MS No.: acp-2020-555

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.

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### RESPONSE TO REVIEWERS

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The authors thank the reviewers for their insightful comments and have made many modifications in response, and to enhance the clarity of the paper.

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### **Anonymous Referee #1**

Received and published: 31 August 2020 12

A couple of decades ago, a number of studies tried to link meteorological variables and gas phase pollutants with NPF. In some cases, the analysis was concise enough to produce evidence that a certain physical parameter played a role in NPF at a specific site.

Since then, studies - mostly chamber-based - have provided evidence on the ruling mechanisms of nucleation and subsequent growth of newly formed particles. These are mainly related to the concentration of low vapor pressure compounds such as sulfuric acid, or ELVOC as well as agents that could stabilize the former (ammonia, amines, iodine) suggesting that NPF is dictated mainly by 20 gas phase chemistry rather than meteorology. However, other parameters such as the ones investigated in this study, play a secondary yet important role. Therefore a summary of observations 21 from European, or even better global sites, is always welcome. During the past 15 years more than 20 compilations of results related to atmospheric NPF have been published, the majority of which are summarized by Kerminen et al., 2018. Even though some of them (eg Kerminen et al., 2018,

Lee et al 2019) provide insight on the parameters this study is also focusing on, none has gone been as detailed as the one presented in this work. Therefore, the compilation of results presented in this work are of interest to the community and would be worthwhile publishing if the manuscript was well written and the analysis provided informative and concise. I am afraid that this is not the case. After reading the article, I was disappointed not to find any information on seasonality for any of

30 the parameters investigated even though multi year data were investigated. 31

**RESPONSE:** It is not clear by the comment what kind of analysis is expected (whether it is the seasonality of the parameters themselves or the seasonality of their effect). The seasonality of the parameters (which was found to favour mainly summer for the growth rate, while the results for the formation rate were more variable) is separately investigated in a previously published paper for the UK sites (Bousiotis et al., 2019) and for the rest of the sites in an already submitted manuscript (Bousiotis et al., 2020) and thus was not discussed in the present study (this is noted in 2.1). The seasonality of their effect was not studied in the present manuscript as this would extend its size too much. It is worth pointing out that the variables most affected by season such as temperature and insolation are considered in this paper, and to break down the data analysis according to season would involve a great deal of repetition.

Furthermore the authors fail to deliver any error metric whatsoever (deviation, error, confidence

level). The lack of the most elementary statistical analysis was striking.

44 **RESPONSE:** Much of this information is included in the paper. Deviation errors are included in the SI figures for every subgroup of every variable studied (reporting these is unrealistic as they are over a thousand). R<sup>2</sup> is reported for every slope calculated of every variable studied on the figures. p-values are reported (when significant) for every variable in every site in tables 3 and 4. We have calculated, but not included, the error of the slope for every variable calculated using the normalised gradients, but have not included this as the normalised slopes do not have any significance other than their absolute value; we include only information on their trend.

The other striking feature is the poor use of English and terminology, which I explain thoroughly 52 53 below. The use of English must be improved as there are many sentences that require revision. **RESPONSE:** Many changes in terminology and corrections were applied throughout the 54 manuscript to improve the level of English. 55

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The major drawback is the generalizations and uncertain phrases used throughout the manuscript.

58 The authors should be concise and specific instead. 59

**RESPONSE:** The manuscript was updated in many cases to reduce uncertainty (whenever there was enough confidence in the statements presented)

As an example in Line 69 (76) it is advised to name the places (exceptions) were NPF is hardly observed. A nice review can be found by Lee et al., 2019 (section 4.8).

64 **RESPONSE:** Exceptions where NPF events are not observed and references were added.

Example 2 Line 270: A few sites presented a strong correlation, which in all cases were background sites (either rural or urban). A few sites (which ones?) presented a strong correlation (nowhere in the manuscript strong, medium weak is defined. The reader has no idea what the author is

69 discussing) which in all cases were background sites (either rural or urban; to the best of my knowledge rural sites are considered as background sites. What do the authors mean?). I assume 70

71 that the authors are trying to point at urban kerbside sites with this sentence, yet I am not really sure

72 what they mean. And the paragraph continues The relation (which one?) found in most cases (how

73 many, percentage?) was positive (does this mean a positive slope? Where is it shown? In which 74 table or graph?) apart from two roadsides (improper terminology) and GREUB, though due to the

low (again low is not defined?) R2 these results cannot be used with confidence (and where do the

75 authors draw the confidence line?). The above lines are just an example of improper phrasing used 76

throughout the manuscript that make it very hard to follow. Similar examples can be found 77

throughout the manuscript. A major drawback of this work is that many trends/relationships 78

reported are not referred to any table or Figure and hence are hard to follow. 79

**RESPONSE:** References to the sites mentioned in each case as well as R<sup>2</sup> values were added

throughout the manuscript to improve readability. References for the results were added in the

83 84 beginning of each section (to avoid repetition). Specific references for unusual trends were also added for the figures in the SI (figure numbering in SI was overhauled). References for SI figures for simple relationships were not added as they are covered by the slopes found in tables 3 and 4. Strong, weak and other characterisations of the correlations are now accompanied with either the R<sup>2</sup> or a range of the R<sup>2</sup>.

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NPF probability sounds to me as if you are trying to predict the occurrence of nucleation events. Based on Line 218 (Equations are not numbered!) a more suited term would be NPF frequency. **RESPONSE:** The term NPF frequency is used within the text for the frequency of the events without taking into account any grouping of the data (into groups of condition ranges e.g NPF probability for RH in the range 60-65%). To separate these the term probability was used instead. Equation numbers were added.

The authors should consider adding reference formation and growth rates from other studies in their figures for comparison. I understand that this is not always possible (especially for formation rates) but is for the other two parameters in question.

**RESPONSE:** Similar to a previous comment, such an analysis was done in other studies either already submitted or published (Bousiotis et al., 2020; 2019).

Table 2 should also include growth rates for this study.

**RESPONSE:** The parameters of NPF are already reported in previously submitted studies (Bousiotis et al., 2020; 2019). The frequency and formation rate are reported here because they are used in the calculation of the normalised slopes, which is not done for the growth rate (see the methodology). Nevertheless, the growth rate and the number of NPF events for each site were added in Table 2.

The authors fail to summarize the seasonality of the parameters they are exploring even though they are having multi year data. This is very disappointing.

**RESPONSE:** This comment has already been addressed.

The statement in Lines 45-46 (49-50) is not true. Please read Kerminen et al., 2018 for example. That work which explicitly states the opposite.

**RESPONSE:** The sentence was rephrased into "without always following" to state that exceptions exist as pointed later in the Introduction part.

I have noted another case (Lines 98-99) (109 – 110) in the manuscript where the authors focus on the exceptions (which always exist) rather than the rule giving a very distorted view to the reader. **RESPONSE:** In the text it is stated that "the negative effect of CS is widely accepted" and follows mentioning the exceptions found in the literature as "cases were found". This does not imply that the exceptions are anything more than that and it is essential that they are mentioned.

8889 The introduction is very poor on references.

90 RESPONSE: More references were added in the introduction and throughout the manuscript

Lines 107-111 (118 – 123) should be rephrased. I cannot make sense of it at all.

**RESPONSE:** This sentence was included to show that the NPF events considered in our study were not driven by combustion products but by secondary formation. A parenthesis was added though which makes the sentence easier to understand.

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Lines 82-84 (94 – 96). Please mention that increasing temperatures also have a negative effect as they increase the energy barrier the clusters have to overcome to become stable and grow in size.

99 **RESPONSE**: The comment has been added.

100

Kerminen, V. M., Chen, X., Vakkari, V., Petäjä, T., Kulmala, M. and Bianchi, F.: Atmo-spheric
new particle formation and growth: Review of field observations, Environ. Res. Lett., 13(10),
doi:10.1088/1748-9326, 2018.

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Lee, S. H., Gordon, H., Yu, H., Lehtipalo, K., Haley, R., Li, Y. and Zhang, R.: New Particle Formation in the Atmosphere: From Molecular Clusters to Global Climate, J. Geophys. Res. Atmos., 124(13), 7098–7146, doi:10.1029/2018JD029356, 2019.

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### Anonymous Referee #2

110 Received and published: 26 September 2020

111 General comments

112 The focus of this study is to investigate the effect of meteorological conditions and atmospheric

113 composition on the occurrence of new particle formation (NPF) events at 16 sites (rural, urban

114 background and roadside) located in 6 European countries. The results are based on more than 85

115 years of meteorological and atmospheric composition data. The authors are using a binned linear

116 regression to find correlations between parameters such as windspeed, temperature, pressure, or

solar radiation intensity, ozone or volatile organic compounds mixing ratios to name a few, and the

118 occurrence of NPF, particle growth and the formation rates. This is an interesting study and of

119 interest to the community, however the following comments should be addressed before publishing.

120 On many occasions the authors claim that certain variables are weakly or strongly correlated but do

121 not provide any numbers or figure to support these statements. Please provide references to the

122 exact figures in the manuscript or in the supplemental material (SM). This is an issue reappearing

123 throughout the manuscript. Following on that, Figure S1 in SM contains many figures and only one

124 caption. These figures are not marked with a number/letter. Please consider adding individual

125 numbering (or introduce letters) so the respective figures corresponding to individual sites when

being discussed in the manuscript could be easily found in SM.

127 **RESPONSE**: This is addressed in a later comment

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It looks like the authors use terms frequency of NPF occurrence and NPF probability 129

interchangeably. NPF probability doesn't really fit here since you do not predict NPF events.

- 131 However, the NPF probability term is explained in the text and in the equation (line 191; also please
- number equations). In results, however, the authors are using term frequency of NPF occurrence 132
- 133 (line 245). Please clarify, review the explanation in text and use the correct term throughout the
- 134 manuscript. I assume what you want to use is NPF frequency.
- 135 **RESPONSE:** This is explained later

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- 137 I understand you identify the number of days with NPF according to the method by Dal Maso et al. (2005) with additional certain criteria. It would be good to report the numbers of NPF events for 139 each site (and season?). Please explain what days with "relevant data" are when calculating the 140 frequency.
- 141 **RESPONSE:** Two additional papers analysing in detail the conditions of the NPF events at all the sites were either published (for UK) or were submitted (for the rest of the sites). This is noted in section 2.1. A figure has been added to the SI to show the seasonality. 143

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145 If I understood correctly to calculate the frequency, you divide the number of days identified as NPF event-days by all days that you have data available or "relevant(?)" data available? I am 147 curious how does the frequency changes when you use the number of all days with all data and not 148 only with "relevant data available"? It would be good to mention this number somewhere in the manuscript or in the SM? Following on the above, please explain what is in e.g. line 191 "available 149 data" and "given group"?

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RESPONSE: The term "relevant data" refers to all the data available at each site and are considered in each analysis (and of course when those are available). At each site the data were almost in their entirety available and the limitation was in most cases the SMPS data (its availability for each site is reported in Table 1). The data is always considered only when they are available for each variable studied (by the code used in the analysis, as they were calculated one by one), so no hours of data with missing values were included. In other words, when e.g. temperature was studied only the hours with both SMPS and temperature data were considered.

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More detail on the site selection criteria would be helpful. Do these sites belong to a network? How were these sites selected? In Line 120: the authors mention "geographical region and type of environment". I suggest adding more description on the sites (e.g. in SM), their characteristic and typical meteorological conditions, e.g. features they have in common/differences, number of NPF studied and identified.

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**RESPONSE:** A justification for the sites chosen is given in the Site description section. As mentioned earlier, the analysis of the events, as well as the typical conditions for all the sites were given in two separate earlier papers.

- 168 Having this extensive dataset, I encourage the authors to discuss variability e.g. seasonal, site to
- 169 site/regional. A figure where you plot frequency of NPF occurrence or number of NPF events for
- 170 each station in each season (e.g. bar plot?) on (y axis), for each site type (x axis) would be helpful.
- 171 Exploring e.g. seasonal variability would add value to the paper.
- 172 **RESPONSE:** This was already done in two earlier papers (Bousiotis et al., 2020; 2019).
- Where there any limitations of the study? If yes, these should be discussed. Further, errors should be included.
- 176 **RESPONSE**: The study is pretty straightforward, and the only limitation was the lack of data for
- 177 some variables at some sites e.g. SO<sub>2</sub> data was not available at all sites. A comment was added for
- 178 this limitation at the end of 2.1 Site Description and Data Availability section.
- 179

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- Are there general trends for these three site types? Maybe you could discuss these more. It would be
- 181 helpful to highlight (e.g. text in bold) data in Table S1 e.g. significant correlations.
- 182 **RESPONSE**: As explained in the response to the first comment, these are provided in other studies
- 183 (Bousiotis et al., 2020; 2019). Stronger correlations were highlighted with bold numbers.
- 184
- 185 What is the importance of the result of this study? The authors could discuss it more e.g. in
- 186 conclusion. I feel that is missing in the current version.
- 187 RESPONSE: The statement: "This study, apart from providing insights into the effect of a number
- 188 of variables on the occurrence and development of NPF events in atmospheric conditions across
- 189 Europe, also shows the differences that climatic, land use and atmospheric composition variations
- 190 cause to those effects. Such variations are probably the cause of the differences found among
- 191 previous studies." was added in the last paragraph of the text (838).
- 192

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- 193 Please improve the language. It is critical to make the text more concise and clearer. It is hard to
- 194 follow the line of thoughts at points. There are some repetitions and long sentences that could be
- 195 shortened (e.g. lines: 76-82, 107-111, 325-330).
  - **RESPONSE:** Many changes were made in the manuscript to improve readability.
- 198 Specific comments
- 199 Line 38-62: in the abstract the authors could also mention: 85 years of data; how good these
- 200 correlations are (r2)' mention "meteorological conditions" e.g. such as ...
- 201 RESPONSE: The information that a combined dataset of 85 years was used was added (52).
- 202 Added the highest  $R^2$  values found for some variables (54 56). Added "(such as solar radiation
- 203 and relative humidity)" (58)
- 204
- 205 Line 42 (46): "except at very clean air sites" more information is needed to this statement.
- 206 Something is missing. Please review or explain.
- 207 **RESPONSE:** This phrase has been removed.

209 Line 54 (60): What "higher values" means there exactly? Provide a number. **RESPONSE:** Added the word "average". No values can be provided as what is implied by the 211 results is that the importance of some variables becomes less as the average values (average 212 conditions) become higher or lower, depending on the general trend. 213 214 Line 61- 62 (67-68): you could give these values in brackets **RESPONSE:** No values were added as the text implies that one increases with the other 215 simultaneously, similar to the meteorological conditions. 216 217 Line 97 (108): "negative effect" on? "on the occurrence of the events" was added. 218 219 Line 99 (110): "average conditions"? What does it mean here? 220 221 **RESPONSE:** No change in the text. It means the average CS which is well covered with the term "average conditions" in this case. 222 223 224 Line 107-111 (118 – 123): hard to follow, please review and shorten 225 **RESPONSE:** Already mentioned by referee #1 and addressed. 226 227 Line 121 (133): please add references to the studies you refer to **RESPONSE:** References were added 228 229 Line 122 (134): NPF probability? Or NPF occurrence? As mentioned above, probability doesn't 230 really fit here 231 232 **RESPONSE:** NPF probability was not changed as every time it is mentioned it implies the results from the analysis/modelling that was done in this study. An explanation for this was provided (213) 233 234 235 Line 124 (138): I suggest calling this section: "2. Methods", 2.1 as is. 2.2 as is or similar. This way you can remove 2.2 Methods so it does not appear twice. In 2.1 the authors could mention which cities/countries/sites were used; which meteorological and atmospheric composition variables did 237 you use in this study already at this point. Which stations had a full set of data and which only some 239 etc. Maybe also mention which are dependent and which independent variables. And what do you 240 consider relevant data days, what do you mean by available data; e.g. in line 189. Please be more specific upfront. You could also add information on how these sites were chosen? Any criteria you applied to select these? Are they belong to a network? Are they similar or different in any respect? 243 **RESPONSE:** Section naming was not changed as it is considered sensible for a chapter named "Data and Methods" to have section 1 named "Sites and data" and section 2 as "Methods". The 244 countries and cities included in the study are mentioned. A list of the data available in each site is 245

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found in Table 1, as mentioned in the text. A justification for the sites chosen was also added. The

sites do not belong to a single network and thus such information is not provided.

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249 Line 127 (140): I feel that the number of events (1950) is already a result so it should go to the

250 result section and not methods. Also, it is mentioned before the description of the NPF selection

251 method itself.

252 **RESPONSE:** The number of events was moved to the beginning of the Results section

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Line 131 (142): it is also referring to the result. I suggest moving this sentence to the result section.

**255 RESPONSE:** The reference for the Table with results was moved to the beginning of the Results

256 section.

257

258 Line 136-143 (156 – 165): please add more details to the approach taken in this study. What "Ia"

259 exactly refers to and which additional criteria was used (line 142).

260 RESPONSE: The process of NPF event extraction was rewritten and more details were added for

**261** the approach taken (156 - 169).

262

263 Line 137 (158): add size range of the nucleation mode you consider in your study

264 **RESPONSE**: Added "(smaller than 20 nm in diameter)".

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266 Line 139 (160): you could mention confidence level in the brackets

**267 RESPONSE:** Changed to level of certainty to avoid misunderstandings.

268

269 Line 151 (178): add "respectively" after "particles". You could already mention there Fuchs

270 correction factor and keep it explained below.

**271 RESPONSE**: Added the word "respectively" (178). Second was not mentioned to avoid repetition

272 and flow distraction.

273

274 Line 149 (176): Formulas need to be numbered

275 **RESPONSE**: Equation numbers were added

276

277 Line 188 (216): given group? please explain

**RESPONSE:** The NPF probability is calculated for the range of data in a specific group (time

279 range, range of a given variable ex. for relative humidity from 50 to 55% etc.). Text was slightly

280 modified to reflect this better.

281

282 Line 191 (219): Again: I am not sure I follow this equation: what are these groups? Is it just a

283 number of days with NPF that was accompany by all relevant data? From explanation you seem

4 only to take days with NPF that were accompanied by relevant data.

**285 RESPONSE:** For the analysis done, data was separated into smaller groups as mentioned earlier.

Thus, the term probability is considered more appropriate than frequency. This is clearly stated in

287 the text.

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Line 196 (224): low significance? Please give a number

RESPONSE: The results found from the analysis of raw data, due to the large spread, almost never
 provided with any significant result (the R² was always very low). A single number cannot be
 provided as the results are numerous. The word "statistical" was added.

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296 297 Line 212 (241): extreme values? Please give a number

**RESPONSE:** As previous. The cases that extreme values that biased the results were many. For example, an extreme value of wind speed (a single very windy day with no event) would result in an NPF probability of zero for that wind speed range. This though would result in biasing the whole analysis by a very limited range of data.

298 299

Line 239 (268): Results and Discussion? You could include here sentence in line 126: You mention
 1950 events studied, could you provide information on how many were identified? It would be
 helpful if authors mention that in the paper a summary of data can be found in the manuscript and in
 the SM data/results for individual sites is presented.

RESPONSE: The sentence mentioned (the number of events extracted) was moved to the Results section as suggested. The events studied were those that all the work was focused on. While other, less clear events (without the expected growth, advected, uncertain etc.) were also extracted, they were considered only as exceptions or special cases in previous works. For further information about the events for each site as well as the comparative study between them, references were added in the Site Description section.

310

Line 245 (277): what is relevant data? Please explain more clearly in methods section and refer to it.
 Diagnostic features – wouldn't these be better in methods?

RESPONSE: Added "depending on the variable studied". Relevant data refer to the data available
depending on the variables studied, e.g. to find the frequency of NPF events, the days with available
SMPS data were only considered. These diagnostic features are used to present the results in Table
and thus were not moved to Methods.

317

Line 252 (286): "slopes and R2" please use correct terms for these or more careful description
 RESPONSE: Changed to the terms gradient (instead of slope) and coefficient of determination for
 R<sup>2</sup>.

- 322 Line 261 (296): very strong? Please provide references to the exact figures in the
- 323 figures/supplemental material when discussing results
- **RESPONSE:** In this case very strong correlations were considered for  $R^2 > 0.75$  as explained in the parenthesis (and a clarification is added to any characterised correlation). The correlations (whether
- 326 weak or strong) are found in Tables 3 and 4 (references added). References to the figures in the SI

are not needed when discussing slopes and R<sup>2</sup> and were only referenced when variable/unusual
 trends were found.

Line 279 (315): low? Please give a number and refer to the figure. Also, you placed all figures in the SM under Figure 1. Maybe it would be better for the reader to have them split into different figure numbers or a,b,c,d? This way it would be easier to find the one you describe at the very

333 moment.

**RESPONSE:** The value of the  $R^2$  was added in a parenthesis. Also, changed the numbering scheme for the figures in SI. References to figures in the SI that present results not in the tables (i.e. variable trends) were added in the text.

337

338 Line 296 (333): reference?

339 **RESPONSE:** A reference was added

340

- Line 301-303 (339): why? Could you explain? When describing results maybe worth mentioning these for various site specifics? Anything in common?
- 343 **RESPONSE**: A possible explanation was added "This may be due to the different seasonality of
  - the events found for the Greek sites (being more balanced within a year), as there was increased probability of NPF events for the seasons with higher RH compared to other sites, making it a less

346 important factor for their occurrence."

347

348 Line 369 (414): which factors remain constant? 349 **RESPONSE**: Added the word "meteorological"

350 351 Line 377 (420): reference?

352 **RESPONSE**: References for this are found in the introduction

353

- 354 Line: 398 (441 443): maximum? Low?
- 355 **RESPONSE**: Maximum changed to greatest. Low wind speeds changed to "close to zero wind

356 speeds".

357

- 358 Line 420 (464): Ethesian: add few words what these are could be added
- 359 **RESPONSE**: A brief description has been added ("a pressure system that develops in the region

360 every summer").

361

Table 3: what is a "p value"? has it been defined somewhere? In tables: the authors could use bold

363 text to highlight significant correlations? So these patterns/trends could be clearly seen?

**364 RESPONSE:** Added the definition of p-value (line 286). Used bold text for all correlation r > 0.50

365

366 Figures: no need to mention in each caption "of the present studies"

367 **RESPONSE:** The phrase was removed 368 369 Line 433 (479): you could already mention here which pollutants (such as...) are studied and 370 described in the upcoming sections. Added the chemical compounds studied in a parenthesis Line 752 (806): "at higher values"? 372 Changed to "at sites with higher average values" 373 374 375 Line 755 (810): "meteorological conditions" such as? 376 "such as temperature or relative humidity" was added 377 378 Line 756-757 (812): is that the only explanation? How about chemistry/composition at such type of 379 site? Anything else that might play a role? 380 RESPONSE: Added "compared to the urban environments and the more complex chemical interactions found there" 382 Line 782-783 (840): seems out of place here; it would be more suitable at the beginning of 383 conclusion section or removed. 384 **RESPONSE:** Moved the sentence to the start of the Conclusions section (line 796). 385 386 Interactive comment on Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2020-555, 2020 387 **RESPONSE:** Additionally, four authors were added in the list of authors 388 389 390 Table 1 was updated 391 Bousiotis, D., Pope, F. D., Beddows, D. C., Dall'Osto, M., Massling, A., Nøjgaard, J. K., 392 Nørdstrom, C., Niemi, J. V., Portin, H., Petäjä, T., Perez, N., Alastuey, A., Querol, X., Kouvarakis, 394 G., Vratolis, S., Eleftheriadis, K., Wiedensohler, A., Weinhold, K., Merkel, M., Tuch, T., and 395 Harrison, R. M.: An Analysis of New Particle Formation (NPF) at Thirteen European Sites, Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2020-414, in review, 2020. 396 397 398 Bousiotis, D., Dall'Osto, M., Beddows, D. C. S., Pope, F. D., and Harrison, R. M.: Analysis of new 399 particle formation (NPF) events at nearby rural, urban background and urban roadside sites, Atmos.

Chem. Phys., 19, 5679–5694, https://doi.org/10.5194/acp-19-5679-2019, 2019.

