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

Author(s): Dimitrios Bousiotis et al.

RESPONSE TO REVIEWERS

Referee #1

General comments

The current version of the manuscript has improved. Readability is better. The authors followed most of the suggestions. However, there are still few minor aspects I encourage to improve.

The readability of the abstract and introduction could still be polished.

RESPONSE: We have done our best to improve these sections.

I am not convinced by the use of term “NPF probability”. You are probably aware that some readers might only (or first) check the abstract and conclusion. Using “NPF probability” term may be confusing. If you still think this is something you would like to keep, I suggest be more specific what it actually represents when discussing it in conclusion.

RESPONSE: As the term “NPF probability” seems to cause a lot of misunderstandings it was replaced by the more conventional “NPF frequency” throughout the manuscript. Small changes were also made in the Methods section to accommodate the change (line 238).

Please specify how the NPF frequency was calculated, and check values presented in Table 2. Which data is considered for this calculation? I think the answer was given in author’s response (AR) file but I missed mentioning of that in the main manuscript. Further, some comments are addressed in AR but these clarifications do not always appear in text. Please double check.

RESPONSE: Due to the aforementioned change (removal of the “NPF probability” term), the term “NPF frequency” is now explained in the Methods section with the inclusion of the “full dataset” as an option of the possible groups for which it is calculated (as the term “NPF frequency” was previously used when full datasets were considered). Additionally, some information about the trends and results included in the AR (pointing to previous work) were also added in the manuscript as they can help in explaining some trends (please see the response on a later comment as well).

How sure are you that NPF occurred at UK sites if you only have SMPS data available >16 nm for these locations? Please comment in the manuscript. It is also one of the limitations of the study that you could mention. What was your approach for analysis of these sites?

RESPONSE: For the extraction of the NPF events in the UK sites additional criteria were set (including the variation of particle number concentration data from 7 nm, the variations of pollutant concentrations, the condensation sink, the effect of the nearby Heathrow Airport, etc.). The text from the Methods section in the paper in which these results were first presented reads: “At this point it should be mentioned that due to the particle size range available, NPF events in which new formed particles failed to grow beyond 16.6 nm (if any) could not be identified. Bursts of new particles in the size range < 16.6 nm that were identified using the CPC data but did not appear in the SMPS dataset were ignored as their development was unknown. This type of development was rare and mainly found at the rural background site, occurring on a few days per year mainly in summer. Its main feature was the short duration of the bursts compared to event days. In the urban sites, this type of development was almost non-existent. High time resolution data for gaseous pollutants and aerosol constituents was used to identify pollution events affecting particle concentrations and these were removed from the data analysis. This analysis took account of the fact that nanoparticle emissions from Heathrow Airport affect size distributions at London sites (Harrison et al., 2018), and such primary emission influences were not included as NPF events.”

As this is a lengthy clarification, a note was added in the Methods section in the present study, mentioning the limitation and referencing the work where this is further explained:

“As the available SMPS datasets for the sites in the U.K. are for particles of diameter greater than 16 nm, additional criteria were set to ensure the correct extraction of NPF events including the variations of the particle number concentrations from a Condensation Particle Counter (CPC – measuring from 7nm), as well as of the concentrations of gaseous pollutants and aerosol constituents (please refer to the Methods section in Bousiotis et al., 2019).” (line 186)

I have noticed that frequently two other papers published/submitted by Bousiotis et al (2019, 2020) are mentioned in AR. It would be good if the authors make sure that these are also mentioned in relevant places throughout the manuscript (mentioning the issue raised by reviewers e.g.: “xx was explored in Bousiotis et al. xxxx and is not the focus in this paper” or “xx can be found in...”).

Also make sure that crucial information on the study is provided in the current manuscript without the need to often look into two other papers to get a complete picture.

RESPONSE: We thank the reviewer for this suggestion. Some references were added in the manuscript that point to results from the previous studies. Also, some crucial information found in these works were also added in several points in the manuscript, to clarify the points made (results about the seasonality of the GR and J, the variation of the temperature, CS etc.).

Table 3: please indicate in the caption what values “in bold” indicate

RESPONSE: A clarification for what the values in bold indicate was added in the captions of Tables 3, 4 and S1.

Figure 7a, 7b: make one *caption* for figure 7(a and b) and only indicate on corresponding plots “a” and “b”.

RESPONSE: The figure was updated with a single caption. Also, the plot 4b was removed from the Figure Legends table as it was moved to the SI

Referee #2

This work compiles already published results from 16 sites located in six European cities. Within this huge task, the effort is to identify relationships between meteorological variables, gas phase composition and aerosol organic content with key NPF variables such as NPF frequency (the authors name it NPF probability), growth rate and formation rate. Several findings within this work justify publication; two most striking is the nonlinear relation of temperature with NPF probability and the fact that increased solar irradiance reduces the probability for NPF.

Some issues require attention though.

Starting from most important to least severe

The authors have chosen to use only Ia events and as a result the probability shown in Table 2 is several factors smaller than those reported in literature for the same sites. However, from a brief search, the difference can be up to a factor of six. It is well understood why the authors made such a choice as formation and growth rates can be calculated reliably from these type of events only. How does this choice reflect to the results shown? I strongly support that for one site (e.g. Hyytiälä) an intercomparison is carried out to indicate to the reader the tentative differences. My recommendation is to do so only for NPF probability. This is critical as most studies in the end

classify NPF as events, undefined and non-events lumping Ia, Ib and II classes into one. The authors should add to the caption of Table 1 the fact that only Ia events are considered.

RESPONSE: According to the results from the analysis of NPF events at the sites of the study it was found that the NPF events that did not meet the criteria for class Ia were up to double the number of those that are characterised as class Ia. Thus, in the methods section the following text was added:

“As only class Ia events were only considered, it is expected that the frequency of the events calculated should be lower than that expected if all types of events were included. This could result in values up to one third of those anticipated if all classes of events were considered. For the extent of this variation please refer to Bousiotis et al., (2019; 2020) in which there is an extended analysis of the NPF events for each site, including the special cases of NPF events that do not comply with the criteria set for class Ia.” (line 316).

Additionally, the text was updated for the Table mentioning that the statistics refer to class Ia NPF events. (We assume that the caption of Table 2 is the one that needs updating).

How coarse is the time resolution of OC and sulfate measurements examined in this study? The first impression for the former is that they are derived from the thermal optical method. For the latter AMS is mentioned. However, AMS is typically used for short-term campaigns and this is a multi-year study. Do the authors mean ACSM? If the resolution of these measurements is coarser than 1 h, are they reliable to be used in NPF studies? The authors should clarify the time resolution of these measurements, both OC and SO₄, in the manuscript and discuss any complications. If the authors indeed use AMS measurements what fraction of the time period discussed do they cover? Also, it would be worthwhile to mention the publications that refer to these measurements.

RESPONSE: ACSM data was used (updated in the text). The measurement resolution for all the sites for which such data was used is 1 hour. Data with 3-hour resolution or more was available but was not used as it would bias the results. The note “For all the sites, the data used in the present study are of either 1-hour resolution or less. Data with coarser resolution were omitted for reliability.” was added in the Methods section for clarification (line 160). References for publications that reported the measurements of this study were added in the Site Description section.

In each pair of variables (e.g. NPF probability and RH; growth rate and temperature) presented in this work, there seems to be a norm and one (or more) site that is an exception. Since the authors cannot fully explain why (and this is perfectly understood), it is worthwhile to mention that in the abstract or the conclusions or in both.

RESPONSE: The note “though exceptions were found among the sites for all the variables studied” was added in the abstract (line 57). The note “in the majority of the sites (though exceptions were found as well, mostly in the southern sites),” was added in the Conclusions section to point out the exceptions found (line 863).

There is little relationship between RH and CS at most sites. Is this because CS was based on dried measurements and was not corrected for hygroscopic growth? This would be understood since chemical composition was lacking on most sites. Please discuss if CS was corrected for hygroscopic growth and how that affects the results presented.

RESPONSE: A note has been added in the methods that CS was not corrected for hygroscopic growth as well as for the effect this has on the results presented.

ANOVA is only valid for normally distributed populations. Have the authors tested for normality? The F-test is typically used. How did the authors treat skewed distributions? Please discuss.

RESPONSE: The Shapiro-Wilk test was used to assess the normality and the vast majority of the variables were found to have $p > 0.05$ and thus were considered as normal. This is probably due to

the removal of the extreme values (for the calculations, 90% of each dataset was kept removing the extremely high and/or low values and the possible outliers included in them). While this was not done to promote the normality of the populations but to reduce the bias from extreme values, it indirectly assisted in making the distributions normal. For the few remaining (e.g. the growth rates associated with SO₂ concentrations for UKRO) for which normality was not present, the square root of the values of the variable were considered to achieve normality and proceed to the ANOVA test. This clarification was added in the Methods section (line 244).

In the supplement, several relationships are clearly non-linear, such as the temperature-NPF probability for a few sites, but the authors insist to use a linear fit (probably for consistency). May I ask the authors to note on the supplemental graphs in which cases linearity is not followed. The authors are better aware of the statistical significance of the related graphs than the reader is. In the case of Denmark Rural (S2b), it is not evident whether the deviation from linearity is statistically significant or not.

RESPONSE: As it is expected that most readers will not read the SI, such deviations are discussed in the text (one example is the case of the Danish rural site mentioned which is discussed in the text). A linear relationship is not always the best to describe the relationships found, but indeed was chosen for consistency (now discussed in the text – line 282). A metric for the consistency of the linearity can be given by the R² and the p-values (e.g. when R² is low then the linearity is not consistent, at least statistically). Apart from that, it is unknown even to the authors whether a trend that starts at the extreme values of a variable (e.g. the decline found in the Danish rural site with the NPF frequency at high temperatures) would consistently continue if the temperature increases further or it is an artefact, and thus it was decided not to be further discussed in detail, apart from the mentions made in the text (as only speculation can be made).

Please define in the methods section what is weak, strong and very strong correlation in this work. It will assist the reader further.

RESPONSE: As there is no specific mention for the relationships in the Methods section, the definition of weak, strong and very strong correlations is mentioned in the first reference to the coefficient of determination (line 334).

The effect of SO₂ on NPF that the authors are discussing in Section 3.2.1 has been presented before. Please check the references below. These works relate particle acidity to NPF.

Experimentally it has also been verified at the site named GRERU in this work.

RESPONSE: The works suggested are mentioned and referenced in the SO₂ section (line 548).

Line 247. “The remaining data” is better use of English than the “data left”

RESPONSE: Text changed to “remaining data” (now line 278).

If the authors prefer the term NPF probability it is fine. But please use it throughout the manuscript. The caption in Table 2 is a bit confusing.

RESPONSE: The “NPF probability” was addressed in an earlier comment. The caption in Table 2 was updated.

References

Jung, J., P. J. Adams, and S. N. Pandis (2006), Simulating the size distribution and chemical composition of ultrafine particles during nucleation events, *Atmos. Environ.*, 40, 2248–2259, doi:10.1016/j.atmosenv.2005.09.082.

Jung, J. G., S. N. Pandis, and P. J. Adams (2008), Evaluation of nucleation theories in a sulfur-rich environment, *Aerosol Sci. Technol.*, 42, 495–504, doi:10.1080/02786820802187085.

1 **The Effect of Meteorological Conditions and Atmospheric**
2 **Composition in the Occurrence and Development of New Particle**
3 **Formation (NPF) Events in Europe**
4

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42 ABSTRACT

43 Although new particle formation (NPF) events have been studied extensively for some decades, the
44 mechanisms that drive their occurrence and development are yet to be fully elucidated. Laboratory
45 studies have done much to elucidate the molecular processes involved in nucleation, but this
46 knowledge has yet to be conclusively linked to NPF events in the atmosphere. There is great
47 difficulty in successful application of the results from laboratory studies to real atmospheric
48 conditions, due to the diversity of atmospheric conditions and observations found, as NPF events
49 occur almost everywhere in the world without always following a clearly defined trend of
50 frequency, seasonality, atmospheric conditions or event development.

