; Wang et al., 2020). The effect of other meteorological variables is even more complex, with studies presenting mixed results on the effect of the wind speed and atmospheric pressure. Extreme values of those variables may be favourable for the occurrence of NPF events, as they are associated with increased mixing in the atmosphere, but at the same time suppress due to increased dilution of precursors (Brines et al., 2015; Rimnácová et al., 2011; Shen et al., 2018; Siakavaras et al., 2016), or favour them due to a reduced condensation sink (CS).

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

It is evident that while a general knowledge of the role of the meteorological and atmospheric variables has been achieved, there is great uncertainty over the extent and variability of their effect (and for some of them even their actual effect) in the mechanisms of NPF in real atmospheric conditions, especially in the more complex urban environment (Harrison, 2017). The present study, using an extensive dataset from 16 sites in six European countries, attempts to elucidate the effect of several meteorological and atmospheric variables not only in general, but also depending on the geographical region or type of environment. While studies with multiple sites have been reported in the past (Dall'Osto et al., 2018; Kulmala et al., 2005; Rivas et al., 2020), to the authors' knowledge this is the first study that focuses directly on the effect of these variables upon the probability of NPF events as well as the formation and growth rates of newly formed particles in real atmospheric conditions.

2. DATA AND METHODS

2.1 Site Description and Data Availability

The present study uses a total of more than 85 years of hourly data from 16 sites from six countries of Europe of various land usage and climates. It was considered very important that at least a rural and an urban site would be available from each country to study the differences between the different land usage on NPF events throughout Europe. The sites were chosen to cover the greatest possible extent of the European continent, with sites from both northern, central and southern Europe, as well as from western and eastern. The sites are located in the UK (London and Harwell), Denmark (Copenhagen greater area), Germany (Leipzig greater area), Finland (Helsinki and Hyytiälä), Spain (Barcelona and Montseny – a site in a mountainous area) and Greece (Athens and

Finokalia). Unfortunately, not all sites had available data for all the variables studied, which to an extent may bias some of the results. An extended analysis of the typical and NPF event conditions, seasonal variations and trends at these sites for the same period is found in other studies (Bousiotis et al., 2019; 2020). A list of the available data and a brief description for each site is found in Table 1 (for the ease of reading the sites are named by the country of the site followed by the last two letters which refer to the type of site, being RU for rural/regional background, UB for urban background and RO for roadside site), while a map of the sites is found in Figure 1.

2.2 Methods

2.2.1 NPF events selection

NPF events were selected using the method proposed by Dal Maso et al (2005). An NPF event is identified by the appearance of a new mode or particles in the nucleation mode (smaller than 20 nm in diameter), which prevails for some hours and shows signs of growth. The events can then be classified into classes I and II according to the level of certainty, while class I events can be further classified to Ia and Ib. Events having both a clear formation of a new mode of particles in the smallest size bins available (thus excluding possible advected events) as well as a distinct and persistent growth of the new mode of particles for at least 3 hours were classified as Ia, while Ib consists of rather clear events that fail though by at least one of the criteria set. Additionally, for the roadside sites, a formation of particles in the nucleation mode accompanied by a significant increase of the concentrations of pollutants was not considered as an NPF event, as it may be associated with mechanisms other than the secondary formation. In the present study, only the events of class Ia were considered with the additional criterion of at least 1 nm h^{-1} growth for at least 3 hours.

2.2.2 Calculation of condensation sink, growth rate, formation rate, and NPF event probability

The condensation sink (CS) is calculated according to the method proposed by Kulmala et al., (2001) as:

$$CS = 4\pi D_{vap} \sum \beta_M \, r \, N \qquad (1)$$

where r and N is the radius and number concentration of the particles respectively and D_{vap} is the diffusion coefficient calculated as (Poling et al., 2001):

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

for T = 293 K and P = 1013.25 mbar. M and D_x are the molar mass and diffusion volume for air and sulphuric acid. β_M is the Fuchs correction factor calculated as (Fuchs and Sutugin, 1971):

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

where K_n is the Knudsen number, calculated as $K_n = 2\lambda_m/d_p$ where λ_m is the mean free path of the gas.

Growth rate (GR) is calculated as (Kulmala et al., 2012):

$$GR = \frac{D_{P_2} - D_{P_1}}{t_2 - t_1} \qquad (4)$$

for the size range between the minimum available particle diameter up to 30 nm (50 nm for the UK sites due to the higher minimum particle size available). The time window used for the calculation of the growth rate was from the start of the event until a) growth stopped, b) GMD reached the upper limit set or c) the day ended.

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

$$J_{d_p} = \frac{dN_{d_p}}{dt} + CoagS_{d_p} \times N_{d_p} + \frac{GR}{\Delta d_p} \times N_{d_p} + S_{losses} \qquad (5)$$

where $CoagS_{dp}$ is the coagulation rate of particles of diameter d_p, calculated as (Kerminen et al., 2001):

$$CoagS_{d_p} = \int K(d_p, d'_p)\, n(d'_p) dd'_p \cong \sum_{d'_p=d_p}^{d'_p=max} K(d_p, d'_p)\, N_{d_p} \qquad (6)$$

$K(d_p, d'_p)$ is the coagulation coefficient of particles with diameters d_p and d'_p, while S_{losses} accounts for additional loss terms (i.e. chamber wall losses), which are not applicable in the present study. For the present study, the formation rate of particles of diameter of 10 nm was calculated for uniformity (16 nm for the UK sites), though most sites had data for particle sizes below 10 nm.

The NPF probability, used instead of NPF frequency when modelled results are presented, was calculated by the number of NPF event days divided by the number of days with available data in the given group (temporal, variable range etc.). The results presented in this study were normalised according to the data availability, as:

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

2.2.3 Calculation of the gradient and intercept for the variables used

Due to the large datasets available and the great spread of the values, a direct comparison between a given variable and any of the characteristics associated with NPF events (NPF probability, growth rate and formation rate) always provided results with low statistical significance. As a result, an alternative method which can provide a reliable result without the dispersion of the large datasets was used in the present study, to investigate the relationships between the variables which are considered to be associated with the NPF events. For this, a timeframe which is more directly associated with the NPF events typically observed in the mid-latitudes was chosen. For NPF probability and GR the timeframe between 05:00 to 17:00 Local Time (LT) was chosen, which is considered the time when the vast majority of NPF events take place and further develop with the growth of the particles. For the formation rate a smaller timeframe was chosen, 09:00 to 15:00 LT which is ± 3 hours from the time of the maximum formation rate found for almost all sites (12:00 LT). This was done to exclude as far as possible the effect of the morning rush at the roadside sites, as well as only to include the time window when the formation rate is mostly relevant to NPF events (negative values that are more probable outside this timeframe and are not associated with the formation of the particles would bias the results).

For the CS the timeframe 05:00 to 10:00 LT was chosen. This was done to avoid including the direct effect of the NPF events (the contribution of newly formed particles to CS), as well as to provide results for the conditions which either promote or suppress the characteristics studied, which specifically for the CS are more important before the start of the events. The extreme values (very high or very low) which bias the results only carrying a very small piece (forming bins of very small size) of information were then removed, though 90% of the available data was used for all the variables. The data left was separated into smaller bins and a minimum of 10 bins was required for each variable (for example if the difference between the minimum and the maximum relative humidity (RH) is 70%, then 14 bins each with a range of 5% were formed). The variables of interest were then averaged for each bin and plotted, and a linear relation was considered for each one of them.

The gradient of these linear relations (a_N, a_G and a_J for NPF probability, growth rate and formation rate J_{10} accordingly) found in this analysis should be used with great caution as apart from the atmospheric conditions (local and meteorological as well as atmospheric composition) it is also affected by the variable in question (e.g. a greater NPF probability will provide a greater gradient), resulting in giving the same trend for all the atmospheric variables tested; the sites with the higher values of these variables (NPF probability and formation rate) always had greater gradient values and vice versa. In order to remove the effect of the variable in question (NPF probability or formation rate – growth rate will provide an unreliable result as it is calculated in a different range for each site due to the lower available size of particles), the gradients were normalised by dividing them by their respective variable (e.g. divide the gradient of the NPF probability with the NPF frequency), providing with a new normalised slope (a_N* for NPF probability or a_J* for the formation rate) that will have no significance other than its absolute value, which can be used for direct comparisons:

$$a_N^* = \frac{a_N}{\text{NPF \%}}$$

Where a_N is the gradient of the relation between the given variable and NPF frequency (NPF %)

$$a_J^* = \frac{a_J}{J_{10}}$$

Where a_J is the gradient of the relation between the given variable and the formation rate of 10 nm particles J_{10} (J_{16} for the UK sites).

3. RESULTS

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

- the probability of events occurring (i.e. days with an event divided by total days with relevant data, depending on the variable and range studied),
- the rate of particle formation at a given size (J_{10} in this case),

- the growth rate of particles from the lower measurement limit to 30 nm (or 50 nm for the UK sites).

From the analysis of the extended dataset a total of 1952 NPF events were extracted and studied. The NPF frequency, growth and formation rate for each site is found in Table 2. The seasonal variation of NPF events is found in Figure S14.

3.1 Meteorological Conditions

The gradients, coefficients of determination (R^2) and the p-values (deriving from one-way ANOVA test) from the analysis of the meteorological variables, as well as the average conditions of these variables are found in Table 3. The results for each site and variable are found in figures S1 – S5.

3.1.1 Solar radiation intensity

As mentioned earlier, solar radiation intensity is considered to be one of the most important variables in NPF occurrence, as it contributes to the production of H_2SO_4 which is a main component of the initial clusters and participates in the early growth of the newly formed particles. Hidy et al. (1994) reported up to six times higher SO_2 oxidation rates into H_2SO_4 in typical summer conditions compared to winter. For almost all sites this relation is confirmed with very strong correlations ($R^2 > 0.75$) between the intensity of solar radiation and the probability for NPF events to occur. The relationship between the solar radiation and NPF probability was positive at all sites and only three sites (FINUB, SPARU and GREUB) presented weak correlations ($R^2 < 0.40$). Weaker correlations were found for the southern European sites, which might be associated with the higher averages for solar radiation intensity, or the interference of other processes (such as coinciding with increased CS by recirculation of air masses (Carnerero et al., 2019)), possibly making it less of an important factor for these areas.

The relationship of solar radiation with the growth rate was weaker in all cases and did not present a clear trend. Only some rural background sites (GERRU, FINRU and GRERU) presented a strong correlation ($R^2 > 0.50$). The relationship found in most cases was positive apart from two roadside sites (GERRO and UKRO) and two urban background sites (GREUB and UKUB), though due to the low R^2 (< 0.10) these results cannot be considered with confidence. It seems though that the solar radiation intensity is probably a more important factor at background sites rather than at roadside sites, where possibly local conditions (such as local emissions) are more important (Olin et al, 2020). Finally, the formation rate has a positive relationship with the solar radiation intensity, with relatively strong correlations in most areas ($R^2 > 0.50$). The correlations were stronger at the rural background sites compared to the roadside sites, which further underlines the increased importance of this factor at this type of site. A negative relationship between the solar radiation intensity and the formation rate was found at the GRERU site but the R^2 is very low ($R^2 = 0.05$).