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#### The Effect of Meteorological Conditions and Atmospheric 403 Composition in the Occurrence and Development of New Particle 404 Formation (NPF) Events in Europe 405 406 Dimitrios Bousiotis<sup>1</sup>, James Brean<sup>1</sup>, Francis Pope<sup>1</sup>, Manuel Dall'Osto<sup>2</sup> 407 Xavier Querol<sup>3</sup>, Andres Alastuey<sup>3</sup>, Noemi Perez<sup>3</sup>, Tuukka Petäjä<sup>4</sup> 408 Andreas Massling<sup>5</sup>, Jacøb Klenø Nøjgaard<sup>5</sup>, Claus Nørdstrom<sup>5</sup> 409 Giorgos Kouvarakis<sup>6</sup>, Stergios Vratolis<sup>7</sup>, Konstantinos Eleftheriadis<sup>7</sup> 410 Jarkko V. Niemi<sup>8</sup>, Harri Portin<sup>8</sup>, Alfred Wiedensohler<sup>9</sup>, Kay Weinhold<sup>9</sup>, Maik 411 Merkel9, Thomas Tuch9 and Roy M. Harrison1a\* 412 413 414 <sup>1</sup>Division of Environmental Health and Risk Management 415 School of Geography, Earth and Environmental Sciences University of Birmingham, Edgbaston, Birmingham B15 2TT, United Kingdom 416 417 418 <sup>2</sup>Institute of Marine Sciences, Passeig Marítim de la Barceloneta, 37-49 E-08003 419 Barcelona, Spain 420 <sup>3</sup>Institute of Environmental Assessment and Water Research (IDAEA - CSIC) 421 422 08034, Barcelona, Spain 423 424 <sup>4</sup>Institute for Atmospheric and Earth System Research (INAR) / Physics, Faculty of Science University of Helsinki, Finland 425 426 <sup>5</sup>Department for Environmental Science, Aarhus University, DK-400, Roskilde, Denmark 427 428 429 <sup>6</sup>Environmental Chemical Processes Laboratory (ECPL), Department of Chemistry, 430 University of Crete, 70013, Heraklion, Greece 431 <sup>7</sup>Environmental Radioactivity Laboratory, Institute of Nuclear and Radiological Science & 432 Technology, Energy & Safety, NCSR Demokritos, Athens, Greece 433 434 435 8Helsinki Region Environmental Services Authority (HSY), FI-00066 HSY, Helsinki, Finland 436 437 438 <sup>9</sup>Leibniz Institute for Tropospheric Research (TROPOS), Permoserstr. 15, 04318 Leipzig, Germany 439 440

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### ABSTRACT

Although new particle formation (NPF) events have been studied extensively for some decades, the mechanisms that drive their occurrence and development are yet to be fully elucidated. Laboratory 446 studies have done much to elucidate the molecular processes involved in nucleation, but this knowledge has yet to be conclusively linked to NPF events in the atmosphere, except at very clean 448 449 air sites. There is great difficulty in successful application of the results from laboratory studies to real atmospheric conditions, due to the diversity of atmospheric conditions and observations found, as NPF events occur almost everywhere in the world without always following a clearly defined 452 trend of frequency, seasonality, atmospheric conditions or event development. The present study 453 seeks common features in nucleation events by applying a binned linear regression over an extensive dataset from 16 sites of various types (combined dataset of 85 years from rural and urban 455 backgrounds as well as roadside sites;) in Europe. At most sites, a clear positive relation is found between the solar radiation intensity (up to  $R^2 = 0.98$ ), temperature (up to  $R^2 = 0.98$ ) and atmospheric pressure (up to  $R^2 = 0.97$ ) with the frequency probability of NPF events, while relative 457 humidity (RH) presents a negative relation (up to  $R^2 = 0.95$ ) with NPF event-probability frequency. 459 Wind speed presents a less consistent relationship which appears to be heavily affected by local 460 conditions. While some meteorological variables (such as the solar radiation intensity and RH) 461 appear to have a crucial effect on the occurrence and characteristics of NPF events, especially at rural sites, it appears that their role becomes less marked when at higher average values. 463

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

### 472 1. INTRODUCTION

New Particle Formation (NPF) events are an important source of particles in the atmosphere (Merikanto et al., 2009; Spracklen et al., 2010), which are known to have adverse effects on human 475 health (Schwartz et al., 1996; Politis et al., 2008; Kim, et al., 2015) as well as affecting the optical and physical properties of the atmosphere (Makkonen et al., 2012; Seinfeld and Pandis, 2012). 476 While they occur almost everywhere in the world (Dall'Osto et al., 2018; Kulmala et al., 2017; O'Dowd et al., 2002; Wiedensohler et al., 2019; Chu et al., 2019; Kerminen et al., 2018), with some exceptions mentioned in the literature in forest (Lee et al., 2016; Pillai et al., 2013; Rizzo et al., 479 480 2010) or high-elevation sites (Bae et al., 2010; Hallar et al., 2016), great diversity is found in the 481 atmospheric conditions within which they take place. Many studies have been done in a large 482 number of different types of locations (urban, traffic, regional background) around the world and 483 differences were found in both the seasonality and intensity of NPF events. To an extent this 484 variability is due to the mix of conditions that are specific to each location, which blurs the general 485 understanding of the conditions that are favourable for the occurrence of NPF events (Berland et al., 486 2017; Bousiotis et al., 2020). For example, solar radiation is considered as one of the most 487 important factors in the occurrence of NPF events (Kulmala and Kerminen, 2008; Kürten et al., 488 2016; Pikridas et al., 2015; Salma et al., 2011), as it is needed for the photochemical reactions that 489 lead to the formation of sulphuric acid (Petäjä et al., 2009; Cheung et al., 2013). Sulphuric acid 490 which is considered as the main component of the formation and growth of the initial clusters (Iida 491 et al., 2008; Stolzenburg et al., 2020; Weber et al., 1995). Nevertheless, although in many cases

NPF events did not occur in the seasons with the highest insolation (Park et al., 2015; Vratolis et al., 2019). Similarly, uncertainty exists over the effect of temperature (Yli-Juuti et al., 2020; Stolzenburg et al., 2018). Hhigher temperatures are considered favourable for the growth of the 494 495 newly formed particles as increased concentrations of both Biogenic Volatile Organic Compounds (BVOCs) and Anthropogenic Volatile Organic Compounds (AVOCs) (Yamada, 2013; Paasonen et 496 al., 2013) and their oxidation products (Ehn et al., 2014) are associated towith the growth of the particles. The negative effect of increasing temperatures in increasing the energy barriers the clusters have to overcome to become stable and grow in size though should not be overlooked 499 500 (Kürten et al., 2018; Zhang et al., 2012). This appears to be true in most cases, as higher growth rates are found in most cases in the local summer (Nieminen et al., 2018), although the actual 501 502 importance of those VOCs in the occurrence of NPF events is still not fully elucidated, with oxidation mechanisms still under intense research (Tröstl et al., 2016; Wang et al., 2020). The effect 503 504 of other meteorological variables is even more complex, with studies presenting mixed results on the effect of the wind speed and atmospheric pressure. Extreme values of those variables may be 505 favourable for the occurrence of NPF events, as they are associated with increased mixing in the 506 507 atmosphere, but at the same time suppress due to increased dilution of precursors (Brines et al., 508 2015; Rimnácová et al., 2011; Shen et al., 2018; Siakavaras et al., 2016), or favour them due to a 509 reduced condensation sink (CS).

The effect of atmospheric composition on NPF events is also a puzzle of mixed results. While the negative effect of the increased CS on the occurrence of the events is widely accepted (Kalkavouras et al., 2017; Kerminen et al., 2004; Wehner et al., 2007), cases are found when NPF events occur 513 on days with higher CS compared to average conditions (Größ et al., 2018; Kulmala et al., 2005). Sulphur dioxide (SO<sub>2</sub>), which is one of the most important contributors to many NPF pathways, in 515 most studies was found in lower concentrations on NPF event days compared to average conditions 516 (Alam et al., 2003; Bousiotis et al., 2019), although there are studies that have reported the opposite 517 (Woo et al., 2001; Charron et al., 2008). Additionally, in a combined study of NPF events in China, 518 519 events were found to be more probable under sulphur-rich conditions rather than sulphur-poor 520 (Jayaratne et al., 2017). Similar is the case with the BVOCs and AVOCs, which present great variability depending the area studied (Dai et al., 2017), and their contribution in the growth of the 522 particles is not fully understood yet. Until recently, it was considered unlikely for NPF events, as they are considered in the present study, (deriving from secondary formation not associated with 524 traffic related processes such as dilution of the exhaust), to occur within the complex urban 525 environment due to the increased presence of compounds, mainly associated with combustion processes, which would suppress the survival of the newly formed particles within this type of 526 527 environment (Kulmala et al., 2017). Despite thiast though, NPF events were found to occur within 528 even the most polluted areas and sometimes with high formation and growth rates (Bousiotis et al., 529 2019; Yao et al., 2018).

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

### 542 2. DATA AND METHODS

### 543 2.1 Site Description and Data Availability

The present study uses a total of more than 85 years of hourly data from 16 sites from six countries
of Europe of various land usage and climates from which 1950 NPF events were extracted and
studied. It was considered very important that at least a rural and an urban site would be available
from each country to study the differences between the different land usage on NPF events
throughout Europe. The sites were chosen to cover the greatest possible areaextent of the European
continent, with sites from both northern, central and southern Europe, as well as from western and

eastern. The sites are located in the UK (London and Harwell), Denmark (Copenhagen greater area), Germany (Leipzig greater area), Finland (Helsinki and Hyytiälä), Spain (Barcelona and Montseny – a site in a mountainous area) and Greece (Athens and Finokalia). Unfortunately, not all sites had available data for all the variables studied, which to an extented may bias some of the results. An extended analysis of the typical and NPF evented conditions, seasonal variations and trends at these sites for the same period is found in other studies (Bousiotis et al., 2019; 2020). A list of the available data and a brief description for each site is found in Table 1 (for the ease of reading the sites are named by the country of the site followed by the last two letters which refer to the type of site, being RU for rural/regional background, UB for urban background and RO for roadside site), while a map of the sites is found in Figure 1. The NPF frequency and formation rate for each site is found in Table 2.

## **2.2** Methods

### 563 2.2.1 NPF events selection

NPF events were selected using the method proposed by Dal Maso et al (2005). As of this, an NPF
event is considered identified when a new mode of particles appears by the appearance of a new
mode or particles in the nucleation mode (smaller than 20 nm in diameter), which prevails for some
hours and shows signs of growth. The events can then be classified into classes I and II according to
the level of confidence certainty, while class I events can be further classified to Ia and Ib. E, with Ia
events having both a clear formation of a by-new mode of particles at in the smallest size bins

available (thus excluding possible advected events) as well as a distinct and persistent growth of the
new mode of particles for at least 3 hours were classified as Ia, while Ib consists of rather clear
events that fail though by at least one of the criteria set. Additionally, for the roadside sites, a
formation of particles in the nucleation mode accompanied withby a significant increase of the
concentrations of pollutants was not considered as an NPF event, as it may be associated towith
mechanisms other than the secondary formation. In the present study, only the events of class Ia
were considered with the additional criterion of at least 1 nm h<sup>-1</sup> growth for at least 3 hours.

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

578 **probability** 

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

580 (2001) as:

581

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

583

where r and N is the radius and number concentration of the particles <u>respectively</u> and D<sub>vap</sub> is the
 diffusion coefficient calculated as (Poling et al., 2001):

587 
$$D_{\text{vap}} = 0.00143 \cdot T^{1.75} \frac{\sqrt{M_{\text{air}}^{-1} + M_{\text{vap}}^{-1}}}{P\left(D_{x,\text{air}}^{\frac{1}{3}} + D_{x,\text{vap}}^{\frac{1}{3}}\right)^2}$$
 (2)

588

for T=293~K and P=1013.25~mbar. M and  $D_x$  are the molar mass and diffusion volume for air and

590 sulphuric acid. β<sub>M</sub> is the Fuchs correction factor calculated as (Fuchs and Sutugin, 1971):

591

592 
$$\beta_{\text{M}} = \frac{1 + K_n}{1 + \left(\frac{4}{3a} + 0.377\right)K_n + \frac{4}{3a}{K_n}^2}$$
 (3)

593

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

595 gas.

596

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

598

$$599 \quad GR = \frac{D_{P_2} - D_{P_1}}{t_2 - t_1} \tag{4}$$

600

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601 for the size range between the minimum available particle diameter up to 30 nm (50 nm for the UK

sites due to the higher minimum particle size available). The time window used for the calculation

of the growth rate was from the start of the event until a) growth stopped, b) GMD reached the

604 upper limit set or c) the day ended.

605 606

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

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

610 where CoagS<sub>dp</sub> is the coagulation rate of particles of diameter d<sub>p</sub>, calculated as (Kerminen et al.,

611 2001):

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

 $615 \quad K(d_p,\,d'_p) \text{ is the coagulation coefficient of particles with diameters } d_p \text{ and } d'_p, \text{ while } S_{losses} \text{ accounts}$ 

for additional loss terms (i.e. chamber wall losses), which are not applicable in the present study.

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

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

The NPF probability, used instead of NPF frequency when modelled results are presented, -was

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

the given group (temporal, variable range wind direction sector etc.). The results presented in this

study were also-normalised according to the data availability, as:

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

2.2.3 Calculation of the slope-gradient and intercept for the variables used

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

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

The slope-gradient of these linear relations ( $a_{N,a_G}$  and  $a_{J}$  for NPF probability, growth rate and formation rate  $J_{10}$  accordingly) found in this analysis should be used with great caution as apart from the atmospheric conditions (local and meteorological as well as atmospheric composition) it is also affected by the variable in question (e.g. a greater NPF probability will provide a greater slopegradient), resulting in giving the same trend for all the atmospheric variables tested; the sites with the higher values of these variables (NPF probability and formation rate) always had greater slope-gradient values and vice versa. In order to remove the effect of the variable in question (NPF probability or formation rate – growth rate will provide an untrustworthy unreliable result as it is

calculated in a different range for each site due to the lower available size of particles), the slopes
gradients were normalised by dividing them by their respective variable (e.g. divide the slope
gradient of the NPF probability with the NPF probability frequency), providing with a new
normalised slope (a<sub>N</sub>\* for NPF probability or a<sub>J</sub>\* for the formation rate) that will have no
significance other than its absolute value, which can be used for direct comparisons:

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

Where a<sub>N</sub> is the slope gradient of the relation between the given variable and NPF probability

frequency (NPF %)

673 
$$a_{J}^{*} = \frac{a_{J}}{J_{10}}$$

Where a<sub>J</sub> is the <u>slope-gradient</u> of the relation between the given variable and the formation rate of 10 nm particles J<sub>10</sub> (J<sub>16</sub> for the UK sites).

### 677 3. **RESULTS**

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In this study NPF events are generally observed as particles grow from a smaller size (typically 3-679 165 nm depending on the size detection limit of instruments used) to 30 nm or larger. They 680 therefore reflect the result both of nucleation, which creates new particles of 1-2 nm (not detected 681 with the instruments used in this study), and growth to larger sizes. In analysing NPF events, we 682 therefore consider three diagnostic features:

683	• the frequency probability of events occurring (i.e. days with an event divided by total days with
684	relevant data, depending on the variable and range studied),
1 685	• the rate of particle formation at a given size $(J_{10}$ in this case),
686	• the growth rate of particles from the lower measurement limit to 30 nm (or 50 nm for the UK
687	sites).
688	From the analysis of the extended dataset a total of 1952 NPF events were extracted and studied.
689	The NPF frequency, growth and formation rate for each site is found in Table 2. The seasonal
690	variation of NPF events is found in Figure S14.
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692 693	3.1 Meteorological Conditions
	3.1 Meteorological Conditions  The slopes-gradients, coefficients of determination (and R <sup>2</sup> ) and the p-values (deriving from one-
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693 694	The slopes gradients, coefficients of determination (and R <sup>2</sup> ) and the p-values (deriving from one-
693 694 695	The slopes-gradients, coefficients of determination (and R <sup>2</sup> ) and the p-values (deriving from one-way ANOVA test) from the analysis of the meteorological variables, as well as the average
693 694 695 696	The slopes-gradients, coefficients of determination (and R <sup>2</sup> ) and the p-values (deriving from one-way ANOVA test) from the analysis of the meteorological variables, as well as the average conditions of these variables are found in Table 3. The results for each site and variable are found in
693 694 695 696	The slopes-gradients, coefficients of determination (and R <sup>2</sup> ) and the p-values (deriving from one-way ANOVA test) from the analysis of the meteorological variables, as well as the average conditions of these variables are found in Table 3. The results for each site and variable are found in
693 694 695 696 697 698	The slopes gradients, coefficients of determination (and $R^2$ ) and the p-values (deriving from one-way ANOVA test) from the analysis of the meteorological variables, as well as the average conditions of these variables are found in Table 3. The results for each site and variable are found in Figures $S1 - S5$ .

702 component of the initial clusters and participates in the early growth of the newly formed particles.

Hidy et al. (1994) reported up to six times higher SO<sub>2</sub> oxidation rates into H<sub>2</sub>SO<sub>4</sub> in typical summer conditions compared to winter). For almost all sites this relation is confirmed with very strong correlations (R<sup>2</sup> > 0.75) between the intensity of solar radiation and the probability for NPF events 705 706 to occur... The relationship between the solar radiation and NPF probability was positive at all sites 707 and only three sites (FINUB, SPARU and GREUB) presented weak correlations (R<sup>2</sup> < below 0.40). Weaker correlations were found for the southern European sites, which might be associated with the 708 higher averages for solar radiation intensity, or the interference of other processes (such as coinciding with increased CS by recirculation of air masses (Carnerero et al., 2019)), possibly 711 making it less of an important factor for these areas. 712 713 The relationship of solar radiation to with the growth rate was weaker in all cases and did not present a clear trend. Only some rural background sites (GERRU, FINRU and GRERU) A few sites 714 presented a strong correlation (R<sup>2</sup> > 0.50)., which in all cases were background sites (either rural or urban). The relationship found in most cases was positive apart from two roadside sites (GERRO and UKRO)s and two urban background sites (GREUB and UKUB), though due to the low R2 (< 0.10) these results cannot be used-considered with confidence. It seems though that the solar 718 719 radiation intensity is probably a more important factor at background sites rather than at roadside 720 sites, where possibly local conditions (such as local emissions) are more important (Olin et al., 2020). Finally, the formation rate has a positive relationship with the solar radiation intensity, with 721 722 relatively strong correlations in most areas ( $R^2 > 0.50$ ). The correlations were stronger at the rural

background sites compared to the roadside sites, which further underlines the increased importance of this factor at this type of site. A negative relationship between the solar radiation intensity and the formation rate was found at the GRERU site but the  $R^2$  is very low ( $R^2 = 0.05$ ). 726 Plotting the normalised slopes gradients for NPF event probability  $a_N^*$  with the average solar radiation intensity at each site (Figure 2) a negative relationship is found ( $R^2 = 0.62$ ), with the 728 southern areas (those with higher average solar intensity) having smaller an\* compared to those in 729 730 higher latitudes (and thus with a lower average solar radiation). This may indicate that while solar 731 radiation is a deciding factor in the occurrence of an NPF event, when in greater intensity its role becomes relatively less important, a finding that was also implied by Wonaschütz et al. (2015). Additionally, the a<sub>J</sub>\* was found to be higher at all rural sites compared to their respective roadside 734 sites (and urban background sites for all but the Greek and German ones), making it a more 735 important factor at this type of site (Figure 3).

### 737 3.1.2 Relative humidity

736

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

number concentrations of new particles by increasing aerosol surface area (Li et al. 2019). 744 Consistent with this, a negative relationship of the RH with NPF probability was found for all the sites of this study with very high  $R^2$  for almost all of them ( $R^2 > 0.80$ ). This is not simple to 746 interpret as solar radiation intensity, temperature, RH and CS are not independent variables, since an increase in temperature of an air mass due to increased solar radiation will be associated with 747 reduced RH, which in turn affects the CS. The sites in Greece presented lower R<sup>2</sup> compared to the 748 other sites while, GRERU was found to have the weakest correlation ( $R^2 = 0.22$ ). This may be due to the different seasonality of the events found for the Greek sites (being more balanced within a 750 751 year), as there was increased frequency of NPF events for the seasons with higher RH compared to other sites, making it a less important factor for their occurrence. Growth rate on the other hand had 752 753 a variable relationship, either positive or negative, with only a handful of background sites having strong correlations. T-Among these the German background sites as well as FINRU, which were 754 755 among the sites with the highest average RH (average RH for GERRU is 81.9%, GERUB is 78.7% and FINUB is 80.1%) presented a negative relationship between the RH and growth rate, while DENRU (average RH at 75.7%) had a positive relationship, which might indicate that the relationship between these two variables may vary depending upon the RH range. Formation rate 759 also appears to have a negative relationship with the RH, though this relationship was significant  $(R^2 > 0.40)$  for only 6 sites, which once again in most cases are sites with higher RH average 760 conditions. Along with the results of the growth rate this might indicate that the RH becomes a 761 more important factor in the development of NPF events as its values increase. 762

The normalised slopes-gradients once again provide some additional information. Regarding the NPF probability, it is found that the an\* was more negative at rural sites compared to roadside sitess. This indicates that the RH has a smaller effect at roadside sitess, as other variables, such as the atmospheric composition, are probably more important within the complex environment in this type of sites. Additionally, the relationship between an\* and average RH at the sites had a negative relationship (R² = 0.46), which further shows that the RH becomes a more important factor at higher values (Figure 4). Furthermore, at the sets of rural and roadside sites with R² higher than 0.40 for the relation between RH and the formation rate (UK and German sites), it was found that the a<sub>J</sub>\* was more negative at the rural sites which indicates that the RH is a more important factor at rural sites compared to their respective roadside sitess.

### 3.1.3 Temperature

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Temperature can have both a direct and indirect effect in the development of NPF events, as it is
directly associated with the abundance of <a href="both">both</a> biogenic-<a href="and anthropogenic">and anthropogenic</a> volatile carbon, which
is an important group of compounds whose oxidation products can participate in nucleation itself
(Lehtipalo et al., 2018; Rose et al., 2018), as well as in the growth of newly formed particles, while
If may affect also have a negative effect on the particle size distributions or number concentrations
through other processes such as particle evaporation. Most of the sites of the present study
presented a strong relationship of NPF probability with temperature, which in most cases was

positive, though in many cases (such as the Danish, Spanish and FinnishFinnish and Spanish sites – 784 figures S2b, d and e) there seems to be a peak in the NPF probability at some temperature, after 785 which a decline starts (though being at the higher end does not greatly affect the results). Sites with 786 smaller R<sup>2</sup> (weaker association with temperature), were mainly those that have a seasonal variation 787 that favoured seasons other than summer. These sites not only had weaker relationship of NPF 788 probability with temperature, but in most cases had a negative relationship (background sites in Finland, Spain and Greece). The Finnish sites, having the lowest average temperatures and a 789 790 sufficient amount of data below zero temperature, show at all three sites the possible presence of a peak in the NPF event probability for temperatures below zero (Figure S2d). This seems to be the cause of the weak relations found there and they seem to be associated with the formation rate J<sub>10</sub>, 792 793 which also seems to have an increasing trend below zero degrees (Figure S2p). This may depend 794 on the nucleation mechanism occurring, as cluster evaporation rates of sulphuric acid clusters are 795 sensitive to the ternary stabilising compound present (Olenius et. al., 2017) be the result of 796 increased stability of molecular clusters at lower temperatures, as well as the possible enhancement of growth mechanisms in at lower temperatures (below 5°C) by other chemical compounds in the 798 atmosphere (i.e. nitric acid and ammonia) as found by Wang et al., (2020). Laboratory experiments 799 show that the characteristics of organic aerosol forming from alpha-pinene is governed by gas phase 800 oxidation (e.g. Ye et al. 2019). In the real atmosphere, the higher temperature enhances the amount of biogenic vapours (e.g. Paasonen et al. 2013), and, although the oxidation can be more efficient in

at higher temperatures, the lower temperatures favour formation of more non-volatile compounds 803 (Quéléver et al., 2019; Stolzenburg et al. 2018; Ye et al. 2019; Stolzenburg et al. 2018). 804 Growth rate had a more uniform trend, with almost all sites having a positive relationship with temperature (apart from GERRO, though with  $R^2 = 0.00$ ). This relationship was very strong for 806 807 most sites ( $\mathbb{R}^2 > 0.60$  for 10 sites), which is also confirming the summer peak found for the growth rate at most of these sites in other studies (Bousiotis et al., 2020; 2019). A rather strong relationship  $(R^2 > 0.50)$  with temperature was also found for the formation rate for most sites, and was positive for almost all sites (apart from FINRO with  $R^2 = 0.01$  and the Greek sites with  $R^2 < 0.47$ ). As with 810 811 the NPF probability, in general the sites with a seasonal variation of events that favoured summer had the strongest relationship (high R<sup>2</sup>) of the formation ratetemperature with temperature formation 813 rate, which might indicate that this variable, either through its direct or indirect effect is an important one for the seasonal variability of NPF events in a given area. 814 815 The normalised slopes-gradients for this variable did not present a clear trend among the areas studied, other than presenting greater a<sub>N</sub>\* for the sites with a summer peak in their NPF event 817 818 seasonal variation. As with other meteorological variables, the importance of this variable became 819 smaller with increased values in the average conditions for both the NPF probability (Figure 5) and J<sub>10</sub>, though these relationships were not significant (biased by the very low average temperatures 820 and different behaviour of the variables at the Finnish sites, without which the relation becomes a 821

lot clearer as pointed indicated in Figure S132). The variation though within the sites of the same area (different sites in same country / region) appears to directly follow the variability of temperature, showing that the temperature directly affects the occurrence of NPF events when other meteorological factors remain constant, having a negative trend for all countries but Finland. The a<sub>J</sub>\* though is found to be greater (positively or negatively) at the rural background sites than at the other two types of sites at all areas studied, showing that it is a more important factor for the formation rate at this type of site compared to others (Figure 6).

### 3.1.4 Wind speed

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

wind speed variability. Three different behaviours were found in the variation of NPF event 843 probability and wind speed which appear to be associated with local conditions as they are almost 844 uniformly found among the sites within close proximity. Some sites presented a steady increase of 845 NPF event probability with wind speed (Danish sites, as well as UKUB, FINRU, SPAUB and GRERU), while others were found to steadily decline with increasing wind speeds (German sites – 846 it should be noted that the German sites are the only ones that are located at a great distance from the sea), while some were found to reach a peak and then decline, which also leads to smaller R<sup>2</sup> 848 849 (UKRU, UKRO, SPARU and to a lesser extent GREUB – figures S4a, e and f). The reasons for 850 these differences between the sites are very hard to distinguish as apart from the wind speed the 851 origin and the characteristics of these air masses play a crucial role. Following this, it appears that 852 NPF probability is very low or zero for wind speeds close to calm for the sites with an increasing 853 trend (as well as those that have a peak and decline after), while the opposite is observed for the 854 German sites where the maximum NPF probability is found for very low wind speeds (fig. S4c). 855 856 Similarly, the effect of different wind speeds upon the growth rate also varied a lot, though it was found to be negative in all the cases where R2 was higher than 0.50 (UKUB, DENRU, DENRO, 857 GERRU, GERUB and GREUB). Finally, the formation rate was found to have a significant 858 859 correlation ( $\mathbb{R}^2 > 0.40$ ) only at two sites (UKRO and DENRU), probably indicating that the 860 variability of the wind speed either does not affect this variable or its effect is rather small. 861

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

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#### 3.1.5 **Pressure**

In almost all the sites with available data (apart from the Spanish), the NPF probability presented a 868 positive relationship with high significance at all types of sites. The greater significance found at the rural sites (apart from SPARU) indicates the increased importance of meteorological conditions in the occurrence of NPF events at this type of site. The growth rate also presented a similar picture, 871 with positive relationships at all the background sites of this study except the ones in Greece (R<sup>2</sup> > 0.71) and FINUB (though with low R<sup>2</sup> at 0.02). This is probably associated with the seasonal 873 variation found in Greece where higher growth rates were found in summer, a period when increased wind speeds and lower atmospheric pressure was found due to the Etesians, a pressure system that develops in the region every summer (Kalkavouras et al., 2017). An interesting finding is the negative slopes gradients found at all the roadside sites, though the significance of these 878 results is relatively low ( $R^2 < 0.43$ ) and always lower compared to the rural sites. The effects of pressure above are not likely to be important. Once again however, this is not an independent 880 variable and higher pressure in summer tends to be associated with higher insolation and

temperatures and lower RH. Since most events occur in the warmer months of the year, this is

probably the explanation for the apparent effects of pressure. The formation rate presented relationships of low significance  $(R^2 < 0.47)$  for the sites of this study. Due to this, pressure should 884 not be an important factor for the formation rate at any type of site. 885 886 The normalised slopes gradients did not present any clear trends, even for the NPF probability for which the results presented significant relations at almost all sites. 887 888 889 3.2 **Atmospheric Composition** The slopes-gradients, and R<sup>2</sup> and p-values from the analysis of a number of air pollutants (SO<sub>2</sub>, NO<sub>x</sub>, O<sub>3</sub>, organic compounds, sulphate and ammonia) and the condensation sinkCS, as well as the 891 892 average conditions of these variables are found in Table 4. The results for each site and variable are 893 found in Figures S6 - S124. 894 3.2.1 Sulphur dioxide (SO<sub>2</sub>) Sulphur dioxide, as a precursor of H<sub>2</sub>SO<sub>4</sub>, is considered as one of the main components that associated towithparticipate in the NPF process. According to nucleation theories and observations, 896 897 H<sub>2</sub>SO<sub>4</sub> is the most important compound from which the initial clusters are formed, as well as one of 898 the candidate compounds for the initial steps of particle growth (Kirkby et al., 2011; Nieminen et 899 al., 2010; Sipila et al., 2010; Stolzenburg et al., 2020). As H<sub>2</sub>SO<sub>4</sub> in the atmosphere is produced from oxidation reactions of SO<sub>2</sub> it would be expected that increased concentrations of the latter 900

would be associated with increased values for all the variables associated with the NPF process.