51 The present study seeks common features in nucleation events by applying a binned linear
52 regression over an extensive dataset from 16 sites of various types (combined dataset of 85 years
53 from rural and urban backgrounds as well as roadside sites) in Europe. At most sites, a clear
54 positive relation is found between the solar radiation intensity (up to $R^2 = 0.98$), temperature (up to
55 $R^2 = 0.98$) and atmospheric pressure (up to $R^2 = 0.97$) with the [probability frequency](#) of NPF events,
56 while relative humidity (RH) presents a negative relation (up to $R^2 = 0.95$) with NPF event

57 [probability frequency, though exceptions were found among the sites for all the variables studied.](#)

58 Wind speed presents a less consistent relationship which appears to be heavily affected by local
59 conditions. While some meteorological variables (such as the solar radiation intensity and RH)
60 appear to have a crucial effect on the occurrence and characteristics of NPF events, especially at
61 rural sites, it appears that their role becomes less marked when at higher average values.

62

63 The analysis of chemical composition data presents interesting results. Concentrations of almost all
64 chemical compounds studied (apart from O₃) and the Condensation Sink (CS) have a negative
65 relationship with NPF event [probabilityfrequency](#), though areas with higher average concentrations
66 of SO₂ had higher NPF event [probabilityfrequency](#). Particulate Organic Carbon (OC), Volatile
67 Organic Compounds (VOCs) and particulate phase sulphate consistently had a positive relation with
68 the growth rate of the newly formed particles. As with some meteorological variables, it appears
69 that at increased concentrations of pollutants or the CS, their influence upon NPF
70 [probabilityfrequency](#) is reduced.

71

72 1. INTRODUCTION

73 New Particle Formation (NPF) events are an important source of particles in the atmosphere
74 (Merikanto et al., 2009; Spracklen et al., 2010). ~~These which~~ are known to have adverse effects on
75 human health (Schwartz et al., 1996; Politis et al., 2008; Kim, et al., 2015), as well as affecting the
76 optical and physical properties of the atmosphere (Makkonen et al., 2012; Seinfeld and Pandis,
77 2012). While ~~they~~ NPF events occur almost everywhere in the world (Dall'Osto et al., 2018;
78 Kulmala et al., 2017; O'Dowd et al., 2002; Wiedensohler et al., 2019; Chu et al., 2019; Kerminen et
79 al., 2018), with some exceptions mentioned in the literature reported in forest (Lee et al., 2016; Pillai
80 et al., 2013; Rizzo et al., 2010) or high-elevation sites (Bae et al., 2010; Hallar et al., 2016), great
81 diversity is found in the atmospheric conditions within which they take place. ~~The M~~ many studies
82 ~~have been done in~~ conducted have included many ~~da large number of~~ different types of locations
83 (urban, traffic, regional background), around the world and differences were found in both the
84 seasonality and intensity of NPF events. ~~To an extent~~ This variability is due may be related to the
85 mix of conditions that are specific to each location, which ~~blurs~~ obscures the general understanding
86 of the conditions that are favourable for the occurrence of NPF events (Berland et al., 2017;
87 Bousiotis et al., 2020). For example, solar radiation is considered as one of the most important
88 factors in the occurrence of NPF events (Kulmala and Kerminen, 2008; Kürten et al., 2016; Pikridas
89 et al., 2015; Salma et al., 2011), as it ~~is needed for~~ drives the photochemical reactions ~~that~~ leading to
90 the formation of sulphuric acid (Petäjä et al., 2009; Cheung et al., 2013), ~~which~~ Sulphuric acid is
91 ~~considered as~~ frequently the main component of the formation and growth of the initial clusters (Iida

92 et al., 2008; Stolzenburg et al., 2020; Weber et al., 1995). Nevertheless, in many cases NPF events
93 ~~did do~~ not occur in the seasons with the highest insolation (Park et al., 2015; Vratolis et al., 2019).
94 Similarly, uncertainty exists over the effect of temperature (Yli-Juuti et al., 2020; Stolzenburg et al.,
95 2018). Higher temperatures are considered favourable for the growth of the newly formed particles
96 as increased concentrations of both Biogenic Volatile Organic Compounds (BVOCs) and
97 Anthropogenic Volatile Organic Compounds (AVOCs) (Yamada, 2013; Paasonen et al., 2013) and
98 their oxidation products (Ehn et al., 2014) ~~are associated to the~~support growth of the particles.
99 ~~Still, On the other hand, t~~The negative effect of increaseded temperatures ~~in increasing the~~
100 ~~energy upon the stability of barriers the~~molecular clusters ~~have to overcome to become stable and~~
101 ~~grow in size though~~ should not be overlooked (Kürten et al., 2018; Zhang et al., 2012). ~~This~~The
102 ~~former factor~~ appears ~~frequently to be true~~be dominant in most cases, as higher growth rates are
103 found in most cases in the local summer (Nieminen et al., 2018), although the actual importance of
104 those VOCs in the occurrence of NPF events is still not fully elucidated, with oxidation mechanisms
105 still under intense research (Tröstl et al., 2016; Wang et al., 2020). The effect of other
106 meteorological variables is even more complex, with studies presenting mixed results on the effect
107 of the wind speed and atmospheric pressure. Extreme values of those variables may be favourable
108 for the occurrence of NPF events, as they are associated with increased mixing in the atmosphere,
109 but at the same time suppress nucleation due to increased dilution of precursors (Brines et al., 2015;
110 Rimnácová et al., 2011; Shen et al., 2018; Siakavaras et al., 2016), or favour ~~them~~it due to a
111 reduced condensation sink (CS).

112

113 The effect of atmospheric composition on NPF events is also a puzzle of mixed results. While the
114 negative effect of the increased CS on the occurrence of the events is widely accepted (Kalkavouras
115 et al., 2017 ; Kerminen et al., 2004; Wehner et al., 2007), cases are found when NPF events occur
116 on days with higher CS compared to average conditions (Größ et al., 2018; Kulmala et al., 2005).
117 Sulphur dioxide (SO₂), which is one of the most important contributors to many NPF pathways, in
118 most studies was found ~~in~~at lower concentrations on NPF event days compared to average
119 conditions (Alam et al., 2003; Bousiotis et al., 2019), although there are studies that have reported
120 the opposite (Woo et al., 2001; Charron et al., 2008). Additionally, in a combined study of NPF
121 events in China, events were found to be more probable under sulphur-rich conditions rather than
122 sulphur-poor (Jayaratne et al., 2017). Similar is the case with the BVOCs and AVOCs, which
123 present great variability depending the area studied (Dai et al., 2017), and their contribution in the
124 growth of the particles is not fully understood yet. Until recently, it was considered unlikely for
125 NPF events, as they are considered in the present study (deriving from secondary formation not
126 associated with traffic related processes such as dilution of the engine exhaust), to occur within the
127 complex urban environment due to the increased presence of compounds, mainly associated with
128 combustion processes, which would suppress the survival of the newly formed particles within this
129 type of environment (Kulmala et al., 2017). Despite this, NPF events were found to occur within
130 even the most polluted areas and sometimes with high formation and growth rates (Bousiotis et al.,
131 2019; Yao et al., 2018).

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

143

144 **2. DATA AND METHODS**

145 **2.1 Site Description and Data Availability**

146 The present study uses a total of more than 85 years of hourly data from 16 sites from six countries
147 of Europe of various land usage and climates. It was considered very important that at least a rural
148 and an urban site would be available from each country to study the differences between the
149 different land usage on NPF events throughout Europe. The sites were chosen to cover the greatest
150 possible extent of the European continent, with sites from both northern, central and southern
151 Europe, as well as from western and eastern. The sites are located in the UK (London and Harwell),

152 Denmark (Copenhagen greater area), Germany (Leipzig greater area), Finland (Helsinki and
153 Hyytiälä), Spain (Barcelona and Montseny – a site in a mountainous area) and Greece (Athens and
154 Finokalia). Unfortunately, not all sites had available data for all the variables studied, which to an
155 extent may bias some of the results. An extended analysis of the typical and NPF event conditions,
156 seasonal variations and trends at these sites for the same period is found in other studies (Bousiotis
157 et al., 2019; 2020). A list of the available data and a brief description for each site is found in Table
158 1 (for the ease of reading the sites are named by the country of the site followed by the last two
159 letters which refer to the type of site, being RU for rural/regional background, UB for urban
160 background and RO for roadside site), while a map of the sites is found in Figure 1. [For all the sites,](#)
161 [the data used in the present study are of either 1-hour resolution or less. Data with coarser](#)
162 [resolutions were omitted for reliability.](#)
163 [Most of the data used in this analysis were also published in previous studies. The data from the UK](#)
164 [were published in Bousiotis et al., \(2019; 2020\), while parts of it were also published in Beddows et](#)
165 [al., \(2015; 2019\). The data for the German sites and parts of the data from UK, Denmark and](#)
166 [Finland were also published in von Bismarck et al., \(2013; 2014; 2015\). Parts of the measurements](#)
167 [for the Spanish sites were used in Carnerero et al., \(2019\) and Brines et al., \(2015\). The data for the](#)
168 [Greek rural background site were published in Kalivitis et al., \(2019\). Finally, the data for the Greek](#)
169 [urban background site were extracted from the European database \(EBAS – ebas.nilu.no\) and to the](#)
170 [authors' knowledge has not been used in previous studies. Additional data for some of the sites](#)
171 [were provided from their respective operators and were also not used in the past.](#)

172

173 2.2 Methods

174 2.2.1 NPF events selection

175 NPF events were selected using the method proposed by Dal Maso et al (2005). An NPF event is
176 identified by the appearance of a new mode or particles in the nucleation mode (smaller than 20 nm
177 in diameter), which prevails for some hours and shows signs of growth. The events can then be
178 classified into classes I and II according to the level of certainty, while class I events can be further
179 classified to Ia and Ib. Events having both a clear formation of a new mode of particles in the
180 smallest size bins available (thus excluding possible advected events) as well as a distinct and
181 persistent growth of the new mode of particles for at least 3 hours were classified as Ia, while Ib
182 consists of rather clear events that fail though by at least one of the criteria set. Additionally, for the
183 roadside sites, a formation of particles in the nucleation mode accompanied by a significant increase
184 of the concentrations of pollutants was not considered as an NPF event, as it may be associated with
185 mechanisms other than the secondary formation. In the present study, only the events of class Ia
186 were considered with the additional criterion of at least 1 nm h^{-1} growth for at least 3 hours. [As the](#)
187 [available SMPS datasets for the sites in the U.K. are for particles of diameter greater than 16 nm,](#)
188 [additional criteria were set to ensure the correct extraction of NPF events, including the variations](#)
189 [of the particle number concentrations from a Condensation Particle Counter \(CPC – measuring](#)
190 [particles with diameter from 7nm\), as well as of the concentrations of gaseous pollutants and](#)
191 [aerosol constituents \(please refer to the Methods section in Bousiotis et al., 2019\).](#)

192 2.2.2 Calculation of condensation sink, growth rate, formation rate, and NPF event

193 probability frequency

194 The condensation sink (CS) is calculated according to the method proposed by Kulmala et al.,
195 (2001) as:

196

$$197 \text{ CS} = 4\pi D_{vap} \sum \beta_M r N \quad (1)$$

198

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

201

$$202 D_{vap} = 0.00143 \cdot T^{1.75} \frac{\sqrt{M_{air}^{-1} + M_{vap}^{-1}}}{P \left(D_{x,air}^{\frac{1}{3}} + D_{x,vap}^{\frac{1}{3}} \right)^2} \quad (2)$$

203

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

206

$$207 \beta_M = \frac{1 + K_n}{1 + \left(\frac{4}{3a} + 0.377 \right) K_n + \frac{4}{3a} K_n^2} \quad (3)$$

208

209 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
210 gas. It should be noted that due to the lack of sufficient chemical composition data for a number of
211 sites, the CS calculated is not corrected for hygroscopic growth. As a result, the values for CS and
212 the results associated to it presented in this work, may be biased between the sites studied due to the
213 great differences in the conditions between them.