Plotting the normalised gradients for NPF event probability a_N^* with the average solar radiation intensity at each site (Figure 2) a negative relationship is found ($R^2 = 0.62$), with the southern areas (those with higher average solar intensity) having smaller a_N* compared to those in higher latitudes (and thus with a lower average solar radiation). This may indicate that while solar radiation is a deciding factor in the occurrence of an NPF event, when in greater intensity its role becomes relatively less important, a finding that was also implied by Wonaschütz et al. (2015). Additionally, the a_J* was found to be higher at all rural sites compared to their respective roadside sites (and urban background sites for all but the Greek and German ones), making it a more important factor at this type of site (Figure 3).

3.1.2 Relative humidity

Relative humidity is considered to have a negative effect on the occurrence of NPF events (Jeong et al., 2010; Hamed et al., 2011; Park et al., 2015; Dada et al., 2017; Li et al., 2019). While water in the atmosphere is one of the main compounds needed for the formation of the initial clusters either on the binary or ternary nucleation theory (Henschel et al., 2016; Korhonen et al., 1999; Mirabel and Katz, 1974), under atmospheric conditions it may also play a negative role suppressing the number concentrations of new particles by increasing aerosol surface area (Li et al. 2019). Consistent with this, a negative relationship of the RH with NPF probability was found for all the sites of this study with very high R^2 for almost all of them ($R^2 > 0.80$). This is not simple to interpret as solar radiation intensity, temperature, RH and CS are not independent variables, since an increase in temperature of an air mass due to increased solar radiation will be associated with reduced RH, which in turn affects the CS. The sites in Greece presented lower R^2 compared to the other sites while, GRERU was found to have the weakest correlation ($R^2 = 0.22$). This may be due to the different seasonality of the events found for the Greek sites (being more balanced within a year), as there was increased frequency of NPF events for the seasons with higher RH compared to other sites, making it a less important factor for their occurrence. Growth rate on the other hand had a variable relationship, either positive or negative, with only a handful of background sites having strong correlations. The German background sites as well as FINRU, which were among the sites with the highest average RH (average RH for GERRU is 81.9%, GERUB is 78.7% and FINUB is 80.1%) presented a negative relationship between the RH and growth rate. DENRU (average RH at 75.7%) had a positive relationship, which might indicate that the relationship between these two variables may vary depending upon the RH range. Formation rate also appears to have a negative relationship with the RH, though this relationship was significant ($R^2 > 0.40$) for only 6 sites, which once again in most cases are sites with higher RH average conditions. Along with the results of the growth rate this might indicate that the RH becomes a more important factor in the development of NPF events as its values increase.

The normalised gradients once again provide some additional information. Regarding the NPF probability, it is found that the a_N* was more negative at rural sites compared to roadside sites. This indicates that the RH has a smaller effect at roadside sites, as other variables, such as the atmospheric composition, are probably more important within the complex environment in this type of site. Additionally, the relationship between a_N* and average RH at the sites had a negative relationship ($R^2 = 0.46$), which further shows that the RH becomes a more important factor at higher values (Figure 4). Furthermore, at the sets of rural and roadside sites with R^2 higher than 0.40 for the relation between RH and the formation rate (UK and German sites), it was found that the a_J* was more negative at the rural sites which indicates that the RH is a more important factor at rural sites compared to their respective roadside sites.

3.1.3 Temperature

Temperature can have both a direct and indirect effect in the development of NPF events, as it is directly associated with the abundance of both biogenic and anthropogenic volatile carbon, which is an important group of compounds whose oxidation products can participate in nucleation itself (Lehtipalo et al., 2018; Rose et al., 2018), as well as in the growth of newly formed particles. It may also have a negative effect on the particle size distributions or number concentrations through other processes such as particle evaporation. Most of the sites of the present study presented a strong relationship of NPF probability with temperature, which in most cases was positive, though in many cases (such as the Danish, Finnish and Spanish sites – figures S2b, d and e) there seems to be a peak in the NPF probability at some temperature, after which a decline starts (though being at the higher end does not greatly affect the results). Sites with smaller R^2 (weaker association with temperature), were mainly those that have a seasonal variation that favoured seasons other than summer. These sites not only had weaker relationship of NPF probability with temperature, but in most cases had a negative relationship (background sites in Finland, Spain and Greece). The Finnish sites, having the lowest average temperatures and a sufficient amount of data below zero temperature, show at all three sites the possible presence of a peak in the NPF event probability for temperatures below zero (Figure S2d). This seems to be the cause of the weak relations found there and they seem to be associated with the formation rate J_{10}, which also seems to have an increasing trend below zero degrees (Figure S2p). This may depend on the nucleation mechanism occurring, as cluster evaporation rates of sulphuric acid clusters are sensitive to the ternary stabilising compound present (Olenius et. al., 2017), as well as the possible enhancement of growth mechanisms at lower temperatures (below 5^{o}C) by other chemical compounds in the atmosphere (i.e. nitric acid and ammonia) as found by Wang et al., (2020). Laboratory experiments show that the characteristics of organic aerosol forming from alpha-pinene is governed by gas phase oxidation (e.g. Ye et al. 2019). In the real atmosphere, the higher temperature enhances the amount of biogenic vapours (e.g. Paasonen et al. 2013) and, although the oxidation can be more efficient at higher temperatures, the lower temperatures favour formation of more non-volatile compounds (Quéléver et al., 2019; Stolzenburg et al. 2018; Ye et al. 2019).

Growth rate had a more uniform trend, with almost all sites having a positive relationship with temperature (apart from GERRO, though with $R^2 = 0.00$). This relationship was very strong for most sites ($R^2 > 0.60$ for 10 sites), which is also confirming the summer peak found for the growth rate at most of these sites in other studies (Bousiotis et al., 2020; 2019). A rather strong relationship ($R^2 > 0.50$) with temperature was also found for the formation rate for most sites, and was positive for almost all sites (apart from FINRO with $R^2 = 0.01$ and the Greek sites with $R^2 < 0.47$). As with the NPF probability, in general the sites with a seasonal variation of events that favoured summer had the strongest relationship (high R^2) of the temperature with formation rate, which might indicate that this variable, either through its direct or indirect effect is an important one for the seasonal variability of NPF events in a given area.

The normalised gradients for this variable did not present a clear trend among the areas studied, other than presenting greater a_N* for the sites with a summer peak in their NPF event seasonal variation. As with other meteorological variables, the importance of this variable became smaller with increased values in the average conditions for both the NPF probability (Figure 5) and J_{10}, though these relationships were not significant (biased by the very low average temperatures and different behaviour of the variables at the Finnish sites, without which the relation becomes a lot clearer as indicated in Figure S13). The variation though within the sites of the same area (different sites in same country / region) appears to directly follow the variability of temperature, showing that the temperature directly affects the occurrence of NPF events when other meteorological factors remain constant, having a negative trend for all countries but Finland. The a_J^* though is found to be greater (positively or negatively) at the rural background sites than at the other two types of sites at all areas studied, showing that it is a more important factor for the formation rate at this type of site compared to others (Figure 6).

3.1.4 Wind speed

Wind speed may have both a positive and a negative effect on the occurrence of NPF events. On one hand, it may promote NPF events by the increased mixing of the condensable compounds in the atmosphere as well as by reducing the CS. On the other hand, high wind speeds may suppress NPF events due to increased dilution. It should be considered that the variability found is also affected by the specific conditions found at each site. The wind speed measurements in many cases, especially in urban sites, can be biased by the local topography or specific conditions found at each site, thus representing the local conditions for this variable rather than the regional ones. Similarly, measurements of wind speed at well sited meteorological stations may be more representative of regional conditions, than of those affecting the sites of nucleation measurement. The sites in this study presented mixed results, both in the importance as well as the effect of the wind speed variability. Three different behaviours were found in the variation of NPF event probability and wind speed which appear to be associated with local conditions as they are almost uniformly found among the sites within close proximity. Some sites presented a steady increase of NPF event probability with wind speed (Danish sites, UKUB, FINRU, SPAUB and GRERU), while others were found to steadily decline with increasing wind speeds (German sites – it should be noted that the German sites are the only ones that are located at a great distance from the sea), while some were found to reach a peak and then decline, which also leads to smaller R^2 (UKRU, UKRO, SPARU and to a lesser extent GREUB – figures S4a, e and f). The reasons for these differences between the sites are very hard to distinguish as apart from the wind speed the origin and the characteristics of these air masses play a crucial role. Following this, it appears that NPF probability is very low or zero for wind speeds close to calm for the sites with an increasing trend (as well as those that have a peak and decline after), while the opposite is observed for the German sites where the maximum NPF probability is found for very low wind speeds (fig. S4c).

Similarly, the effect of different wind speeds upon the growth rate also varied a lot, though it was found to be negative in all the cases where R^2 was higher than 0.50 (UKUB, DENRU, DENRO, GERRU, GERUB and GREUB). Finally, the formation rate was found to have a significant correlation ($R^2 > 0.40$) only at two sites (UKRO and DENRU), probably indicating that the variability of the wind speed either does not affect this variable or its effect is rather small.

The normalised gradients did not have any notable relation to either the NPF probability or the formation rate further confirming that the effect of the different wind speeds is not due to its variability only, but it is also influenced by the characteristics of the incoming air masses as well as specific local conditions found at each site.

3.1.5 Pressure

In almost all the sites with available data (apart from the Spanish), the NPF probability presented a positive relationship with high significance at all types of sites. The greater significance found at the rural sites (apart from SPARU) indicates the increased importance of meteorological conditions in the occurrence of NPF events at this type of site. The growth rate also presented a similar picture, with positive relationships at all the background sites of this study except the ones in Greece ($R^2 > 0.71$) and FINUB (though with low R^2 at 0.02). This is probably associated with the seasonal variation found in Greece where higher growth rates were found in summer, a period when increased wind speeds and lower atmospheric pressure was found due to the Etesians, a pressure system that develops in the region every summer (Kalkavouras et al., 2017). An interesting finding is the negative gradients found at all the roadside sites, though the significance of these results is relatively low ($R^2 < 0.43$) and always lower compared to the rural sites. The effects of pressure above are not likely to be important. Once again however, this is not an independent variable and higher pressure in summer tends to be associated with higher insolation and temperatures and lower RH. Since most events occur in the warmer months of the year, this is probably the explanation for the apparent effects of pressure. The formation rate presented relationships of low significance ($R^2 < 0.47$) for the sites of this study. Due to this, pressure should not be an important factor for the formation rate at any type of site.

The normalised gradients did not present any clear trends, even for the NPF probability for which the results presented significant relations at almost all sites.

3.2 Atmospheric Composition

The gradients, R^2 and p-values from the analysis of a number of air pollutants (SO_2, NO_x, O_3, organic compounds, sulphate and ammonia) and the CS, as well as the average conditions of these variables are found in Table 4. The results for each site and variable are found in Figures S6 – S12.