Contrary to this though, the relationship of SO<sub>2</sub> concentrations with NPF probability was found to be negative at all the sites in this study with available data. This relationship was relatively strong  $(R^2 > 0.50)$  in most areas with an increased significance at roadside sites compared to their respective rural sites. As this is a negative relationship, this may indicate that SO<sub>2</sub> is in sufficient concentrations for H<sub>2</sub>SO<sub>4</sub> formation, thus not suppressing the occurrence of NPF events, as well as showing that in increased concentrations, it is a more important factor (or surrogate for a factor) in preventing the occurrence of NPF events within the urban environment, as probably higher SO<sub>2</sub> is likely associated with increased co-emitted particle pollution and hence CS. The growth rate on the other hand, presented mixed results and the significance of the relationships is low in most cases, which makes these results untrustworthyunreliable. Finally, the relationship of SO<sub>2</sub> concentrations with the formation rate was found to be positive at all sites but SPARU and FINRU (which had the lowest concentrations across the sites of this study with available data). The significance of this relationship was rather low  $(R^2 < 0.40)$  for all but the roadside sitess. This suggests that higher H<sub>2</sub>SO<sub>4</sub> concentrations favour increased greater formation rates (i.e. more particles can be formed), rather than necessarily promoting nucleation itself because of the competing effect of condensation onto the pre-existing particle population.

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The normalised slopes gradients  $a_N^*$  were found to be more negative at the background sites compared to their respective roadside sites, as well as being less negative in the UK (where SO<sub>2</sub> is in greater abundance) compared to the other sites with relatively significant relationships. Plotting

the average  $SO_2$  concentrations with the normalised slopes-gradients  $a_N^*$  for the all sites (though not all had significant relations), a positive relationship with relatively high  $R^2$  (when the extreme values from Marylebone Road-UKRO are removed) is found which might indicate that while increased concentrations are a negative factor in NPF event occurrence at a given site, in general the sites with higher  $SO_2$  concentrations on average present higher probability for NPF events (Figures 7a and 7b). This appears to be in agreement with Dall'Osto et al. (2018) who discussed the variable role of  $SO_2$  depending on its concentrations. No significant relations were found for the values of  $a_J^*$  as in most cases these relationships were rather weak.

# 3.2.2 Nitrogen oxides or nitrogen dioxide (NO<sub>x</sub> or NO<sub>2</sub>)

 $NO_x$  and  $NO_2$  are directly associated with pollution, which can be a limiting factor for NPF events as it increases the CS and may suppress the events (An et al., 2015), though with the reduction of  $SO_2$  concentrations achieved the last couple of decades, there is <u>a</u>-possibility for oxidation products of  $NO_x$  to become an important component for NPF (Wang et al., 2020). For almost all sites (apart from GRERU) with available data a negative relation<u>ship</u> between the NPF probability and  $NO_x$  concentrations (or  $NO_2$  depending on the available data) concentrations (depending on what data was available) was found. Similarly, for all the sites but SPARU and GRERU, the correlations were strong with  $R^2 > 0.43$ . The rural background sites had a weaker relation<u>ship</u> between the two variables compared to the urban sites, which is probably associated with them having rather low

concentrations and variability of NO<sub>x</sub> (or NO<sub>2</sub>) and variability, making the variations of this factor

less important. Growth rate had weaker correlations with NO<sub>x</sub> and different trends between the sites, either being positive or negative. The variable effect of NO<sub>xNOX</sub> on particle growth, shifting 944 HOMs<sup>2</sup> volatility, was previously discussed by Yan et al. (2020). While variability was found for 945 the background sites, all roadside sitess regardless of the strength of the relationship had a positive relation between NO<sub>x</sub> and the growth rate. This may indicate the different components associated 946 947 with the growth process at each type of site which, as found in other studies, can be related to compounds associated with combustion processes that take place within the urban environment 948 949 (Guo et al., 2020; Wang et al., 2017a). The formation rate presents few cases of strong relationships, with variable trends (positive and negative). While much effort was made to isolate 951 the effect of NPF events by taking a shorter time frame before the event, the effect of local pollution is still included, especially at the urban sites (which probably explains the positive effect found). 953 954 The normalised slopes-gradients do not provide a significant result for the relationship of this 955 variable with either the probability of the events or the formation rate. The only noteworthy points are the more negative  $a_N^*$  at the rural background sites compared to the roadside sites in all the 957 areas studied, which shows the increased importance of a clean environment for NPF events to 958 occur in areas where condensable compounds are in lesser abundance, such as a rural environment. 959 Additionally, the negative slopes gradients found at all the roadside sites, which increases the confidence that the events extracted at the roadside sitess are not pollution incidents but NPF 960

events. However, it appears that traffic pollution favours higher particle growth rates, although the components responsible for this effect are unknown.

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## 3.2.3 Ozone (O<sub>3</sub>)

Ozone is typically the result of atmospheric photochemistry and is itself a source of hydroxyl radical through photolysis, or ozonolysis of alkenes both during daytime and night-time (Fenske et al., 2000). It might therefore be expected to act as an indicator of photochemical activity which promotes the oxidation of SO<sub>2</sub> and VOCs. Ozone concentrations may be directly related to the solar radiation intensity as well as the pollution levels in the area studied, and O<sub>3</sub> is considered as a positive factor in the occurrence of NPF events (Woo et al., 2001; Berndt et al., 2006). As withfor the solar radiation intensity, there is a strong relationship between O<sub>3</sub> concentration and the probability for NPF events. This positive relationship was found to be stronger for the sites in northern Europe ( $\mathbb{R}^2 > 0.51$ ), while it was not significant ( $\mathbb{R}^2 < 0.38$ ) for the sites from in southern Europe (Spanish sites and GRERU), possibly indicating that  $O_3$  is a less important factor at the southern sites. Specifically for the Spanish sites which have the highest average concentrations of O<sub>3</sub> with some extreme values (Querol et al., 2017), the relationship of O<sub>3</sub> concentrations with the NPF probability presents a unique trend (Figure S8d), having a clear peak then a steady decline at both sites (though at different O<sub>3</sub> concentrations), which is also responsible for the low correlations found (this trend seems to also occur at SPARU for the growth rate and to a lesser extent for the formation rate as well, though for different O<sub>3</sub> concentration ranges – figures S8i and n). The

specific variability found at the Spanish sites was also studied by Carnerero et al., (2019). For sites with a marked seasonal variation in ozone, associations with NPF may be artefactual due to correlations with other variables such as temperature, RH and solar radiation intensity. Unlike the solar radiation intensity though, the growth rate presents a negative relationship at the sites where the relationship between these two variables was significant (UKRU, UKUB, DENUB and FINRU), which might either be an indication of a polluted background that may have a negative effect in the growth of the newly formed particles (though the trends found for NO<sub>x</sub> indicate differently) or specific chemical processes which cannot be identified due to the lack of detailed chemical composition data. A significant relationship between O<sub>3</sub> and the formation rate was only found for two a few sites (UKRO and DENRO, though the trends become a lot clearer if some values are removed from the extreme lower or higher end). This way the relationships become strong, but positive, for some areas and negative for some others without any clear trend (type or location of the site, O<sub>3</sub> concentrations etc.). No clear relationship between these two variables was found as the sites with strong relationship have both positive (DENRO) and negative (UKRO) relationships and as a result no confident conclusions can be drawn. As the correlations found were strong the normalised slopes gradients for NPF probability, when plotted against the average concentrations of O<sub>3</sub>, present a negative correlation with relatively high  $R^2$  (0.64), indicating that the  $O_3$  is a more important factor in the occurrence of NPF events when in

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lower concentrations (Figure 8). Finally, though with a low level of confidence for the southern

sites, the  $a_N^*$  were smaller at the southern sites compared to those in the north, up to one order of magnitude between the FINRU (furthest north rural background) and GRERU (furthest south rural background).

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## 3.2.4 Organic compounds

## 3.2.4.1 Particulate organic carbon (OC)

Organic carbon (OC) compounds are considered as components with importance in the growth of newly formed particles in the secondary aerosol typically enter the particles via condensational 1008 1009 processes, with a role that becomes increasingly important as the size of the particles becomes 1010 larger (Nieminen et al., 2010; Zhang et al., 2012; Shrivastava et al., 2017). Particulate OC, the data for which are is available in the present study, can be associated with pollution, especially in the 1012 urban environment. Only a few of the sites of the present study were found to have a relatively 1013 strong negative relationship ( $R^2 > 0.50$ ) of particulate OC with the NPF probability (UKUB, UKRO 1014 and DENRU). Regardless though of the strength of this relationship, all other sites (apart from 1015 FINRU) had a negative relationship between these two variables as well, consistent with increased concentrations of particulate OC being associated with increased pollution, which elevate the CS, is 1017 a-suppressing factor in the occurrence of NPF events. Growth rate on the other hand was found to have a slight positive relationship  $(R^2 > 0.40)$  for most of the sites. This relationship appeared to be 1018 stronger (higher R<sup>2</sup>) at the roadside sitess with available data compared to their respective rural 1020 background sites. The relationship between particulate OC and the growth rate was positive at all

the sites with available data regardless of their significance showing that, despite its effect in the occurrence of NPF events, it is still a favourable variable for the growth of the particles. The formation rate was found to have a significant relationship with particulate OC concentrations at half of the sites with available data (UKUB, UKRO, DENRU, DENRO).

1026 The normalised slopes gradients for this variable did not present any noteworthy relations with
 1027 either the type of site or the concentrations of OC at a given site.

# 3.2.4.2 Volatile organic compounds (VOCs)

Many volatile organic compounds have been found to be associated with the NPF process. Benzene, toluene, ethylbenzene, m\_+p-xylene, o-xylene and trimethylbenzenes have been reported to be able to form Highly Oxygenated Organic Molecules (HOMs) in flow tubes (Wang et al., 2017a; Molteni et al., 2018), which may act as contributors to particle nucleation and/or growth. Xylenes, and to a lesser extent trimethylbenzenes, are the most efficient at forming HOMs. Benzene and toluene are less efficient and will form more volatile HOMs. These HOMs may all be too volatile to form new particles, though this is not yet confirmed. Chamber studies involving H<sub>2</sub>SO<sub>4</sub> and trimethylbenzene oxidation products were associated with high formation rates when measuring J<sub>1.5</sub> (Metzger et al., 2010). All these HOMs though will be sufficiently involatile to contribute to particle growth. Those with higher oxygen content or carbon number will be classed as LVOC and if they dimerise, they will form ELVOC (Bianchi et al., 2019). Monoterpenes can also form HOMs which drive both the

1041 formation (Ehn et al., 2014; Riccobono et al., 2014) and growth (Tröstl et al., 2016), while isoprene 1042 can act as a sink for hydroxyl radical (Kiendler-Scharr et al., 2009) and is not as effective in HOM 1043 and secondary organic aerosol formation compared to monoterpenes (McFiggans et al., 2019). 1044 Volatile organic compound data were available for three of the sites of this study (Table S2). Two 1045 1046 of the sites with VOC data were from the rural background and the roadside site in the UK. Most of 1047 the compounds are associated with combustion sources and were found to have a negative relationship with NPF event occurrence at both sites, with high  $R^2$  ( $R^2 > 0.50$ ) in most cases. 1048 1049 Additionally, isoprene, which may have either biogenic or anthropogenic sources (Wagner and 1050 Kuttler, 2014) was also found to have a negative relationship with NPF event occurrence at Marylebone Road-UKRO, though with low  $R^2(0.07)$ . This result is in line with the VOCs being 1052 strongly correlated with particulate OC (which presented a negative relationship with NPF event 1053 probability, as discussed in Section 3.2.4.1), as well as with the CS (which also presented a negative 1054 relationship with NPF event probability, as mentioned in Section 3.2.6), further associating these 1055 compounds with combustion emissions. 1056 1057 Growth rate was found to have a positive relationship with VOCs in almost all cases for both UK 1058 sites. Few exceptions were found (with only 1,3 butadiene having a relatively high R<sup>2</sup>) which 1059 presented a negative relationship with the growth rate in rural Harwell-UKRU. Finally, the 1060 formation rate presented a different behaviour between the two sites. At Harwell-UKRU, the

1061 relationship was unclear in most cases, with a group of VOCs presenting a negative relationship 1062 with the formation rate (ethane, ethene, propane, 1,3 butadiene, toluene, ethylbenzene, o-xylene and 1063 1,2,4 trimethylbenzene – with  $R^2 > 0.40$ ), two VOCs presented a rather clear positive relationship 1064 with the formation rate (iso-pentane and 2-methylbenzene) and the rest of the VOCs had an unclear 1065 relationship. At Marylebone Road UKRO though, VOCs presented a positive relationship with the 1066 formation rate (for particles of diameter 16 nm). This is probably due to the fact that these VOCs 1067 are associated with pollution emissions (as mentioned earlier) and though a smaller time window 1068 was chosen to avoid including the effect of the morning rush hour traffic, this is very difficult in the 1069 traffic polluted environment of Marylebone Road-UKRO. 1070 1071 As Hyvtiälä (FINRU) is a rural background site far from the direct effect of combustion emissions, 1072 different VOCs were measured, which mainly originate from biogenic sources rather than 1073 anthropogenic ones. The results were mixed and less clear compared to those from the UK sites 1074 (mainly due to the smaller dataset), and three groups were found depending on their relationship with NPF probability. The first group, including acetonitrile, acetic acid and mMethyl elethyl 1076 **k**Ketone (MEK) presented a slight positive relationship. The second group presented a negative 1077 relationship, with the VOCs in this group being MEK, monoterpenes, methacroleine, benzene, 1078 isoprene and toluene (only the last two have  $R^2 > 0.50$ ). Finally, the third group included VOCs that

presented a peak and then a decline for higher concentrations including methanol, and acetone. Two

groups of VOCs were found depending on their relationship with the growth rate. The ones with a

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positive relationship being methanol, acetonitrile, acetone, acetic acid, isoprene, MEK methacroleine, monoterpenes and toluene, while acetaldehyde, MEK and benzene had a negative relationship, with relatively high R<sup>2</sup> in most cases. Finally, the results with the formation rate were unclear with only a handful presenting weak ( $\mathbb{R}^2 < 0.21$ ) positive (methanol, acetic acid and benzene) or negative (MEK) relationships that do not appear to be significant. The normalised slopes gradients cannot be used for VOCs as there are very few sites with available data.

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#### 3.2.5 Sulphate (SO<sub>4</sub>-2)

1089 Sulphate (SO<sub>4</sub><sup>2-</sup>) is a major secondary constituent of aerosols. Secondary SO<sub>4</sub><sup>2-</sup> aerosols largely arise 1090 from either gas phase reaction between SO<sub>2</sub> and OH, or in the aqueous phase by the reaction of SO<sub>2</sub> 1091 and O<sub>3</sub> or H<sub>2</sub>O<sub>2</sub>, or NO<sub>2</sub> (Hidy et al., 1994). In environments where SO<sub>4</sub><sup>2-</sup> chemistry is dominant 1092 (i.e. remote areas),  $SO_4^2$  and ammonium (bi) sulphate ((NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub> and NH<sub>4</sub>HSO<sub>4</sub>) particles are a 1093 large relative contributor to aerosol mass, while this contribution is lower in environments where 1094 other emissions are also significant (i.e. urban areas where the secondary NO<sub>3</sub>- relative contribution 1095 is a lot higher). While not well established, a possible relationship of SO<sub>4</sub><sup>2</sup>-containing compounds 1096 and variables of NPF events was found in previous studies (Beddows et al., 2015; Minguillón et al., 2015; Wang et al., 2017b). In the present study, only a few sites had SO<sub>4</sub><sup>2-</sup> data available, for PM<sub>1</sub> 1098 (FINRU), PM<sub>2.5</sub> (Danish sites) or PM<sub>10</sub> (rest of the sites). While this data cannot be considered as directly associated with the ultrafine particles, for two sites with available AMS data for ultrafine 1100 particles, the direct comparison between SO<sub>4</sub><sup>2</sup>- aerosol in PM and in the range of particles of about

50 nm, very high correlations were found (results not included). For all the sites with available data the NPF probability presented a negative relationship. The significance of this relationship was 1103 found to be relatively high ( $R^2 > 0.50$ ) only for background sites (apart from GERRU, which has 1104 rather low concentrations and probably different mechanisms for the NPF events). Similarly, the 1105 growth rate presented a more significant relationship  $(R^2 > 0.40)$  for the same background sites 1106 (apart from FINRU), though this relationship was found to be positive at all sites regardless of its 1107 significance. Finally, the formation rate did not present a clear trend as it was found to have both negative and positive relationships for different sites. This relationship was significant only for two rural sites (UKRU and DENRU) and as a result no assumptions conclusions can be made reached. 1110 The normalised slopes gradients cannot be used for any analysis on sulphate as the measurements 1112 available are from different particle size ranges. 1113 1114

#### 3.2.6 Gaseous ammonia (NH<sub>3</sub>)

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Ammonia (NH<sub>3</sub>) can be an important compound in the nucleation process according to the ternary theory (Kirkby et al., 2011; Napari et al., 2002). It was found that elevations in NH<sub>3</sub> concentrations can lead to elevations to NPF rate (Lehtipalo et al., 2018) and it was also found to be an important factor for NPF event occurrence even when stronger bases are present in high concentrations (Glasoe et al., 2015). No significant variation was found though between event and non-event days in a previous study in Harwell - Harwell - UKRU (Bousiotis et al., 2019). Data for gaseous ammonia were-was only available for Harwell-UKRU and presented a positive relationship with NPF probability, until reaching a peak point. Further increase in NH<sub>3</sub> concentrations presented a decline with NPF probability (Figure S11a), which might be due to its association with increased pollution levels. Interesting though is that iIt presented a clear positive relationship with both the growth rate (though it also appears to decline at high concentrations) and the formation rate, consistent with its well-established role in accelerating both of these processes (Kirkby et al. 2011; Stolzenburg et al., 2020).

## 1129 3.2.7 Condensation sink (CS)

The CS is a measure of the rate at which molecules will condense onto pre-existing aerosols (Lehtinen et al., 2003). It is highly dependent on the number and size of the particles in the atmosphere and as a result it is expected to be affected by both the local emissions within the urban environment as well as the formation and growth of the particles due to NPF events. As a result, for the specific metric a time frame before the events are in full development was chosen (05:00 to 10:00 LT) to avoid including the effect of the NPF events and provide a picture of the atmospheric conditions that preceded the NPF events. With this data, the NPF probability presented very strong relationships with the condensation sink. Two groups of sites were found though; those which had a positive relationship and those with a negative relationship. In the first group are the sites in Germany and Greece while all others had a negative relationship. This grouping follows the trend between the countries, the sites of which presented a greater (the ones with the positive slopes) or

smaller CS on NPF event days (having positive or negative gradients respectively), though it is unknown what causes this behaviour (at the German sites and GREUB it may be associated with the very high formation rates on NPF event days). While the slopes gradients from this analysis cannot be used for direct comparisons, a trend was found for which the slopes gradients were more positive or negative at the rural sites compared to their respective roadside sites, which might indicate the greater importance of the variability of the CS at the rural sites in the occurrence of NPF events. The growth rate was positively correlated with the CS for most of the sites, with strong relationships (high- $R^2 > 0.40$ ) for about half of them. As the CS is a metric of pre-existing particles, it is also associated with the level of pollution in a given area. The increased significance and slope gradient found at the rural sites probably indicates the importance of enhanced presence of condensable compounds in a cleaner environment, which in many cases are associated with the moderate presence of pollution. The formation rate was also found to have a positive relationship with the CS. This relationship was more significant at the roadside sites of this study, a result which to some extent is biased by the presence of increased traffic emissions found in the timeframe chosen. While to an extent, increased presence of condensable compounds can be favourable for greater formation rates, this result should be considered with great caution. The normalised slopes gradients an followed a similar trend as those found with the initial analysis. These slopes gradients were found to be more positive or negative, depending on the trend of the

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given area, at the rural sites compared to their roadside <u>sitess</u>. The urban background sites did not always have a uniform behaviour (though in UK, Denmark and Finland these were between the rural site and the roadside <u>site</u>), due to their more diverse character compared to the other two types of sites.

# 3.3 Association of the Effect of the Variables

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

The relatively strong relationship between the solar radiation intensity, temperature and O<sub>3</sub> found, as well as their anticorrelation with the RH may lead to the conclusion that not all these factors play a role in NPF events, but their visible effect is the result of their relationship with each other. There is a similar case with the association of the CS and NO<sub>x</sub> (or NO<sub>2</sub>), and OC, as well as SO<sub>2</sub>, especially at urban sites. However, the factors affect different outcomes differently, as for example the solar radiation intensity does not seem to be as important a factor for the growth rate as temperature, or O<sub>3</sub> does not seem to be strongly associated with either the formation or the growth rate. This is further established by the fact that some of these variables do not correlate well at the southern sites, but still appear to be associated with either the probability of NPF events or the growth or nucleation rate. The effects of all of these factors have been demonstrated in both laboratory and atmospheric studies in the past and were discussed earlier in this paper. By the analysis provided in the present study, the effect of each of these variables is further established, providing an association of each one of these variables with either the formation or the growth

mechanism. However, RH does not seem to be a consistent factor in any mechanism, and it appears that its effect is dependent on location specific conditions, although it was the variable with the most consistent relation with NPF event probability at almost all sites.

## 3.4 Relationship to a previous-multi-station European study

The findings of our study in respect of the background sites show many similarities with the conclusions drawn in the previous multi-station study in Europe by Dall'Osto et al. (2018) despite the two studies using several different sampling stations as well as some in common. Both studies point towards the influence of variables such as solar radiation intensity and CS upon the occurrence of NPF events. The previous study suggested that different compounds participate in the growth of the particles, depending on the area considered. Thus, for northern and southern sites the growth of the particles is suggested to be driven mainly by organic compounds, while for the sites in central Europe sulphate plays a more important role. These findings are confirmed by the present study, as the growth rate was found to correlate better with organic compounds for the rural sites in Finland and Greece, while SO<sub>4</sub><sup>2-</sup> presented a stronger relationship with the growth rate for the Danish and German sites (the latter presented high slope-gradient values but low R<sup>2</sup> due to a decline at higher SO<sub>4</sub><sup>2-</sup> concentrations — figure S10i, probably associated with NPF events being suppressed by increased pollution). The growth of the particles at the rural background site in the UK, characterised as "Overlap" in the previous study, was found to be strongly associated with both organic compounds and sulphate, consistent with it being in the central group.

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

## 4. CONCLUSIONS

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The present study attempts to explain the effect of several meteorological and atmospheric variables on the occurrence and development of NPF events, by using a large-scale dataset. More than 85 1212 1213 site-years of data from 16 sites from six countries in Europe were analysed for NPF events. A total of 19520 NPF events with consequent growth of the newly formed particles were extracted and with the use of binned linear regression, the relationship between three variables associated with 1216 NPF events (NPF event probability, formation and growth rate) with meteorological conditions and 1217 atmospheric composition was studied. Among the meteorological conditions, solar radiation 1218 intensity, temperature and atmospheric pressure presented a positive relationship with the occurrence of NPF events-occurrence, and either promoting the formation or growth rate. Relative 1220 humidityRH presented a negative relationship with NPF event probability which in most cases was

associated with it being a limiting factor on particle formation at higher average values. Wind speed on the other hand presented variable results, appearing to depend on the location of the sites rather than their type. This shows that while wind speed can be a factor in NPF event occurrence, the origin of the incoming air masses also plays a very important role. In most cases, meteorological conditions, such as temperature or RH appeared to be more important factors in NPF event occurrence at rural sites compared to urban sites, suggesting that NPF events are driven more by them at this type of site compared to urban environments and the more complex chemical interactions found there. Additionally, while some meteorological variables appeared to play a crucial role in the occurrence of NPF events, this role appears to become less important at higher values when a positive relation was found (or lower when a negative relation was found). The results for the levels of atmospheric pollutants presented a more interesting picture as most of these, which appear to be either directly or indirectly associated with the NPF process were found to have negative relationships with NPF probability. This is probably due to the fact that increased concentrations of such compounds are associated with more polluted conditions, which are a limiting factor in the occurrence of NPF events, as was found with the negative relationship between the CS and NPF probability in most cases. Thus, SO<sub>2</sub>, NO<sub>x</sub> (or NO<sub>2</sub>), particulate OC and SO<sub>4</sub><sup>2-</sup> concentrations were negatively correlated with NPF probability in most cases. Average SO<sub>2</sub> concentrations though appeared to correlate positively with the normalised NPF event probability slopes gradients with a relatively significant correlation, indicating that while increasing

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sites with higher SO<sub>2</sub> concentrations have higher probability for NPF events. On the other hand though Conversely, these compounds in many cases had a positive relationship (not always though with high significance) with the other variables considered. Thus, particulate OC (and VOCs where data were was available) and SO<sub>4</sub><sup>2</sup> consistently had a positive relationship with the growth rate, while SO<sub>2</sub> was positively associated with both the formation and growth rate in most cases. Finally, O<sub>3</sub> was positively correlated with NPF event probability at all sites in this study, though it presented variable results with the other two variables. As with some meteorological conditions it was found that at sites with increased concentrations of O<sub>3</sub>, its importance as a factor was decreased, which to an some extent can be related with the high CS associated with peak summer O<sub>3</sub> days in southern 1251 Europe. The present study attempts to explain the effect of several meteorological and atmospheric variables on the occurrence and development of NPF events, by using a large scale dataset. It should be noted that the variables considered are in many cases inter-related (e.g. temperature and RH) and this considerably complicates considerably the interpretation in terms of causal factors. Large datasets are very useful in providing with more uniform results by removing the possible bias of

concentrations have a negative impact in the occurrence of NPF events at a given site, in general

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short period extremities, which may lead to wrong assumptions. This study, apart from providing

insights into the effect of a number of variables on the occurrence and development of NPF events

in atmospheric conditions across Europe, also shows the differences that climatic, land use and

1261	atmospheric composition variations cause to those effects. Such variations are probably the cause of
1262	the differences found among previous studies. Following from this, the importance of a high-
1263	$resolution\ measurement\ network,\ both\ \underline{\text{site-}\underline{\text{spatially}}}\ and\ \underline{\text{timewise-}\underline{\text{temporally}}}\ is\ underlined,\ as\ it\ can$
1 1264	help in elucidating the mechanisms of new particle formation in the real atmosphere.
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1266	DATA ACCESSIBILITY
1267	Data supporting this publication are openly available from the UBIRA eData repository at
1268	https://doi.org/
1269	
1270	AUTHOR CONTRIBUTIONS
1271	The study was conceived and planned by RMH who also contributed to the final manuscript, and
1272	DB who also carried out the analysis and prepared the first draft of the manuscript. AM, JKN, CN,
1273	JVN, HP, NP, AA, GK, SV and KE have provided with the data for the analysis. JB provided help
1274	with analysis of the data. FDP provided advice on the analysis. MDO, XQ and TP contributed to the
1275	final manuscript.
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1277	COMPETING INTERESTS
1278	The authors have no conflict of interests.
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1280	ACKNOWLEDGMENTS

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## REFERENCES

1284 1285

- 1286 Aalto, P., Hämeri, K., Becker, E. D. O., Weber, R., Salm, J., Mäkelä, J. M., Hoell, C., O'Dowd, C.
- 1287 D., Karlsson, H., Hansson, H., Väkevä, M., Koponen, I. K., Buzorius, G. and Kulmala, M.: Physical
- 1288 characterization of aerosol particles during nucleation events, Tellus, Ser. B Chem. Phys. Meteorol..
- 1289 53(4), 344–358, doi:10.3402/tellusb.v53i4.17127, 2001.