214

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

216

$$217 \quad GR = \frac{D_{P_2} - D_{P_1}}{t_2 - t_1} \quad (4)$$

218

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

223

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

225

$$226 \quad J_{d_p} = \frac{dN_{d_p}}{dt} + \text{Coag}S_{d_p} \times N_{d_p} + \frac{GR}{\Delta d_p} \times N_{d_p} + S_{\text{losses}} \quad (5)$$

227

228 where CoagS_{d_p} is the coagulation rate of particles of diameter d_p , calculated as (Kerminen et al.,
229 2001):

230

$$231 \quad \text{CoagS}_{d_p} = \int K(d_p, d'_p) n(d'_p) dd'_p \cong \sum_{d'_p=d_p}^{d'_p=\max} K(d_p, d'_p) N_{d_p} \quad (6)$$

232

233 $K(d_p, d'_p)$ is the coagulation coefficient of particles with diameters d_p and d'_p , while S_{losses} accounts
234 for additional loss terms (i.e. chamber wall losses), which are not applicable in the present study.

235 For the present study, the formation rate of particles of diameter of 10 nm was calculated for

236 uniformity (16 nm for the UK sites), though most sites had data for particle sizes below 10 nm.

237

238 The NPF ~~probability frequency, used instead of NPF frequency when modelled results are presented,~~

239 was calculated by the number of NPF event days divided by the number of days with available data

240 in the given group (full dataset or temporal, variable ranges etc.). The results presented in this study

241 were normalised according to the data availability, as:

242

$$243 \quad NPF_{\text{probability frequency}} = \frac{N_{NPF \text{ event days for group of days } X}}{N_{\text{days with available data for group of days } X}} \quad (76)$$

244 Finally, the p-values reported in the analysis derive from the ANOVA one-way test. As the

245 normality of the variables is required for such an analysis, the Shapiro-Wilk test was used to assess

246 the normality and the vast majority of the variables were found to have $p > 0.05$ and thus were
247 considered as normal. This is probably due to the removal of the extreme values (as mentioned in
248 section 2.2.3, for the calculations 90% of each dataset was kept removing the extremely high and/or
249 low values and the possible outliers included in them). While this was not done to promote the
250 normality of the populations but to reduce the bias from extreme values, it indirectly assisted in
251 making the distributions normal. For the few remaining (e.g. the growth rates associated with SO₂
252 concentrations for UKRO) for which normality was not present, the square root of the values of the
253 variable were considered to achieve normality and proceed to the ANOVA test.

254

255 **2.2.3 Calculation of the gradient and intercept for the variables used**

256 Due to the large datasets available and the great spread of the values, a direct comparison between a
257 given variable and any of the characteristics associated with NPF events (NPF
258 probabilityfrequency, growth rate and formation rate) always provided results with low statistical
259 significance. As a result, an alternative method which can provide a reliable result without the
260 dispersion of the large datasets was used in the present study, to investigate the relationships
261 between the variables which are considered to be associated with the NPF events. For this, a
262 timeframe which is more directly associated with the NPF events typically observed in the mid-
263 latitudes was chosen. For NPF probabilityfrequency and GR the timeframe between 05:00 to 17:00
264 Local Time (LT) was chosen, which is considered the time when the vast majority of NPF events
265 take place and further develop with the growth of the particles. For the formation rate a smaller

266 timeframe was chosen, 09:00 to 15:00 LT which is ± 3 hours from the time of the maximum
267 formation rate found for almost all sites (12:00 LT). This was done to exclude as far as possible the
268 effect of the morning rush at the roadside sites, as well as only to include the time window when the
269 formation rate is mostly relevant to NPF events (negative values that are more probable outside this
270 timeframe and are not associated with the formation of the particles would bias the results).

271

272 For the CS the timeframe 05:00 to 10:00 LT was chosen. This was done to avoid including the
273 direct effect of the NPF events (the contribution of newly formed particles to CS), as well as to
274 provide results for the conditions which either promote or suppress the characteristics studied,
275 which specifically for the CS are more important before the start of the events. The extreme values
276 (very high or very low) which bias the results only carrying a very small piece (forming bins of very
277 small size) of information were then removed, though 90% of the available data was used for all the
278 variables. The [data left remaining data](#) was separated into smaller bins and a minimum of 10 bins
279 was required for each variable (for example if the difference between the minimum and the
280 maximum relative humidity (RH) is 70%, then 14 bins each with a range of 5% were formed). The
281 variables of interest were then averaged for each bin and plotted, and a linear relation was
282 considered for each one of them. [While it is evident that not all relationships are linear, the specific
283 type was chosen in the present analysis for all the variables studied. This was done because the aim
284 was to elucidate the general positive or negative effect of the variables studied. Furthermore, the
285 effect of many variables appears to vary between sites with great differences \(either geographical or](#)

286 type of land use) and the choice of a single method to describe these relationships ensures the
287 uniformity of the results, as it appears to better describe them in most cases.

288

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

302

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

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

304

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

305
306 Where a_J is the gradient of the relation between the given variable and the formation rate of 10 nm
307 particles J_{10} (J_{16} for the UK sites).

308

309 3. RESULTS

310 In this study NPF events are generally observed as particles grow from a smaller size (typically 3-
311 16 nm depending on the size detection limit of instruments used) to 30 nm or larger. They therefore
312 reflect the result both of nucleation, which creates new particles of 1-2 nm (not detected with the
313 instruments used in this study), and growth to larger sizes. In analysing NPF events, we therefore
314 consider three diagnostic features:

- 315 • the probabilityfrequency of events occurring (i.e. days with an event divided by total days with
316 relevant data, depending on the variable and range studied), As only class Ia events were only
317 considered, it is expected that the frequency of the events calculated should be lower than the
318 expected one if all types of events were included. This could result in values up to one third of
319 those anticipated if all types of events were considered. For the extent of this variation please
320 refer to Bousiotis et al., (2019; 2020) in which there is an extended analysis of the NPF events
321 for each site, including the special cases of NPF events that do not comply for the criteria set for
322 class Ia. ;

- 323 • the rate of particle formation at a given size (J_{10} in this case), which was found to have unclear
324 seasonal trends among the sites and was higher for urban sites compared to rural in most cases
325 (Bousiotis, 2019; 2020)
- 326 • the growth rate of particles from the lower measurement limit to 30 nm (or 50 nm for the UK
327 sites), which was found to be greater during summer months for most of the sites, also studied in
328 the aforementioned works.-

329 From the analysis of the extended dataset a total of 1952 NPF events were extracted and studied.

330 The NPF frequency, growth and formation rate for each site is found in Table 2. The seasonal
331 variation of NPF events is found in Figure S14.

332

333 **3.1 Meteorological Conditions**

334 The gradients, coefficients of determination (R^2 – the relationships found are characterised as weak
335 for $R^2 < 0.50$, strong for $0.50 < R^2 < 0.75$ and very strong for $R^2 > 0.75$) and the p-values (~~deriving~~
336 ~~from one way ANOVA test~~) from the analysis of the meteorological variables, as well as the
337 average conditions of these variables are found in Table 3. The results for each site and variable are
338 found in figures S1 – S5.

339

340 **3.1.1 Solar radiation intensity**

341 As mentioned earlier, solar radiation intensity is considered to be one of the most important
342 variables in NPF occurrence, as it contributes to the production of H_2SO_4 which is a main

343 component of the initial clusters and participates in the early growth of the newly formed particles.
344 Hidy et al. (1994) reported up to six times higher SO₂ oxidation rates into H₂SO₄ in typical summer
345 conditions compared to winter. For almost all sites this relation is confirmed with very strong
346 correlations ($R^2 > 0.75$) between the intensity of solar radiation and the [probabilityfrequency](#) for
347 NPF events to occur. The relationship between the solar radiation and NPF [probabilityfrequency](#)
348 was positive at all sites and only three sites (FINUB, SPARU and GREUB) presented weak
349 correlations ($R^2 < 0.40$). Weaker correlations were found for the southern European sites, which
350 might be associated with the higher averages for solar radiation intensity, or the interference of
351 other processes (such as coinciding with increased CS by recirculation of air masses (Carnerero et
352 al., 2019)), possibly making it less of an important factor for these areas.

353
354 The relationship of solar radiation with the growth rate was weaker in all cases and did not present a
355 clear trend. Only some rural background sites (GERRU, FINRU and GRERU)- presented a strong
356 correlation ($R^2 > 0.50$). The relationship found in most cases was positive apart from two roadside
357 sites (GERRO and UKRO) and two urban background sites (GREUB and UKUB), though due to
358 the low $R^2 (< 0.10)$ these results cannot be considered with confidence. It seems though that the
359 solar radiation intensity is probably a more important factor at background sites rather than at
360 roadside sites, where possibly local conditions (such as local emissions) are more important (Olin et
361 al, 2020). Finally, the formation rate has a positive relationship with the solar radiation intensity,
362 with relatively strong correlations in most areas ($R^2 > 0.50$). The correlations were stronger at the

363 rural background sites compared to the roadside sites, which further underlines the increased
364 importance of this factor at this type of site. A negative relationship between the solar radiation
365 intensity and the formation rate was found at the GRERU site but the R^2 is very low ($R^2 = 0.05$).

366
367 Plotting the normalised gradients for NPF event [probability frequency](#) a_N^* with the average solar
368 radiation intensity at each site (Figure 2) a negative relationship is found ($R^2 = 0.62$), with the
369 southern areas (those with higher average solar intensity) having smaller a_N^* compared to those in
370 higher latitudes (and thus with a lower average solar radiation). This may indicate that while solar
371 radiation is a deciding factor in the occurrence of an NPF event, when in greater intensity its role
372 becomes relatively less important, a finding that was also implied by Wonaschütz et al. (2015).
373 Additionally, the a_r^* was found to be higher at all rural sites compared to their respective roadside
374 sites (and urban background sites for all but the Greek and German ones), making it a more
375 important factor at this type of site (Figure 3).

376

377 3.1.2 Relative humidity

378 Relative humidity is considered to have a negative effect on the occurrence of NPF events (Jeong et
379 al., 2010; Hamed et al., 2011; Park et al., 2015; Dada et al., 2017; Li et al., 2019). While water in
380 the atmosphere is one of the main compounds needed for the formation of the initial clusters either
381 on the binary or ternary nucleation theory (Henschel et al., 2016; Korhonen et al., 1999; Mirabel
382 and Katz, 1974), under atmospheric conditions it may also play a negative role suppressing the

383 number concentrations of new particles by increasing aerosol surface area (Li et al. 2019).

384 Consistent with this, a negative relationship of the RH with NPF [probability frequency](#) was found
385 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
386 to interpret as solar radiation intensity, temperature, RH and CS are not independent variables, since
387 an increase in temperature of an air mass due to increased solar radiation will be associated with
388 reduced RH, which in turn affects the CS. The sites in Greece presented lower R^2 compared to the
389 other sites while, GRERU was found to have the weakest correlation ($R^2 = 0.22$). This may be due
390 to the different seasonality of the events found for the Greek sites (being more balanced within a
391 year), as there was increased frequency of NPF events for the seasons with higher RH compared to
392 other sites, making it a less important factor for their occurrence [as found in the previous study by](#)
393 [Bousiotis et al., \(2020\)](#). Growth rate on the other hand had a variable relationship, either positive or
394 negative, with only a handful of background sites having strong correlations. The German
395 background sites as well as FINRU, which were among the sites with the highest average RH
396 (average RH for GERRU is 81.9%, GERUB is 78.7% and FINUB is 80.1%) presented a negative
397 relationship between the RH and growth rate. DENRU (average RH at 75.7%) had a positive
398 relationship, which might indicate that the relationship between these two variables may vary
399 depending upon the RH range. Formation rate also appears to have a negative relationship with the
400 RH, though this relationship was significant ($R^2 > 0.40$) for only 6 sites, which once again in most
401 cases are sites with higher RH average conditions. Along with the results of the growth rate this

402 might indicate that the RH becomes a more important factor in the development of NPF events as
403 its values increase.