3.2.1 Sulphur dioxide (SO_2)

Sulphur dioxide, as a precursor of H_2SO_4, is considered as one of the main components associated with the NPF process. According to nucleation theories and observations, H_2SO_4 is the most important compound from which the initial clusters are formed, as well as one of the candidate compounds for the initial steps of particle growth (Kirkby et al., 2011; Nieminen et al., 2010; Sipila et al., 2010; Stolzenburg et al., 2020). As H_2SO_4 in the atmosphere is produced from oxidation reactions of SO_2 it would be expected that increased concentrations of the latter would be associated with increased values for all the variables associated with the NPF process. Contrary to this though, the relationship of SO_2 concentrations with NPF probability was found to be negative at all the sites in this study with available data. This relationship was relatively strong ($R^2 > 0.50$) in most areas with an increased significance at roadside sites compared to their respective rural sites. As this is a negative relationship, this may indicate that SO_2 is in sufficient concentrations for H_2SO_4 formation, thus not suppressing the occurrence of NPF events, as well as showing that in increased concentrations, it is a more important factor (or surrogate for a factor) in preventing the occurrence of NPF events within the urban environment, as higher SO_2 is likely associated with increased co-emitted particle pollution and hence CS. The growth rate on the other hand, presented mixed results and the significance of the relationships is low in most cases, which makes these results unreliable. Finally, the relationship of SO_2 concentrations with the formation rate was found to be positive at all sites but SPARU and FINRU (which had the lowest concentrations across the sites with available data). The significance of this relationship was rather low ($R^2 < 0.40$) for all but the roadside sites. This suggests that higher H_2SO_4 concentrations favour greater formation rates (i.e. more particles can be formed), rather than necessarily promoting nucleation itself because of the competing effect of condensation onto the pre-existing particle population.

The normalised gradients a_N^* were found to be more negative at the background sites compared to their respective roadside sites, as well as being less negative in the UK (where SO_2 is in greater abundance) compared to the other sites with relatively significant relationships. Plotting the average SO_2 concentrations with the normalised gradients a_N^* for the all sites (though not all had significant relations), a positive relationship with relatively high R^2 (when the extreme values from Marylebone Road-UKRO are removed) is found which might indicate that while increased concentrations are a negative factor in NPF event occurrence at a given site, in general the sites with higher SO_2 concentrations on average present higher probability for NPF events (Figures 7a and

7b). This appears to be in agreement with Dall'Osto et al. (2018) who discussed the variable role of SO_2 depending on its concentrations. No significant relations were found for the values of a_J^* as in most cases these relationships were rather weak.

3.2.2 Nitrogen oxides or nitrogen dioxide (NO_x or NO_2)

NO_x and NO_2 are directly associated with pollution, which can be a limiting factor for NPF events as it increases the CS and may suppress the events (An et al., 2015), though with the reduction of SO_2 concentrations achieved the last couple of decades, there is a possibility for oxidation products of NO_x to become an important component for NPF (Wang et al., 2020). For almost all sites (apart from GRERU) with available data a negative relationship between the NPF probability and NO_x concentrations (or NO_2 depending on the available data) was found. Similarly, for all the sites but SPARU and GRERU, the correlations were strong with $R^2 > 0.43$. The rural background sites had a weaker relationship between the two variables compared to the urban sites, which is probably associated with them having rather low concentrations and variability of NO_x (or NO_2), making the variations of this factor less important. Growth rate had weaker correlations with NO_x and different trends between the sites, either being positive or negative. The variable effect of NO_x on particle growth, shifting HOMs volatility, was previously discussed by Yan et al. (2020). While variability was found for the background sites, all roadside sites regardless of the strength of the relationship had a positive relation between NO_x and the growth rate. This may indicate the different components associated with the growth process at each type of site which, as found in other studies, can be related to compounds associated with combustion processes that take place within the urban environment (Guo et al., 2020; Wang et al., 2017a). The formation rate presents few cases of strong relationships, with variable trends (positive and negative). While much effort was made to isolate the effect of NPF events by taking a shorter time frame before the event, the effect of local pollution is still included, especially at the urban sites (which probably explains the positive effect found).

The normalised gradients do not provide a significant result for the relationship of this variable with either the probability of the events or the formation rate. The only noteworthy points are the more negative a_N^* at the rural background sites compared to the roadside sites in all the areas studied, which shows the increased importance of a clean environment for NPF events to occur in areas where condensable compounds are in lesser abundance, such as a rural environment. Additionally, the negative gradients found at all the roadside sites, which increases the confidence that the events extracted at the roadside sites are not pollution incidents but NPF events. However, it appears that traffic pollution favours higher particle growth rates, although the components responsible for this effect are unknown.

3.2.3 Ozone (O_3)

Ozone is typically the result of atmospheric photochemistry and is itself a source of hydroxyl radical through photolysis, or ozonolysis of alkenes both during daytime and night-time (Fenske et al., 2000). It might therefore be expected to act as an indicator of photochemical activity which promotes the oxidation of SO_2 and VOCs. Ozone concentrations may be directly related to the solar radiation intensity as well as the pollution levels in the area studied, and O_3 is considered as a positive factor in the occurrence of NPF events (Woo et al., 2001; Berndt et al., 2006). As with the solar radiation intensity, there is a strong relationship between O_3 concentration and the probability for NPF events. This positive relationship was found to be stronger for the sites in northern Europe ($R^2 > 0.51$), while it was not significant ($R^2 < 0.38$) for the sites in southern Europe (Spanish sites and GRERU), possibly indicating that O_3 is a less important factor at the southern sites. Specifically for the Spanish sites which have the highest average concentrations of O_3 with some extreme values (Querol et al., 2017), the relationship of O_3 concentrations with the NPF probability presents a unique trend (Figure S8d), having a clear peak then a steady decline at both sites (though at different O_3 concentrations), which is also responsible for the low correlations found (this trend seems to also occur at SPARU for the growth rate and to a lesser extent for the formation rate as well, though for different O_3 concentration ranges – figures S8i and n). The specific variability found at the Spanish sites was also studied by Carnerero et al., (2019). For sites with a marked seasonal variation in ozone, associations with NPF may be artefactual due to correlations with other variables such as temperature, RH and solar radiation intensity.

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

As the correlations found were strong the normalised gradients for NPF probability, when plotted against the average concentrations of O_3, present a negative correlation with relatively high R^2 (0.64), indicating that the O_3 is a more important factor in the occurrence of NPF events when in lower concentrations (Figure 8). Finally, though with a low level of confidence for the southern sites, the a_N^* were smaller at the southern sites compared to those in the north, up to one order of magnitude between FINRU (furthest north rural background) and GRERU (furthest south rural background).

3.2.4 Organic compounds

3.2.4.1 Particulate organic carbon (OC)

Organic carbon (OC) compounds in the secondary aerosol typically enter the particles via condensational processes, with a role that becomes increasingly important as the size of the particles becomes larger (Nieminen et al., 2010; Zhang et al., 2012; Shrivastava et al., 2017). Particulate OC, the data for which is available in the present study, can be associated with pollution, especially in the urban environment. Only a few of the sites of the present study were found to have a relatively strong negative relationship ($R^2 > 0.50$) of particulate OC with the NPF probability (UKUB, UKRO and DENRU). Regardless though of the strength of this relationship, all other sites (apart from FINRU) had a negative relationship between these two variables as well, consistent with increased concentrations of particulate OC being associated with increased pollution, which elevate the CS, suppressing the occurrence of NPF events. Growth rate on the other hand was found to have a positive relationship ($R^2 > 0.40$) for most of the sites. This relationship appeared to be stronger (higher R^2) at the roadside sites with available data compared to their respective rural background sites. The relationship between particulate OC and the growth rate was positive at all the sites with available data regardless of their significance showing that, despite its effect in the occurrence of NPF events, it is still a favourable variable for the growth of the particles. The formation rate was found to have a significant relationship with particulate OC concentrations at half of the sites with available data (UKUB, UKRO, DENRU, DENRO).

The normalised gradients for this variable did not present any noteworthy relations with either the type of site or the concentrations of OC at a given site.

3.2.4.2 Volatile organic compounds (VOCs)

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

Volatile organic compound data were available for three of the sites of this study (Table S2). Two of the sites with VOC data were from the rural background and the roadside site in the UK. Most of the compounds are associated with combustion sources and were found to have a negative relationship with NPF event occurrence at both sites, with high R^2 ($R^2 > 0.50$) in most cases. Additionally, isoprene, which may have either biogenic or anthropogenic sources (Wagner and Kuttler, 2014) was also found to have a negative relationship with NPF event occurrence at Marylebone Road-UKRO, though with low R^2 (0.07). This result is in line with the VOCs being strongly correlated with particulate OC (which presented a negative relationship with NPF event probability, as discussed in Section 3.2.4.1), as well as with the CS (which also presented a negative relationship with NPF event probability, as mentioned in Section 3.2.6), further associating these compounds with combustion emissions.

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

As Hyytiälä (FINRU) is a rural background site far from the direct effect of combustion emissions, different VOCs were measured, which mainly originate from biogenic sources rather than anthropogenic ones. The results were mixed and less clear compared to those from the UK sites (mainly due to the smaller dataset), and three groups were found depending on their relationship with NPF probability. The first group, including acetonitrile, acetic acid and methyl ethyl ketone (MEK) presented a slight positive relationship. The second group presented a negative relationship, with the VOCs in this group being monoterpenes, methacroleine, benzene, isoprene and toluene (only the last two have $R^2 > 0.50$). Finally, the third group included VOCs that presented a peak and then a decline for higher concentrations including methanol, and acetone. Two groups of VOCs were found depending on their relationship with the growth rate. The ones with a positive relationship being methanol, acetonitrile, acetone, acetic acid, isoprene, methacroleine, monoterpenes and toluene, while acetaldehyde, MEK and benzene had a negative relationship, with relatively high R^2 in most cases. Finally, the results with the formation rate were unclear with only a handful presenting weak ($R^2 < 0.21$) positive (methanol, acetic acid and benzene) or negative

(MEK) relationships that do not appear to be significant. The normalised gradients cannot be used for VOCs as there are very few sites with available data.

3.2.5 Sulphate (SO_4^{-2})

Sulphate (SO_4^{2-}) is a major secondary constituent of aerosols. Secondary SO_4^{2-} aerosols largely arise from either gas phase reaction between SO_2 and OH, or in the aqueous phase by the reaction of SO_2 and O_3 or H_2O_2, or NO_2 (Hidy et al., 1994). In environments where SO_4^{2-} chemistry is dominant (i.e. remote areas), SO_4^{2-} and ammonium (bi) sulphate ($(NH_4)_2SO_4$ and NH_4HSO_4) particles are a large relative contributor to aerosol mass, while this contribution is lower in environments where other emissions are also significant (i.e. urban areas where the secondary NO_3^- relative contribution is a lot higher). While not well established, a possible relationship of SO_4^{2-}-containing compounds and variables of NPF events was found in previous studies (Beddows et al., 2015; Minguillón et al., 2015; Wang et al., 2017b). In the present study, only a few sites had SO_4^{2-} data available, for PM_1 (FINRU), $PM_{2.5}$ (Danish sites) or PM_{10} (rest of the sites). While this data cannot be considered as directly associated with the ultrafine particles, for two sites with available AMS data for ultrafine particles, the direct comparison between SO_4^{2-} aerosol in PM and in the range of particles of about 50 nm, very high correlations were found (results not included). For all the sites with available data the NPF probability presented a negative relationship. The significance of this relationship was found to be relatively high ($R^2 > 0.50$) only for background sites (apart from GERRU, which has rather low concentrations and probably different mechanisms for the NPF events). Similarly, the growth rate presented a significant relationship ($R^2 > 0.40$) for the same background sites (apart from FINRU), though this relationship was found to be positive at all sites regardless of its significance. Finally, the formation rate did not present a clear trend as it was found to have both negative and positive relationships for different sites. This relationship was significant only for two rural sites (UKRU and DENRU) and as a result no conclusions can be reached.