1290

- 1291 Alam, A., Shi, J. P. and Harrison, R. M.: Observations of new particle formation in urban air, J.
- 1292 Geophys. Res. Atmos., 108(D3), n/a-n/a, doi:10.1029/2001JD001417, 2003.

1293

- 1294 An, J., Wang, H., Shen, L., Zhu, B., Zou, J., Gao, J. and Kang, H.: Characteristics of new particle
- 1295 formation events in Nanjing, China: Effect of water-soluble ions, Atmos. Environ., 108, 32–40,
- 1296 doi:10.1016/j.atmosenv.2015.01.038, 2015.

1297 1298

- Bae, M.-S., Schwab, J. J., Hogrefe, O., Frank, B. P., Lala, G. G. and Demerjian, K. L.:
- 1299 <u>Characteristics of size distributions at urban and rural locations in New York, Atmos. Chem. Phys.</u>
- 1800 Discuss., 10(1), 69–108, doi:10.5194/acpd-10-69-2010, 2010.

1301

- 1302 Beddows, D. C. S., Harrison, R. M., Green, D. C. and Fuller, G. W.: Receptor modelling of both
- 1303 particle composition and size distribution from a background site in London, UK, Atmos. Chem.
- 1304 Phys., 15(17), 10107–10125, doi:10.5194/acp-15-10107-2015, 2015.

1305

- 1306 Berland, K., Rose, C., Pey, J., Culot, A., Freney, E., Kalivitis, N., Kouvarakis, G., Cerro, J. C., Mallet,
- 1307 M., Sartelet, K., Beckmann, M., Bourriane, T., Roberts, G., Marchand, N., Mihalopoulos, N. and
- 1308 Sellegri, K.: Spatial extent of new particle formation events over the Mediterranean Basin from
- 1309 multiple ground-based and airborne measurements, Atmos. Chem. Phys., 17(15), 9567-9583,
- 1310 doi:10.5194/acp-17-9567-2017, 2017.

1311

- 1312 Berndt, T., Böge, O. and Stratmann, F.: Formation of atmospheric H2SO4H2O particles in the
- 1313 absence of organics: A laboratory study, Geophys. Res. Lett., 33(15), 2-6,
- 1314 doi:10.1029/2006GL026660, 2006.

1315

- 1316 Bianchi, F., Kurtén, T., Riva, M., Mohr, C., Rissanen, M. P., Roldin, P., Berndt, T., Crounse, J. D.,
- 1317 Wennberg, P. O., Mentel, T. F., Wildt, J., Junninen, H., Jokinen, T., Kulmala, M., Worsnop, D. R.,
- 1318 Thornton, J. A., Donahue, N., Kjaergaard, H. G. and Ehn, M.: Highly oxygenated organic molecules
- 1319 (HOM) from gas-phase autoxidation involving peroxy radicals: A key contributor to atmospheric
- aerosol, Chem. Rev., 119, 3472–3509, doi:10.1021/acs.chemrev.8b00395, 2019.

- 1322 Bigi, A. and Harrison, R. M.: Analysis of the air pollution climate at a central urban background site,
- 1323 Atmos. Environ., 44(16), 2004–2012, doi:10.1016/j.atmosenv.2010.02.028, 2010.

1324

- 1325 Birmili, W., Weinhold, K., Rasch, F., Sonntag, A., Sun, J., Merkel, M., Wiedensohler, A., Bastian,
- 1326 S., Schladitz, A., Löschau, G., Cyrys, J., Pitz, M., Gu, J., Kusch, T., Flentje, H., Quass, U., Kaminski,
- 1327 H., Kuhlbusch, T. A. J., Meinhardt, F., Schwerin, A., Bath, O., Ries, L., Wirtz, K. and Fiebig, M.:
- 1328 Long-term observations of tropospheric particle number size distributions and equivalent black
- carbon mass concentrations in the German Ultrafine Aerosol Network (GUAN), Earth Syst. Sci. Data,
- 1330 8(2), 355–382, doi:10.5194/essd-8-355-2016, 2016.

1331

- 1832 Bousiotis, D., Pope, F. D., Beddows, D. C., Dall'Osto, M., Massling, A., Nøjgaard, J. K.,
- 1833 Nørdstrom, C., Niemi, J. V., Portin, H., Petäjä, T., Perez, N., Alastuey, A., Querol, X., Kouvarakis,
- 1834 G., Vratolis, S., Eleftheriadis, K., Wiedensohler, A., Weinhold, K., Merkel, M., Tuch, T., and
- 1835 <u>Harrison, R. M.: An Analysis of New Particle Formation (NPF) at Thirteen European Sites, Atmos.</u>
- 1836 <u>Chem. Phys. Discuss., https://doi.org/10.5194/acp-2020-414, in review, 2020.</u>
- 1837 Bousiotis, D., Pope, F. D., Dall'Osto, M., Massling A., Nøjgaard, J. K., Nørdstrom C., Niemi J. V.,
- 1338 Portin H., Petäjä T., Perez, N., Alastuey, A., Querol, X., Kouvarakis, G., Vratolis, S., Eleftheriadis,
- 1339 K., and Harrison, R. M.: An Analysis of new particle formation (NPF) at several European Sites,
- 1340 Atmos. Chem. Phys., submitted, 2020

1341

- 1342 Bousiotis, D., Osto, M., Beddows, D. C. S., Pope, F. D. and Harrison, R. M.: Analysis of new
- 1343 particle formation (NPF) events at nearby rural, urban background and urban roadside sites, Atmos.
- 1344 Chem. Phys., 19, 5679-5694, 2019.

1345

- 1346 Brines, M., Dall'Osto, M., Beddows, D. C. S., Harrison, R. M., Gómez-Moreno, F., Núñez, L.,
- $1347 \quad Art \'inano, B., Costabile, F., Gobbi, G. P., Salimi, F., Morawska, L., Sioutas, C. and Querol, X.: Traffic$
- 1348 and nucleation events as main sources of ultrafine particles in high-insolation developed world cities,
- 1349 Atmos. Chem. Phys., 15(10), 5929–5945, doi:10.5194/acp-15-5929-2015, 2015.

1350

- 1351 Carnerero, C., Pérez, N., Petäjä, T., Laurila, T. M., Ahonen, L. R., Kontkanen, J., Ahn, K. H.,
- 1352 Alastuey, A. and Querol, X.: Relating high ozone, ultrafine particles, and new particle formation
- episodes using cluster analysis, Atmos. Environ. X, 4(October), doi:10.1016/j.aeaoa.2019.100051,
- 1354 2019.

1355

- 1356 Charron, A. and Harrison, R. M.: Primary particle formation from vehicle emissions during exhaust
- 1357 dilution in the roadside atmosphere, Atmos. Environ., 37(29), 4109-4119, doi:10.1016/S1352-
- 1358 2310(03)00510-7, 2003.

1359

- 1360 Charron, A., Birmili, W. and Harrison, R. M.: Fingerprinting particle origins according to their size
- 1361 distribution at a UK rural site, J. Geophys. Res. Atmos., 113(7), 1–15, doi:10.1029/2007JD008562,
- 1362 2008.

- 1364 Charron, A., Degrendele, C., Laongsri, B. and Harrison, R. M.: Receptor modelling of secondary and
- 1365 carbonaceous particulate matter at a southern UK site, Atmos. Chem. Phys., 13(4), 1879-1894,
- 1366 doi:10.5194/acp-13-1879-2013, 2013.
- 1367
- 1368 Cheung, H. C., Chou, C. C.-K., Huang, W.-R. and Tsai, C.-Y.: Characterization of ultrafine particle
- 1369 number concentration and new particle formation in an urban environment of Taipei, Taiwan, Atmos.
- 1370 Chem. Phys., 13(17), 8935–8946, doi:10.5194/acp-13-8935-2013, 2013.
- 1371
- 1372 Chu, B., Kerminen, V., Bianchi, F., Yan, C., Petäjä, T. and Kulmala, M.: Atmospheric new particle
- 1373 formation in China, Atmos. Chem. Phys., 19, 115-138, doi:10.5194/acp-2018-612, 2019
- 1374
- 1875 Costabile, F., Birmili, W., Klose, S., Tuch, T., Wehner, B., Wiedensohler, A., Franck, U., Konig, K.
- 1376 and Sonntag, A.: Spatio temporal variability and principal components of the particle number size
- 1377 distribution in an urban atmosphere, Atmos. Chem. Phys., 9(9), 3163-3195, doi:10.5194/aep-9-3163-
- 1378 <del>2009, 2009.</del>
- 1379
- 1380 Dada, L., Paasonen, P., Nieminen, T., Buenrostro Mazon, S., Kontkanen, J., Peräkylä, O.,
- 1381 Lehtipalo, K., Hussein, T., Petäjä, T., Kerminen, V. M., Bäck, J. and Kulmala, M.: Long-term
- analysis of clear-sky new particle formation events and nonevents in Hyytiälä, Atmos. Chem. Phys.,
- 1383 17(10), 6227–6241, doi:10.5194/acp-17-6227-2017, 2017.
- 1384
- 1385 Dai, L., Wang, H., Zhou, L., An, J., Tang, L., Lu, C., Yan, W., Liu, R., Kong, S., Chen, M., Lee, S.
- 1386 and Yu, H.: Regional and local new particle formation events observed in the Yangtze River Delta
- 1387 region, China, J. Geophys. Res., 122(4), 2389-2402, doi:10.1002/2016JD026030, 2017.
- 1388 1389
- 1390 Dal Maso, M., Kulmala, M., Riipinen, I., Wagner, R., Hussein, T., Aalto, P. P. and Lehtinen, K. E.
- 1391 J.: Formation and growth of fresh atmospheric aerosols: Eight years of aerosol size distribution data
- 1392 from SMEAR II, Hyytiälä, Finland, Boreal Environ, Res., 10(5), 323–336,
- 1393 doi:10.1016/j.ijpharm.2012.03.044, 2005.
- 1394
- 1395 Dall'Osto, M., Beddows, D. C. S., Asmi, A., Poulain, L., Hao, L., Freney, E., Allan, J. D.,
- 1396 Canagaratna, M., Crippa, M., Bianchi, F., De Leeuw, G., Eriksson, A., Swietlicki, E., Hansson, H.
- 1397 C., Henzing, J. S., Granier, C., Zemankova, K., Laj, P., Onasch, T., Prevot, A., Putaud, J. P., Sellegri,
- 1398 K., Vidal, M., Virtanen, A., Simo, R., Worsnop, D., O'Dowd, C., Kulmala, M. and Harrison, R. M.:
- 1399 Novel insights on new particle formation derived from a pan-european observing system, Sci. Rep.,
- 1400 8(1), 1–11, doi:10.1038/s41598-017-17343-9, 2018.
- 1401
- 1402 Dall'Osto, M., Querol, X., Alastuey, A., O'Dowd, C., Harrison, R. M., Wenger, J. and Gómez-
- 1403 Moreno, F. J.: On the spatial distribution and evolution of ultrafine particles in Barcelona, Atmos.

- 1404 Chem. Phys., 13(2), 741–759, doi:10.5194/acp-13-741-2013, 2013.
- 1406 Dall'Osto, M., Beddows, D. C. S., Pey, J., Rodriguez, S., Alastuey, A., M. Harrison, R. and Querol,
- 1407 X.: Urban aerosol size distributions over the Mediterranean city of Barcelona, NE Spain, Atmos.
- 1408 Chem. Phys., 12(22), 10693–10707, doi:10.5194/acp-12-10693-2012, 2012.
- 1409 1410 Eh

1405

- 1410 Ehn, M., Thornton, J. A., Kleist, E., Sipilä, M., Junninen, H., Pullinen, I., Springer, M., Rubach, F.,
- 1411 Tillmann, R., Lee, B., Lopez-Hilfiker, F., Andres, S., Acir, I. H., Rissanen, M., Jokinen, T.,
- 1412 Schobesberger, S., Kangasluoma, J., Kontkanen, J., Nieminen, T., Kurtén, T., Nielsen, L. B.,
- 1413 Jørgensen, S., Kjaergaard, H. G., Canagaratna, M., Maso, M. D., Berndt, T., Petäjä, T., Wahner, A.,
- 1414 Kerminen, V. M., Kulmala, M., Worsnop, D. R., Wildt, J. and Mentel, T. F.: A large source of low-
- 1415 volatility secondary organic aerosol, Nature, 506(7489), 476–479, doi:10.1038/nature13032, 2014.
- 1416
- 1417 Engler, C., Rose, D., Wehner, B., Wiedensohler, A., Bruggermann, E., Gnauk, T., Spindler, G., Tuch,
- 1418 T. and Birmili, W.: Size distributions of non-volatile particle residuals (Dp <800 nm) at a rural site
- 1419 in Germany and relation to air mass origin, Atmos. Chem. Phys., 7, 5785 5802, 2007.
- 1420

1426

- 1421 Fenske, J. D., Hasson, A.S., Paulson, S. E., Kuwata, K. T., Ho, A., Houk, K. N.: The Pressure
- 1422 Dependence of the OH Radical Yield from Ozone Alkene Reactions J Phys Chem A, 104 7821, 2000 1423
- 1424 Fuchs, N. A. and Sutugin, A. G.: Highly dispersed aerosols, Top. Curr. Aerosol Res., 1,
- 1425 doi:https://doi.org/10.1016/B978-0-08-016674-2.50006-6, 1971.
- 1427 Glasoe, W. a, Volz, K., Panta, B., Freshour, N., Bachman, R., Hanson, D. R., Mcmurry, P. H. and
- 1428 Jen, C.: Sulfuric acid nucleation: An experimental study of the effect of seven bases, 1933–1950,
- 1429 doi:10.1002/2014JD022730, 2015.
- 1431 Größ, J., Hamed, A., Sonntag, A., Spindler, G., Manninen, H. E., Nieminen, T., Kulmala, M.,
- 1432 Hõrrak, U., Plass-Dülmer, C., Wiedensohler, A., and Birmili, W.: Atmospheric new particle
- 1433 formation at the research station Melpitz, Germany: connection with gaseous precursors and
- 1434 meteorological parameters, Atmos. Chem. Phys., 18, 1835–1861, https://doi.org/10.5194/acp-18-
- 1435 1835-2018, 2018.
- 1436 Größ, J., Hamed, A., Sonntag, A., Spindler, G. and Manninen, H. E.: Atmospheric new particle
- 1437 formation at the research station Melpitz, Germany: connection with gaseous precursors and
- 1438 meteorological parameters, , 1835 1861, doi:10.5194/acp-18-1835-2018, 2018.
- 1439
- 1440 Guo, S., Hu, M., Peng, J., Wu, Z., Zamora, M. L., Shang, D., Du, Z., Zheng, J., Fang, X., Tang, R.,
- 1441 Wu, Y., Zeng, L., Shuai, S., Zhang, W., Wang, Y., Ji, Y., Li, Y., Zhang, A. L., Wang, W., Zhang, F.,
- 1442 Zhao, J., Gong, X., Wang, C., Molina, M. J. and Zhang, R.: Remarkable nucleation and growth of
- 1443 ultrafine particles from vehicular exhaust, Proc. Nat. Acad. Sci. U. S. A., 117(7), 3427-3432,

1444 doi:10.1073/pnas.1916366117, 2020.

1445

- 1446 Hallar, A. G., Petersen, R., McCubbin, I. B., Lowenthal, D., Lee, S., Andrews, E. and Yu, F.:
- 1447 Climatology of new particle formation and corresponding precursors at storm peak laboratory,
- 1448 Aerosol Air Qual. Res., 16(3), 816–826, doi:10.4209/aagr.2015.05.0341, 2016.

1449

- 1450 Hamed, A., Korhonen, H., Sihto, S. L., Joutsensaari, J., Jrvinen, H., Petäjä, T., Arnold, F.,
- 1451 Nieminen, T., Kulmala, M., Smith, J. N., Lehtinen, K. E. J. and Laaksonen, A.: The role of relative
- 1452 humidity in continental new particle formation, J. Geophys. Res. Atmos., 116(3), 1–12,
- 1453 doi:10.1029/2010JD014186, 2011.

1454

Harrison, R. M.: Urban atmospheric chemistry: A very special case for study, npj Clim. Atmos. Sci., 1456 1(1), 5, doi:10.1038/s41612-017-0010-8, 2017.

1457

- 1458 Henschel, H., Kurtén, T., Vehkamäki, H.: Computational study on the effect of hydration on new
- particle formation in the sulfuric acid/ammonia and sulfuric acid/dimethylamine systems, J. Phys.
- 1460 <u>Chem. A 2016, 120, 11, 1886–1896, 2016.</u> 1461

1462

- 1463 Hidy, G. M.: Atmospheric sulfur and nitrogen oxides, Academic Press, ISBN: 9781483288666,
- 1464 1994

1465

- 1466 Hietikko, R., Kuuluvainen, H., Harrison, R. M., Portin, H., Timonen, H., Niemi, J. V and Rönkkö,
- 1467 T.: Diurnal variation of nanocluster aerosol concentrations and emission factors in a street canyon,
- 1468 Atmos. Environ., 189, 98–106, doi:10.1016/j.atmosenv.2018.06.031, 2018.

1469

- 1470 Iida, K., Stolzenburg, M. R., McMurry, P. H. and Smith, J. N.: Estimating nanoparticle growth rates
- 1471 from size-dependent charged fractions: Analysis of new particle formation events in Mexico City, J.
- 1472 Geophys. Res. Atmos., 113(5), 1–15, doi:10.1029/2007JD009260, 2008.

1473

- 1474 Järvi, L., Hannuniemi, H., Hussein, T., Junninen, H., Aalto, P., Hillamo, R., Mäkelä, T., Keronen, P.
- 1475 and Siivola, E.: The urban measurement station SMEAR III: Continuous monitoring of air pollution
- and surface atmosphere interactions in Helsinki, Finland, 14(April), 86–109, 2009.

1477

- 1478 Jayaratne, R., Pushpawela, B., He, C., Li, H., Gao, J., Chai, F. and Morawska, L.: Observations of
- 1479 particles at their formation sizes in Beijing, China, Atmos. Chem. Phys., 17(14), 8825-8835,
- 1480 doi:10.5194/acp-17-8825-2017, 2017.

- 1482 Jeong, C.-H. H., Evans, G. J., McGuire, M. L., Y.-W. Chang, R., Abbatt, J. P. D. D., Zeromskiene,
- 1483 K., Mozurkewich, M., Li, S.-M. M., Leaitch, W. R., Chang, R. Y.-W., Abbatt, J. P. D. D.,

- 1484 Zeromskiene, K., Mozurkewich, M., Li, S.-M. M. and Leaitch, W. R.: Particle formation and growth
- 1485 at five rural and urban sites, Atmos. Chem. Phys., 10(16), 7979-7995, doi:10.5194/acp-10-7979-
- 1486 2010, 2010.
- 1487
- 1488 Kalkavouras, P., Bossioli, E., Bezantakos, S., Bougiatioti, A., Kalivitis, N., Stavroulas, I.,
- 1489 Kouvarakis, G., Protonotariou, A. P., Dandou, A., Biskos, G., Mihalopoulos, N., Nenes, A. and
- 1490 Tombrou, M.: New particle formation in the southern Aegean Sea during the Etesians: Importance
- 1491 for CCN production and cloud droplet number, Atmos. Chem. Phys., 17(1), 175-192,
- 1492 doi:10.5194/acp-17-175-2017, 2017.
- 1493
- 1494 Kerminen, V., Lehtinen, K. E. J., Anttila, T., Kulmala, M., Lehtinen, K. E. J., Anttila, T. and Kulmala,
- 1495 M.: Dynamics of atmospheric nucleation mode particles: a timescale analysis, Tellus, 56B, 135–146,
- 1496 doi:10.3402/tellusb.v56i2.16411, 2004.
- 1497
- 1498 Kerminen, V. M., Pirjola, L. and Kulmala, M.: How significantly does coagulational scavenging limit
- 1499 atmospheric particle production?, J. Geophys. Res. Atmos., 106(D20), 24119-24125,
- 1500 doi:10.1029/2001JD000322, 2001.
- 1501

- 1502 Kerminen, V. M., Kulmala, M., Worsnop, D. R., Wildt, J. and Mentel, T. F.: A large source of low-
- 1503 volatility secondary organic aerosol, Nature, 506(7489), 476–479, doi:10.1038/nature13032, 2014.
- 1505 Ketzel, M., Wåhlin, P., Kristensson, A., Swietlicki, E., Berkowicz, R., Nielsen, O. J. and Palmgren,
- 1506 F.: Particle size distribution and particle mass measurements at urban, near-city and rural level in the
- 1507 Copenhagen area and Southern Sweden, Atmos. Chem. Phys. Discuss., 3(6), 5513-5546,
- 1508 doi:10.5194/acpd-3-5513-2003, 2004.
- 1509
- 1510 Kiendler-Scharr, A., Wildt, J., Dal Maso, M., Hohaus, T., Kleist, E., Mentel, T. F., Tillmann, R.,
- 1511 Uerlings, R., Schurr, U. and Wahner, A.: New particle formation in forests inhibited by isoprene
- 1512 emissions, 461, 381–384, 2009.
- 1513
- 1514 Kim, K. H., Kabir, E. and Kabir, S.: A review on the human health impact of airborne particulate
- 1515 matter, Environ. Int., 74, 136–143, doi:10.1016/j.envint.2014.10.005, 2015.
- 1516
- 1517 Kirkby, J., Curtius, J., Almeida, J., Dunne, E., Duplissy, J., Ehrhart, S., Franchin, A., Gagné, S., Ickes,
- 1518 L., Kürten, A., Kupc, A., Metzger, A., Riccobono, F., Rondo, L., Schobesberger, S., Tsagkogeorgas,
- 1519 G., Wimmer, D., Amorim, A., Bianchi, F., Breitenlechner, M., David, A., Dommen, J., Downard, A.,
- 1520 Ehn, M., Flagan, R. C., Haider, S., Hansel, A., Hauser, D., Jud, W., Junninen, H., Kreissl, F., Kvashin,
- 1521 A., Laaksonen, A., Lehtipalo, K., Lima, J., Lovejoy, E. R., Makhmutov, V., Mathot, S., Mikkilä, J.,
- 1522 Minginette, P., Mogo, S., Nieminen, T., Onnela, A., Pereira, P., Petäjä, T., Schnitzhofer, R., Seinfeld,
- 1523 J. H., Sipilä, M., Stozhkov, Y., Stratmann, F., Tomé, A., Vanhanen, J., Viisanen, Y., Vrtala, A.,

- 1524 Wagner, P. E., Walther, H., Weingartner, E., Wex, H., Winkler, P. M., Carslaw, K. S., Worsnop, D.
- 1525 R., Baltensperger, U. and Kulmala, M.: Role of sulphuric acid, ammonia and galactic cosmic rays in
- 1526 atmospheric aerosol nucleation, Nature, 476(7361), 429–435, doi:10.1038/nature10343, 2011.
- 1528 Korhonen, P., Kulmala, M., Laaksonen, A., Viisanen, Y., Mcgraw, R. and Seinfeld, J. H.: Ternary
- 1529 nucleation of H2SO4, NH3 and H2O in the atmosphere, J. Geophys. Res., 104(D21), 26349–26353,
- 1530 1999. 1531

- 1532 Kulmala, M., Petäjä, T., Mönkkönen, P., Koponen, I. K., Dal Maso, M., Aalto, P. P., Lehtinen, K.
- 1533 E. J. and Kerminen, V.-M.: On the growth of nucleation mode particles: source rates of condensable
- 1534 vapor in polluted and clean environments, Atmos. Chem. Phys. Discuss., 4(5), 6943–6966,
- 1535 doi:10.5194/acpd-4-6943-2004, 2005.
- 1536
- 1537 Kulmala, M. and Kerminen, V. M.: On the formation and growth of atmospheric nanoparticles,
- $1538 \quad Atmos. \ Res., 90(2-4), 132-150, \\ doi: 10.1016/j. \\ atmosres. 2008. \\ 01.005, 2008. \\$
- 1539
- 1540 Kulmala, M., Kerminen, V.-M. M., Petäjä, T., Ding, A. J. and Wang, L.: Atmospheric gas-to-particle
- 1541 conversion: Why NPF events are observed in megacities?, Faraday Discuss., 200, 271-288,
- 1542 doi:10.1039/c6fd00257a, 2017.
- 1543
- 1544 Kulmala, M., Petäjä, T., Nieminen, T., Sipilä, M., Manninen, H. E., Lehtipalo, K., Dal Maso, M.,
- 1545 Aalto, P. P., Junninen, H., Paasonen, P., Riipinen, I., Lehtinen, K. E. J., Laaksonen, A. and Kerminen,
- 1546 V. M.: Measurement of the nucleation of atmospheric aerosol particles, Nat. Protoc., 7(9), 1651–
- 1547 1667, doi:10.1038/nprot.2012.091, 2012.
- 1548
- 1549 Kulmala, M., Petäjä, T., Mönkkönen, P., Koponen, I. K., Dal Maso, M., Aalto, P. P., Lehtinen, K. E.
- 1550 J. and Kerminen, V.-M.: On the growth of nucleation mode particles: source rates of condensable
- 1551 vapor in polluted and clean environments, Atmos. Chem. Phys. Discuss., 4(5), 6943-6966,
- 1552 doi:10.5194/acpd-4-6943-2004, 2005.
- 1553
- 1554 Kulmala, M., Dal Maso, M., Mäkelä, J. M., Pirjola, L., Väkevä, M., Aalto, P., Miikkulainen, P.,
- 1555 Hämeri, K. and O'Dowd, C. D.: On the formation, growth and composition of nucleation mode
- 1556 particles, Tellus, Ser. B Chem. Phys. Meteorol., 53(4), 479-490, doi:10.3402/tellusb.v53i4.16622,
- **1557** 2001.
- 1558
- 1559 Kürten, A., Li, C., Bianchi, F., Curtius, J., Dias, A., Donahue, N. M., Duplissy, J., Flagan, R. C.,
- 1560 Hakala, J., Jokinen, T., Kirkby, J., Kulmala, M., Laaksonen, A., Lehtipalo, K., Makhmutov, V.,
- 1561 Onnela, A., Rissanen, M. P., Simon, M., Sipilä, M., Stozhkov, Y., Tröstl, J., Ye, P., and McMurry, P.
- 1562 H.: New particle formation in the sulfuric acid—dimethylamine—water system: reevaluation of
- 1563 CLOUD chamber measurements and comparison to an aerosol nucleation and growth model, Atmos.

1564 Chem. Phys., 18, 845–863, https://doi.org/10.5194/acp-18-845-2018, 2018.

1565 1566 Kü

- 1566 Kürten, A., Bergen, A., Heinritzi, M., Leiminger, M., Lorenz, V., Piel, F., Simon, M., Sitals, R.,
- 1567 Wagner, A. C. and Curtius, J.: Observation of new particle formation and measurement of sulfuric
- 1568 acid, ammonia, amines and highly oxidized organic molecules at a rural site in central Germany,
- 1569 Atmos, Chem. Phys., 16(19), 12793–12813, doi:10.5194/acp-16-12793-2016, 2016.