404

405 The normalised gradients once again provide some additional information. Regarding the NPF
406 [probability frequency](#), it is found that the a_N^* was more negative at rural sites compared to roadside
407 sites. This indicates that the RH has a smaller effect at roadside sites, as other variables, such as the
408 atmospheric composition, are probably more important within the complex environment in this type
409 of site. Additionally, the relationship between a_N^* and average RH at the sites had a negative
410 relationship ($R^2 = 0.46$), which further shows that the RH becomes a more important factor at
411 higher values (Figure 4). Furthermore, at the sets of rural and roadside sites with R^2 higher than
412 0.40 for the relation between RH and the formation rate (UK and German sites), it was found that
413 the a_J^* was more negative at the rural sites which indicates that the RH is a more important factor at
414 rural sites compared to their respective roadside sites.

415

416 3.1.3 Temperature

417 Temperature can have both a direct and indirect effect in the development of NPF events, as it is
418 directly associated with the abundance of both biogenic and anthropogenic volatile carbon, which is
419 an important group of compounds whose oxidation products can participate in nucleation itself
420 (Lehtipalo et al., 2018; Rose et al., 2018), as well as in the growth of newly formed particles. It may
421 also have a negative effect on the particle size distributions or number concentrations through other

422 processes such as particle evaporation. Most of the sites of the present study presented a strong
423 relationship of NPF [probabilityfrequency](#) with temperature, which in most cases was positive,
424 though in many cases (such as the Danish, Finnish and Spanish sites – figures S2b, d and e) there
425 seems to be a peak in the NPF [probabilityfrequency](#) at some temperature, after which a decline
426 starts (though being at the higher end does not greatly affect the results). Sites with smaller R^2
427 (weaker association with temperature), were mainly those that have a seasonal variation that
428 favoured seasons other than summer. These sites not only had weaker relationship of NPF
429 [probabilityfrequency](#) with temperature, but in most cases had a negative relationship (background
430 sites in Finland, Spain and Greece). The Finnish sites, having the lowest average temperatures and a
431 sufficient amount of data below zero temperature, show at all three sites the possible presence of a
432 peak in the NPF event [probabilityfrequency](#) for temperatures below zero (Figure S2d). This seems
433 to be the cause of the weak relationships found there and they seem to be associated with the
434 formation rate J_{10} , which also seems to have an increasing trend below zero degrees (Figure S2p).
435 This may depend on the nucleation mechanism occurring, as cluster evaporation rates of sulphuric
436 acid clusters are sensitive to the ternary stabilising compound present (Olenius et. al., 2017), as well
437 as the possible enhancement of growth mechanisms at lower temperatures (below 5°C) by other
438 chemical compounds in the atmosphere (i.e. nitric acid and ammonia) as found by Wang et al.,
439 (2020). Laboratory experiments show that the characteristics of organic aerosol forming from
440 alpha-pinene is governed by gas phase oxidation (e.g. Ye et al. 2019). In the real atmosphere, the
441 higher temperature enhances the amount of biogenic vapours (e.g. Paasonen et al. 2013) and,

442 although the oxidation can be more efficient at higher temperatures, the lower temperatures favour
443 formation of more non-volatile compounds (Quéléver et al., 2019; Stolzenburg et al. 2018; Ye et al.
444 2019).

445

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

456

457 The normalised gradients for this variable did not present a clear trend among the areas studied,
458 other than presenting greater a_N^* for the sites with a summer peak in their NPF event seasonal
459 variation. As with other meteorological variables, the importance of this variable became smaller
460 with increased values in the average conditions for both the NPF [probabilityfrequency](#) (Figure 5)
461 and J_{10} , though these relationships were not significant (biased by the very low average

462 temperatures and different behaviour of the variables at the Finnish sites, without which the
463 relationship becomes a lot clearer as indicated in Figure S13). The variation though within the sites
464 of the same area (different sites in same country / region) appears to directly follow the variability
465 of temperature, showing that the temperature directly affects the occurrence of NPF events when
466 other meteorological factors remain constant, having a negative trend for all countries but Finland.
467 The a_j^* though is found to be greater (positively or negatively) at the rural background sites than at
468 the other two types of sites at all areas studied, showing that it is a more important factor for the
469 formation rate at this type of site compared to others (Figure 6).

470

471

472 **3.1.4 Wind speed**

473 Wind speed may have both a positive and a negative effect on the occurrence of NPF events. On
474 one hand, it may promote NPF events by the increased mixing of the condensable compounds in the
475 atmosphere as well as by reducing the CS. On the other hand, high wind speeds may suppress NPF
476 events due to increased dilution. It should be considered that the variability found is also affected by
477 the specific conditions found at each site. The wind speed measurements in many cases, especially
478 in urban sites, can be biased by the local topography or specific conditions found at each site, thus
479 representing the local conditions for this variable rather than the regional ones. Similarly,
480 measurements of wind speed at well sited meteorological stations may be more representative of
481 regional conditions, than of those affecting the sites of nucleation measurement. The sites in this

482 study presented mixed results, both in the importance as well as the effect of the wind speed
483 variability. Three different behaviours were found in the variation of NPF event
484 [probabilityfrequency](#) and wind speed which appear to be associated with local conditions as they
485 are almost uniformly found among the sites within close proximity. Some sites presented a steady
486 increase of NPF event [probabilityfrequency](#) with wind speed (Danish sites, UKUB, FINRU,
487 SPAUB and GRERU), while others were found to steadily decline with increasing wind speeds
488 (German sites – it should be noted that the German sites are the only ones that are located at a great
489 distance from the sea), while some were found to reach a peak and then decline, which also leads to
490 smaller R^2 (UKRU, UKRO, SPARU and to a lesser extent GREUB – figures S4a, e and f). The
491 reasons for these differences between the sites are very hard to distinguish as apart from the wind
492 speed the origin and the characteristics of these air masses play a crucial role. Following this, it
493 appears that NPF [probabilityfrequency](#) is very low or zero for wind speeds close to calm for the
494 sites with an increasing trend (as well as those that have a peak and decline after), while the
495 opposite is observed for the German sites where the maximum NPF [probabilityfrequency](#) is found
496 for very low wind speeds (fig. S4c).

497
498 Similarly, the effect of different wind speeds upon the growth rate also varied a lot, though it was
499 found to be negative in all the cases where R^2 was higher than 0.50 (UKUB, DENRU, DENRO,
500 GERRU, GERUB and GREUB). Finally, the formation rate was found to have a significant

501 correlation ($R^2 > 0.40$) only at two sites (UKRO and DENRU), probably indicating that the
502 variability of the wind speed either does not affect this variable or its effect is rather small.

503

504 The normalised gradients did not have any notable relationship to either the NPF
505 [probabilityfrequency](#) or the formation rate further confirming that the effect of the different wind
506 speeds is not due to its variability only, but it is also influenced by the characteristics of the
507 incoming air masses as well as specific local conditions found at each site.

508

509 3.1.5 Pressure

510 In almost all the sites with available data (apart from the Spanish), the NPF [probabilityfrequency](#)
511 presented a positive relationship with high significance at all types of sites. The greater significance
512 found at the rural sites (apart from SPARU) indicates the increased importance of meteorological
513 conditions in the occurrence of NPF events at this type of site. The growth rate also presented a
514 similar picture, with positive relationships at all the background sites of this study except the ones
515 in Greece ($R^2 > 0.71$) and FINUB (though with low R^2 at 0.02). This is probably associated with the
516 seasonal variation found in Greece where higher growth rates were found in summer, a period when
517 increased wind speeds and lower atmospheric pressure was found due to the Etesians, a pressure
518 system that develops in the region every summer (Kalkavouras et al., 2017). An interesting finding
519 is the negative gradients found at all the roadside sites, though the significance of these results is
520 relatively low ($R^2 < 0.43$) and always lower compared to the rural sites. The effects of pressure

521 above are not likely to be important. Once again however, this is not an independent variable and
522 higher pressure in summer tends to be associated with higher insolation and temperatures and lower
523 RH. Since most events occur in the warmer months of the year, this is probably the explanation for
524 the apparent effects of pressure. The formation rate presented relationships of low significance (R^2
525 < 0.47) for the sites of this study. Due to this, pressure should not be an important factor for the
526 formation rate at any type of site.

527

528 The normalised gradients did not present any clear trends, even for the NPF [probability frequency](#)
529 for which the results presented significant relationships at almost all sites.

530

531 **3.2 Atmospheric Composition**

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

535 **3.2.1 Sulphur dioxide (SO_2)**

536 Sulphur dioxide, as a precursor of H_2SO_4 , is considered as one of the main components associated
537 with the NPF process. According to nucleation theories and observations, H_2SO_4 is the most
538 important compound from which the initial clusters are formed, as well as one of the candidate
539 compounds for the initial steps of particle growth (Kirkby et al., 2011; Nieminen et al., 2010; Sipila
540 et al., 2010; Stolzenburg et al., 2020). As H_2SO_4 in the atmosphere is produced from oxidation

541 reactions of SO₂ it would be expected that increased concentrations of the latter would be associated
542 with increased values for all the variables associated with the NPF process. Contrary to this though,
543 the relationship of SO₂ concentrations with NPF [probabilityfrequency](#) was found to be negative at
544 all the sites in this study with available data. [This is expected as the average concentrations of SO₂](#)
545 [on NPF event days was found to be lower compared to the average conditions in most cases as](#)
546 [found by Bousiotis et al., \(2019; 2020\).](#) This relationship was relatively strong ($R^2 > 0.50$) in most
547 areas with an increased significance at roadside sites compared to their respective rural sites. As this
548 is a negative relationship, this may indicate that SO₂ is in sufficient concentrations for H₂SO₄
549 formation, thus not suppressing the occurrence of NPF events, as well as showing that in increased
550 concentrations, it is a more important factor (or surrogate for a factor) in preventing the occurrence
551 of NPF events within the urban environment, as higher SO₂ is likely associated with increased co-
552 emitted particle pollution and hence CS. The growth rate on the other hand, presented mixed results
553 and the significance of the relationships is low in most cases, which makes these results unreliable.
554 Finally, the relationship of SO₂ concentrations with the formation rate was found to be positive at
555 all sites but SPARU and FINRU (which had the lowest concentrations across the sites with
556 available data). The significance of this relationship was rather low ($R^2 < 0.40$) for all but the
557 roadside sites. This suggests that higher H₂SO₄ concentrations favour greater formation rates (i.e.
558 more particles can be formed), rather than necessarily promoting nucleation itself because of the
559 competing effect of condensation onto the pre-existing particle population.
560

561 The normalised gradients a_N^* were found to be more negative at the background sites compared to
562 their respective roadside sites, as well as being less negative in the UK (where SO_2 is in greater
563 abundance) compared to the other sites with relatively significant relationships. Plotting the average
564 SO_2 concentrations with the normalised gradients a_N^* for the all sites (though not all had significant
565 relationships), a positive relationship with relatively high R^2 (when the extreme values from
566 Marylebone Road-UKRO are removed) is found which might indicate that while increased
567 concentrations are a negative factor in NPF event occurrence at a given site, in general the sites with
568 higher SO_2 concentrations on average present higher [probabilityfrequency](#) for NPF events (Figures
569 7a and 7b). This appears to be in agreement with Dall'Osto et al. (2018) who discussed the variable
570 role of SO_2 depending on its concentrations. [Similar findings for the effect of \$SO_2\$ were also found](#)
571 [in previous worksstudies \(Jung et al., 2006; 2008\), relating particle acidity to NPF. Finally, nNo](#)
572 significant relationships were found for the values of a_j^* as in most cases these relationships were
573 rather weak.