The normalised gradients cannot be used for any analysis on sulphate as the measurements available are from different particle size ranges.

3.2.6 Gaseous ammonia (NH_3)

Ammonia (NH_3) can be an important compound in the nucleation process according to the ternary theory (Kirkby et al., 2011; Napari et al., 2002). It was found that elevations in NH_3 concentrations can lead to elevations to NPF rate (Lehtipalo et al., 2018) and it was also found to be an important factor for NPF event occurrence even when stronger bases are present in high concentrations (Glasoe et al., 2015). No significant variation was found though between event and non-event days in a previous study in Harwell - UKRU (Bousiotis et al., 2019). Data for gaseous ammonia was only available for UKRU and presented a positive relationship with NPF probability, until reaching a peak point. Further increase in NH_3 concentrations presented a decline with NPF probability (Figure S11a), which might be due to its association with increased pollution levels. It presented a clear positive relationship with both the growth rate (though it also appears to decline at high concentrations) and the formation rate, consistent with its well-established role in accelerating both of these processes (Kirkby et al. 2011; Stolzenburg et al., 2020).

3.2.7 Condensation sink (CS)

The CS is a measure of the rate at which molecules will condense onto pre-existing aerosols (Lehtinen et al., 2003). It is highly dependent on the number and size of the particles in the atmosphere and as a result it is expected to be affected by both the local emissions within the urban environment as well as the formation and growth of the particles due to NPF events. As a result, for the specific metric a time frame before the events are in full development was chosen (05:00 to 10:00 LT) to avoid including the effect of the NPF events and provide a picture of the atmospheric conditions that preceded the NPF events. With this data, the NPF probability presented very strong relationships with the condensation sink. Two groups of sites were found though; those which had a positive relationship and those with a negative relationship. In the first group are the sites in Germany and Greece while all others had a negative relationship. This grouping follows the trend between the countries, the sites of which presented a greater or smaller CS on NPF event days (having positive or negative gradients respectively), though it is unknown what causes this behaviour (at the German sites and GREUB it may be associated with the very high formation rates on NPF event days). While the gradients from this analysis cannot be used for direct comparisons, a trend was found for which the gradients were more positive or negative at the rural sites compared to their respective roadside sites, which might indicate the greater importance of the variability of the CS at the rural sites in the occurrence of NPF events.

The growth rate was positively correlated with the CS for most of the sites, with strong relationships ($R^2 > 0.40$) for about half of them. As the CS is a metric of pre-existing particles, it is also associated with the level of pollution in a given area. The increased significance and gradient found at the rural sites probably indicates the importance of enhanced presence of condensable compounds in a cleaner environment, which in many cases are associated with the moderate presence of pollution. The formation rate was also found to have a positive relationship with the CS. This relationship was more significant at the roadside sites of this study, a result which to some extent is biased by the presence of increased traffic emissions found in the timeframe chosen. While to an extent, increased presence of condensable compounds can be favourable for greater formation rates, this result should be considered with great caution.

The normalised gradients a_N^* followed a similar trend as those found with the initial analysis. These gradients were found to be more positive or negative, depending on the trend of the given area, at the rural sites compared to their roadside sites. The urban background sites did not always have a uniform behaviour (though in UK, Denmark and Finland these were between the rural site and the roadside site), due to their more diverse character compared to the other two types of sites.

3.3 Association of the Effect of the Variables

The Pearson correlation coefficients for the variables studied on each site are found in Table S1. The relatively strong relationship between the solar radiation intensity, temperature and O_3 found, as well as their anticorrelation with the RH may lead to the conclusion that not all these factors play a role in NPF events, but their visible effect is the result of their relationship with each other. There is a similar case with the association of the CS and NO_x (or NO_2), and OC, as well as SO_2, especially at urban sites. However, the factors affect different outcomes differently, as for example the solar radiation intensity does not seem to be as important a factor for the growth rate as temperature, or O_3 does not seem to be strongly associated with either the formation or the growth rate. This is further established by the fact that some of these variables do not correlate well at the southern sites, but still appear to be associated with either the probability of NPF events or the growth or nucleation rate. The effects of all of these factors have been demonstrated in both laboratory and atmospheric studies in the past and were discussed earlier in this paper. By the analysis provided in the present study, the effect of each of these variables is further established, providing an association of each one of these variables with either the formation or the growth mechanism. However, RH does not seem to be a consistent factor in any mechanism, and it appears that its effect is dependent on location specific conditions, although it was the variable with the most consistent relation with NPF event probability at almost all sites.

3.4 Relationship to a previous multi-station European study

The findings of our study in respect of the background sites show many similarities with the conclusions drawn in the previous multi-station study in Europe by Dall'Osto et al. (2018) despite the two studies using several different sampling stations as well as some in common. Both studies point towards the influence of variables such as solar radiation intensity and CS upon the occurrence of NPF events. The previous study suggested that different compounds participate in the growth of the particles, depending on the area considered. Thus, for northern and southern sites the growth of the particles is suggested to be driven mainly by organic compounds, while for the sites in central Europe sulphate plays a more important role. These findings are confirmed by the present study, as the growth rate was found to correlate better with organic compounds for the rural sites in Finland and Greece, while SO_4^{2-} presented a stronger relationship with the growth rate for the Danish and German sites (the latter presented high gradient values but low R^2 due to a decline at higher SO_4^{2-} concentrations – figure S10i, probably associated with NPF events being suppressed by increased pollution). The growth of the particles at the rural background site in the UK, characterised as "Overlap" in the previous study, was found to be strongly associated with both organic compounds and sulphate, consistent with it being in the central group.

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

4. CONCLUSIONS

The present study attempts to explain the effect of several meteorological and atmospheric variables on the occurrence and development of NPF events, by using a large-scale dataset. More than 85 site-years of data from 16 sites from six countries in Europe were analysed for NPF events. A total of 1952 NPF events with consequent growth of the newly formed particles were extracted and with the use of binned linear regression, the relationship between three variables associated with NPF events (NPF event probability, formation and growth rate) with meteorological conditions and atmospheric composition was studied. Among the meteorological conditions, solar radiation intensity, temperature and atmospheric pressure presented a positive relationship with the occurrence of NPF events, either promoting the formation or growth rate. RH presented a negative relationship with NPF event probability which in most cases was associated with it being a limiting factor on particle formation at higher average values. Wind speed on the other hand presented variable results, appearing to depend on the location of the sites rather than their type. This shows that while wind speed can be a factor in NPF event occurrence, the origin of the incoming air masses also plays a very important role. In most cases, meteorological conditions, such as temperature or RH appeared to be more important factors in NPF event occurrence at rural sites compared to urban sites, suggesting that NPF events are driven more by them at this type of site compared to urban environments and the more complex chemical interactions found there. Additionally, while some meteorological variables appeared to play a crucial role in the occurrence of NPF events, this role appears to become less important at higher values when a positive relation was found (or lower when a negative relation was found).

The results for the levels of atmospheric pollutants presented a more interesting picture as most of these, which appear to be either directly or indirectly associated with the NPF process were found to have negative relationships with NPF probability. This is probably due to the fact that increased concentrations of such compounds are associated with more polluted conditions, which are a limiting factor in the occurrence of NPF events, as was found with the negative relationship between the CS and NPF probability in most cases. Thus, SO_2, NO_x (or NO_2), particulate OC and SO_4^{2-} concentrations were negatively correlated with NPF probability in most cases. Average SO_2 concentrations appeared to correlate positively with the normalised NPF event probability gradients with a relatively significant correlation, indicating that while increasing concentrations have a negative impact in the occurrence of NPF events at a given site, in general sites with higher SO_2 concentrations have higher probability for NPF events. Conversely, these compounds in many cases had a positive relationship (not always though with high significance) with the other variables considered. Thus, particulate OC (and VOCs where data was available) and SO_4^{2-} consistently had a positive relationship with the growth rate, while SO_2 was positively associated with both the formation and growth rate in most cases. Finally, O_3 was positively correlated with NPF event probability at all sites in this study, though it presented variable results with the other two variables. As with some meteorological conditions it was found that at sites with increased concentrations of O_3, its importance as a factor was decreased, which to some extent can be related with the high CS associated with peak summer O_3 days in southern Europe.

It should be noted that the variables considered are in many cases inter-related (e.g. temperature and RH) and this considerably complicates the interpretation in terms of causal factors. Large datasets are very useful in providing more uniform results by removing the possible bias of short period extremities, which may lead to wrong assumptions. This study, apart from providing insights into the effect of a number of variables on the occurrence and development of NPF events in atmospheric conditions across Europe, also shows the differences that climatic, land use and atmospheric composition variations cause to those effects. Such variations are probably the cause of the differences found among previous studies. Following from this, the importance of a high-resolution measurement network, both spatially and temporally is underlined, as it can help in elucidating the mechanisms of new particle formation in the real atmosphere.

DATA ACCESSIBILITY

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

AUTHOR CONTRIBUTIONS

The study was conceived and planned by RMH who also contributed to the final manuscript, and DB who also carried out the analysis and 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. JB provided help with analysis of the data. FDP provided advice on the analysis. MDO, XQ and TP contributed to the final manuscript.

COMPETING INTERESTS

The authors have no conflict of interests.

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TABLE LEGENDS

Table 1: Location and data availability of the sites.

Table 2: Frequency (and number of NPF events), growth and formation rate of NPF events.

Table 3: Normalised gradients (non-normalised for growth rate), R^2 and p-values (- for values >0.05) for the relationship between meteorological conditions and NPF event variables.

Table 4: Normalised gradients (non-normalised for growth rate), R^2 and p-values (- for values >0.05) for the relationship between atmospheric composition variables and NPF event variables.

FIGURE LEGENDS

Figure 1: Map of the sites of the present study.

Figure 2: Relation of average downward incoming solar radiation (K↓) and normalised gradients a_N^*.

Figure 3: Normalised gradients a_J^* for K↓ (*UK sites are calculated with solar irradiance).

Figure 4a: Relationship of average relative humidity and normalised gradients a_N^*.

Figure 4b: Relationship of average relative humidity and normalised gradients a_N^* (SPAUB not included).

Figure 5: Relationship of average temperature and normalised gradients a_N^*.

Figure 6: Normalised gradients a_J^* for temperature.

Figure 7a: Relationship of average SO_2 concentrations and normalised gradients a_N^*.

Figure 7b: Relationship of average SO_2 concentrations and normalised gradients a_N^* (UKRO not included).

Figure 8: Relationship of average O_3 concentrations and normalised gradients a_N^*.

Table 1: Location and data availability of the sites.

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

Table 2: Frequency (and number of NPF events), growth and formation rate of NPF events.

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

* GR up to 50 nm calculated

** J_{16} calculated

Table 3: Normalised gradients (non-normalised for growth rate), R^2 and p-values (- for values >0.05) for the relation between meteorological conditions and NPF event variables.