1570

- 1571 Lee, S.-H. H., Uin, J., Guenther, A. B., de Gouw, J. A., Yu, F., Nadykto, A. B., Herb, J., Ng, N. L.,
- 1572 Koss, A., Brune, W. H., Baumann, K., Kanawade, V. P., Keutsch, F. N., Nenes, A., Olsen, K.,
- 1573 Goldstein, A. and Ouyang, Q.: Isoprene suppression of new particle formation: Potential
- 1574 mechanisms and implications, J. Geophys. Res. Atmos., 121(24), 14,621-14,635,
- **1575** doi:10.1002/2016JD024844, 2016.

1576

Lehtinen, K. E. J., Korhonen, H., Dal Maso, M. and Kulmala, M.: On the concept of condensation sink diameter, Boreal Environ. Res., 8(4), 405–411, 2003.

1579

- 1580 Lehtipalo, K., Yan, C., Dada, L., Bianchi, F., Xiao, M., Wagner, R., Stolzenburg, D., Ahonen, L. R.,
- 1581 Amorim, A., Baccarini, A., Bauer, P. S., Baumgartner, B., Bergen, A., Bernhammer, A.,
- 1582 Breitenlechner, M., Brilke, S., Buchholz, A., Mazon, S. B., Chen, D., Chen, X., Dias, A., Dommen,
- 1583 J., Draper, D. C., Duplissy, J., Ehn, M., Finkenzeller, H., Fischer, L., Frege, C., Fuchs, C., Garmash,
- 1584 O., Gordon, H., Hakala, J., He, X., Heikkinen, L., Heinritzi, M., Helm, J. C., Hofbauer, V., Hoyle, C.
- 1585 R., Jokinen, T., Ojdanic, A., Onnela, A., Passananti, M., Petäjä, T., Piel, F., Sarnela, N., Schallhart,
- 1586 S., Schuchmann, S., Sengupta, K. and Simon, M.: Multicomponent new particle formation from
- 1587 sulfuric acid, ammonia, and biogenic vapors, (3), 1–10, 2018.

1588

- 1589 Li, X., Chee, S., Hao, J., Abbatt, J. P. D., Jiang, J. and Smith, J. N.: Relative humidity effect on the
- 1590 formation of highly oxidized molecules and new particles during monoterpene oxidation, Atmos.
- 1591 Chem. Phys., 19(3), 1555–1570, doi:10.5194/acp-19-1555-2019, 2019.

1592

- 1593 Makkonen, R., Asmi, A., Kerminen, V. M., Boy, M., Arneth, A., Hari, P. and Kulmala, M.: Air
- 1594 pollution control and decreasing new particle formation lead to strong climate warming, Atmos.
- 1595 Chem. Phys., 12(3), 1515–1524, doi:10.5194/acp-12-1515-2012, 2012.

- 1597 McFiggans, G., Mentel, T. F., Wildt, J., Pullinen, I., Kang, S., Kleist, E., Schmitt, S., Springer, M.,
- 1598 Tillmann, R., Wu, C., Zhao, D., Hallquist, M., Faxon, C., Le Breton, M., Hallquist, A. M., Simpson,
- 1599 D., Bergström, R., Jenkin, M. E., Ehn, M., Thornton, J. A., Alfarra, M. R., Bannan, T. J., Percival, C.
- 1500 J., Priestley, M., Topping, D. and Kiendler-Scharr, A.: Secondary organic aerosol reduced by mixture
- 1601 of atmospheric vapours, Nature, 565(7741), 587–593, doi:10.1038/s41586-018-0871-y, 2019.
- 1602 Merikanto, J., Spracklen, D. V., Mann, G. W., Pickering, S. J. and Carslaw, K. S.: Impact of
- 1603 nucleation on global CCN, Atmos. Chem. Phys., 9(21), 8601–8616, doi:10.5194/acp-9-8601-2009,

1604 <u>2009</u>.

- 1605 McFiggans, G., Mentel, T. F., Wildt, J., Pullinen, I., Kang, S., Kleist, E., Schmitt, S., Springer, M.,
- 1606 Tillmann, R., Wu, C., Zhao, D., Hallquist, M., Faxon, C., Le Breton, M., Hallquist, Å. M., Simpson,
- 1607 D., Bergström, R., Jenkin, M. E., Ehn, M., Thornton, J. A., Alfarra, M. R., Bannan, T. J., Percival, C.
- 1508 J., Priestley, M., Topping, D. and Kiendler-Scharr, A.: Secondary organic aerosol reduced by mixture
- 1609 of atmospheric vapours, Nature, 565(7741), 587-593, doi:10.1038/s41586-018-0871-y, 2019.

1610

- 1611 Merikanto, J., Spracklen, D. V., Mann, G. W., Pickering, S. J. and Carslaw, K. S.: Impact of nucleation on global CCN, Atmos. Chem. Phys., 9(21), 8601–8616, doi:10.5194/acp-9-8601-2009,
- 1613 2009.

1614

- 1615 Metzger, A., Verheggen, B., Dommen, J., Duplissy, J., Prevot, A. S. H., Weingartner, E., Riipinen,
- 1616 I., Kulmala, M., Spracklen, D. V., Carslaw, K. S. and Baltensperger, U.: Evidence for the role of
- 1617 organics in aerosol particle formation under atmospheric conditions, Proc. Nat. Acad. Sci., 107(15),
- 1618 6646–6651, doi:10.1073/pnas.0911330107, 2010.

1619

- 1620 Minguillón, M. C., Brines, M., Pérez, N., Reche, C., Pandolfi, M., Fonseca, A. S., Amato, F.,
- 1621 Alastuey, A., Lyasota, A., Codina, B., Lee, H. K., Eun, H. R., Ahn, K. H. and Querol, X.: New particle
- 1622 formation at ground level and in the vertical column over the Barcelona area, Atmos. Res., 164–165,
- 1623 118–130, doi:10.1016/j.atmosres.2015.05.003, 2015.

1624 1625

- Mirabel, P. and Katz, J. L.: Binary homogeneous nucleation as a mechanism for the formation of
- 1626 aerosols, J. Chem. Phys., 60(3), 1138–1144, doi:10.1063/1.1681124, 1974.

1627

- 1628 Mølgaard, B., Birmili, W., Clifford, S., Massling, A., Eleftheriadis, K., Norman, M., Vratolis, S.,
- 1629 Wehner, B., Corander, J., Hämeri, K. and Hussein, T.: Evaluation of a statistical forecast model for
- 1630 size-fractionated urban particle number concentrations using data from five European cities, J.
- 1631 Aerosol Sci., 66, 96–110, doi:10.1016/j.jaerosci.2013.08.012, 2013.

1632

- 1633 Molteni, U., Bianchi, F., Klein, F., El Haddad, I., Frege, C., Rossi, M. J., Dommen, J. and
- 1634 Baltensperger, U.: Formation of highly oxygenated organic molecules from aromatic compounds,
- 1635 Atmos. Chem. Phys., 18(3), 1909–1921, doi:10.5194/acp-18-1909-2018, 2018.

1636

- 1637 Napari, I., Noppel, M., Vehkamäki, H. and Kulmala, M.: An improved model for ternary nucleation
- 1638 of sulfuric acid-ammonia-water, J. Chem. Phys., 116(10), 4221–4227, doi:10.1063/1.1450557, 2002.
- 1640 Nieminen, T., Kerminen, V.-M., Petäjä, T., Aalto, P. P., Arshinov, M., Asmi, E., Baltensperger, U.,
- 1641 Beddows, D. C. S., Beukes, J. P., Collins, D., Ding, A., Harrison, R. M., Henzing, B., Hooda, R., Hu,
- 1642 M., Hõrrak, U., Kivekäs, N., Komsaare, K., Krejci, R., Kristensson, A., Laakso, L., Laaksonen, A.,
- 1643 Leaitch, W. R., Lihavainen, H., Mihalopoulos, N., Németh, Z., Nie, W., O'dowd, C., Salma, I.,

- 1644 Sellegri, K., Svenningsson, B., Swietlicki, E., Tunved, P., Ulevicius, V., Vakkari, V., Vana, M.,
- 1645 Wiedensohler, A., Wu, Z., Virtanen, A., Kulmala, M., O'Dowd, C., Salma, I., Sellegri,
- 1646 K., Svenningsson, B., Swietlicki, E., Tunved, P., Ulevicius, V., Vakkari, V., Vana, M., Wiedensohler,
- 1647 A., Wu, Z., Virtanen, A., Kulmala, M., O 'dowd, C., Salma, I., Sellegri, K., Svenningsson, B.,
- 1648 Swietlicki, E., Tunved, P., Ulevicius, V., Vakkari, V., Vana, M., Wiedensohler, A., Wu, Z., Virtanen,
- A. and Kulmala, M.: Global analysis of continental boundary layer new particle formation based on
- 1650 long-term measurements, Atmos. Chem. Phys. Discuss, 5194, 2018–304, doi:10.5194/acp-2018-304,
- 1651 2018.
- 1652
- Nieminen, T., Lehtinen, K. E. J. and Kulmala, M.: Sub-10 nm particle growth by vapor condensation-
- effects of vapor molecule size and particle thermal speed, Atmos. Chem. Phys., 10(20), 9773–9779,
- 1655 doi:10.5194/acp-10-9773-2010, 2010.
- 1656
- 1657 O'Dowd, C. D., Jimenez, J. L., Bahreini, R., Flagan, R. C., Seinfeld, J. H., Hameri Kaarle, Pirjola,
- 1658 L., Kulmala, M., Jennings, S. G. and Hoffmann, T.: Marine aerosol formation from biogenic iodine
- 1659 emissions, Lett. to Nat., 417(June), 1–5, doi:10.1038/nature00773.1.2.3.4.5.6.7.8.9.10., 2002.
- 1660
- Olenius, T., Halonen, R., Kurten, T., Henschel, H., Maatta, O. K., Ortega, I. K., Jen, C.,
- 1662 Vehkamaki, H. and Riipinen, I.: New particle formation from sulfuric acid amines: Comparison of
- monomethylamine, dimethylamine, and trimethylamine, J. Geophys. Res. Atmos., 7103–7118,
- 1664 doi:10.1002/2017JD026501, 2017.
- 1665
- 1666 Olin, M., Kuuluvainen, H., Aurela, M., Kalliokoski, J., Kuittinen, N., Isotalo, M., Timonen, H. J.,
- 1667 Niemi, J. V., Rönkkö, T., and Dal Maso, M.: Traffic-originated nanocluster emission exceeds
- 1668 <u>H2SO4-driven photochemical new particle formation in an urban area, Atmos. Chem. Phys., 20, 1–</u>
- 1669 13, https://doi.org/10.5194/acp-20-1-2020, 2020.
- 1670 1671
- 1672 Paasonen, P., Asmi, A., Petäjä, T., Kajos, M. K., Äijälä, M., Junninen, H., Holst, T., Abbatt, J. P.
- 1673 D., Arneth, A., Birmili, W., Van Der Gon, H. D., Hamed, A., Hoffer, A., Laakso, L., Laaksonen,
- 1674 A., Richard Leaitch, W., Plass-Dülmer, C., Pryor, S. C., Räisänen, P., Swietlicki, E., Wiedensohler,
- 1675 A., Worsnop, D. R., Kerminen, V. M. and Kulmala, M.: Warming-induced increase in aerosol
- number concentration likely to moderate climate change, Nat. Geosci., 6(6), 438–442,
- 1677 doi:10.1038/ngeo1800, 2013.
- 1678
- 1679 Park, M., Yum, S. S. and Kim, J. H.: Characteristics of submicron aerosol number size distribution
- 1680 and new particle formation events measured in Seoul, Korea, during 2004-2012, Asia-Pacific J.
- 1681 Atmos. Sci., 51(1), 1–10, doi:10.1007/s13143-014-0055-0, 2015.
- 1682
- 1683 Petäjä, T., Mauldin, R. L., Kosciuch, E., McGrath, J., Nieminen, T., Paasonen, P., Boy, M.,

- 1684 Adamov, A., Kotiaho, T. and Kulmala, M.: Sulfuric acid and OH concentrations in a boreal forest
- site, Atmos. Chem. Phys., 9(19), 7435–7448, doi:10.5194/acp-9-7435-2009, 2009.
- 1686
- 1687 Pikridas, M., Sciare, J., Freutel, F., Crumeyrolle, S., Von Der Weiden-Reinmüller, S. L., Borbon, A.,
- 1688 Schwarzenboeck, A., Merkel, M., Crippa, M., Kostenidou, E., Psichoudaki, M., Hildebrandt, L.,
- 1689 Engelhart, G. J., Petäjä, T., Prévôt, A. S. H., Drewnick, F., Baltensperger, U., Wiedensohler, A.,
- 1690 Kulmala, M., Beekmann, M. and Pandis, S. N.: In situ formation and spatial variability of particle
- 1691 number concentration in a European megacity, Atmos. Chem. Phys., 15(17), 10219-10237,
- 1692 doi:10.5194/acp-15-10219-2015, 2015.

1693

Pillai, P., Khlystov, A., Walker, J. and Aneja, V.: Observation and analysis of particle nucleation at a forest site in southeastern US, Atmosphere (Basel)., 4(2), 72–93, doi:10.3390/atmos4020072, 2013.

1696

1697 Poling, B. E., Prausnitz, J. M. and O'Connell, J. P.: The properties of gases and liquids, 5th ed., 1698 McGraw-Hill Education., 2001.

1699

1700 Politis, M., Pilinis, C. and Lekkas, T. D.: Ultrafine particles (UFP) and health effects. Dangerous.
1701 Like no other PM? Review and analysis, Glob. Nest J., 10(3), 439–452, 2008.

1702

- Quéléver, L. L. J., Kristensen, K., Normann Jensen, L., Rosati, B., Teiwes, R., Daellenbach, K. R.,
- 1704 Peräkylä, O., Roldin, P., Bossi, R., Pedersen, H. B., Glasius, M., Bilde, M. and Ehn, M.: Effect of
- 1705 temperature on the formation of highly oxygenated organic molecules (HOMs) from alpha-pinene
- 1706 ozonolysis, Atmos. Chem. Phys., 19(11), 7609–7625, doi:10.5194/acp-19-7609-2019, 2019.

1707

- 1708 Querol, X., Gangoiti, G., Mantilla, E., Alastuey, A., Minguillón, M. C., Amato, F., Reche, C., Viana,
- 1709 M., Moreno, T., Karanasiou, A., Rivas, I., Pérez, N., Ripoll, A., Brines, M., Ealo, M., Pandolfi, M.,
- 1710 Lee, H. K., Eun, H. R., Park, Y. H., Escudero, M., Beddows, D., Harrison, R. M., Bertrand, A.,
- 1711 Marchand, N., Lyasota, A., Codina, B., Olid, M., Udina, M., Jiménez-Esteve, B. B., Jiménez-Esteve,
- 1712 B. B., Alonso, L., Millán, M. and Ahn, K. H.: Phenomenology of high-ozone episodes in NE Spain,
- 1713 Atmos. Chem. Phys., 17(4), 2817–2838, doi:10.5194/acp-17-2817-2017, 2017.

1714

- 1715 Riccobono, F., Schobesberger, S., Scott, C. E., Dommen, J., Ortega, I. K., Rondo, L., Almeida, J.,
- 1716 Amorim, A., Bianchi, F., Breitenlechner, M., David, A., Downard, A., Dunne, E. M., Duplissy, J.,
- 1717 Ehrhart, S., Flagan, R. C., Franchin, A., Hansel, A., Junninen, H., Kajos, M., Keskinen, H., Kupc, A.,
- 1718 Makhmutov, V., Mathot, S., Nieminen, T., Onnela, A., Petäjä, T., Tsagkogeorgas, G., Vaattovaara,
- 1719 P., Viisanen, Y., Vrtala, A. and Wagner, P. E.: Oxidation Products of biogenic atmospheric particles,
- 1720 Science, 717, 717–722, doi:10.1126/science.1243527, 2014.

- 1722 Rimnácová, D., Ždímal, V., Schwarz, J., Smolík, J. and Rimnác, M.: Atmospheric aerosols in suburb
- 1723 of Prague: The dynamics of particle size distributions, Atmos. Res., 101(3), 539-552,

- 1724 doi:10.1016/j.atmosres.2010.10.024, 2011.
- 1725 Rose, C., Zha, Q., Dada, L., Yan, C., Lehtipalo, K., Junninen, H., Mazon, S. B., Jokinen, T., Sarnela,
- 1726 N., Sipilä, M., Petäjä, T., Kerminen, V. M., Bianchi, F. and Kulmala, M.: Observations of biogenic
- 1727 ion-induced cluster formation in the atmosphere, Sci. Adv., 4(4), 1–10, doi:10.1126/sciadv.aar5218,
- 1728 2018.
- 1729
- 1730 Rivas, I., Beddows, D. C. S., Amato, F., Green, D. C., Järvi, L., Hueglin, C., Reche, C., Timonen, H.,
- Fuller, G. W., Niemi, J. V, Pérez, N., Aurela, M., Hopke, P. K., Alastuey, A., Kulmala, M., Harrison,
- 1732 R. M., Querol, X. and Kelly, F. J.: Source apportionment of particle number size distribution in urban
- 1733 <u>background and traffic stations in four European cities, Environ. Int., 135, 105345,</u>
- 1734 doi:10.1016/j.envint.2019.105345, 2020.
- 1735
- 1736 Rizzo, L. V., Artaxo, P., Karl, T., Guenther, A. B. and Greenberg, J.: Aerosol properties, in-canopy
- 1737 gradients, turbulent fluxes and VOC concentrations at a pristine forest site in Amazonia, Atmos.
- 1738 Environ., 44(4), 503–511, doi:10.1016/j.atmosenv.2009.11.002, 2010.
- 1739
- 1740 Salma, I., Borsòs, T., Weidinger, T., Aalto, P., Hussein, T., Dal Maso, M. and Kulmala, M.:
- 1741 Production, growth and properties of ultrafine atmospheric aerosol particles in an urban environment,
- 1742 Atmos. Chem. Phys., 11(3), 1339–1353, doi:10.5194/acp-11-1339-2011, 2011.
- 1743
- 1744 Schwartz, J., Dockery, D. W. and Neas, L. M.: Is Daily Mortality Associated Specifically with Fine
- 1745 Particles?, J. Air Waste Manag. Assoc., 46(10), 927–939, doi:10.1080/10473289.1996.10467528,
- 1746 1996.
- 1747
- 1748 Seinfeld, J. H. and Pandis, S. N.: Atmospheric Chemistry and Physics: From Air Pollution to Climate
- 1749 Change, 3rd Editio., John Wiley & Sons, Inc, New Jersey, Canada, 2012.
- 1750
- 1751 Shen, X., Sun, J., Kivekäs, N., Kristensson, A., Zhang, X., Zhang, Y., Zhang, L., Fan, R., Qi, X., Ma,
- 1752 Q. and Zhou, H.: Spatial distribution and occurrence probability of regional new particle formation
- 1753 events in eastern China, Atmos. Chem. Phys., 18(2), 587–599, doi:10.5194/acp-18-587-2018, 2018.
- 1754
- 1755 Shrivastava, M., Cappa, C. D., Fan, J., Goldstein, A. H., Guenther, A. B., Jimenez, J. L., Kuang, C.,
- 1756 Laskin, A., Martin, S. T., Ng, N. L., Petaja, T., Pierce, J. R., Rasch, P. J., Roldin, P., Seinfeld, J. H.,
- 1757 Shilling, J., Smith, J. N., Thornton, J. A., Volkamer, R., Wang, J., Worsnop, D. R., Zaveri, R. A.,
- 1758 Zelenyuk, A. and Zhang, Q.: Recent advances in understanding secondary organic aerosol:
- 1759 Implications for global climate forcing, Rev. Geophys., 55(2), 509-559,
- 1760 doi:10.1002/2016RG000540, 2017.
- 1761
- 1762 Siakavaras, D., Samara, C., Petrakakis, M. and Biskos, G.: Nucleation events at a coastal city during
- 1763 the warm period: Kerbside versus urban background measurements, Atmos. Environ., 140, 60–68,

1764 doi:10.1016/j.atmosenv.2016.05.054, 2016.

1765

- 1766 Sipila, M., Berndt, T., Petaja, T., Brus, D., Vanhanen, J., Stratmann, F., Patokoski, J., Mauldin III,
- 1767 R. L., Hyvarinen, A. P., Lihavainen, H. and Kulmala, M.: The Role of Sulfuric Acid in
- 1768 Atmospheric Nucleation, Science, 327, 1243–1246, doi:10.1126/science.1180315, 2010.

1769

- 1770 Spracklen, D. V., Carslaw, K. S., Merikanto, J., Mann, G. W., Reddington, C. L., Pickering, S., Ogren,
- 1771 J. A., Andrews, E., Baltensperger, U., Weingartner, E., Boy, M., Kulmala, M., Laakso, L., Lihavainen, H., Kivekäs, N., Komppula, M., Mihalopoulos, N., Kouvarakis, G., Jennings, S. G., 1772
- 1773 O'Dowd, C., Birmili, W., Wiedensohler, A., Weller, R., Gras, J., Laj, P., Sellegri, K., Bonn, B.,
- Krejci, R., Laaksonen, A., Hamed, A., Minikin, A., Harrison, R. M., Talbot, R. and Sun, J.: Explaining 1774
- 1775 global surface aerosol number concentrations in terms of primary emissions and particle formation.
- 1776 Atmos. Chem. Phys., 10(10), 4775–4793, doi:10.5194/acp-10-4775-2010, 2010.

1777

- 1778 Stolzenburg, D., Simon, M., Ranjithkumar, A., Kürten, A., Lehtipalo, K., Gordon, H., Ehrhart, S.,
- 1779 Finkenzeller, H., Pichelstorfer, L., Nieminen, T., He, X.-C., Brilke, S., Xiao, M., Amorim, A.,
- 1780 Baalbaki, R., Baccarini, A., Beck, L., Bräkling, S., Caudillo Murillo, L., Chen, D., Chu, B., Dada,
- 1781 L., Dias, A., Dommen, J., Duplissy, J., El Haddad, I., Fischer, L., Gonzalez Carracedo, L., Heinritzi,
- 1782 M., Kim, C., Koenig, T. K., Kong, W., Lamkaddam, H., Lee, C. P., Leiminger, M., Li, Z.,
- 1783 Makhmutov, V., Manninen, H. E., Marie, G., Marten, R., Müller, T., Nie, W., Partoll, E., Petäjä, T.,
- 1784 Pfeifer, J., Philippov, M., Rissanen, M. P., Rörup, B., Schobesberger, S., Schuchmann, S., Shen, J.,
- 1785 Sipilä, M., Steiner, G., Stozhkov, Y., Tauber, C., Tham, Y. J., Tomé, A., Vazquez-Pufleau, M.,
- 1786 Wagner, A. C., Wang, M., Wang, Y., Weber, S. K., Wimmer, D., Wlasits, P. J., Wu, Y., Ye, O.,
- 1787 Zauner-Wieczorek, M., Baltensperger, U., Carslaw, K. S., Curtius, J., Donahue, N. M., Flagan, R.
- C., Hansel, A., Kulmala, M., Lelieveld, J., Volkamer, R., Kirkby, J., and Winkler, P. M.: Enhanced 1788
- 1789 growth rate of atmospheric particles from sulfuric acid, Atmos. Chem. Phys., 20, 7359-7372,
- 1790 https://doi.org/10.5194/acp-20-7359-2020, 2020.

- 1792 Stolzenburg, D., Fischer, L., Vogel, A. L., Heinritzi, M., Schervish, M. and Simon, M., Wagner, A.
- 1793 C., Dada, L., Ahonen, L. R., Amorim, A., Baccarini, A., Bauer, P. S., Baumgartner, B., Bergen, A.,
- 1794 Bianchi, F., Breitenlechner, M., Brilke, S., Buenorstro Mazon, S., Chen, D., Dias, A., Draper, D. C.,
- 1795 Duplissy, J., El Haddad, I., Finkenzeller, H., Frege, C., Fuchs, C., Garmash, O., Gordon, H., He, X.,
- 1796 Helm., J., Hofbauer, V., Hoyle, C. R., Kim, C., Kirkby, J., Kontkanen, J., Kürten, A., Lampilahti, J.,
- 1797 Lawler, M., Lehtipalo, K., Leiminger, M., Mai, H., Mathot, S., Mentler, B., Molteni, U., Nie, W.,
- 1798 Nieminen, T., Nowak, J. B., Ojdanic, A., Onnela, A., Passananti, M., Petäjä, T., Quéléver, L. L. J.,
- 1799 Rissanen, M. P., Sarnela, N., Schallhart, S., Tauber, C., Tome, A., Wagner, R., Wang, M., Weitz,
- 1800 L., Wimmer, D., Xiao, M., Yan, C., Ye, P., Zha, Q., Baltensperger, U., Curtius, J., Dommen, J.,
- 1801 Flagan, R. C., Kulmala, M., Smith, J. N., Worsnop, D. R., Hansel, A., Donahue, N. M., Winkler, P.
- 1802 M.: Rapid growth of organic aerosol nanoparticles over a wide tropospheric temperature range,
- 1803 PNAS, 115(37), doi:10.1073/pnas.1807604115, 2018.

1805 Stolzenburg, D., Fischer, L., Vogel, A. L., Heinritzi, M., Schervish, M. and Simon, M., Wagner, A. C., Dada, L., Ahonen, L. R., Amorim, A., Baccarini, A., Bauer, P. S., Baumgartner, B., Bergen, A., 1807 Bianchi, F., Breitenlechner, M., Brilke, S., Buenorstro Mazon, S., Chen, D., Dias, A., Draper, D. C., 1808 Duplissy, J., El Haddad, I., Finkenzeller, H., Frege, C., Fuchs, C., Garmash, O., Gordon, H., He, X., 1809 Helm., J., Hofbauer, V., Hoyle, C. R., Kim, C., Kirkby, J., Kontkanen, J., Kürten, A., Lampilahti, J., 1810 Lawler, M., Lehtipalo, K., Leiminger, M., Mai, H., Mathot, S., Mentler, B., Molteni, U., Nie, W., 1811 Nieminen, T., Nowak, J. B., Oidanic, A., Onnela, A., Passananti, M., Petäjä, T., Quéléver, L. L. J., Rissanen, M. P., Sarnela, N., Schallhart, S., Tauber, C., Tome, A., Wagner, R., Wang, M., Weitz, 1812 1813 L., Wimmer, D., Xiao, M., Yan, C., Ye, P., Zha, Q., Baltensperger, U., Curtius, J., Dommen, J., 1814 Flagan, R. C., Kulmala, M., Smith, J. N., Worsnop, D. R., Hansel, A., Donahue, N. M., Winkler, P. 1815 M.: Rapid growth of organic aerosol nanoparticles over a wide tropospheric temperature range. 1816 PNAS, 115(37), doi:10.1073/pnas.1807604115, 2018.