574

575 3.2.2 Nitrogen oxides or nitrogen dioxide (NO_x or NO_2)

576 NO_x and NO_2 are directly associated with pollution, which can be a limiting factor for NPF events
577 as it increases the CS and may suppress the events (An et al., 2015), though with the reduction of
578 SO_2 concentrations achieved the last couple of decades, there is a possibility for oxidation products
579 of NO_x to become an important component for NPF (Wang et al., 2020). For almost all sites (apart
580 from GRERU) with available data a negative relationship between the NPF [probabilityfrequency](#)

581 and NO_x concentrations (or NO₂ depending on the available data) was found. Similarly, for all the
582 sites but SPARU and GRERU, the correlations were ~~strong~~relatively strong with R² > 0.43. The
583 rural background sites had a weaker relationship between the two variables compared to the urban
584 sites, which is probably associated with them having rather low concentrations and variability of
585 NO_x (or NO₂), making the variations of this factor less important. Growth rate had weaker
586 correlations with NO_x and different trends between the sites, either being positive or negative. The
587 variable effect of NO_x on particle growth, shifting HOMs volatility, was previously discussed by
588 Yan et al. (2020). While variability was found for the background sites, all roadside sites regardless
589 of the strength of the relationship had a positive relation~~ship~~ship between NO_x and the growth rate. This
590 may indicate the different components associated with the growth process at each type of site
591 which, as found in other studies, can be related to compounds associated with combustion processes
592 that take place within the urban environment (Guo et al., 2020; Wang et al., 2017a). The formation
593 rate presents few cases of strong relationships, with variable trends (positive and negative). While
594 much effort was made to isolate the effect of NPF events by taking a shorter time frame before the
595 event, the effect of local pollution is still included, especially at the urban sites (which probably
596 explains the positive effect found).

597

598 The normalised gradients do not provide a significant result for the relationship of this variable with
599 either the ~~probability~~frequency of the events or the formation rate. The only noteworthy points are
600 the more negative a_N^{*} at the rural background sites compared to the roadside sites in all the areas

601 studied, which shows the increased importance of a clean environment for NPF events to occur in
602 areas where condensable compounds are in lesser abundance, such as a rural environment.
603 Additionally, the negative gradients found at all the roadside sites, which increases the confidence
604 that the events extracted at the roadside sites are not pollution incidents but NPF events. However,
605 it appears that traffic pollution favours higher particle growth rates, although the components
606 responsible for this effect are unknown.

607

608 3.2.3 Ozone (O₃)

609 Ozone is typically the result of atmospheric photochemistry and is itself a source of hydroxyl
610 radical through photolysis, or ozonolysis of alkenes both during daytime and night-time (Fenske et
611 al., 2000). It might therefore be expected to act as an indicator of photochemical activity which
612 promotes the oxidation of SO₂ and VOCs. Ozone concentrations may be directly related to the
613 solar radiation intensity as well as the pollution levels in the area studied, and O₃ is considered as a
614 positive factor in the occurrence of NPF events (Woo et al., 2001; Berndt et al., 2006). As with the
615 solar radiation intensity, there is a strong relationship between O₃ concentration and the
616 [probability/frequency](#) for NPF events. This positive relationship, [which is in agreement with the](#)
617 [higher concentrations of O₃ found on NPF event days compared to average conditions for all sites in](#)
618 [Bousiotis et al., \(2019; 2020\)](#), was found to be stronger for the sites in northern Europe ($R^2 > 0.51$),
619 while it was not significant ($R^2 < 0.38$) for the sites in southern Europe (Spanish sites and GRERU),
620 possibly indicating that O₃ is a less important factor at the southern sites. Specifically for the

621 Spanish sites which have the highest average concentrations of O₃ with some extreme values
622 (Querol et al., 2017), the relationship of O₃ concentrations with the NPF ~~probability~~frequency
623 presents a unique trend (Figure S8d), having a clear peak then a steady decline at both sites (though
624 at different O₃ concentrations), which is also responsible for the low correlations found (this trend
625 seems to also occur at SPARU for the growth rate and to a lesser extent for the formation rate as
626 well, though for different O₃ concentration ranges – figures S8i and n). The specific variability
627 found at the Spanish sites was also studied by Carnerero et al., (2019). For sites with a marked
628 seasonal variation in ozone, associations with NPF may be artefactual due to correlations with other
629 variables such as temperature, RH and solar radiation intensity.

630

631 Unlike the solar radiation intensity though, the growth rate presents a negative relationship at the
632 sites where the relationship between these two variables was significant (UKRU, UKUB, DENUB
633 and FINRU), which might either be an indication of a polluted background that may have a
634 negative effect in the growth of the newly formed particles (though the trends found for NO_x
635 indicate differently) or specific chemical processes which cannot be identified due to the lack of
636 detailed chemical composition data. A significant relationship between O₃ and the formation rate
637 was only found for two sites (UKRO and DENRO, though the trends become a lot clearer if some
638 values are removed from the extreme lower or higher end). This way the relationships become
639 strong, but positive, for some areas and negative for some others without any clear trend (type or
640 location of the site, O₃ concentrations etc.). No clear relationship between these two variables was

641 found as the sites with strong relationship have both positive (DENRO) and negative (UKRO)
642 relationships and as a result no confident conclusions can be drawn.

643 As the correlations found were strong the normalised gradients for NPF [probabilityfrequency](#), when
644 plotted against the average concentrations of O₃, present a negative correlation with relatively high
645 R² (0.64), indicating that the O₃ is a more important factor in the occurrence of NPF events when in
646 lower concentrations (Figure 8). Finally, though with a low level of confidence for the southern
647 sites, the a_N^{*} were smaller at the southern sites compared to those in the north, up to one order of
648 magnitude between FINRU (furthest north rural background) and GRERU (furthest south rural
649 background).

650

651 **3.2.4 Organic compounds**

652 **3.2.4.1 Particulate organic carbon (OC)**

653 Organic carbon (OC) compounds in the secondary aerosol typically enter the particles via
654 condensational processes, with a role that becomes increasingly important as the size of the
655 particles becomes larger (Nieminen et al., 2010; Zhang et al., 2012; Shrivastava et al., 2017).

656 Particulate OC, the data for which is available in the present study, can be associated with pollution,
657 especially in the urban environment. Only a few of the sites of the present study were found to have
658 a relatively strong negative relationship (R² > 0.50) of particulate OC with the NPF

659 [probabilityfrequency](#) (UKUB, UKRO and DENRU). Regardless though of the strength of this
660 relationship, all other sites (apart from FINRU) had a negative relationship between these two

661 variables as well, consistent with increased concentrations of particulate OC being associated with
662 increased pollution, which elevate the CS, suppressing the occurrence of NPF events. Growth rate
663 on the other hand was found to have a positive relationship ($R^2 > 0.40$) for most of the sites. This
664 relationship appeared to be stronger (higher R^2) at the roadside sites with available data compared
665 to their respective rural background sites. The relationship between particulate OC and the growth
666 rate was positive at all the sites with available data regardless of their significance showing that,
667 despite its effect in the occurrence of NPF events, it is still a favourable variable for the growth of
668 the particles. The formation rate was found to have a significant relationship with particulate OC
669 concentrations at half of the sites with available data (UKUB, UKRO, DENRU, DENRO).

670

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

673

674 **3.2.4.2 Volatile organic compounds (VOCs)**

675 Many volatile organic compounds have been found to be associated with the NPF process. Benzene,
676 toluene, ethylbenzene, m-p-xylene, o-xylene and trimethylbenzenes have been reported to be able to
677 form Highly Oxygenated Organic Molecules (HOMs) in flow tubes (Wang et al., 2017a; Molteni et
678 al., 2018), which may act as contributors to particle nucleation and/or growth. Xylenes, and to a
679 lesser extent trimethylbenzenes, are the most efficient at forming HOMs. Benzene and toluene are
680 less efficient and will form more volatile HOMs. These HOMs may all be too volatile to form new

681 particles, though this is not yet confirmed. Chamber studies involving H₂SO₄ and trimethylbenzene
682 oxidation products were associated with high formation rates when measuring J_{1.5} (Metzger et al.,
683 2010). All these HOMs though will be sufficiently involatile to contribute to particle growth. Those
684 with higher oxygen content or carbon number will be classed as LVOC and if they dimerise, they
685 will form ELVOC (Bianchi et al., 2019). Monoterpenes can also form HOMs which drive both the
686 formation (Ehn et al., 2014; Riccobono et al., 2014) and growth (Tröstl et al., 2016), while isoprene
687 can act as a sink for hydroxyl radical (Kiendler-Scharr et al., 2009) and is not as effective in HOM
688 and secondary organic aerosol formation compared to monoterpenes (McFiggans et al., 2019).

689

690 Volatile organic compound data were available for three of the sites of this study (Table S2). Two
691 of the sites with VOC data were from the rural background and the roadside site in the UK. Most of
692 the compounds are associated with combustion sources and were found to have a negative
693 relationship with NPF event occurrence at both sites, with high R² (R² > 0.50) in most cases.
694 Additionally, isoprene, which may have either biogenic or anthropogenic sources (Wagner and
695 Kuttler, 2014) was also found to have a negative relationship with NPF event occurrence at
696 Marylebone Road-UKRO, though with low R² (0.07). This result is in line with the VOCs being
697 strongly correlated with particulate OC (which presented a negative relationship with NPF event
698 [probability frequency](#), as discussed in Section 3.2.4.1), as well as with the CS (which also presented
699 a negative relationship with NPF event [probability frequency](#), as mentioned in Section 3.2.6), further
700 associating these compounds with combustion emissions.

701

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

715

716 As Hyytiälä (FINRU) is a rural background site far from the direct effect of combustion emissions,
717 different VOCs were measured, which mainly originate from biogenic sources rather than
718 anthropogenic ones. The results were mixed and less clear compared to those from the UK sites
719 (mainly due to the smaller dataset), and three groups were found depending on their relationship
720 with NPF [probabilityfrequency](#). The first group, including acetonitrile, acetic acid and methyl ethyl

721 ketone (MEK) presented a slight positive relationship. The second group presented a negative
722 relationship, with the VOCs in this group being monoterpenes, methacroleine, benzene, isoprene
723 and toluene (only the last two have $R^2 > 0.50$). Finally, the third group included VOCs that
724 presented a peak and then a decline for higher concentrations including methanol, and acetone. Two
725 groups of VOCs were found depending on their relationship with the growth rate. The ones with a
726 positive relationship being methanol, acetonitrile, acetone, acetic acid, isoprene, methacroleine,
727 monoterpenes and toluene, while acetaldehyde, MEK and benzene had a negative relationship, with
728 relatively high R^2 in most cases. Finally, the results with the formation rate were unclear with only a
729 handful presenting weak ($R^2 < 0.21$) positive (methanol, acetic acid and benzene) or negative
730 (MEK) relationships that do not appear to be significant. The normalised gradients cannot be used
731 for VOCs as there are very few sites with available data.

732

733 **3.2.5 Sulphate (SO_4^{2-})**

734 Sulphate (SO_4^{2-}) is a major secondary constituent of aerosols. Secondary SO_4^{2-} aerosols largely arise
735 from either gas phase reaction between SO_2 and OH, or in the aqueous phase by the reaction of SO_2
736 and O_3 or H_2O_2 , or NO_2 (Hidy et al., 1994). In environments where SO_4^{2-} chemistry is dominant
737 (i.e. remote areas), SO_4^{2-} and ammonium (bi) sulphate ($(\text{NH}_4)_2\text{SO}_4$ and NH_4HSO_4) particles are a
738 large relative contributor to aerosol mass, while this contribution is lower in environments where
739 other emissions are also significant (i.e. urban areas where the secondary NO_3^- relative contribution
740 is a lot higher). While not well established, a possible relationship of SO_4^{2-} -containing compounds

741 and variables of NPF events was found in previous studies (Beddows et al., 2015; Minguillón et al.,
742 2015; Wang et al., 2017b). In the present study, only a few sites had SO_4^{2-} data available, for PM_{10}
743 (FINRU), $\text{PM}_{2.5}$ (Danish sites) or PM_{10} (rest of the sites). While this data cannot be considered as
744 directly associated with the ultrafine particles, for two sites with available [AMS-ACSM](#) data for
745 ultrafine particles, the direct comparison between SO_4^{2-} aerosol in PM and in the range of particles
746 of about 50 nm, very high correlations were found (results not included). For all the sites with
747 available data the NPF [probability frequency](#) presented a negative relationship. The significance of
748 this relationship was found to be relatively high ($R^2 > 0.50$) only for background sites (apart from
749 GERRU, which has rather low concentrations and probably different mechanisms for the NPF
750 events). Similarly, the growth rate presented a significant relationship ($R^2 > 0.40$) for the same
751 background sites (apart from FINRU), though this relationship was found to be positive at all sites
752 regardless of its significance. Finally, the formation rate did not present a clear trend as it was found
753 to have both negative and positive relationships for different sites. This relationship was significant
754 only for two rural sites (UKRU and DENRU) and as a result no conclusions can be reached.