Downward shortwave solar radiation K↓ (W m^{-2})										
Site	**a_N* (W^{-1} m^2)**	**R^2**	**p**	**a_G**	**R^2**	**p**	**a_J* (W^{-1} m^2)**	**R^2**	**p**	**Average**
UKRU*	**1.21E-03**	0.94	<0.001	6.53E-05	0.11	-	**6.28E-04**	0.93	<0.001	443
UKUB*	**6.81E-04**	0.90	<0.001	-8.26E-05	0.10	-	1.49E-04	0.19	-	448
UKRO*	**8.69E-04**	0.98	<0.001	-7.75E-06	0.00	-	**2.66E-04**	0.64	<0.005	464
DENRU	**2.22E-03**	0.88	<0.001	4.24E-04	0.20	-	**1.38E-03**	0.64	<0.001	115
DENUB	**1.87E-03**	0.91	<0.001	1.47E-04	0.03	-	8.98E-04	0.48	<0.01	115
DENRO	**2.46E-03**	0.95	<0.001	1.27E-04	0.01	-	**6.77E-04**	0.50	<0.005	117
GERRU	**2.87E-03**	0.98	<0.001	**9.88E-04**	0.72	<0.01	**1.45E-03**	0.81	<0.001	130
GERUB	**3.18E-03**	0.97	<0.001	**7.28E-04**	0.51	<0.005	**1.53E-03**	0.69	<0.001	114
GERRO	**2.40E-03**	0.95	<0.001	-5.89E-04	0.09	-	**9.95E-04**	0.59	<0.005	114
FINRU	**2.63E-03**	0.76	<0.001	**1.01E-03**	0.57	<0.01	**2.04E-03**	0.82	<0.001	91.5
FINUB	1.38E-03	0.37	-	1.81E-04	0.08	-	8.99E-04	0.25	-	111
FINRO	**1.76E-03**	0.59	<0.005	9.15E-04	0.34	<0.005	4.45E-04	0.03	-	114
SPARU	3.46E-04	0.35	<0.05	5.68E-04	0.13	-	**1.97E-03**	0.74	<0.001	162
SPAUB	**5.92E-04**	0.58	<0.05	6.98E-04	0.23	-	**1.58E-03**	0.81	<0.001	180
GRERU	**4.10E-04**	0.52	<0.001	**7.14E-04**	0.55	<0.001	-6.30E-04	0.05	-	201
GREUB	3.49E-04	0.31	-	-1.10E-04	0.02	-	8.97E-04	0.34	<0.05	183

* Global solar irradiation measurements in kJ m^{-2}

Relative Humidity (%)										
Site	**a_N* (%$^{-1}$)**	**R^2**	**p**	**a_G**	**R^2**	**p**	**a_J* (%$^{-1}$)**	**R^2**	**p**	**Average**
UKRU	**-5.89E-02**	0.85	<0.001	1.69E-03	0.02	-	**-3.35E-02**	0.85	<0.001	79.7
UKUB	**-3.42E-02**	0.94	<0.001	8.23E-03	0.24	-	-5.66E-03	0.19	-	75.3
UKRO	**-5.09E-02**	0.85	<0.001	7.03E-03	0.25	-	-1.49E-02	0.46	<0.05	74.5
DENRU	**-3.90E-02**	0.95	<0.001	**9.42E-03**	0.74	<0.001	5.45E-04	0.00	-	75.7
DENUB	**-3.14E-02**	0.94	<0.001	3.64E-03	0.06	-	2.57E-03	0.00	-	75.7
DENRO	**-3.64E-02**	0.95	<0.001	-1.21E-02	0.22	-	-3.91E-03	0.10	-	75.7
GERRU	**-5.08E-02**	0.88	<0.001	**-1.30E-02**	0.72	<0.001	**-2.46E-02**	0.91	<0.001	81.9
GERUB	**-5.35E-02**	0.86	<0.001	**-6.34E-03**	0.67	<0.001	**-2.25E-02**	0.86	<0.001	78.7
GERRO	**-2.83E-02**	0.90	<0.001	3.98E-03	0.05	-	**-1.72E-02**	0.81	<0.001	78.7
FINRU	**-4.48E-02**	0.94	<0.001	**-7.07E-03**	0.65	<0.001	**-2.16E-02**	0.87	<0.001	80.1
FINUB	**-5.89E-02**	0.95	<0.001	1.04E-02	0.26	-	-6.52E-03	0.18	-	76.5
FINRO	**-3.34E-02**	0.92	<0.001	-1.47E-03	0.01	-	7.39E-03	0.10	-	71.1
SPARU	**-1.54E-02**	0.90	<0.001	-4.67E-03	0.08	-	-7.12E-03	0.14	-	66.4
SPAUB	**-4.84E-02**	0.93	<0.001	**2.43E+02**	0.50	<0.01	-9.83E-03	0.19	-	69.2
GRERU	-7.72E-03	0.22	-	1.06E-02	0.06	-	-1.83E-01	0.15	-	70.0
GREUB	**-1.42E-02**	0.62	<0.001	2.83E-03	0.06	-	4.85E-04	0.00	-	60.5

Temperature (ºC)										
Site	**a_N* (ºC^{-1})**	**R^2**	**p**	**a_G**	**R^2**	**p**	**a_J* (ºC^{-1})**	**R^2**	**p**	**Average**
UKRU	**1.10E-01**	0.93	<0.001	**7.85E-02**	0.94	<0.001	**8.72E-02**	0.84	<0.001	10.6
UKUB	**9.04E-02**	0.98	<0.001	**1.39E-01**	0.96	<0.001	**6.34E-02**	0.73	<0.005	11.8
UKRO	**8.22E-02**	0.98	<0.001	**3.51E-02**	0.52	<0.05	4.32E-02	0.44	<0.05	12.1
DENRU	**6.68E-02**	0.83	<0.001	1.54E-02	0.08	-	**6.68E-02**	0.92	<0.001	9.80
DENUB	**2.50E-02**	0.45	<0.05	2.40E-02	0.33	-	3.05E-02	0.45	<0.05	9.82
DENRO	**6.64E-02**	0.88	<0.001	3.51E-03	0.00	-	**2.96E-02**	0.58	<0.005	10.0
GERRU	**7.27E-02**	0.92	<0.001	**5.65E-02**	0.92	<0.001	**5.37E-02**	0.93	<0.001	10.3
GERUB	**8.20E-02**	0.93	<0.001	**3.38E-02**	0.62	<0.001	**4.28E-02**	0.54	<0.005	11.1
GERRO	**5.08E-02**	0.89	<0.001	-3.33E-03	0.00	-	1.61E-02	0.11	-	11.1
FINRU	-2.01E-02	0.17	-	**1.13E-01**	0.79	<0.001	**4.27E-02**	0.72	<0.001	4.79
FINUB	-4.21E-03	0.00	-	**7.42E-02**	0.83	<0.001	1.67E-02	0.28	-	6.52
FINRO	**6.24E-02**	0.65	<0.005	**9.28E-02**	0.87	<0.001	-1.09E-02	0.05	-	7.72
SPARU	-2.51E-02	0.41	<0.05	**1.23E-01**	0.92	<0.001	**9.11E-02**	0.71	<0.001	13.9
SPAUB	-3.43E-03	0.02	-	**6.67E-02**	0.66	<0.005	1.18E-02	0.08	-	18.2
GRERU	**-4.66E-02**	0.75	<0.001	**1.74E-01**	0.75	<0.001	-9.45E-02	0.47	<0.05	18.2
GREUB	-1.00E-02	0.25	-	**4.67E-02**	0.62	<0.005	-2.85E-02	0.20	-	17.6

Wind Speed (m s^{-1})										
Site	**a_N* (m^{-1} s)**	**R^2**	**p**	**a_G**	**R^2**	**p**	**a_J* (m^{-1} s)**	**R^2**	**p**	**Average**
UKRU	5.72E-02	0.20	-	-3.04E-02	0.07	-	6.87E-03	0.00	-	3.96
UKUB	**1.72E-01**	0.87	<0.001	**-1.91E-01**	0.71	<0.001	3.56E-03	0.00	-	4.16
UKRO	6.34E-02	0.19	-	3.21E-02	0.02	-	7.28E-02	0.45	<0.005	4.14
DENRU	**1.08E-01**	0.88	<0.001	**-2.33E-01**	0.74	<0.001	1.28E-01	0.44	<0.01	4.17
DENUB	**1.50E-01**	0.90	<0.001	-3.33E-02	0.10	-	8.31E-02	0.19	-	4.17
DENRO	**1.65E-01**	0.89	<0.001	-1.51E-01	0.49	<0.001	9.08E-03	0.00	-	4.16
GERRU	**-1.06E-01**	0.57	<0.005	**-2.26E-01**	0.83	<0.001	-5.32E-03	0.00	-	2.58
GERUB	**-1.27E-01**	0.52	<0.01	**-1.41E-01**	0.60	<0.005	-3.32E-02	0.04	-	2.33
GERRO	**-2.40E-01**	0.56	-	-2.54E-01	0.38	-	-1.30E-01	0.22	-	2.33
FINRU	**1.62E-01**	0.63	<0.005	-1.29E-01	0.16	<0.05	7.99E-02	0.07	-	1.31
FINUB	-3.17E-02	0.08	-	7.26E-02	0.20	<0.05	-9.74E-02	0.17	-	3.43
FINRO	**8.62E-02**	0.51	<0.05	-1.60E-01	0.32	<0.05	-1.86E-01	0.32	-	4.26
SPARU	-2.20E-02	0.02	-	3.80E-01	0.31	-	5.74E-02	0.02	-	0.94
SPAUB	**2.90E-01**	0.93	<0.001	7.71E-02	0.24	-	-5.90E-02	0.05	-	2.05
GRERU	**4.37E-02**	0.54	<0.001	1.01E-01	0.36	<0.005	1.73E-03	0.00	-	6.06
GREUB	-1.13E-01	0.47	<0.01	**-1.88E-01**	0.50	<0.005	-3.78E-02	0.01	-	1.87

Atmospheric Pressure (mbar)										
Site	**a_N* ($mbar^{-1}$)**	**R^2**	**p**	**a_G**	**R^2**	**p**	**a_J* ($mbar^{-1}$)**	**R^2**	**p**	**Average**
UKRU	**4.26E-02**	0.83	<0.005	**3.93E-02**	0.58	<0.005	2.95E-02	0.47	<0.05	1007.7
UKUB	**1.90E-02**	0.50	-	1.17E-02	0.05	<0.05	4.16E-03	0.04	-	1011.7
UKRO	**6.33E-02**	0.95	<0.001	-1.21E-01	0.40	-	-2.98E-02	0.17	-	1012
GERRU	**5.10E-02**	0.97	-	**8.95E-02**	0.85	<0.001	2.16E-02	0.21	-	1007.0
GERUB	**6.27E-02**	0.97	-	**4.00E-02**	0.76	-	2.00E-02	0.37	<0.05	995.5
GERRO	**4.57E-02**	0.79	-	-9.61E-02	0.43	-	-2.80E-02	0.21	-	995.5
FINRU	**3.46E-02**	0.88	<0.001	**2.90E-02**	0.57	<0.001	1.05E-02	0.14	-	985.1
FINUB	**2.61E-02**	0.55	<0.005	-3.57E-03	0.02	-	4.38E-03	0.05	-	1004.4
FINRO	**4.91E-02**	0.70	-	-2.67E-02	0.17	-	1.43E-02	0.26	-	1008.8
SPARU	-2.02E-02	0.09	-	4.79E-02	0.14	-	2.89E-02	0.08	-	939.3
SPAUB	-2.83E-02	0.44	<0.05	1.86E-02	0.08	-	1.68E-02	0.21	-	1006.3
GRERU	6.00E-02	0.46	<0.001	-1.50E-01	0.73	-	8.14E-02	0.33	-	1014.5
GREUB	9.42E-03	0.10	<0.05	-1.00E-01	0.71	-	1.58E-02	0.04	-	1015.7

Table 4: Normalised gradients (non-normalised for growth rate), R^2 and p-values (- for values >0.05) for the relation between atmospheric composition variables and NPF event variables.