1\$17
1818 Tröstl, J., Chuang, W. K., Gordon, H., Heinritzi, M., Yan, C., Molteni, U., Ahlm, L., Frege, C.,
1819 Bianchi, F., Wagner, R., Simon, M., Lehtipalo, K., Williamson, C., Craven, J. S., Duplissy, J.,

1820 Adamov, A., Almeida, J., Bernhammer, A. K., Breitenlechner, M., Brilke, S., Dias, A., Ehrhart, S.,

1821 Flagan, R. C., Franchin, A., Fuchs, C., Guida, R., Gysel, M., Hansel, A., Hoyle, C. R., Jokinen, T., 1822 Junninen, H., Kangasluoma, J., Keskinen, H., Kim, J., Krapf, M., Kürten, A., Laaksonen, A., Lawler,

Junninen, H., Kangasluoma, J., Keskinen, H., Kim, J., Krapf, M., Kurten, A., Laaksonen, A., Lawler, M., Leiminger, M., Mathot, S., Möhler, O., Nieminen, T., Onnela, A., Petäjä, T., Piel, F. M.,

Miettinen, P., Rissanen, M. P., Rondo, L., Sarnela, N., Schobesberger, S., Sengupta, K., Sipilä, M.,

1825 Smith, J. N., Steiner, G., Tomè, A., Virtanen, A., Wagner, A. C., Weingartner, E., Wimmer, D., 1826 Winkler, P. M., Ye, P., Carslaw, K. S., Curtius, J., Dommen, J., Kirkby, J., Kulmala, M., Riipinen, I.,

Winkler, P. M., Ye, P., Carslaw, K. S., Curtius, J., Dommen, J., Kirkby, J., Kulmala, M., Riipinen, I., Worsnop, D. R., Donahue, N. M. and Baltensperger, U.: The role of low-volatility organic compounds

worshop, D. R., Donande, N. M. and Banensperger, U.: The role of low-volutinty organic compounds in initial particle growth in the atmosphere, Nature, 533(7604), 527–531, doi:10.1038/nature18271,

1829 2016.

1804

1830
1831 Vratolis, S., Gini, M. I., Bezantakos, S., Stavroulas, I., Kalivitis, N., Kostenidou, E., Louvaris, E.,
1832 Siakavaras, D., Biskos, G., Mihalopoulos, N., Pandis, S. N. N., Pilinis, C., Papayannis, A. and
1833 Eleftheriadis, K.: Particle number size distribution statistics at City-Centre Urban Background, urban
1834 background, and remote stations in Greece during summer, Atmos. Environ., 213(May), 711–726,
1835 doi:10.1016/j.atmosenv.2019.05.064, 2019.

1836

Wagner, P. and Kuttler, W.: Biogenic and anthropogenic isoprene in the near-surface urban atmosphere - A case study in Essen, Germany, Sci. Total Environ., 475, 104–115, doi:10.1016/j.scitotenv.2013.12.026, 2014.

1840 1**8**41

Wang, D., Fu, Q., Geng, F., Li, L., Wang, H., Qiao, L., Yang, X., Chen, J., Kerminen, V. M.,
 Petäjä, T., Worsnop, D. R., Kulmala, M. and Wang, L.: Atmospheric new particle formation from

- 1843 <u>sulfuric acid and amines in a Chinese megacity, Science, 361(6399), 278–281,</u>
   1844 <u>doi:10.1126/science.aao4839, 2018.</u>
- 1846 Wang, S., Wu, R., Berndt, T., Ehn, M. and Wang, L.: Formation of Highly Oxidized Radicals and
- 1847 Multifunctional Products from the Atmospheric Oxidation of Alkylbenzenes,
- 1848 doi:10.1021/acs.est.7b02374, 2017a.
- 1849

- 1850 Wang, Z., Wu, Z., Yue, D., Shang, D., Guo, S., Sun, J., Ding, A., Wang, L., Jiang, J., Guo, H., Gao,
- 1851 J., Cheung, H. C., Morawska, L., Keywood, M. and Hu, M.: New particle formation in China: Current
- 1852 knowledge and further directions, Sci. Total Environ., 577, 258–266,
- 1853 doi:10.1016/j.scitotenv.2016.10.177, 2017b.
- 1854
- 1855 Wang, F., Ketzel, M., Ellermann, T., Wåhlin, P., Jensen, S. S., Fang, D. and Massling, A.: Particle
- 1856 number, particle mass and  $NO_x$  emission factors at a highway and an urban street in Copenhagen,
- 1857 Atmos. Chem. Phys., 10(6), 2745–2764, doi:10.5194/acp-10-2745-2010, 2010.
- 1858
- 1859 Wang, M., Kong, W., Marten, R., He, X. C., Chen, D., Pfeifer, J., Heitto, A., Kontkanen, J., Dada,
- 1860 L., Kürten, A., Yli-Juuti, T., Manninen, H. E., Amanatidis, S., Amorim, A., Baalbaki, R., Baccarini,
- 1861 A., Bell, D. M., Bertozzi, B., Bräkling, S., Brilke, S., Murillo, L. C., Chiu, R., Chu, B., De
- 1862 Menezes, L. P., Duplissy, J., Finkenzeller, H., Carracedo, L. G., Granzin, M., Guida, R., Hansel, A.,
- 1863 Hofbauer, V., Krechmer, J., Lehtipalo, K., Lamkaddam, H., Lampimäki, M., Lee, C. P.,
- 1864 Makhmutov, V., Marie, G., Mathot, S., Mauldin, R. L., Mentler, B., Müller, T., Onnela, A., Partoll,
- 1865 E., Petäjä, T., Philippov, M., Pospisilova, V., Ranjithkumar, A., Rissanen, M., Rörup, B., Scholz,
- 1866 W., Shen, J., Simon, M., Sipilä, M., Steiner, G., Stolzenburg, D., Tham, Y. J., Tomé, A., Wagner,
- 1867 A. C., Wang, D. S., Wang, Y., Weber, S. K., Winkler, P. M., Wlasits, P. J., Wu, Y., Xiao, M., Ye,
- 1868 Q., Zauner-Wieczorek, M., Zhou, X., Volkamer, R., Riipinen, I., Dommen, J., Curtius, J.,
- 1869 Baltensperger, U., Kulmala, M., Worsnop, D. R., Kirkby, J., Seinfeld, J. H., El-Haddad, I., Flagan,
- 1870 R. C. and Donahue, N. M.: Rapid growth of new atmospheric particles by nitric acid and ammonia
- 1871 condensation, Nature, 581(7807), 184–189, doi:10.1038/s41586-020-2270-4, 2020.
- 1872
  1873 Weber, R. J., McMurry, P. H., Eisele, F. L. and Tanner, D. J.: Measurement of expected nucleation
- 1874 precursor species and 3-500-nm diameter particles at Mauna Loa Observatory, Hawaii, J. Atmos.
- 1875 Sci., 52(12), 2242–2257, doi:10.1175/1520-0469(1995)052<2242;MOENPS>2.0.CO:2, 1995.
- 1876
- 1877 Wehner, B., Siebert, H., Stratmann, F., Tuch, T., Wiedensohler, A., Petäjä, T., Dal Maso, M. and
- 1878 Kulmala, M.: Horizontal homogeneity and vertical extent of new particle formation events, Tellus,
- 1879 Ser. B Chem. Phys. Meteorol., 59(3), 362–371, doi:10.1111/j.1600-0889.2007.00260.x, 2007.
- 1880
- 1881 Wiedensohler, A., Ma, N., Birmili, W., Heintzenberg, J., Ditas, F., Andreae, M. O. and Panov, A.:
- 1882 Infrequent new particle formation over the remote boreal forest of Siberia, Atmos. Environ., 200,

- 1883 167–169, doi:10.1016/j.atmosenv.2018.12.013, 2019.
- 1884 1885 W
- 1885 Wonaschütz, A., Demattio, A., Wagner, R., Burkart, J., Zíková, N., Vodička, P., Ludwig, W., Steiner,
- 1886 G., Schwarz, J. and Hitzenberger, R.: Seasonality of new particle formation in Vienna, Austria
- 1887 Influence of air mass origin and aerosol chemical composition, Atmos. Environ., 118, 118-126,
- 1888 doi:10.1016/j.atmosenv.2015.07.035, 2015.
- 1889
- 1890 Woo, K. S., Chen, D. R., Pui, D. Y. H. H. and McMurry, P. H.: Measurement of Atlanta aerosol
- 1891 size distributions: Observations of lutrafine particle events, Aerosol Sci. Technol., 34, 75–87,
- 1892 doi:10.1080/02786820120056, 2001.
- 1893
- 1894 Yamada, H.: Contribution of evaporative emissions from gasoline vehicles toward total VOC 1895 emissions in Japan, Sci. Total Environ., 449, 143–149, doi:10.1016/j.scitotenv.2013.01.045, 2013.
- 1896
- 1897 Yan, C., Nie, W., Vogel, A. L., Dada, L., Lehtipalo, K., Stolzenburg, D. and Wagner, R.: Size-
- dependent influence of NOx on the growth rates of organic aerosol particles, , Sci. Adv., 6, 1–10,
- 1899 <u>2020.</u>
- 1900 1901
- 1902 Yan, C., Dada, L., Rose, C., Jokinen, T., Nie, W., Schobesberger, S., Junninen, H., Lehtipalo, K.,
- 1903 Sarnela, N., Makkonen, U., Garmash, O., Wang, Y., Zha, Q., Paasonen, P., Bianchi, F., Sipilä, M.,
- 1904 Ehn, M., Petäjä, T., Kerminen, V.-M., Worsnop, D. R. and Kulmala, M.: The role of H<sub>2</sub>SO<sub>4</sub>-
- 1905 NH<sub>3</sub> anion clusters in ion-induced aerosol nucleation mechanisms in the boreal forest, Atmos.
- 1906 Chem. Phys., 18, 13231–13243, doi:10.5194/acp-18-13231-2018, 2018.
- 1907
- 1908 Yan, C., Nie, W., Vogel, A. L., Dada, L., Lehtipalo, K., Stolzenburg, D. and Wagner, R.: Size
- 1909 dependent influence of NOx on the growth rates of organic acrosol particles, , Sci. Adv., 6, 1-10
- 1910 <del>2020.</del>
- 1911 1912
- 1912 Yao, L., Garmash, O., Bianchi, F., Zheng, J., Yan, C., Kontkanen, J., Junninen, H., Mazon, S. B.,
- 1913 Ehn, M., Paasonen, P., Sipilä, M., Wang, M., Wang, X., Xiao, S., Chen, H., Lu, Y., Zhang, B.,
- 1914 Wang, M., Chen, D., Xiao, M., Ye, Q., Stolzenburg, D., Hofbauer, V., Ye, P., Vogel, A. L.,
- 1915 Mauldin, R. L., Amorim, A., Baccarini, A., Baumgartner, B., Brilke, S., Dada, L., Dias, A.,
- 1916 Duplissy, J., Finkenzeller, H., Garmash, O., He, X. C., Hoyle, C. R., Kim, C., Kvashnin, A.,
- 1917 Lehtipalo, K., Fischer, L., Molteni, U., Petäjä, T., Pospisilova, V., Quéléver, L. L. J., Rissanen, M.,
- 1918 Simon, M., Tauber, C., Tomé, A., Wagner, A. C., Weitz, L., Volkamer, R., Winkler, P. M., Kirkby,
- 1919 J., Worsnop, D. R., Kulmala, M., Baltensperger, U., Dommen, J., El-Haddad, I. and Donahue, N.
- 5., Worshop, D. K., Kunnara, W., Bartensperger, C., Bohimen, J., El-Traddad, I. and Bohande,
- 1920 M.: Photo-oxidation of Aromatic Hydrocarbons Produces Low-Volatility Organic Compounds,
- Environ, Sci. Technol., 54(13), 7911–7921, doi:10.1021/acs.est.0c02100, 2020.

1923 Ehn, M., Paasonen, P., Sipilä, M., Wang, M., Wang, X., Xiao, S., Chen, H., Lu, Y., Zhang, B., Wang, D., Fu, Q., Geng, F., Li, L., Wang, H., Qiao, L., Yang, X., Chen, J., Kerminen, V. M., 1925 Petäjä, T., Worsnop, D. R., Kulmala, M. and Wang, L.: Atmospheric new particle formation from sulfuric acid and amines in a Chinese megacity, Science, 361(6399), 278 281. 1926 1927 doi:10.1126/science.aao4839, 2018. 1928 1929 Ye, J., Abbatt, J. P. D., Chan, A. W.H., Novel pathway of SO<sub>2</sub> oxidation in the atmosphere: 1930 reactions with monoterpene ozonolysis intermediates and secondary organic aerosol, Atmos. Chem. 1931 Phys., 18, 5549-5565, 2018 1932 1933 Yli-Juuti, T., Mohr, C. and Riipinen, I.: Open questions on atmospheric nanoparticle growth, Commun. Chem., 3(1), 2–5, doi:10.1038/s42004-020-00339-4, 2020. 1934 1935 1936 Zhang, R., Khalizov, A., Wang, L., Hu, M. and Xu, W.: Nucleation and growth of nanoparticles in

the atmosphere, Chem. Rev., 112(3), 1957–2011, doi:10.1021/cr2001756, 2012.

1937

1938 1939

1922 Yao, L., Garmash, O., Bianchi, F., Zheng, J., Yan, C., Kontkanen, J., Junninen, H., Mazon, S. B.,

1940	TABLE LEG	GENDS
1941 1942	Table 1:	Location and data availability of the cites
1942	Table 1:	Location and data availability of the sites.
1944	Table 2:	Frequency (and number of NPF events), growth and formation rate of NPF events for
1945	the sites of th	the study.
1946		
1947	Table 3:	Normalised slopes gradients (non-normalised for growth rate), R <sup>2</sup> and p-values (- for
1948		values >0.05) for the relationship between meteorological conditions and NPF event
1949		variables.
1950		
1951	Table 4:	Normalised gradientsslopes (non-normalised for growth rate), R <sup>2</sup> and p-values (- for
1952		values >0.05) for the relationship between atmospheric composition variables and
1953		NPF event variables.
1954		
1955 1956	FIGURE LE	CCENDC
1950	FIGURE LE	CGENDS
1958	Figure 1:	Map of the sites of the present study.
1959	rigure r.	Map of the sites of the present study.
1960	Figure 2:	Relation of average downward incoming solar radiation $(K\downarrow)$ and normalised
1961	8	gradients <del>slopes</del> a <sub>N</sub> *-for the sites of the present study.
1962		
1963	Figure 3:	Normalised gradients slopes a <sub>J</sub> * for K\ for the sites of the present study (*UK sites
1964		are calculated with solar irradiance).
1965		
1966	Figure 4a:	Relation <u>ship</u> of average relative humidity and normalised <u>gradients</u> slopes a <sub>N</sub> * for the
1967		sites of the present study.
1968	E* 41	
1969	Figure 4b:	Relationship of average relative humidity and normalised gradientsslopes a <sub>N</sub> * for the sites of the present study (SPAUB not included).
1970 1971		sites of the present study (SFAOB not included).
1972	Figure 5:	Relationship of average temperature and normalised gradientsslopes a <sub>N</sub> * for the sites
1973	riguit c.	of the present study.
1974		of the present study.
1975	Figure 6:	Normalised gradientsslopes a <sub>J</sub> * for temperature for the sites of the present study.
1976	J	
1977	Figure 7a:	Relation <u>ship</u> of average SO <sub>2</sub> concentrations and normalised <u>gradients</u> slopes a <sub>N</sub> * for
1978		the sites of the present study.
1979		

1980	rigure /b:	Relationship of average SO <sub>2</sub> concentrations and normalised gradients <del>slopes</del> an Hor
1981		the sites of the present study (UKRO not included).
1982		
1983	Figure 8:	Relationship of average O <sub>3</sub> concentrations and normalised gradientsslopes a <sub>N</sub> * for the
1984		sites of the present study.

## 1985 <u>Table 1: Location and data availability of the sites.</u>

Site	Location		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 <sub>x</sub> , SO <sub>2</sub> , O <sub>3</sub> , OC, SO <sub>4</sub> <sup>2-</sup> , gaseous ammonia		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 <sub>x</sub> , SO <sub>2</sub> , O <sub>3</sub> , OC, SO <sub>4</sub> <sup>2</sup> ·	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 <sub>x</sub> , SO <sub>2</sub> , O <sub>3</sub> , OC, SO <sub>4</sub> <sup>2-</sup>	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)		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)	availability), NO <sub>x</sub> , O <sub>3</sub>	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 <sub>x</sub> , SO <sub>2</sub> , O <sub>3</sub> , OC, SO <sub>4</sub> <sup>2-</sup>	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 <sub>4</sub> <sup>2-</sup>	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 <sub>x</sub> , SO <sub>2</sub> , O <sub>3</sub> , 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 <sub>2</sub> , SO <sub>2</sub> , O <sub>3</sub>	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 <sub>2</sub> , SO <sub>2</sub> , O <sub>3</sub>	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 <sub>2</sub> , O <sub>3</sub> , 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

			Meteorological	Data	
Site I	ocation	Available data	data location	availability	Reference

## Field Code Changed

<del>UKRU</del>	Harwell Science Centre, Oxford, 80 km W of London, UK (51o 34' 15" N; 1o 19' 31" W)	SMPS (16.6 604 nm, 76.5% availability), NOx, SO2, O3, OC, SO42, gaseous ammonia	On site	<del>2009 2015</del>	Charron et al., 2013
UKUB	North Kensington, 4 km W of London eity centre, UK (51o 31' 15" N; 0o 12' 48" W)	SMPS (16.6—604 nm, 83.3% availability), NOx, SO2, O3, OC, SO42—	Heathrow airport	<del>2009 2015</del>	Bigi and Harrison, 2010
UKRO	Marylebone Road, London, UK (51o 31' 21" N; 0o 9' 16" W)	SMPS (16.6 - 604 nm, 74.3% availability), NOx, SO2, O3, OC, SO42-	Heathrow airport	2009 2015	Charron and Harrison, 2003
DENRU	Lille Valby, 25 km W of Copenhagen, (55o 41' 41" N; 12o 7' 7" E) (2008– 6/2010) Risø, 7 km north of Lille Valby, (55° 38' 40" N; 12° 5' 19" E) (7/2010 – 2017)	DMPS and CPC (5.8 – 700 nm, 68.3% availability), NOx, SO2, O3, OC, SO42-	H.C. Ørsted Institute station	<del>2008 2017</del>	<del>Ketzel et al.,</del> <del>2004</del>
DENUB	H.C. Ørsted Institute, 2 km NE of the city centre, Copenhagen, Denmark (55o 42' 1" N; 12o 33' 41" E)	availability),	<del>On site</del>	2008 2017	Wang et al., 2010
DENRO	H.C. Andersens Boulevard, Copenhagen, Denmark (55o 40' 28" N; 12o 34' 16" E)	DMPS and CPC (5.8 700 nm, 65.7% availability), NOx, SO2, O3, OC, SO42-	H.C. Ørsted Institute station	2008 2017	Wang et al., 2010
GERRU	Melpitz, 40 km NE of Leipzig, Germany (51o-31' 31.85" N; 12o-26' 40.30" E)	TDMPS with CPC (4.8 800 nm, 87.2% availability), OC, SO42	On site	2008 2011	Engler et al., 2007
GERUB	Tropos, 3 km NE from the city centre of Leipzig, Germany (51o 21' 9.1" N; 12o 26' 5.1" E)	TDMPS with CPC (3 800 nm, 90.4% availability)	On site	2008 2011	Costabile et al., 2009
GERRO	Eisenbahnstraße, Leipzig, Germany (51o 20' 43.80" N; 12o 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 (61o 50' 50.70" N; 24o 17' 41.20" E)	TDMPS with CPC (3—1000 nm, 98.2% availability), NOx, SO2, O3, VOCs	On site	2008 2011 & 2015 2018	Aalto et al., 2001
FINUB	Kumpula Campus 4 km N of the city centre; Helsinki, Finland (60o-12' 10.52" N; 24o-57' 40.20" E)	TDMPS with CPC (3.4—1000 nm, 99.7% availability)	On site	2008 2011 & 2015 2018	Järvi et al., 2009
FINRO	Mäkelänkatu street, Helsinki, Finland (60e-11' 47.57" N; 24e-57' 6.01" E)	DMPS (6 800 nm, 90.0% availability), NOx, O3	Pasila station and on site	2015 2018	Hietikko et al., 2018
SPARU	Montseny, 50 km NNE from Barcelona, Spain (41o 46' 45" N; 2o 21' 29" E)	SMPS (9 – 856 nm, 53.7% availability), NO2, SO2, O3	On site	2012 2015	Dall'Osto et al., 2013
SPAUB	Palau Reial, Barcelona, Spain (41o-23'-14" N; 2o-6'-56" E)	SMPS (11 – 359 nm, 88.1% availability), NO2, SO2, O3	<del>On site</del>	<del>2012 – 2015</del>	Dall'Osto et al., 2012
GRERU	Finokalia, 70 km E of Heraklion, Greece (35o 20' 16.8" N; 25o 40' 8.4" E)	SMPS (8.77 849 nm, 85.0% availability), NO2, O3, OC	<del>On site</del>	2012 2018	Kalkavouras et al., 2017
GREUB	"Demokritos", 12 km NE from the city centre,	SMPS (10 550 nm, 88.0% availability)	<del>On site</del>	2015 2018	Mølgaard et al., 2013

Athens, Greece (37o 59' 41.96" N;		
23o 48' 57.56" E)		
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Table 2: Frequency (and number of NPF events), growth and formation rate of NPF events for the sites of the study.

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

<sup>\*</sup> GR up to 50 nm calculated \*\* J<sub>16</sub> calculated

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.03) 101	the relation be			d shortwave				28.		-//	Formatted	
ite	<u>a<sub>N</sub>* (W<sup>-1</sup> m<sup>2</sup>)</u>	$\mathbb{R}^2$	<u>p</u>	a <sub>G</sub>	$\mathbb{R}^2$	<u>p</u>	$a_{J}^{*} (W^{-1} m^{2})$	$\mathbb{R}^2$	<u>p</u>	Avera	Formatted	
KRU*	1.21E-03	0.94	<0.001	6.53E-05	0.11	=	6.28E-04	0.93	< 0.001	/443/	Formatted	
KUB*	6.81E-04	0.90	< 0.001	-8.26E-05	0.11		1.49E-04	0.19		448	Formatted	
KRO*	8.69E-04	0.98	< 0.001	-7.75E-06	0.00		2.66E-04	0.19		464	Formatted	
ENRU	2.22E-03	0.88	< 0.001	4.24E-04	0.20		1.38E-03	0.64	< 0.001	115	Formatted	
ENUB	1.87E-03	0.91	< 0.001	1.47E-04	0.03	=	8.98E-04	0.48	<0.01	115	Formatted	
ENRO	2.46E-03	0.91	< 0.001	1.47E-04 1.27E-04	0.03		6.77E-04	0.50		/1/7/	Formatted	
ERRU	2.87E-03	0.93	< 0.001	9.88E-04	0.72	< 0.01	1.45E-03	0.81	< 0.001	130	Formatted	
ERUB	3.18E-03	0.97	< 0.001	7.28E-04	0.72	< 0.005	1.53E-03	0.69	< 0.001	11/4	Formatted	
ERRO	2.40E-03	0.97	< 0.001	-5.89E-04	0.09	=	9.95E-04	0.59	< 0.005	114	Formatted	
INRU	2.63E-03	0.76	< 0.001	1.01E-03	0.09	< 0.01	2.04E-03	0.82	< 0.001	91.5		
INUB	1.38E-03	0.70	=	1.81E-04	0.08	=	8.99E-04	0.82	=	/11/1		
INRO	1.76E-03	0.57	<0.005	9.15E-04	0.08	< 0.005	4.45E-04	0.23		114	Formatted	
PARU	3.46E-04	0.35	< 0.05	5.68E-04	0.13		1.97E-03	0.03	0.004	162	Formatted	
PAKU PAUB	5.40E-04 5.92E-04	0.58	< 0.05	6.98E-04	0.13	<u> </u>	1.58E-03	0.74	< 0.001	180	Formatted	
		0.52	< 0.001		0.23	< 0.001		0.05		201	Formatted	
RERU	4.10E-04 3.49E-04	0.32		7.14E-04	0.02		-6.30E-04 8.97E-04		<0.05	7 /	Formatted	
REUB	olar irradiation		- irements	-1.10E-04	0.02	=	6.97E-04	0.34	<u>&lt;0.03</u>	183	Formatted Table	
Jiouai si	<u>Jiai iiradiatioi</u>	1 IIICast	<u>irements</u>	III KJ III							Formatted	
				Relative I	<u> Iumidit</u>	ty (%)				4//	Formatted	
<u>ite</u>	<u>a<sub>N</sub>* (%-1)</u>	$\mathbb{R}^2$	<u>p</u>	<u>a</u> <sub>G</sub>	$\mathbb{R}^2$	<u>p</u>	<u>a<sub>J</sub>* (% -1)</u>	$\mathbb{R}^2$	<u>p</u>	Averag	Formatted	
KRU	<u>-5.89E-02</u>	0.85	<0.001	1.69E-03	0.02	Ξ	-3.35E-02	0.85	<u>&lt;0.001</u>	<u>79,7</u>	Formatted	
KUB	-3.42E-02	0.94	<0.001	8.23E-03	0.24	=	<u>-5.66E-03</u>	0.19	=	<u>75,3</u>	Formatted	
<u>KRO</u>	-5.09E-02	0.85	<0.001	7.03E-03	0.25	=	-1.49E-02	0.46	<u>&lt;0.05</u>	<u>74,5</u>	Formatted	
<u>ENRU</u>	-3.90E-02	0.95	<0.001	9.42E-03	0.74	< 0.001	5.45E-04	0.00	=	<u> 75.7</u>	Formatted	
ENUB	-3.14E-02	0.94	<0.001	3.64E-03	0.06	=	2.57E-03	0.00	Ξ	<u>75.7</u>	Formatted	
ENRO	-3.64E-02	0.95	<u>&lt;0.001</u>	<u>-1.21E-02</u>	0.22	=	-3.91E-03	0.10	=	<u>75.7</u>	Formatted	
ERRU	-5.08E-02	0.88	< 0.001	-1.30E-02	0.72	< 0.001	-2.46E-02	0.91	< 0.001	<u>81.9</u>		
ERUB	-5.35E-02	0.86	< 0.001	-6.34E-03	0.67	< 0.001	-2.25E-02	0.86	<0.001	<u>78.7</u> /	Formatted	
ERRO	-2.83E-02	0.90	< 0.001	3.98E-03	0.05	=	-1.72E-02	0.81	<u>&lt;0.001</u>	<u> 78.7</u> /	Formatted	
INRU	-4.48E-02	0.94	<0.001	-7.07E-03	0.65	< 0.001	-2.16E-02	0.87	<0.001	80.1/	Formatted	
INUB	-5.89E-02	0.95	<0.001	1.04E-02	0.26	=	-6.52E-03	0.18	=	76.5/	Formatted	
INRO	-3.34E-02	0.92	<0.001	-1.47E-03	0.01	Ξ	7.39E-03	0.10	Ξ	71.1/	Formatted	
PARU	-1.54E-02	0.90	<0.001	-4.67E-03	0.08	=	-7.12E-03	0.14	=	66.4	Formatted	
PAUB	-4.84E-02	0.93	<0.001	2.43E+02	0.50	< 0.01	-9.83E-03	0.19	=	69.2/	Formatted	
RERU	-7.72E-03	0.22	=	1.06E-02	0.06	=	-1.83E-01	0.15	=	70.0	Formatted	
REUB	-1.42E-02	0.62	< 0.001	2.83E-03	0.06	Ξ	4.85E-04	0.00	=	60.5	Formatted	
				Tempe	rature (	(°C)				4/	Formatted Table	
	a <sub>N</sub> * (°C <sup>-1</sup> )	$\mathbb{R}^2$	<u>p</u>	$\underline{\mathbf{a}_{\mathrm{G}}}$	$\mathbb{R}^2$	p	<u>a<sub>J</sub>* (°C-1)</u>	$\mathbb{R}^2$	<u>p</u>	Average	Formatted	
<u>ite</u>	1.10E-01	0.93		7.85E-02	0.94	< 0.001	8.72E-02	0.84	< 0.001	10.6	Formatted	
		0.98	< 0.001	1.39E-01	0.96	< 0.001	6.34E-02	0.73	< 0.005	11.8	Formatted	
KRU	9.04E-02	0.70			0.52	< 0.05	4.32E-02	0.44	<0.05	12.1	Formatted	
KRU KUB			< 0.001	3.51E-02						-		
KRU KUB KRO ENRU	8.22E-02	0.98	<0.001 <0.001	3.51E-02 1.54E-02		_	6.68E-02	0.92	< 0.001	9.80		
KRU KUB KRO ENRU	8.22E-02 6.68E-02	0.98 0.83		1.54E-02	0.08	=		0.72	<0.001 <0.05	9.80 9.82	Formatted	
KRU KUB KRO	8.22E-02 6.68E-02 2.50E-02	0.98	<0.001			= = =	6.68E-02 3.05E-02 2.96E-02	0.45		9.80 9.82 10.0		