755

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

758

759 **3.2.6 Gaseous ammonia (NH_3)**

760 Ammonia (NH₃) can be an important compound in the nucleation process according to the ternary
761 theory (Kirkby et al., 2011; Napari et al., 2002). It was found that elevations in NH₃ concentrations
762 can lead to elevations to NPF rate (Lehtipalo et al., 2018) and it was also found to be an important
763 factor for NPF event occurrence even when stronger bases are present in high concentrations
764 (Glasoe et al., 2015). No significant variation was found though between event and non-event days
765 in a previous study in Harwell - UKRU (Bousiotis et al., 2019). Data for gaseous ammonia was only
766 available for UKRU and presented a positive relationship with NPF [probabilityfrequency](#), until
767 reaching a peak point. Further increase in NH₃ concentrations presented a decline with NPF
768 [probabilityfrequency](#) (Figure S11a), which might be due to its association with increased pollution
769 levels. It presented a clear positive relationship with both the growth rate (though it also appears to
770 decline at high concentrations) and the formation rate, consistent with its well-established role in
771 accelerating both of these processes (Kirkby et al. 2011; Stolzenburg et al., 2020).

772

773 3.2.7 Condensation sink (CS)

774 The CS is a measure of the rate at which molecules will condense onto pre-existing aerosols
775 (Lehtinen et al., 2003). It is highly dependent on the number and size of the particles in the
776 atmosphere and as a result it is expected to be affected by both the local emissions within the urban
777 environment as well as the formation and growth of the particles due to NPF events. As a result, for
778 the specific metric a time frame before the events are in full development was chosen (05:00 to
779 10:00 LT) to avoid including the effect of the NPF events and provide a picture of the atmospheric

780 conditions that preceded the NPF events. With this data, the NPF [probability/frequency](#) presented
781 very strong relationships with the condensation sink. Two groups of sites were found though; those
782 which had a positive relationship and those with a negative relationship. In the first group are the
783 sites in Germany and Greece while all others had a negative relationship. This grouping follows the
784 trend between the countries, the sites of which presented a greater or smaller CS on NPF event days
785 [according to the findings in Bousiotis et al., \(2019; 2020\)](#) (having positive or negative gradients
786 respectively), though it is unknown what causes this behaviour (at the German sites and GREUB it
787 may be associated with the very high formation rates on NPF event days). While the gradients from
788 this analysis cannot be used for direct comparisons, a trend was found for which the gradients were
789 more positive or negative at the rural sites compared to their respective roadside sites, which might
790 indicate the greater importance of the variability of the CS at the rural sites in the occurrence of
791 NPF events.

792

793 The growth rate was positively correlated with the CS for most of the sites, with [strong-relatively](#)
794 [strong](#) relationships ($R^2 > 0.40$) for about half of them. As the CS is a metric of pre-existing
795 particles, it is also associated with the level of pollution in a given area. The increased significance
796 and gradient found at the rural sites probably indicates the importance of enhanced presence of
797 condensable compounds in a cleaner environment, which in many cases are associated with the
798 moderate presence of pollution. The formation rate was also found to have a positive relationship
799 with the CS. This relationship was more significant at the roadside sites of this study, a result which

800 to some extent is biased by the presence of increased traffic emissions found in the timeframe
801 chosen. While to an extent, increased presence of condensable compounds can be favourable for
802 greater formation rates, this result should be considered with great caution.

803

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

809

810 **3.3 Association of the Effect of the Variables**

811 The Pearson correlation coefficients for the variables studied on each site are found in Table S1.

812 The relatively strong relationship between the solar radiation intensity, temperature and O_3 found,
813 as well as their anticorrelation with the RH may lead to the conclusion that not all these factors play
814 a role in NPF events, but their visible effect is the result of their relationship with each other. There
815 is a similar case with the association of the CS and NO_x (or NO_2), and OC, as well as SO_2 ,
816 especially at urban sites. However, the factors affect different outcomes differently, as for example
817 the solar radiation intensity does not seem to be as important a factor for the growth rate as
818 temperature, or O_3 does not seem to be strongly associated with either the formation or the growth
819 rate. This is further established by the fact that some of these variables do not correlate well at the

820 southern sites, but still appear to be associated with either the [probabilityfrequency](#) of NPF events
821 or the growth or nucleation rate. The effects of all of these factors have been demonstrated in both
822 laboratory and atmospheric studies in the past and were discussed earlier in this paper. By the
823 analysis provided in the present study, the effect of each of these variables is further established,
824 providing an association of each one of these variables with either the formation or the growth
825 mechanism. However, RH does not seem to be a consistent factor in any mechanism, and it appears
826 that its effect is dependent on location specific conditions, although it was the variable with the
827 most consistent relation with NPF event [probabilityfrequency](#) at almost all sites.

829 **3.4 Relationship to a previous multi-station European study**

830 The findings of our study in respect of the background sites show many similarities with the
831 conclusions drawn in the previous multi-station study in Europe by Dall'Osto et al. (2018) despite
832 the two studies using several different sampling stations as well as some in common. Both studies
833 point towards the influence of variables such as solar radiation intensity and CS upon the
834 occurrence of NPF events. The previous study suggested that different compounds participate in the
835 growth of the particles, depending on the area considered. Thus, for northern and southern sites the
836 growth of the particles is suggested to be driven mainly by organic compounds, while for the sites
837 in central Europe sulphate plays a more important role. These findings are confirmed by the present
838 study, as the growth rate was found to correlate better with organic compounds for the rural sites in
839 Finland and Greece, while SO_4^{2-} presented a stronger relationship with the growth rate for the

840 Danish and German sites (the latter presented high gradient values but low R^2 due to a decline at
841 higher SO_4^{2-} concentrations – figure S10i, probably associated with NPF events being suppressed
842 by increased pollution). The growth of the particles at the rural background site in the UK,
843 characterised as “Overlap” in the previous study, was found to be strongly associated with both
844 organic compounds and sulphate, consistent with it being in the central group.

845

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

853

854 4. CONCLUSIONS

855 The present study attempts to explain the effect of several meteorological and atmospheric variables
856 on the occurrence and development of NPF events, by using a large-scale dataset. More than 85
857 site-years of data from 16 sites from six countries in Europe were analysed for NPF events. A total
858 of 1952 NPF events with consequent growth of the newly formed particles were extracted and with
859 the use of binned linear regression, the relationship between three variables associated with NPF

860 events (NPF event [probabilityfrequency](#), formation and growth rate) with meteorological conditions
861 and atmospheric composition was studied. Among the meteorological conditions, solar radiation
862 intensity, temperature and atmospheric pressure presented a positive relationship with the
863 occurrence of NPF events [in the majority of the sites \(though exceptions were found as well, mostly](#)
864 [in the southern sites\)](#), either promoting the formation or growth rate. RH presented a negative
865 relationship with NPF event [probabilityfrequency](#) which in most cases was associated with it being
866 a limiting factor on particle formation at higher average values. Wind speed on the other hand
867 presented variable results, appearing to depend on the location of the sites rather than their type.
868 This shows that while wind speed can be a factor in NPF event occurrence, the origin of the
869 incoming air masses also plays a very important role. In most cases, meteorological conditions,
870 such as temperature or RH appeared to be more important factors in NPF event occurrence at rural
871 sites compared to urban sites, suggesting that NPF events are driven more by them at this type of
872 site compared to urban environments and the more complex chemical interactions found there.
873 Additionally, while some meteorological variables appeared to play a crucial role in the occurrence
874 of NPF events, this role appears to become less important at higher values when a positive relation
875 was found (or lower when a negative relation was found).

876

877 The results for the levels of atmospheric pollutants presented a more interesting picture as most of
878 these, which appear to be either directly or indirectly associated with the NPF process were found to
879 have negative relationships with NPF [probabilityfrequency](#). This is probably due to the fact that

880 increased concentrations of such compounds are associated with more polluted conditions, which
881 are a limiting factor in the occurrence of NPF events, as was found with the negative relationship
882 between the CS and NPF [probabilityfrequency](#) in most cases. Thus, SO₂, NO_x (or NO₂), particulate
883 OC and SO₄²⁻ concentrations were negatively correlated with NPF [probabilityfrequency](#) in most
884 cases. Average SO₂ concentrations appeared to correlate positively with the normalised NPF event
885 [probabilityfrequency](#) gradients with a relatively significant correlation, indicating that while
886 increasing concentrations have a negative impact in the occurrence of NPF events at a given site, in
887 general sites with higher SO₂ concentrations have higher [probabilityfrequency](#) for NPF events.
888 Conversely, these compounds in many cases had a positive relationship (not always though with
889 high significance) with the other variables considered. Thus, particulate OC (and VOCs where data
890 was available) and SO₄²⁻ consistently had a positive relationship with the growth rate, while SO₂
891 was positively associated with both the formation and growth rate in most cases. Finally, O₃ was
892 positively correlated with NPF event [probabilityfrequency](#) at all sites in this study, though it
893 presented variable results with the other two variables. As with some meteorological conditions it
894 was found that at sites with increased concentrations of O₃, its importance as a factor was
895 decreased, which to some extent can be related with the high CS associated with peak summer O₃
896 days in southern Europe.

897

898 It should be noted that the variables considered are in many cases inter-related (e.g. temperature and
899 RH) and this considerably complicates the interpretation in terms of causal factors. Large datasets

900 are very useful in providing more uniform results by removing the possible bias of short period
901 extremities, which may lead to wrong assumptions. This study, apart from providing insights into
902 the effect of a number of variables on the occurrence and development of NPF events in
903 atmospheric conditions across Europe, also shows the differences that climatic, land use and
904 atmospheric composition variations cause to those effects. Such variations are probably the cause of
905 the differences found among previous studies. Following from this, the importance of a high-
906 resolution measurement network, both spatially and temporally is underlined, as it can help in
907 elucidating the mechanisms of new particle formation in the real atmosphere.

908

909 **DATA ACCESSIBILITY**

910 Data supporting this publication are openly available from the UBIRA eData repository at

911 <https://doi.org/10.25500/edata.bham.00000491>.~~https://doi.org/~~

912

913 **AUTHOR CONTRIBUTIONS**

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

919

920 **COMPETING INTERESTS**

921 The authors have no conflict of interests.

922

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926

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1567 **TABLE LEGENDS**

1568

1569 **Table 1:** Location and data availability of the sites.

1570

1571 **Table 2:** Frequency (and number of NPF events), growth and formation rate of NPF events.

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1573 **Table 3:** Normalised gradients (non-normalised for growth rate), R^2 and p-values (- for values
1574 >0.05) for the relationship between meteorological conditions and NPF event
1575 variables. Gradients of $R^2 > 0.50$ are in bold.

1576

1577 **Table 4:** Normalised gradients (non-normalised for growth rate), R^2 and p-values (- for values
1578 >0.05) for the relationship between atmospheric composition variables and NPF
1579 event variables. Gradients of $R^2 > 0.50$ are in bold.

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1582 **FIGURE LEGENDS**

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1584 **Figure 1:** Map of the sites of the present study.

1585

1586 **Figure 2:** Relation of average downward incoming solar radiation (K_{\downarrow}) and normalised
1587 gradients a_N^* .

1588

1589 **Figure 3:** Normalised gradients a_J^* for K_{\downarrow} (*UK sites are calculated with solar irradiance).

1590

1591 **Figure 4a:** Relationship of average relative humidity and normalised gradients a_N^* .

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1593 ~~**Figure 4b:** Relationship of average relative humidity and normalised gradients a_N^* (SPAUB not
1594 included).~~

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1596 **Figure 5:** Relationship of average temperature and normalised gradients a_N^* .

1597

1598 **Figure 6:** Normalised gradients a_J^* for temperature.

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1600 **Figure 7a:** Relationship of average SO_2 concentrations and normalised gradients a_N^* for the
1601 sites with available data (a) and for the sites with available data excluding UKRO
1602 (b).