SO_2 (μg m^{-3})										
Site	**a_N* (μg^{-1} m^3)**	**R^2**	**p**	**a_G**	**R^2**	**p**	**a_J* (μg^{-1} m^3)**	**R^2**	**p**	**Average**
UKRU	-1.97E-01	0.38	<0.05	-6.17E-02	0.02	-	3.30E-01	0.06	-	1.64
UKUB	**-2.57E-01**	0.62	<0.001	1.93E-02	0.00	-	4.18E-01	0.40	-	2.04
UKRO	**-1.03E-01**	0.82	<0.001	6.90E-02	0.34	<0.01	**8.43E-02**	0.77	<0.001	7.46
DENRU	**-9.77E-01**	0.53	<0.05	2.84E+00	0.37	-	4.38E-01	0.09	-	0.52
DENRO	**-4.20E-01**	0.91	<0.001	**6.42E-01**	0.54	<0.005	**5.66E-01**	0.62	<0.001	0.97
FINRU	-5.66E-01	0.05	-	-1.42E+00	0.19	-	-6.30E-02	0.00	-	0.09
SPARU	**-3.62E-01**	0.74	<0.001	-1.33E-01	0.02	-	-3.55E-02	0.01	-	0.95
SPAUB	-2.93E-02	0.04	-	**4.12E-01**	0.59	-	1.07E-01	0.29	-	1.99

NO_x or NO_2 (ppb)										
Site	**a_N* (ppb^{-1})**	**R^2**	**p**	**a_G**	**R^2**	**p**	**a_J* (ppb^{-1})**	**R^2**	**p**	**Average**
UKRU	**-4.99E-02**	0.67	<0.005	**4.52E-02**	0.58	<0.05	**-4.51E-02**	0.70	<0.005	11.7
UKUB	**-8.75E-03**	0.83	<0.001	-3.97E-04	0.00	-	-1.09E-02	0.43	<0.05	53.6
UKRO	**-3.22E-03**	0.72	<0.001	1.44E-03	0.39	<0.05	**2.19E-03**	0.66	<0.001	299
DENRU	-9.41E-02	0.43	<0.005	-4.89E-03	0.00	<0.001	**-6.47E-02**	0.55	<0.01	5.42
DENUB	**-4.99E-02**	0.68	<0.001	2.85E-02	0.26	-	8.55E-04	0.00	-	10.5
DENRO	**-5.10E-03**	0.75	<0.001	**1.10E-02**	0.69	<0.001	**8.33E-03**	0.88	<0.001	68.5
FINRU	**-7.27E-01**	0.54	<0.001	-2.74E-01	0.11	-	1.95E-01	0.05	-	0.72
FINRO	**-6.24E-03**	0.68	<0.001	1.70E-03	0.12	-	3.25E-03	0.03	-	88.1
SPARU*	-1.53E-02	0.05	-	2.54E-02	0.01	-	1.25E-01	0.21	-	3.26
SPAUB*	**-2.59E-02**	0.62	<0.005	**2.23E-02**	0.70	<0.001	2.57E-03	0.01	-	31.4
GRERU*	3.01E-01	0.19	-	**-1.40E+00**	0.75	<0.001	5.23E-01	0.13	-	0.52

***** NO_2 measurements

O_3 (ppb)										
Site	**a_N* (ppb^{-1})**	**R^2**	**p**	**a_G**	**R^2**	**p**	**a_J* (ppb^{-1})**	**R^2**	**p**	**Average**
UKRU	**2.27E-02**	0.88	<0.001	**-4.89E-02**	0.53	<0.005	-3.53E-03	0.01	-	54.4
UKUB	**1.37E-02**	0.87	<0.001	**-3.45E-02**	0.68	<0.001	-5.95E-03	0.05	-	39.3
UKRO	**7.46E-02**	0.95	<0.001	-1.06E-02	0.09	-	**-2.44E-02**	0.63	<0.005	16.2
DENRU	**4.97E-02**	0.92	<0.001	-1.32E-02	0.15	-	1.23E-02	0.08	-	30.1
DENUB	**5.85E-02**	0.84	<0.001	**-1.69E-02**	0.58	-	2.77E-02	0.32	<0.05	28.2
DENRO	**6.42E-02**	0.51	<0.05	1.39E-02	0.03	-	**3.24E-02**	0.91	<0.05	31.1
FINRU	**6.76E-02**	0.77	<0.05	**-4.23E-02**	0.60	-	3.92E-02	0.37	<0.05	27.4
FINRO	**2.38E-02**	0.91	<0.001	6.11E-03	0.24	-	-1.83E-02	0.29	-	37.1
SPARU	1.57E-02	0.02	-	4.34E-02	0.11	-	1.31E-02	0.31	-	75.9
SPAUB	7.99E-03	0.38	<0.05	-5.83E-03	0.30	-	-1.13E-03	0.01	-	54.9
GRERU	7.55E-03	0.04	-	3.68E-02	0.17	-	-3.01E-02	0.15	-	49.5

Particulate Organic Carbon (μg m^{-3})										
Site	**a_N* (μg^{-1} m^3)**	**R^2**	**p**	**a_G**	**R^2**	**p**	**a_J* (μg^{-1} m^3)**	**R^2**	**p**	**Average**
UKRU	-3.30E-02	0.00	-	1.13E+00	0.42	<0.005	2.13E-01	0.16	-	1.96
UKUB	**-2.76E-01**	0.59	<0.005	**6.63E-01**	0.58	<0.05	**2.19E-01**	0.55	<0.05	3.63
UKRO	**-3.78E-01**	0.89	<0.001	**8.12E-01**	0.57	<0.005	**4.60E-01**	0.75	<0.001	6.24
DENRU	**-4.44E-01**	0.75	<0.001	2.24E-01	0.11	-	**-3.17E-01**	0.68	<0.01	1.48
DENRO	-7.80E-02	0.11	-	**1.10E+00**	0.77	<0.005	**4.02E-01**	0.81	<0.005	2.59
GERRU	-1.26E-01	0.24	-	1.35E-01	0.09	-	3.14E-02	0.03	-	2.18
FINRU	2.27E-02	0.00	-	**3.39E-01**	0.60	<0.005	-3.46E-01	0.16	-	1.78
GRERU	-2.08E-01	0.11	-	7.87E-01	0.41	<0.05	8.94E-01	0.11	-	1.58

Sulphate (μg m^{-3})										
Site	**a_N* (μg^{-1} m^3)**	**R^2**	**p**	**a_G**	**R^2**	**p**	**a_J* (μg^{-1} m^3)**	**R^2**	**p**	**Average**
UKRU[1]	**-2.62E-01**	0.57	<0.001	**7.34E-01**	0.77	<0.001	7.99E-01	0.44	<0.05	1.97
UKUB[1]	**-3.57E-01**	0.89	<0.001	9.28E-01	0.44	<0.01	9.72E-01	0.16	-	1.58
UKRO[1]	-6.05E-02	0.24	-	3.04E-01	0.34	<0.05	-6.22E-02	0.04	-	1.98
DENRU[2]	-7.81E-01	0.34	<0.05	**1.02E+00**	0.60	<0.05	**-1.03E+00**	0.63	<0.01	0.52
DENRO[2]	-8.23E-01	0.28	-	1.99E+00	0.22	-	2.82E-01	0.12	-	0.55
GERRU[1]	-3.37E-02	0.00	-	5.89E-01	0.11	-	-4.89E-02	0.01	-	0.92
FINRU[3]	**-1.18E+00**	0.65	<0.001	2.35E-01	0.09	-	-2.53E-01	0.17	-	1.02

[1] Measurements in PM_{10}
[2] Measurements in $PM_{2.5}$
[3] Measurements in PM_1

Condensation Sink (s^{-1})										
Site	**a_N* (s)**	**R^2**	**p**	**a_G**	**R^2**	**p**	**a_J* (s)**	**R^2**	**p**	**Average**
UKRU	**-2.28E+02**	0.72	<0.001	**2.64E+02**	0.60	<0.001	7.58E+01	0.22	-	3.38E-03
UKUB	**-1.66E+02**	0.78	<0.001	2.49E+02	0.41	<0.05	1.73E+02	0.35	<0.05	7.41E-03
UKRO	**-4.03E+01**	0.75	<0.001	2.33E+01	0.18	-	**8.94E+01**	0.91	<0.001	2.12E-02
DENRU	**-4.48E+01**	0.91	<0.001	6.90E+01	0.49	<0.05	5.37E+01	0.24	-	9.46E-03
DENUB	**-3.78E+01**	0.75	<0.001	3.58E+01	0.25	-	1.55E+01	0.56	<0.005	1.42E-02
DENRO	**-1.06E+01**	0.73	<0.001	**2.53E+01**	0.56	<0.005	**2.72E+01**	0.79	<0.001	3.10E-02
GERRU	**1.54E+02**	0.86	<0.001	**1.33E+02**	0.56	<0.001	**6.67E+01**	0.63	<0.001	7.02E-03
GERUB	**3.59E+01**	0.56	<0.005	3.63E+01	0.17	-	**4.74E+01**	0.75	<0.001	9.11E-03
GERRO	3.89E+01	0.22	<0.05	-2.21E+01	0.03	<0.005	3.54E+01	0.45	<0.005	1.20E-02
FINRU	**-1.80E+02**	0.59	<0.005	**4.01E+02**	0.74	<0.001	4.98E+01	0.10	-	2.32E-03
FINUB	**-1.51E+02**	0.63	<0.005	8.14E+01	0.31	-	2.01E+02	0.41	<0.05	6.34E-03
FINRO	**-6.99E+01**	0.77	<0.001	-1.56E+01	0.05	-	**2.42E+02**	0.83	<0.001	8.96E-03
SPARU	**-2.15E+02**	0.65	<0.005	1.86E+01	0.00	-	8.60E+01	0.47	<0.05	5.49E-03
SPAUB	**-1.18E+02**	0.65	<0.005	3.74E+01	0.38	<0.05	**9.51E+01**	0.52	<0.01	1.00E-02
GRERU	4.33E+00	0.00	-	**2.86E+02**	0.70	<0.001	**1.77E+02**	0.56	<0.005	4.66E-03
GREUB	**1.64E+02**	0.65	<0.001	9.31E+01	0.28	<0.05	**1.73E+02**	0.83	<0.001	7.55E-03