FINRU	<u>-2.01E-02</u>	0.17	=	1.13E-01	0.79	<0.001	4.27E-02	0.72	<0.001	4.79	Formatted: Font: (Default) Times New Roman
FINUB	<u>-4.21E-03</u>	0.00	=	7.42E-02	0.83	<0.001	1.67E-02	0.28	=	6.52	Formatted: Font: (Default) Times New Roman
FINRO	6.24E-02	0.65	<0.005	9.28E-02	0.87	<0.001	-1.09E-02	0.05	= -0.001	7.72	Formatted: Font: (Default) Times New Roman
SPARU	-2.51E-02	0.41	<0.05	1.23E-01	0.92	<0.001	9.11E-02	0.71	<0.001	13.9	Formatted: Font: (Default) Times New Roman
SPAUB	-3.43E-03	0.02	= <0.001	6.67E-02	0.66	<0.005	1.18E-02	0.08	= <0.05	18.2	Formatted: Font: (Default) Times New Roman
GRERU	-4.66E-02	0.75	<0.001	1.74E-01	0.75	<0.001 <0.005	-9.45E-02	0.47		18.2	Formatted: Font: (Default) Times New Roman
GREUB	<u>-1.00E-02</u>	0.25	Ξ	4.67E-02	0.62	<0.003	<u>-2.85E-02</u>	0.20	Ξ	17.6	Formatted: Font: (Default) Times New Roman
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				Wind S	Speed (1	m s <sup>-1</sup> )					
<u>Site</u>	$\underline{\mathbf{a}}_{\mathrm{N}}^{*}$ (m <sup>-1</sup> s)	<u>R</u> <sup>2</sup>	<u>p</u>	<u>a</u> G	$\mathbb{R}^2$	<u>p</u>	<u>a</u> <sub>J</sub> * (m <sup>-1</sup> s)	$\mathbf{R}^2$	<u>p</u>	Avera	Formatted: Font: (Default) Times New Roman
<u>UKRU</u>	5.72E-02	0.20	=	-3.04E-02	0.07	=	6.87E-03	0.0	0 =	3.96	Formatted: Font: (Default) Times New Roman
UKUB	<u>1.72E-01</u>	0.87	<0.001	-1.91E-01	0.71	<0.001	3.56E-03	0.0	0.005	4.16	Tormatteet Form (Bendun) Times Frew Homan
<u>UKRO</u>	6.34E-02	0.19		3.21E-02	0.02		7.28E-02	0.4	0.04	4.14	1 or matteet: 1 ont. (Belautt) Times I'ew Roman
<u>DENRU</u>	1.08E-01	0.88	<0.001	-2.33E-01	0.74	<0.001	1.28E-01	0.4	_	4.17	Formatted: Font: (Default) Times New Roman
<u>DENUB</u>	1.50E-01	0.90	<0.001	-3.33E-02	0.10	= 0.001	8.31E-02	0.1		4.17	Formatted: Font: (Default) Times New Roman
DENRO	1.65E-01	0.89	<0.001	-1.51E-01	0.49	<0.001	9.08E-03	0.0		4.16	Formatted: Font: (Default) Times New Roman
GERRU	-1.06E-01	0.57	<0.005	-2.26E-01	0.83	<0.001	-5.32E-03	0.0		2.58	Formatted: Font: (Default) Times New Roman
GERUB	-1.27E-01	0.52	<0.01	-1.41E-01	0.60	<0.005	-3.32E-02	0.0	_	2.33	Formatted: Font: (Default) Times New Roman
GERRO	-2.40E-01	0.56		-2.54E-01	0.38	-0.05	-1.30E-01	0.2		2.33	Formatted: Font: (Default) Times New Roman
FINRU	1.62E-01	0.63	<0.005	-1.29E-01	0.16	<0.05	7.99E-02	0.0		1.31	Formatted: Font: (Default) Times New Roman
FINUB	-3.17E-02	0.08	= <0.05	7.26E-02	0.20	<0.05	<u>-9.74E-02</u>	0.1		3.43	Formatted: Font: (Default) Times New Roman
FINRO	8.62E-02	0.51	<0.05	-1.60E-01	0.32	<0.05	-1.86E-01	0.3	_	4.26	Formatted: Font: (Default) Times New Roman
SPARU	-2.20E-02	0.02	<0.001	3.80E-01	0.31	Ξ	5.74E-02	0.0	_	0.94	Formatted: Font: (Default) Times New Roman
SPAUB	2.90E-01	0.93	<0.001	7.71E-02	0.24	< 0.005	-5.90E-02	0.0		2.05	Formatted: Font: (Default) Times New Roman
GRERU	4.37E-02	0.54	<0.01	1.01E-01	0.36	< 0.005	1.73E-03	0.0		6.06 1.87	Formatted: Font: (Default) Times New Roman
GREUB	<u>-1.13E-01</u>	0.47	<u> </u>	<u>-1.88E-01</u>	0.50	<u>&lt;0.003</u>	<u>-3.78E-02</u>	0.0	<u>1</u> <u>=</u>	1.07	Formatted: Font: (Default) Times New Roman
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				Atmospheri	c Press	ure (mbai	<u>r)</u>				
Site	a <sub>N</sub> * (mbar <sup>-1</sup> )	$\mathbb{R}^2$	<u>p</u>	$\mathbf{a}_{\mathrm{G}}$	$\mathbb{R}^2$	<u>p</u>	a <sub>J</sub> * (mbar	·-1) <b>F</b>	<u>p</u>	Avera	Formatted: Font: (Default) Times New Roman
UKRU	4.26E-02	0.83	< 0.005	3.93E-02	0.58	< 0.005	2.95E-02	<u>0</u> .	<u>47</u> <u>&lt;0.05</u>	1007	Formatted: Font: (Default) Times New Roman
UKUB	1.90E-02	0.50	Ξ	1.17E-02	0.05	< 0.05	4.16E-03	<u>0.</u>	04 =	1011	
<u>UKRO</u>	6.33E-02	0.95	<u>&lt;0.001</u>	<u>-1.21E-01</u>	0.40	=	-2.98E-02	<u>0</u> .	<u>17</u> =	101	Formatted: Font: (Default) Times New Roman
GERRU	5.10E-02	0.97	Ξ	8.95E-02	0.85	<0.001	2.16E-02	<u>0.</u>	21 =	1007	Formatted: Font: (Default) Times New Roman
GERUB	6.27E-02	0.97	Ξ	4.00E-02	0.76	Ξ	2.00E-02	<u>0.</u>	<u>≤0.05</u>	995.	Formatted: Font: (Default) Times New Roman
<b>GERRO</b>	4.57E-02	0.79	=	<u>-9.61E-02</u>	0.43	=	-2.80E-02	<u>0</u> .	<u>21</u> =	995.	Formatted: Font: (Default) Times New Roman
FINRU	3.46E-02	0.88	< 0.001	2.90E-02	0.57	<0.001	1.05E-02	<u>0.</u>	<u>14</u> <u>-</u>	985.	Formatted: Font: (Default) Times New Roman
FINUB	2.61E-02	0.55	<u>&lt;0.005</u>	-3.57E-03	0.02	=	4.38E-03	<u>0.</u>	05 =	1004	Formatted: Font: (Default) Times New Roman
FINRO	<u>4.91E-02</u>	0.70	Ξ	<u>-2.67E-02</u>	0.17	Ξ	1.43E-02	<u>0.</u>	<u> 26</u> <u> </u>	1008	Formatted: Font: (Default) Times New Roman  Formatted: Font: (Default) Times New Roman
<u>SPARU</u>	-2.02E-02	0.09	Ξ	4.79E-02	0.14	Ξ	2.89E-02	<u>0.</u>	<u>08</u> <u>=</u>	939.	
<b>SPAUB</b>	<u>-2.83E-02</u>	0.44	<u>&lt;0.05</u>	1.86E-02	0.08	Ξ	1.68E-02	<u>0.</u>	21 =	1006	Formatted: Font: (Default) Times New Roman
GRERU	6.00E-02	0.46	<0.001	-1.50E-01	0.73	=	8.14E-02	<u>0.</u>	33 =	1014	Formatted: Font: (Default) Times New Roman
GREUB	9.42E-03	0.10	<u>&lt;0.05</u>	<u>-1.00E-01</u>	0.71	Ξ	1.58E-02	<u>0.</u>	04 =	1015	Formatted: Font: (Default) Times New Roman
			<del>Downwar</del>	<del>d shortwave</del>	<del>solar r</del>	adiation l	<del>K↓ (W m<sup>-2</sup>)</del>				Formatted: Font: (Default) Times New Roman

a<sub>J</sub>\* (W-1-m<sup>2</sup>)

Average

4.28E-02

1.61E-02

4.27E-02

0.54

0.11

0.72

< 0.001

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4.79 **Formatted:** Font: (Default) Times New Roman

 $\mathbb{R}^2$ 

**GERUB** 

GERRO

FINRU

Site

 $a_N * (W^{-1} - m^2)$ 

8.20E-02

5.08E-02

-2.01E-02

0.93

0.89

0.17

< 0.001

3.38E-02

-3.33E-03

1.13E-01

0.62

0.00

0.79

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	1.49E 04	0.19	1	448
<del>UKRO*</del>	8.69E 04	0.98	<0.001	-7.75E-06	0.00	-	2.66E 04	0.64	<0.005	464
<b>DENRU</b>	2.22E 03	0.88	<0.001	4.24E 04	0.20	1	1.38E 03	0.64	<0.001	115
<b>DENUB</b>	1.87E 03	0.91	<0.001	1.47E 04	0.03	-	8.98E 04	0.48	<0.01	115
<b>DENRO</b>	2.46E 03	0.95	<0.001	1.27E 04	0.01	-	6.77E 04	0.50	<0.005	117
<b>GERRU</b>	2.87E 03	0.98	<0.001	9.88E 04	0.72	<0.01	1.45E 03	0.81	<0.001	<del>130</del>
<b>GERUB</b>	3.18E 03	0.97	<0.001	7.28E 04	0.51	<del>&lt;0.005</del>	1.53E 03	0.69	<0.001	114
<b>GERRO</b>	2.40E 03	0.95	<0.001	-5.89E 04	0.09	1	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	ı	8.99E 04	0.25	1	111
FINRO	1.76E-03	0.59	<0.005	9.15E-04	0.34	<0.005	4.45E-04	0.03	1	114
SPARU	3.46E 04	0.35	<del>&lt;0.05</del>	5.68E 04	0.13	ı	1.97E 03	0.74	<0.001	<del>162</del>
SPAUB	5.92E 04	0.58	<0.05	6.98E 04	0.23	-	1.58E 03	0.81	<0.001	<del>180</del>
GRERU	4.10E 04	0.52	<0.001	7.14E 04	0.55	<0.001	-6.30E-04	0.05	_	201
<b>GREUB</b>	3.49E 04	0.31	_	-1.10E 04	0.02	-	8.97E 04	0.34	<0.05	183

<sup>\*</sup> Solar irradiation measurements

				Relative I	Iumidi	ty (%)				
Site	a <sub>N</sub> * (%-1)	$\mathbb{R}^2$	Ð	<del>a</del> G	$\mathbb{R}^2$	p	aj* (%-1)	$\mathbb{R}^2$	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
<b>DENRU</b>	-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
<b>DENRO</b>	-3.64E-02	0.95	<0.001	-1.21E 02	0.22		-3.91E 03	0.10	-	<del>75.7</del>
<b>GERRU</b>	-5.08E-02	0.88	<0.001	-1.30E 02	0.72	<0.001	-2.46E 02	0.91	<0.001	81.9
<b>CERUB</b>	-5.35E-02	0.86	<0.001	-6.34E 03	0.67	<0.001	<u> -2.25E 02</u>	0.86	<0.001	78.7
<b>CERRO</b>	<del>-2.83E-02</del>	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	-	<del>76.5</del>
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	<del>-4.84E 02</del>	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
<b>GREUB</b>	-1.42E 02	0.62	<0.001	2.83E 03	0.06	-	4.85E 04	0.00	-	60.5
				Tempe	rature	<del>(°C)</del>				
Site	$a_N * (^{\circ}C^{-1})$	$\mathbb{R}^2$	P	<del>a</del> G	$\mathbb{R}^2$	P	$a_{J}^{*}$ (°C <sup>-1</sup> )	$\mathbb{R}^2$	P	Average
<del>UKRU</del>	1.10E 01	0.93	<0.001	7.85E-02	0.94	<0.001	8.72E 02	0.84	<0.001	<del>10.6</del>
<del>UKUB</del>	9.04E 02	0.98	<0.001	1.39E 01	0.96	<0.001	6.34E-02	0.73	<del>&lt;0.005</del>	11.8
<b>UKRO</b>	8.22E-02	0.98	<0.001	3.51E-02	0.52	<del>&lt;0.05</del>	4.32E-02	0.44	<0.05	<del>12.1</del>
<b>DENRU</b>	6.68E 02	0.83	<del>&lt;0.001</del>	1.54E 02	0.08	-	6.68E-02	0.92	<0.001	9.80
<b>DENUB</b>	2.50E 02	0.45	<0.05	2.40E-02	0.33	-	3.05E 02	0.45	<0.05	9.82
<b>DENRO</b>	6.64E 02	0.88	<0.001	3.51E-03	0.00	-	2.96E 02	0.58	<0.005	<del>10.0</del>
<b>GERRU</b>	7.27E 02	0.92	<0.001	5.65E-02	0.92	<0.001	5.37E 02	0.93	<0.001	10.3
<b>GERUB</b>	8.20E-02	0.93	<0.001	3.38E-02	0.62	<0.001	4.28E 02	0.54	<0.005	<del>11.1</del>
<b>GERRO</b>	5.08E-02	0.89	<0.001	3.33E 03	0.00	-	1.61E-02	0.11	-	<del>11.1</del>
FINRU	2.01E 02	0.17	-	1.13E 01	0.79	<0.001	4.27E 02	0.72	<0.001	4.79
FINUR	4.21E.03	0.00	_	7.42F.02	0.83	<0.001	1.67F.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	<del>13.9</del>
SPAUB	-3.43E 03	0.02	-	6.67E 02	0.66	<0.005	1.18E 02	0.08	-	<del>18.2</del>
<b>GRERU</b>	<del>4.66E 02</del>	0.75	<0.001	1.74E 01	0.75	<0.001	<del>9.45E 02</del>	0.47	<0.05	<del>18.2</del>
<b>GREUB</b>	-1.00E-02	0.25	_	4.67E 02	0.62	<0.005	-2.85E 02	0.20	-	<del>17.6</del>

Wind Speed (m·s <sup>-1</sup> )													
Site	a <sub>N</sub> * (m <sup>-1</sup> s)	$\mathbb{R}^2$	Ð	<del>a</del> G	$\mathbb{R}^2$	<del>P</del>	a <sub>J</sub> * (m <sup>-1</sup> s)	$\mathbb{R}^2$	P	Average			
UKRU	5.72E 02	0.20	-	-3.04E-02	0.07	1	6.87E 03	0.00	-	3.96			
<del>UKUB</del>	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			
<b>DENUB</b>	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			
<b>GERUB</b>	<del>-1.27E-01</del>	0.52	<0.01	-1.41E-01	0.60	<0.005	-3.32E-02	0.04	-	2.33			
<b>GERRO</b>	-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	<del>-9.74E 02</del>	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	<del>&lt;0.005</del>	-3.78E 02	0.01	-	1.87			

Atmospheric Pressure (mbar)											
Site	a <sub>N</sub> * (mbar <sup>-1</sup> )	$\mathbb{R}^2$	P	<del>a</del> G	$\mathbb{R}^2$	Ð	a <sub>J</sub> * (mbar <sup>-1</sup> )	$\mathbb{R}^2$	Ð	Average	
UKRU	4.26E 02	0.83	<del>&lt;0.005</del>	3.93E 02	0.58	<del>&lt;0.005</del>	2.95E 02	0.47	<del>&lt;0.05</del>	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	
<b>GERRU</b>	5.10E 02	0.97	_	8.95E 02	0.85	<0.001	2.16E 02	0.21	-	1007.0	
<b>GERUB</b>	6.27E-02	0.97	_	4.00E-02	0.76	_	2.00E-02	0.37	<del>&lt;0.05</del>	995.5	
<b>GERRO</b>	4.57E 02	0.79	-	<del>-9.61E 02</del>	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	<del>&lt;0.05</del>	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	

 $\begin{tabular}{ll} \textbf{Table 4:} Normalised $\frac{slopes-gradients}{slopes-gradients}$ (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. \end{tabular}$ 

SO₂(µg m³)											
Site	$a_{N}^{*} (\mu g^{-1} - m^{3})$	$\mathbb{R}^2$	Ð	₽G	$\mathbb{R}^2$	P	a <sub>J</sub> * (μg <sup>-1</sup> -m <sup>3</sup> )	$\mathbb{R}^2$	P	Average	
<b>UKRU</b>	-1.97E-01	0.38	<del>&lt;0.05</del>	<del>-6.17E 02</del>	0.02	-	3.30E 01	0.06	-	1.64	
<b>UKUB</b>	-2.57E-01	0.62	<0.001	1.93E 02	0.00	1	4.18E 01	0.40	-	2.04	
<b>UKRO</b>	-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	
<b>DENRO</b>	-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	-	<del>-6.30E-02</del>	0.00	_	0.09	
SPARU	-3.62E-01	0.74	<0.001	-1.33E 01	0.02	ı	-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 <sub>x</sub> or NO <sub>2</sub> (ppb)												
Site	a <sub>N</sub> * (ppb <sup>-1</sup> )	$\mathbb{R}^2$	p	<del>a</del> c	$\mathbb{R}^2$	p	a <sub>J</sub> * (ppb <sup>-1</sup> )	$\mathbb{R}^2$	p	Average		
UKRU	-4.99E-02	0.67	<0.005	4.52E 02	0.58	<del>&lt;0.05</del>	-4.51E-02	0.70	<0.005	11.7		
<del>UKUB</del>	-8.75E-03	0.83	<0.001	3.97E 04	0.00	-	-1.09E-02	0.43	<0.05	<del>53.6</del>		
<del>UKRO</del>	-3.22E-03	0.72	<0.001	1.44E 03	0.39	<0.05	2.19E 03	0.66	<0.001	<del>299</del>		
<b>DENRU</b>	9.41E 02	0.43	<0.005	4.89E 03	0.00	<0.001	-6.47E-02	0.55	<0.01	5.42		
<b>DENUB</b>	<del>-4.99E-02</del>	0.68	<0.001	2.85E-02	0.26	-	8.55E-04	0.00	-	<del>10.5</del>		
<b>DENRO</b>	-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	<del>-6.24E-03</del>	0.68	<0.001	1.70E 03	0.12	1	3.25E 03	0.03	-	88.1		
SPARU*	-1.53E-02	0.05	1	2.54E 02	0.01	1	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		

<sup>\*</sup> NO<sub>2</sub> measurements

O <sub>3.</sub> ( <del>ppb)</del>												
Site	a <sub>N</sub> * (ppb <sup>-1</sup> )	$\mathbb{R}^2$	Ð	<del>a</del> G	$\mathbb{R}^2$	Ð	a <sub>J</sub> * (ppb <sup>-1</sup> )	$\mathbb{R}^2$	Ð	Average		
<del>UKRU</del>	2.27E 02	0.88	<0.001	4.89E 02	0.53	<del>&lt;0.005</del>	-3.53E-03	0.01	-	54.4		
<del>UKUB</del>	1.37E 02	0.87	<0.001	-3.45E 02	0.68	<0.001	-5.95E-03	0.05	-	39.3		
<b>UKRO</b>	7.46E 02	0.95	<0.001	-1.06E-02	0.09	-	-2.44E 02	0.63	<0.005	<del>16.2</del>		
<b>DENRU</b>	4.97E 02	0.92	<0.001	-1.32E 02	0.15	1	1.23E 02	0.08	-	30.1		
<b>DENUB</b>	5.85E 02	0.84	<0.001	-1.69E 02	0.58	1	2.77E 02	0.32	<0.05	28.2		
<b>DENRO</b>	6.42E 02	0.51	<del>&lt;0.05</del>	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	1	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	-	<del>54.9</del>		
<b>GRERU</b>	7.55E 03	0.04	-	3.68E 02	0.17	•	-3.01E 02	0.15	-	49.5		

	Particulate Organic Carbon (µg m³)												
Site	a <sub>N</sub> * (μg <sup>-1</sup> -m <sup>3</sup> )	$\mathbb{R}^2$	<del>P</del>	<del>a</del> e	$\mathbb{R}^2$	<del>P</del>	α <sub>J</sub> * (μg <sup>-1</sup> m³)	$\mathbf{R}^2$	<del>P</del>	Average			
UKRU	<del>-3.30E-02</del>	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	<del>&lt;0.05</del>	2.19E 01	0.55	<del>&lt;0.05</del>	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			
<b>GERRU</b>	-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 m³)											
Site	$a_N * (\mu g^{-1} - m^3)$	$\mathbb{R}^2$	P	$\mathbf{a}_{\mathbf{G}}$	$\mathbb{R}^2$	P	$a_{J}^{*} (\mu g^{-1} - m^{3})$	$\mathbb{R}^2$	Ð	Average	
UKRU <sup>1</sup>	-2.62E-01	0.57	<0.001	7.34E 01	0.77	<0.001	7.99E 01	0.44	<0.05	1.97	
UKUB <sup>1</sup>	-3.57E-01	0.89	<0.001	9.28E 01	0.44	<0.01	9.72E 01	0.16	-	1.58	
UKRO <sup>1</sup>	-6.05E-02	0.24	-	3.04E 01	0.34	<0.05	-6.22E-02	0.04	-	1.98	
DENRU <sup>2</sup>	7.81E 01	0.34	<0.05	1.02E+00	0.60	<0.05	-1.03E+00	0.63	<0.01	0.52	
DENRO <sup>2</sup>	-8.23E-01	0.28	-	1.99E+00	0.22	_	2.82E 01	0.12	_	0.55	
GERRU <sup>1</sup>	-3.37E-02	0.00	-	5.89E 01	0.11	-	4.89E-02	0.01	-	0.92	
FINRU <sup>3</sup>	-1.18E+00	0.65	<0.001	2.35E 01	0.09	-	-2.53E-01	0.17	-	1.02	

Condensation Sink (s-1)												
				Condensat	<del>ion Sir</del>	<del>ık (s<sup>-1</sup>)</del>						
Site	an* (s)	$\mathbb{R}^2$	P	ag	$\mathbb{R}^2$	Ð	<b>а</b> ј <del>* (s)</del>	$\mathbb{R}^2$	Ð	Average		
UKRU	-2.28E+02	0.72	<0.001	2.64E+02	0.60	<0.001	7.58E+01	0.22	-	3.38E 03		
<del>UKUB</del>	-1.66E+02	0.78	<0.001	2.49E+02	0.41	<0.05	1.73E+02	0.35	<0.05	7.41E 03		
<b>UKRO</b>	-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		
<b>DENRO</b>	-1.06E+01	0.73	<0.001	2.53E+01	0.56	<0.005	2.72E+01	0.79	<0.001	3.10E-02		
<b>GERRU</b>	1.54E+02	0.86	<del>&lt;0.001</del>	1.33E+02	0.56	<0.001	6.67E+01	0.63	<0.001	7.02E 03		
<b>GERUB</b>	3.59E+01	0.56	<0.005	3.63E+01	0.17	-	4.74E+01	0.75	<0.001	9.11E-03		
<b>GERRO</b>	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		
<b>GRERU</b>	4.33E+00	0.00	-	2.86E+02	0.70	<0.001	1.77E+02	0.56	<0.005	4.66E 03		
<b>GREUB</b>	1.64E+02	0.65	<0.001	9.31E+01	0.28	<0.05	1.73E+02	0.83	<0.001	7.55E 03		
OILLIOD.	1.0.2102	0.05		7.012101	0.20		11,02102	0.00		7.000		

<u>SO<sub>2</sub> (μg m<sup>-3</sup>)</u>												
Site	$a_{\rm N}^* (\mu g^{-1} m^3)$	$\mathbb{R}^2$	<u>p</u>	$\mathbf{a}_{\mathrm{G}}$	$\mathbb{R}^2$	<u>p</u>	$a_{\rm J}^* (\mu g^{-1} m^3)$	$\mathbb{R}^2$	<u>p</u>	Average		
<u>UKRU</u>	-1.97E-01	0.38	<u>&lt;0.05</u>	<u>-6.17E-02</u>	0.02	П	3.30E-01	0.06	Ξ	1.64		
<u>UKUB</u>	-2.57E-01	0.62	<0.001	1.93E-02	0.00	П	4.18E-01	0.40	Ξ	2.04		
<b>UKRO</b>	-1.03E-01	0.82	< 0.001	6.90E-02	0.34	< 0.01	8.43E-02	0.77	< 0.001	<u>7.46</u>		
<b>DENRU</b>	<u>-9.77E-01</u>	0.53	<u>&lt;0.05</u>	2.84E+00	0.37	П	4.38E-01	0.09	Ξ	0.52		
<b>DENRO</b>	-4.20E-01	0.91	< 0.001	6.42E-01	0.54	< 0.005	5.66E-01	0.62	< 0.001	0.97		
<b>FINRU</b>	-5.66E-01	0.05	Ξ	-1.42E+00	0.19	П	-6.30E-02	0.00	=	0.09		
<b>SPARU</b>	-3.62E-01	0.74	<0.001	-1.33E-01	0.02	=	-3.55E-02	0.01	Ξ	0.95		
<b>SPAUB</b>	-2.93E-02	0.04	Ξ	4.12E-01	0.59		1.07E-01	0.29	Ξ	<u>1.99</u>		