1603

1604 ~~**Figure 7b:** Relationship of average SO_2 concentrations and normalised gradients a_N^* (UKRO
1605 not included).~~

1606

1607 **Figure 8:** Relationship of average O₃ 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

1610 **Table 2:** Frequency (and number of NPF events), growth and formation rate of [class Ia](#) 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

** J₁₆ 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. Gradients of $R^2 > 0.50$ are in bold.

Downward shortwave solar radiation K_{\downarrow} ($W\ m^{-2}$)										
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 (°C)										
Site	a_N^* (°C ⁻¹)	R ²	P	a_G	R ²	P	a_J^* (°C ⁻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

1620

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. [Gradients of \$R^2 > 0.50\$ are in bold.](#)

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

1630 * NO₂ 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

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¹ Measurements in PM₁₀² Measurements in PM_{2.5}³ Measurements in PM₁

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

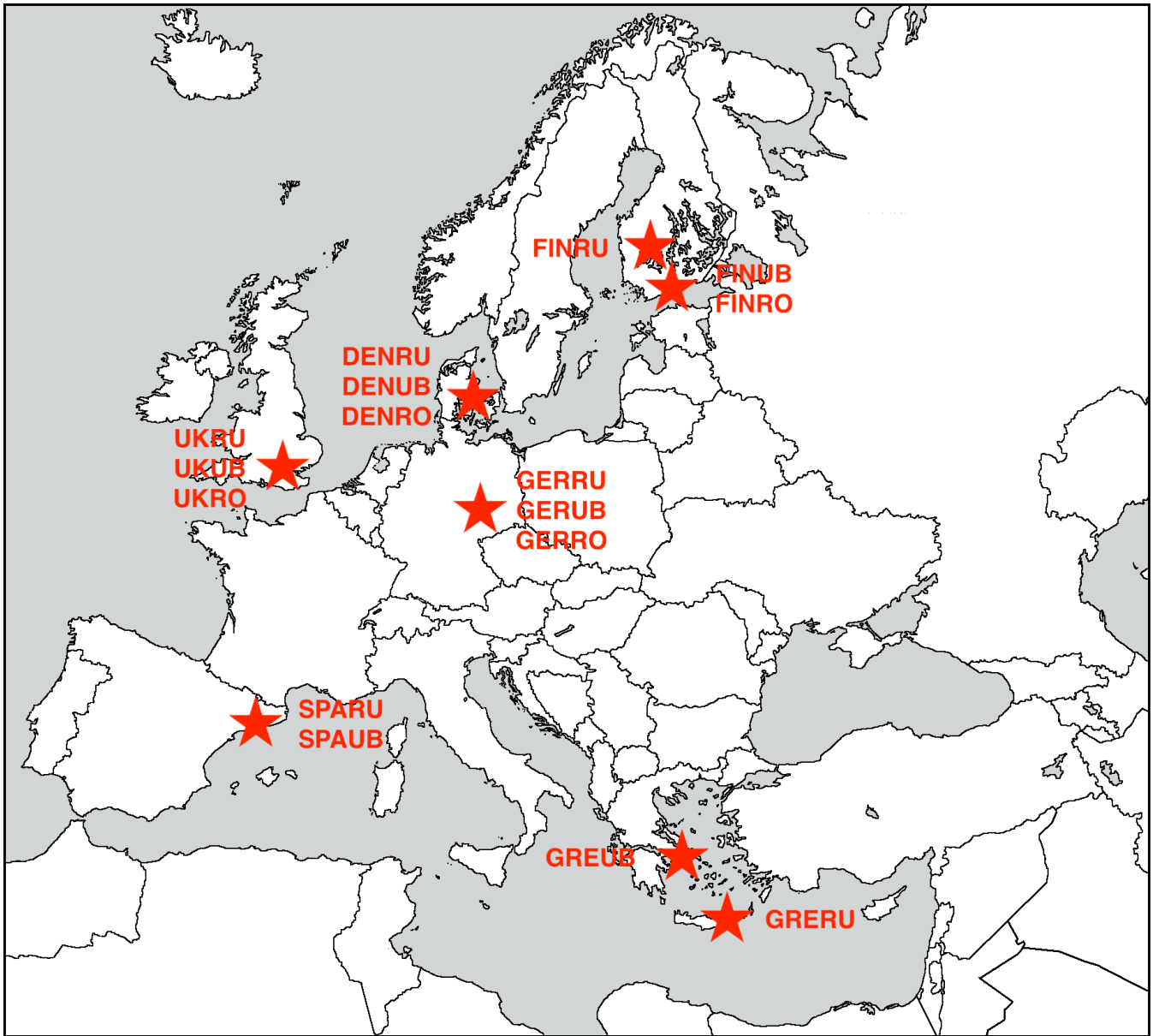


Figure 1: Map of the sites of the present study.

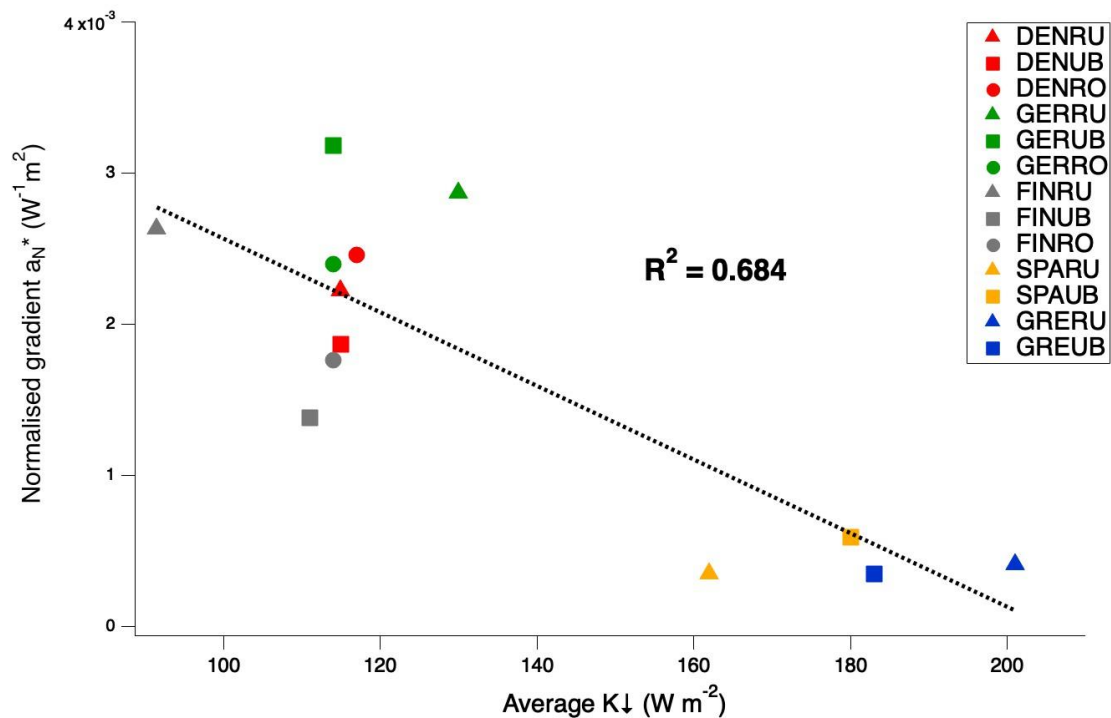


Figure 2: Relationship of average downward incoming solar radiation (K_{\downarrow}) and normalised gradients a_N^* .

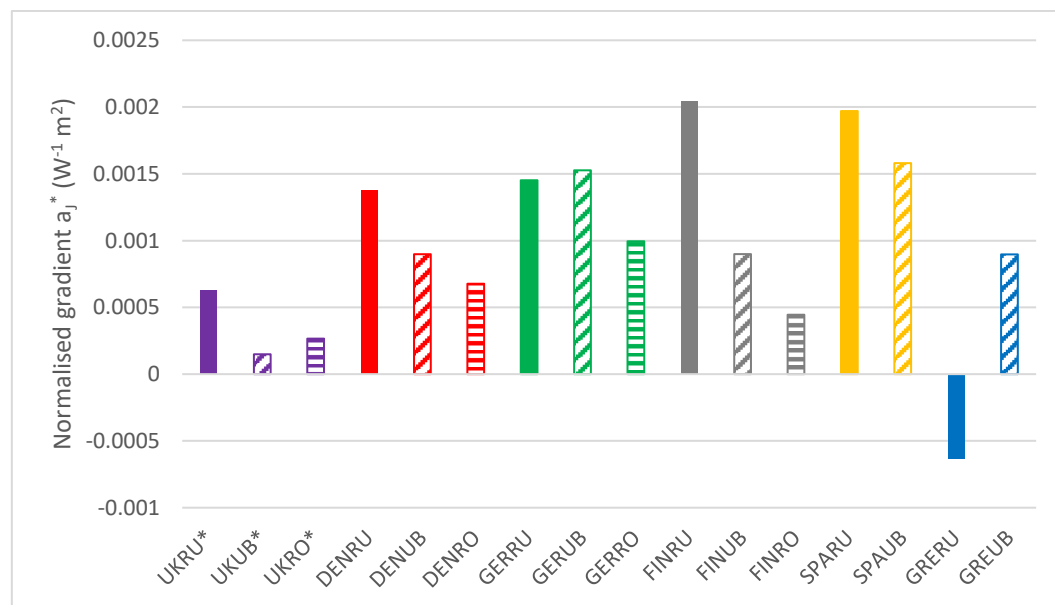


Figure 3: Normalised slopes a_j^* for K_{\downarrow} (*UK sites are calculated with solar irradiance).

1650

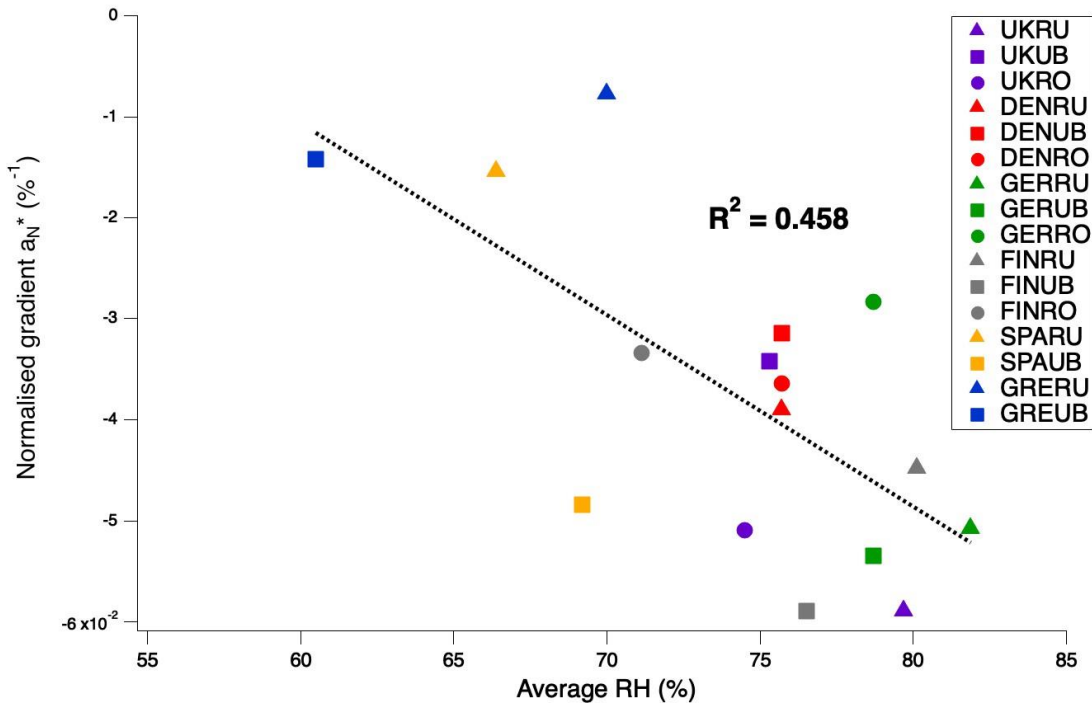


Figure 4: Relationship of average relative humidity and normalised gradients a_N^* .