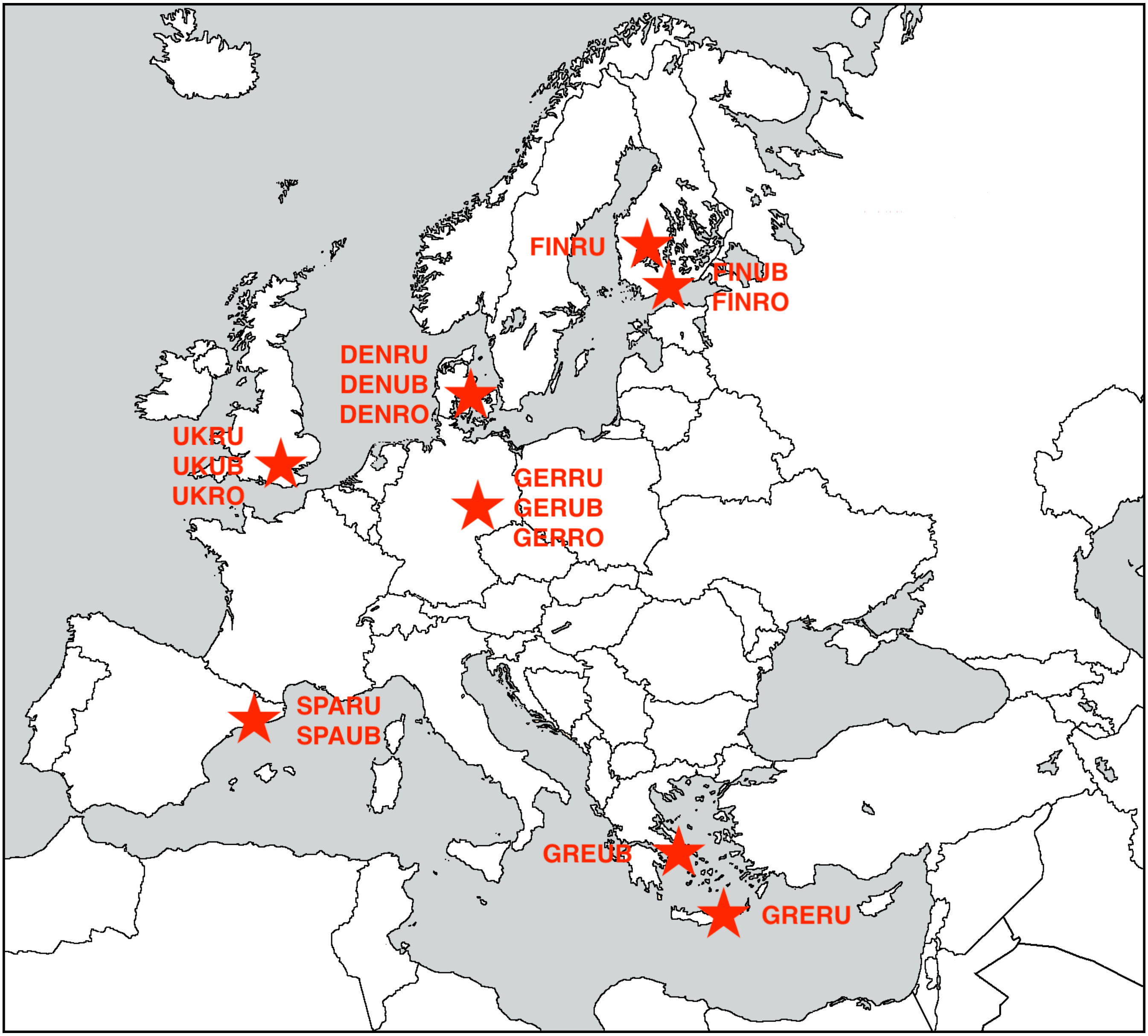

Figure 1: Map of the sites of the present study.

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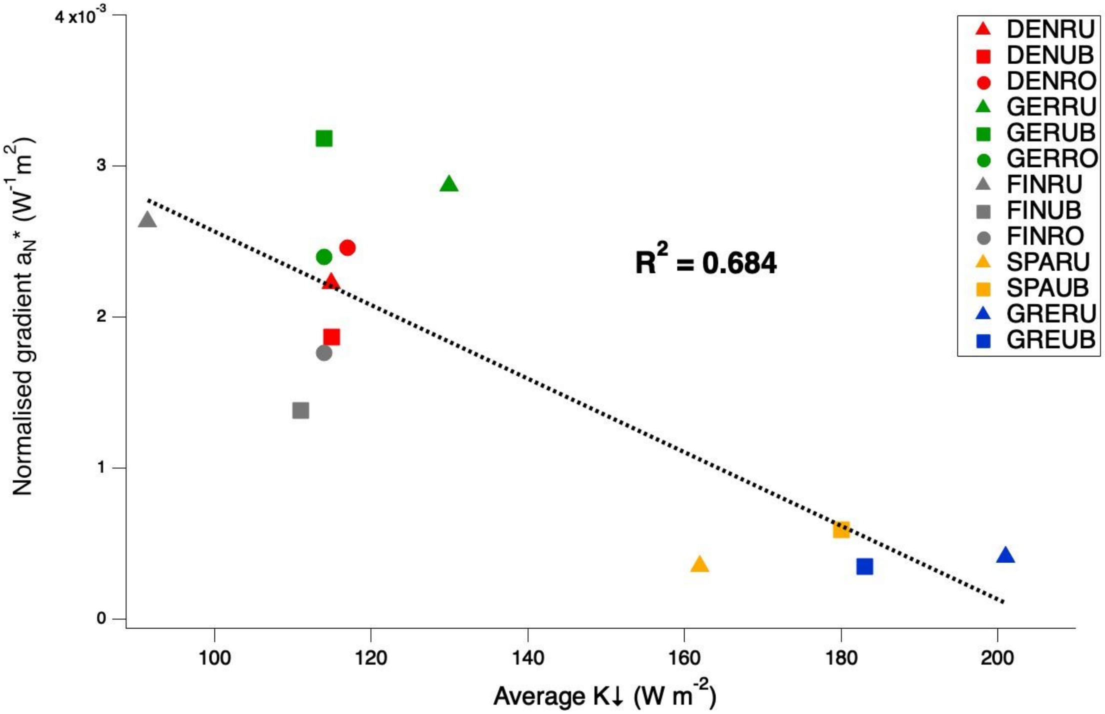

Figure 2: Relationship of average downward incoming solar radiation (K↓) and normalised gradients a_N^*.

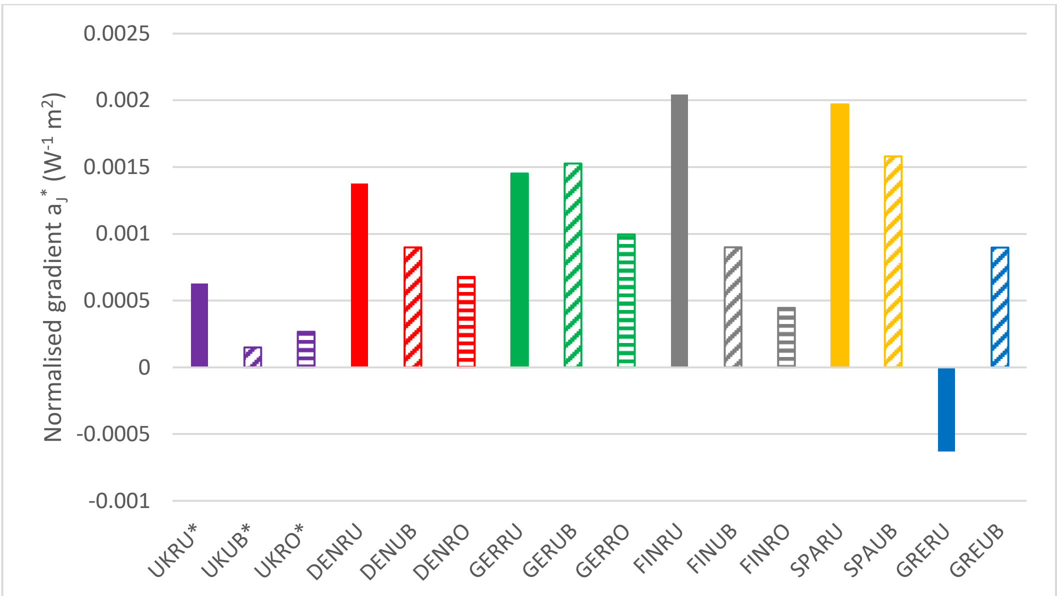

Figure 3: Normalised slopes a_J^* for K↓ (*UK sites are calculated with solar irradiance).

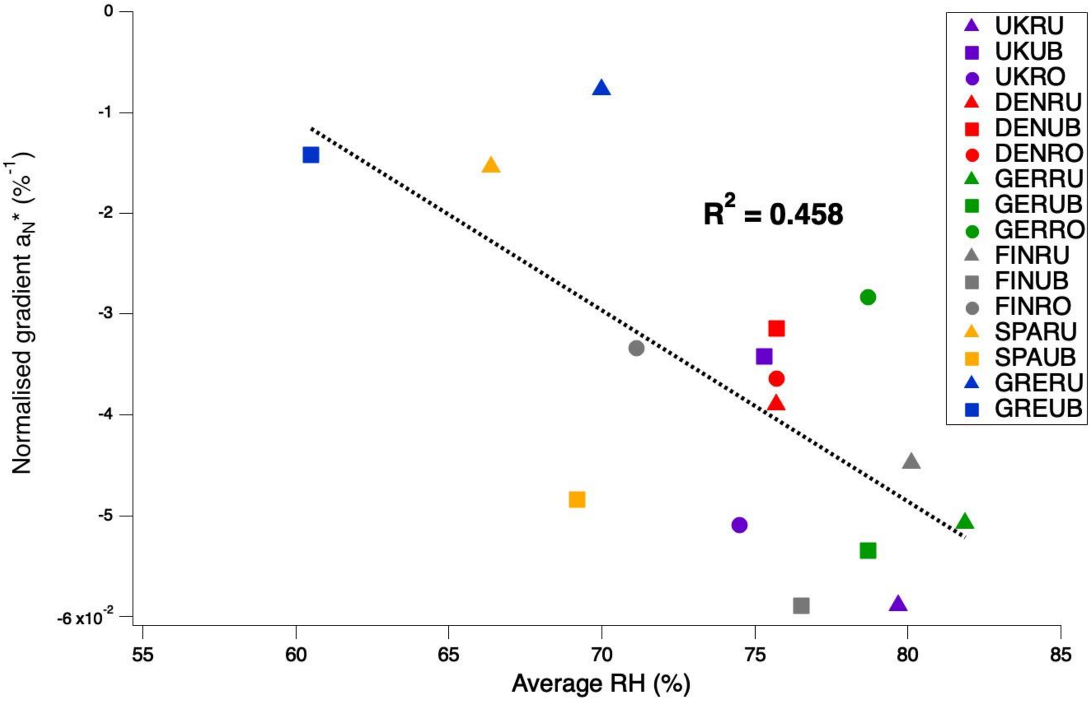

Figure 4: Relationship of average relative humidity and normalised gradients a_N^*.