1655

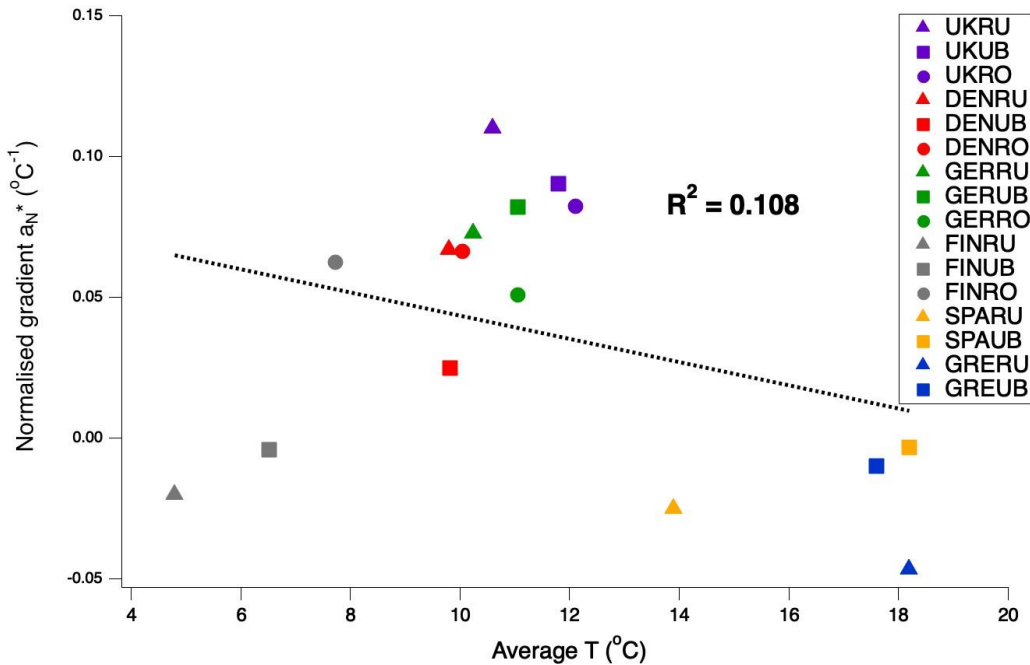
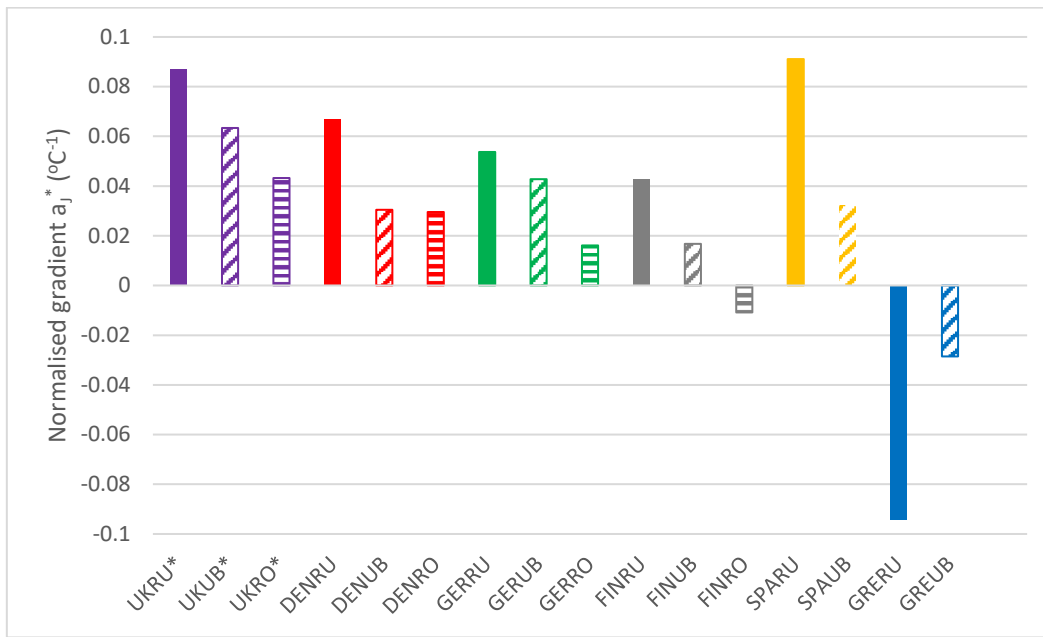


Figure 5: Relationship of average temperature and normalised gradients a_N^* .



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Figure 6: Normalised gradients a_j^* for temperature.

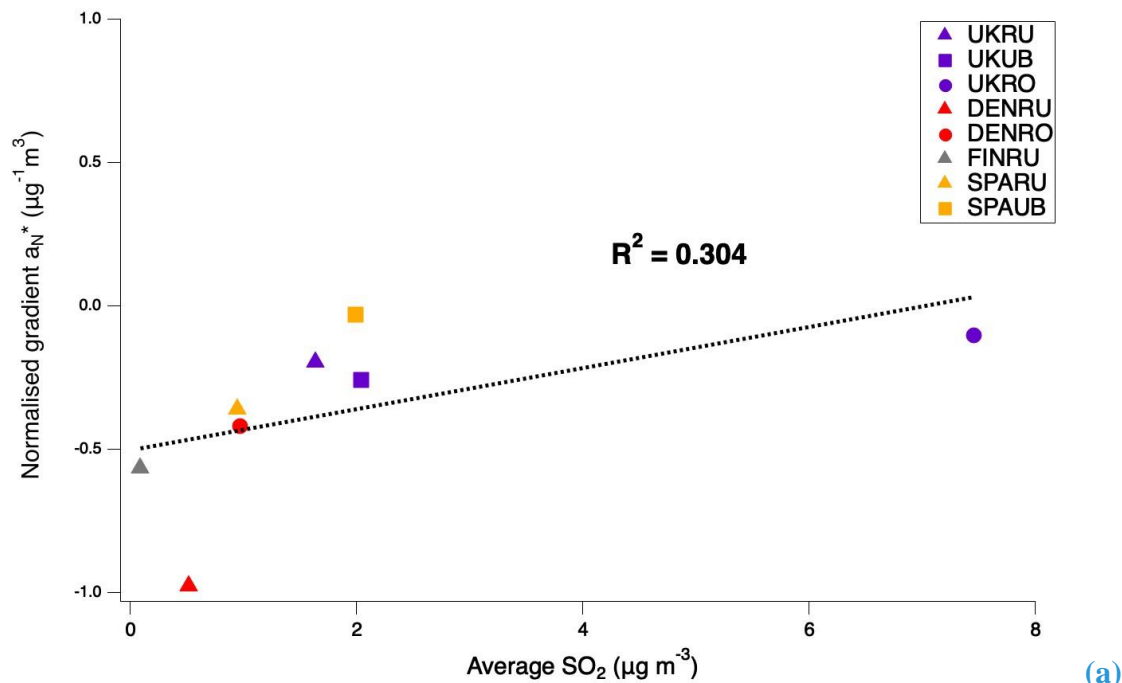


Figure 7a: Relationship of average SO₂ concentrations and normalised gradients a_N^{*} for the sites with available data (a) and for the sites with available data excluding UKRO (b).

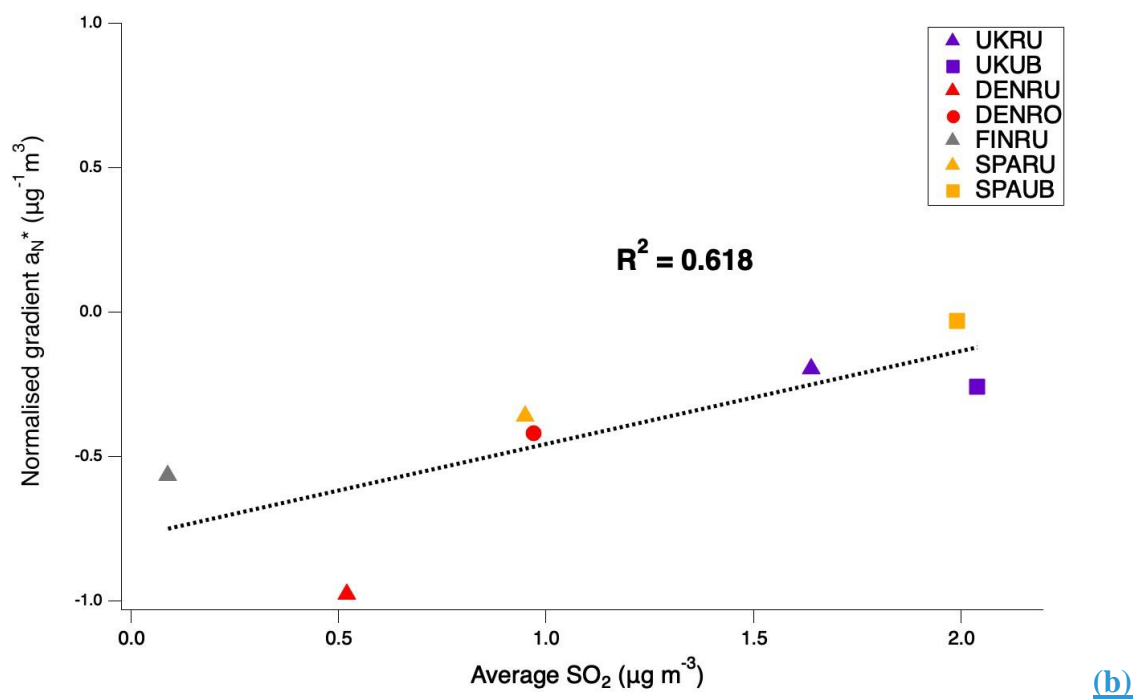
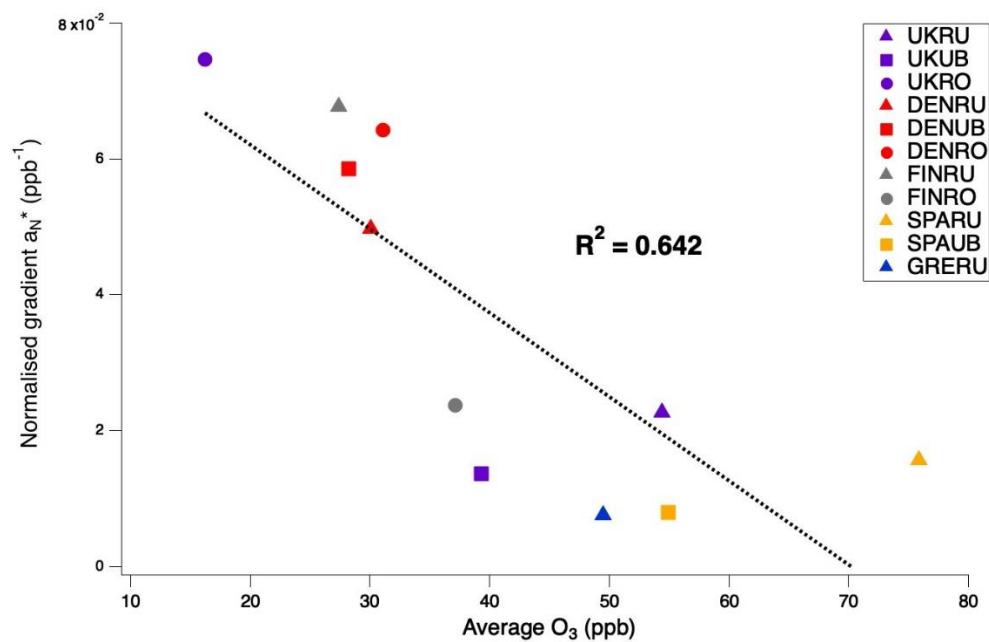


Figure 7b: Relationship of average SO₂ concentrations and normalised gradients a_N^{*} for the sites with available data (a) and for the sites with available data excluding UKRO (b).

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



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

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