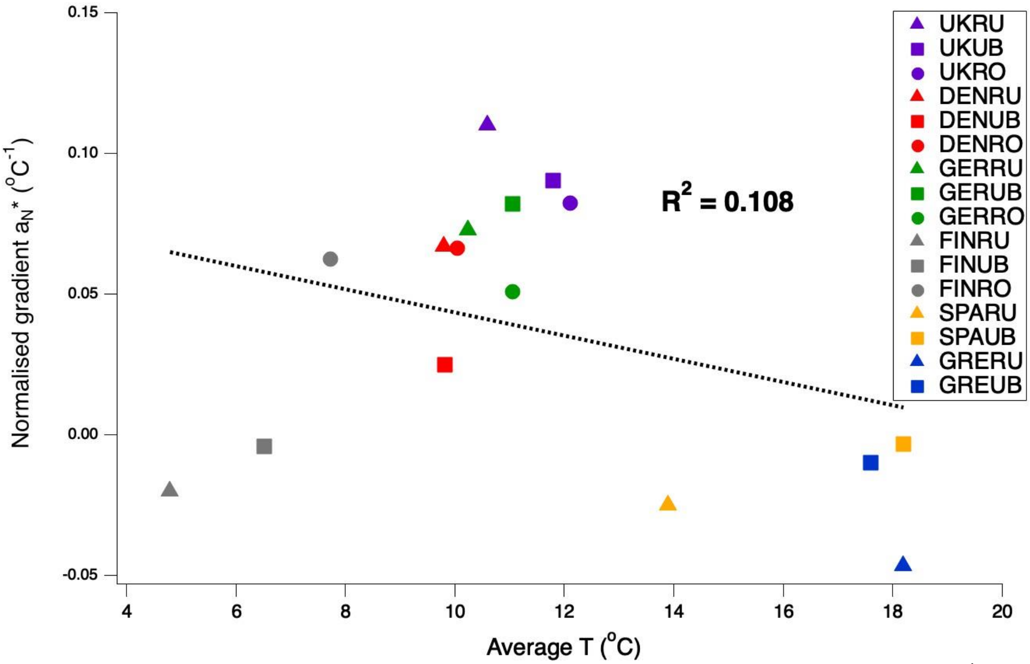

Figure 5: Relationship of average temperature and normalised gradients a_N^*.

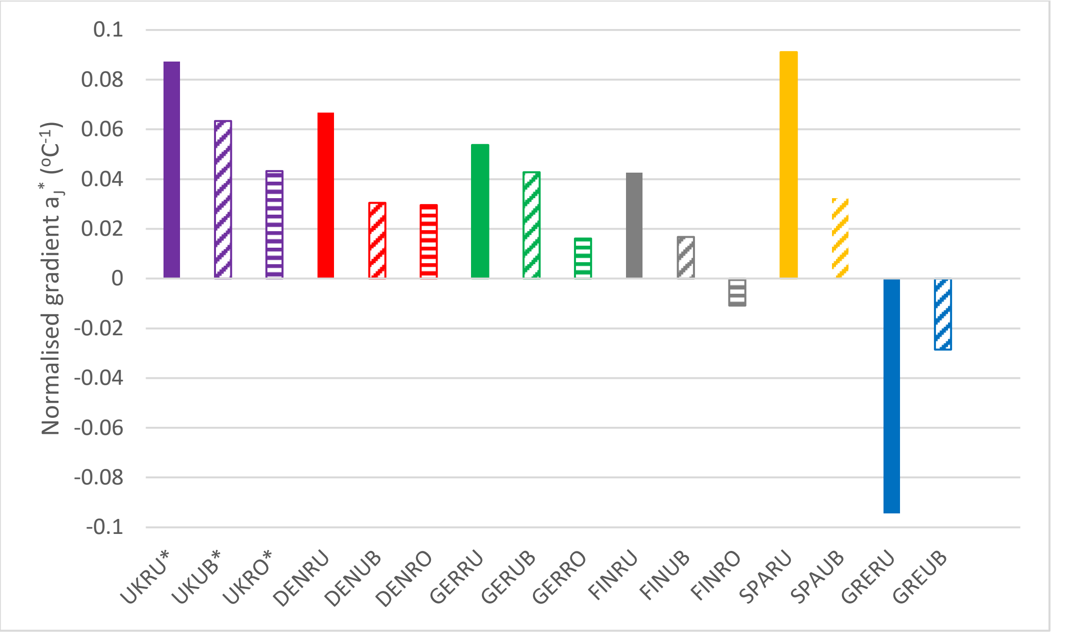

Figure 6: Normalised gradients a_J^* for temperature**.**

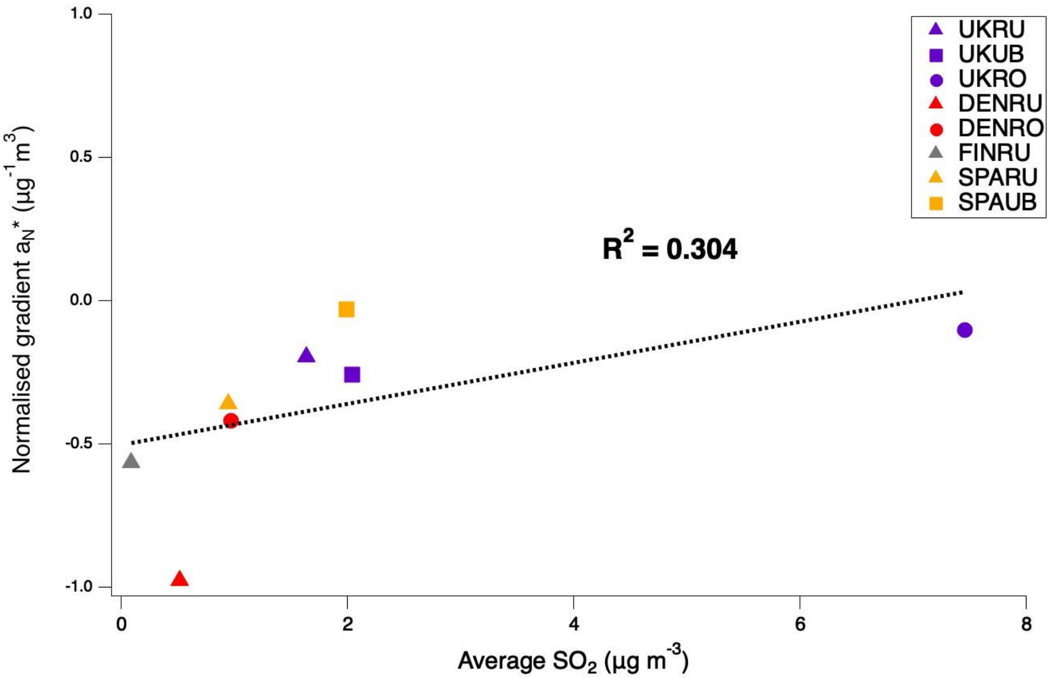

Figure 7a: Relationship of average SO_2 concentrations and normalised gradients a_N^*.

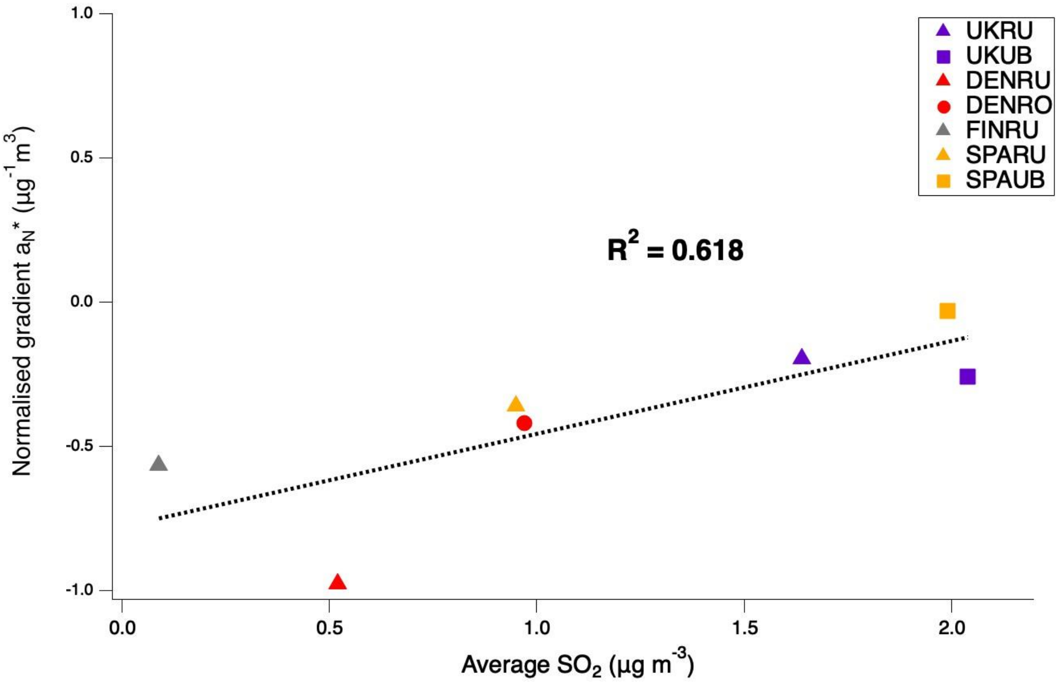

Figure 7b: Relationship of average SO_2 concentrations and normalised gradients a_N^* (UKRO not included).

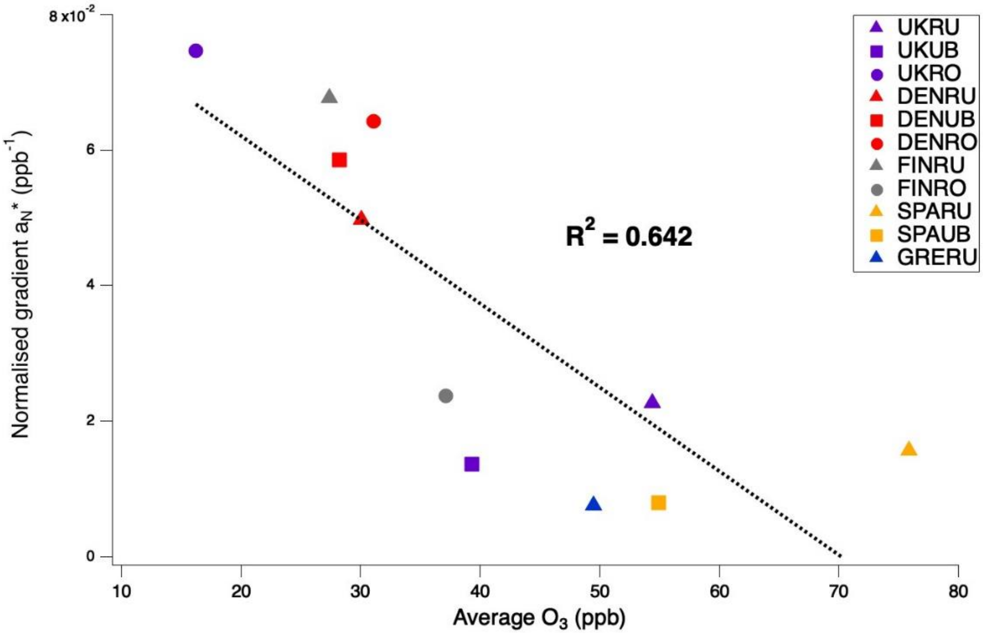

Figure 8: Relationship of average O_3 concentrations and normalised gradients a_N^*.