NO <sub>x</sub> or NO <sub>2</sub> (ppb)												
Site	<u>a<sub>N</sub>* (ppb<sup>-1</sup>)</u>	$\mathbb{R}^2$	<u>p</u>	$\mathbf{a}_{\mathbf{G}}$	$\mathbb{R}^2$	<u>p</u>	<u>a<sub>J</sub>* (ppb<sup>-1</sup>)</u>	$\mathbb{R}^2$	<u>p</u>	Average		
<u>UKRU</u>	-4.99E-02	0.67	<0.005	4.52E-02	0.58	< 0.05	-4.51E-02	0.70	< 0.005	<u>11.7</u>		
<u>UKUB</u>	-8.75E-03	0.83	<0.001	-3.97E-04	0.00	П	-1.09E-02	0.43	< 0.05	<u>53.6</u>		
<u>UKRO</u>	-3.22E-03	0.72	<0.001	1.44E-03	0.39	< 0.05	2.19E-03	0.66	< 0.001	<u>299</u>		
<b>DENRU</b>	-9.41E-02	0.43	<0.005	-4.89E-03	0.00	< 0.001	-6.47E-02	0.55	< 0.01	<u>5.42</u>		
<b>DENUB</b>	-4.99E-02	0.68	<0.001	2.85E-02	0.26	П	8.55E-04	0.00	Ξ	<u>10.5</u>		
<b>DENRO</b>	-5.10E-03	0.75	< 0.001	1.10E-02	0.69	< 0.001	8.33E-03	0.88	< 0.001	<u>68.5</u>		
<b>FINRU</b>	-7.27E-01	0.54	<0.001	-2.74E-01	0.11	П	1.95E-01	0.05	Ξ	0.72		
<b>FINRO</b>	-6.24E-03	0.68	< 0.001	1.70E-03	0.12	Ξ	3.25E-03	0.03	Ξ	88.1		
SPARU*	-1.53E-02	0.05		2.54E-02	0.01	П	1.25E-01	0.21	Ξ	<u>3.26</u>		
SPAUB*	-2.59E-02	0.62	<u>&lt;0.005</u>	2.23E-02	0.70	<0.001	2.57E-03	0.01	Ξ	<u>31.4</u>		
<b>GRERU*</b>	3.01E-01	0.19	Ξ	-1.40E+00	0.75	< 0.001	5.23E-01	0.13	Ξ	0.52		

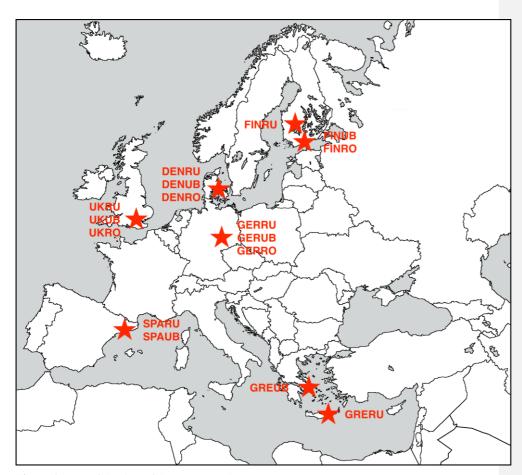
<sup>\*</sup> NO<sub>2</sub> measurements

<sup>&</sup>lt;sup>1</sup>-Measurements in PM<sub>10</sub> <sup>2</sup>-Measurements in PM<sub>2.5</sub> <sup>3</sup>-Measurements in PM<sub>1</sub>

Site	$a_{N}^{*} (ppb^{-1})$	$\mathbb{R}^2$	<u>p</u>	$\mathbf{a}_{\mathbf{G}}$	$\mathbb{R}^2$	<u>p</u>	a <sub>J</sub> * (ppb <sup>-1</sup> )	$\mathbb{R}^2$	<u>p</u>	Aver	Formatted: Font: (Default) Times New Roman
UKRU	2.27E-02	0.88	< 0.001	-4.89E-02	0.53	< 0.005		0.01	=	54.	Formatted: Font: (Default) Times New Roman
UKUB	1.37E-02	0.87	<0.001	-3.45E-02	0.68	< 0.001	-5.95E-03	0.05		39.	Formatted: Font: (Default) Times New Roman
UKRO	7.46E-02	0.95	< 0.001	-1.06E-02	0.09	=	-2.44E-02	0.63	0.00		Formatted: Font: (Default) Times New Roman
DENRU	4.97E-02	0.92	< 0.001	-1.32E-02	0.15	=	1.23E-02	0.08		30.	Formatted: Font: (Default) Times New Roman
DENUB	5.85E-02	0.84	< 0.001	-1.69E-02	0.58		2.77E-02	0.32		28.	, , ,
DENRO	6.42E-02	0.51	< 0.05	1.39E-02	0.03	-	3.24E-02	0.91	< 0.05	31.	Formatted: Font: (Default) Times New Roman
FINRU	6.76E-02	0.77	< 0.05	-4.23E-02	0.60	=	3.92E-02	0.37	< 0.05	27.	Formatted: Font: (Default) Times New Roman
FINRO	2.38E-02	0.91	< 0.001	6.11E-03	0.24	=	-1.83E-02	0.29		37.	Formatted: Font: (Default) Times New Roman
SPARU	1.57E-02	0.02	=	4.34E-02	0.11	=	1.31E-02	0.31		75.	Formatted: Font: (Default) Times New Roman
SPAUB	7.99E-03	0.38	< 0.05	-5.83E-03	0.30	=	-1.13E-03	0.01		54.	Formatted: Font: (Default) Times New Roman
GRERU	7.55E-03	0.04	=	3.68E-02	0.17	=	-3.01E-02	0.15		49.	Formatted: Font: (Default) Times New Roman
											Formatted: Font: (Default) Times New Roman
			-			• (	2.				Formatted: Font: (Default) Times New Roman
				iculate Orga	nic Car						_
Site	$a_{N}^{*} (\mu g^{-1} m^{3})$	$\mathbb{R}^2$	<u>p</u>	$\mathbf{a}_{\mathbf{G}}$	$\mathbb{R}^2$	<u>p</u>	$\frac{\mathbf{a_{J}^*} (\mu \mathbf{g}^{-1})}{\mathbf{m}^3}$	$\mathbb{R}^2$	<u>p</u>	Avorse	Formatted: Font: (Default) Times New Roman
UKRU	-3.30E-02	0.00	_	1.13E+00	0.42	< 0.005	2.13E-01	0.16	_	1.96	
UKUB	-3.36E-02 -2.76E-01	0.59	<0.005	6.63E-01	0.58	< 0.05	2.19E-01	0.55	< 0.05	3.63	Formatted: Font: (Default) Times New Roman
UKRO	-3.78E-01	0.89	< 0.001	8.12E-01	0.57	< 0.005	4.60E-01	0.75	< 0.001	6.24	Formatted: Font: (Default) Times New Roman
DENRU	-4.44E-01	0.75	< 0.001	2.24E-01	0.11	=======================================	-3.17E-01	0.68	< 0.01	1.48	Formatted: Font: (Default) Times New Roman
DENRO	-7.80E-02	0.11	-	1.10E+00	0.77	<0.005	4.02E-01	0.81	< 0.005	2.59	Formatted: Font: (Default) Times New Roman
GERRU	-1.26E-01	0.24	= =	1.35E-01	0.09		3.14E-02	0.03		2.18	Formatted: Font: (Default) Times New Roman
FINRU	2.27E-02	0.00	= =	3.39E-01	0.60	<0.005	-3.46E-01	0.05		1.78	Formatted: Font: (Default) Times New Roman
GRERU	-2.08E-01	0.11	= =	7.87E-01	0.41	< 0.05	8.94E-01	0.10	= =	1.58	Formatted: Font: (Default) Times New Roman
GRERO	-2.00L-01	0.11	_	7.07L-01	0.71		0.74L-01	0.11	_	1.50	Formatted: Font: (Default) Times New Roman
				Sulpha	te (μg 1	m <sup>-3</sup> )					Formatted: Font: (Default) Times New Roman
Site	$a_{N}^{*} (\mu g^{-1} m^{3})$	$\mathbb{R}^2$	<u>p</u>	$\mathbf{a}_{\mathrm{G}}$	$\mathbb{R}^2$	<u>p</u>	$a_{J}^{*} (\mu g^{-1} m^{3})$	$\mathbb{R}^2$	<u>p</u>	Avera	Formatted: Font: (Default) Times New Roman
UKRU <sup>1</sup>	-2.62E-01	0.57	< 0.001	7.34E-01	0.77	< 0.001	7.99E-01	0.44	< 0.05	1.97	Formatted: Font: (Default) Times New Roman
UKUB <sup>1</sup>	-3.57E-01	0.89	<0.001	9.28E-01	0.44	<u>&lt;0.01</u>	9.72E-01	0.16	Ξ	1.58	Formatted: Font: (Default) Times New Roman
UKRO1	-6.05E-02	0.24	Ξ	3.04E-01	0.34	<0.05	-6.22E-02	0.04	Ξ	1.98	Formatted: Font: (Default) Times New Roman
DENRU <sup>2</sup>	-7.81E-01	0.34	<0.05	1.02E+00	0.60	<u>&lt;0.05</u>	-1.03E+00	0.63	< 0.01	0.52	Formatted: Font: (Default) Times New Roman
DENRO <sup>2</sup>	-8.23E-01	0.28	=	1.99E+00	0.22	=	2.82E-01	0.12	Ξ	0.55	Formatted: Font: (Default) Times New Roman
GERRU <sup>1</sup>	-3.37E-02	0.00	Ξ	5.89E-01	0.11	Ξ	-4.89E-02	0.01	Ξ	0.92	
FINRU <sup>3</sup>	-1.18E+00	0.65	<0.001	2.35E-01	0.09	=	-2.53E-01	0.17	=	1.02	Formatted: Font: (Default) Times New Roman
			•								Formatted: Font: (Default) Times New Roman
	nents in PM <sub>10</sub>										Formatted: Font: (Default) Times New Roman
	nents in PM <sub>2.5</sub>										
<sup>3</sup> Measuren	nents in PM <sub>1</sub>										
	1		1	Condensati		k (s <sup>-1</sup> )	1		Т		
Site	<u>a<sub>N</sub>* (s)</u>	$\mathbb{R}^2$	<u>p</u>	<u>a</u> G	$\mathbb{R}^2$	<u>p</u>	$\underline{\mathbf{a}_{\mathbf{J}^{*}}(\mathbf{s})}$ R	2		verage	
<u>UKRU</u>	-2.28E+02	0.72	<0.001	2.64E+02	0.60	<0.001	7.58E+01 0.2	22	<u>3</u> .	38E-03	Formatted: Font: (Default) Times New Roman
<u>UKUB</u>	-1.66E+02	0.78	<0.001	2.49E+02	0.41	<u>&lt;0.05</u>	1.73E+02 0.3		0.05 7.	41E-03	Formatted: Font: (Default) Times New Roman
<u>UKRO</u>		0110	<0.001	2.33E+01	0.18	0.05	8.94E+01 0.9	91 <0	0.001 2.	12E-02	Formatted: Font: (Default) Times New Roman
<b>DENRU</b>			<0.001	6.90E+01	0.49		5.37E+01 <u>0.2</u>			46E-03	Formatted: Font: (Default) Times New Roman
<b>DENUB</b>	<u>-3.78E+01</u>	0110	<0.001	3.58E+01	0.25	0.00=	1.55E+01 0.5		004	42E-02	Formatted: Font: (Default) Times New Roman
<b>DENRO</b>		0170	<0.001	2.53E+01	0.00	0.004	2.72E+01 <u>0.</u>			10E-02	Formatted: Font: (Default) Times New Roman
<u>GERRU</u>	1.54E+02	0.86	<0.001	1.33E+02	0.56	<0.001	6.67E+01 0.0	63 <0	<u>7.</u>	02E-03	Formatted: Font: (Default) Times New Roman
				89							

 $O_3$  (ppb)

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 Formatted: Font: (Default) Times New	
Tormateur Font. (Bendut) Times Nev	w Roman
FINRU -1.80E+02 0.59 <0.005 4.01E+02 0.74 <0.001 4.98E+01 0.10 = 2.32E-03 Formatted: Font: (Default) Times New	w Roman
FINUB -1.51E+02 0.63 <0.005 8.14E+01 0.31 = 2.01E+02 0.41 <0.05 6.34E-03 Formatted: Font: (Default) Times New	w Roman
FINRO -6.99E+01 0.77 <0.001 -1.56E+01 0.05 = 2.42E+02 0.83 <0.001 8.96E-03 Formatted: Font: (Default) Times New	w Roman
SPARU -2.15E+02 0.65 <0.005 1.86E+01 0.00 = 8.60E+01 0.47 <0.05 5.49E-03 Formatted: Font: (Default) Times New	w Roman
SPAUB         -1.18E+02         0.65         <0.005	w Roman
GRERU 4.33E+00 0.00 = 2.86E+02 0.70 <0.001 1.77E+02 0.56 <0.005 4.66E-03 Formatted: Four: (Default) Times New	
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   Formatted: Font: (Default) Times New	



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

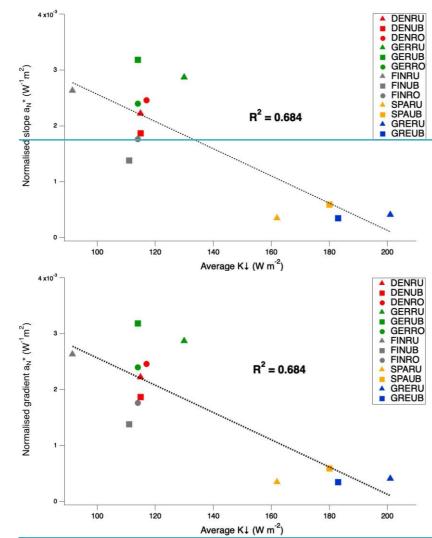
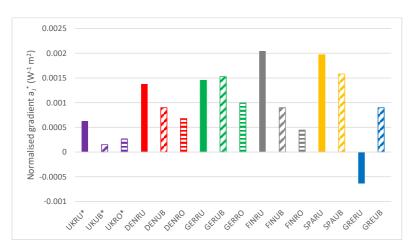


Figure 2: Relationship of average downward incoming solar radiation  $(K\downarrow)$  and normalised gradientsslopes  $a_N^*$  for the sites of the present study.



**Figure 3:** Normalised slopes  $a_J^*$  for  $K\downarrow$  for the sites of the present study (\*UK sites are calculated with solar irradiance).

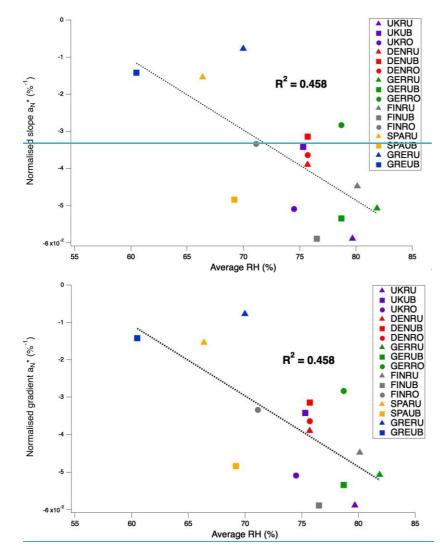
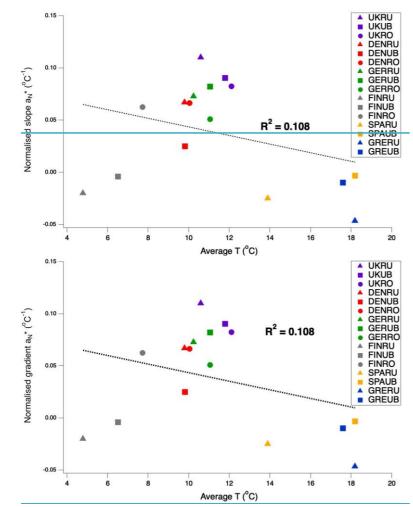


Figure 4: Relation<u>ship</u> of average relative humidity and normalised slopes gradients an\* for the sites of the present study.



**Figure 5:** Relation<u>ship</u> of average temperature and normalised <u>slopes\_gradients</u>  $a_N^*$  <u>for the sites of the present study</u>.

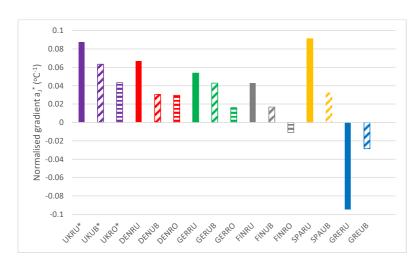
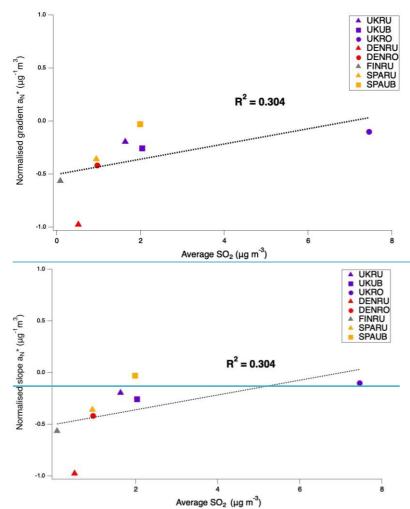
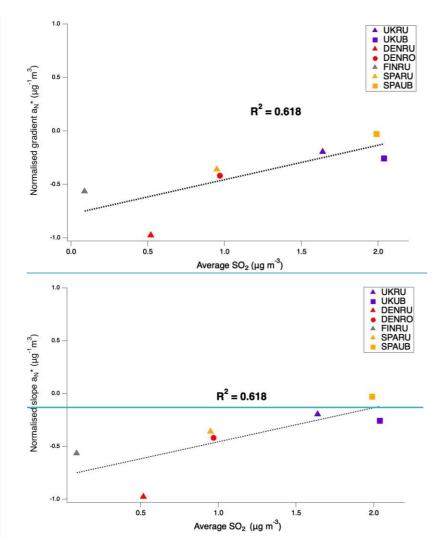


Figure 6: Normalised slopes gradients  $a_J^*$  for temperature for the sites of the present study.



Average  $SO_2$  ( $\mu g \, m^3$ )

Figure 7a: Relation ship of average  $SO_2$  concentrations and normalised slopes gradients  $a_N^*$  for the sites of the present study.



**Figure 7b:** Relation<u>ship</u> of average  $SO_2$  concentrations and normalised <u>slopes-gradients</u>  $a_N^*$  for the <u>sites of the present study</u> (UKRO not included).

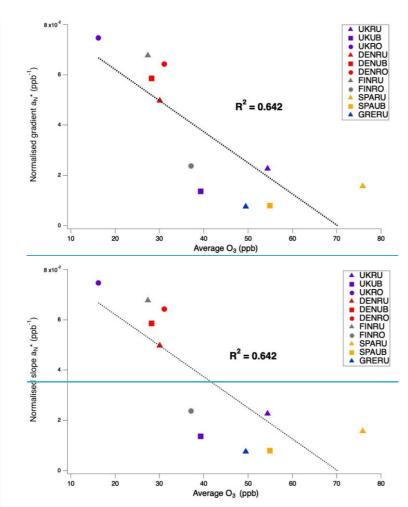


Figure 8: Relationship of average  $O_3$  concentrations and normalised slopes-gradients  $a_N^*$ -for the sites of the present study.

## SUPPLEMENTARY INFORMATION

4

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

8

- 9 Dimitrios Bousiotis, James Brean, Francis Pope, Manuel Dall'Osto, Xavier
- 10 Querol, Andres Alastuey, Noemi Perez, Tuukka Petäjä, Andreas Massling,
- Jacøb Klenø Nøjgaard, Claus Nørdstrom, Giorgos Kouvarakis, Stergios
- 12 Vratolis, Konstantinos Eleftheriadis, Jarkko V. Niemi, Harri Portin, Alfred
- 13 Wiedensohler, Kay Weinhold, Maik Merkel, Thomas Tuch and Roy M.
- 14 Harrison

**Table S1:** Correlation matrices of the meteorological and atmospheric variables.

UKRU	SR	RH	Т	WS	P	$SO_2$	NOx	O <sub>3</sub>	OC	SO <sub>4</sub> <sup>2-</sup>	CS
SR	1.00	-0.70	0.55	0.12	0.13	0.05	-0.12	0.45	0.07	0.05	0.00
RH	-0.70	1.00	-0.55	-0.29	-0.10	-0.10	0.20	-0.59	0.01	-0.04	0.01
Т	0.55	-0.55	1.00	0.12	0.09	-0.01	-0.26	0.37	0.15	0.11	-0.03
WS	0.12	-0.29	0.12	1.00	-0.42	0.04	-0.19	0.41	-0.29	-0.12	-0.32
P	0.13	-0.10	0.09	-0.42	1.00	-0.07	0.03	-0.09	0.13	0.15	0.23
SO <sub>2</sub>	0.05	-0.10	-0.01	0.04	-0.07	1.00	0.05	0.06	0.03	0.37	0.31
NOx	-0.12	0.20	-0.26	-0.19	0.03	0.05	1.00	-0.58	0.48	0.16	0.54
O <sub>3</sub>	0.45	-0.59	0.37	0.41	-0.09	0.06	-0.58	1.00	-0.30	-0.07	-0.34
OC	0.07	0.01	0.15	-0.29	0.13	0.03	0.48	-0.30	1.00	0.37	0.59
SO <sub>4</sub> <sup>2</sup> -	0.05	-0.04	0.11	-0.12	0.15	0.37	0.16	-0.07	0.37	1.00	0.44
CS	0.00	0.01	-0.03	-0.32	0.23	0.31	0.54	-0.34	0.59	0.44	1.00

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UKUB	SR	RH	Т	WS	P	$SO_2$	NOx	$O_3$	OC	SO <sub>4</sub> <sup>2-</sup>	CS
SR	1.00	-0.70	0.53	0.22	0.07	0.08	-0.15	0.47	0.01	0.02	-0.14
RH	-0.70	1.00	-0.56	-0.22	-0.19	-0.08	0.20	-0.66	-0.10	-0.01	0.08
Т	0.53	-0.56	1.00	0.21	-0.05	-0.05	-0.38	0.52	-0.12	0.01	-0.18
WS	0.22	-0.22	0.21	1.00	-0.33	-0.16	-0.44	0.41	-0.43	-0.25	-0.50
P	0.07	-0.19	-0.05	-0.33	1.00	0.18	0.22	-0.06	0.31	0.25	0.26
SO <sub>2</sub>	0.08	-0.08	-0.05	-0.16	0.18	1.00	0.44	-0.16	0.29	0.40	0.39
NOx	-0.15	0.20	-0.38	-0.44	0.22	0.44	1.00	-0.56	0.57	0.29	0.79
O <sub>3</sub>	0.47	-0.66	0.52	0.41	-0.06	-0.16	-0.56	1.00	-0.14	-0.14	-0.40
OC	0.01	-0.10	-0.12	-0.43	0.31	0.29	0.57	-0.14	1.00	0.46	0.63
SO <sub>4</sub> <sup>2</sup> -	0.02	-0.01	0.01	-0.25	0.25	0.40	0.29	-0.14	0.46	1.00	0.36
CS	-0.14	0.08	-0.18	-0.50	0.26	0.39	0.79	-0.40	0.63	0.36	1.00

UKRO	SR	RH	T	WS	P	$SO_2$	NOx	$O_3$	OC	SO <sub>4</sub> <sup>2-</sup>	CS
SR	1.00	-0.68	0.51	0.11	0.15	0.14	0.17	0.16	-0.03	0.03	0.06
RH	-0.68	1.00	-0.49	-0.14	-0.24	0.01	-0.01	-0.35	0.09	-0.01	0.06
T	0.51	-0.49	1.00	0.16	0.21	0.18	0.15	-0.02	0.02	0.01	0.15
WS	0.11	-0.14	0.16	1.00	-0.34	0.17	0.17	0.08	-0.16	-0.19	-0.05
P	0.15	-0.24	0.21	-0.34	1.00	-0.10	-0.05	0.04	0.15	0.08	0.01
SO <sub>2</sub>	0.14	0.01	0.18	0.17	-0.10	1.00	0.91	-0.65	0.36	-0.13	0.72
NOx	0.17	-0.01	0.15	0.17	-0.05	0.91	1.00	-0.63	0.34	-0.04	0.81
$O_3$	0.16	-0.35	-0.02	0.08	0.04	-0.65	-0.63	1.00	-0.43	0.02	-0.64
OC	-0.03	0.09	0.02	-0.16	0.15	0.36	0.34	-0.43	1.00	0.24	0.47
SO <sub>4</sub> <sup>2-</sup>	0.03	-0.01	0.01	-0.19	0.08	-0.13	-0.04	0.02	0.24	1.00	0.18
CS	0.06	0.06	0.15	-0.05	0.01	0.72	0.81	-0.64	0.47	0.18	1.00

DENRU	SR	RH	Т	WS	$SO_2$	NOx	O <sub>3</sub>	OC	SO <sub>4</sub> <sup>2-</sup>	CS
SR	1.00	-0.56	0.44	0.07	-0.05	-0.12	0.43	-0.04	-0.09	0.05
RH	-0.56	1.00	-0.39	0.02	0.02	0.17	-0.54	0.01	0.18	-0.08
Т	0.44	-0.39	1.00	-0.18	-0.09	-0.19	0.37	-0.13	-0.06	0.22
WS	0.07	0.02	-0.18	1.00	0.02	-0.28	0.22	-0.09	0.02	-0.32
$SO_2$	-0.05	0.02	-0.09	0.02	1.00	0.18	-0.06	0.48	0.51	0.34
NOx	-0.12	0.17	-0.19	-0.28	0.18	1.00	-0.58	0.34	0.22	0.54
O <sub>3</sub>	0.43	-0.54	0.37	0.22	-0.06	-0.58	1.00	-0.17	-0.18	-0.17
OC	-0.04	0.01	-0.13	-0.09	0.48	0.34	-0.17	1.00	0.65	0.58
SO <sub>4</sub> <sup>2-</sup>	-0.09	0.18	-0.06	0.02	0.51	0.22	-0.18	0.65	1.00	0.41
CS	0.05	-0.08	0.22	-0.32	0.34	0.54	-0.17	0.58	0.41	1.00

DENUB	SR	RH	Т	WS	NOx	$O_3$	CS
SR	1.00	-0.55	0.45	0.06	-0.02	0.39	0.04
RH	-0.55	1.00	-0.40	-0.02	0.15	-0.58	-0.04
T	0.45	-0.40	1.00	-0.13	-0.11	0.40	0.18
WS	0.06	-0.02	-0.13	1.00	-0.37	0.26	-0.35
NOx	-0.02	0.15	-0.11	-0.37	1.00	-0.59	0.55
O <sub>3</sub>	0.39	-0.58	0.40	0.26	-0.59	1.00	-0.23
CS	0.04	-0.04	0.18	-0.35	0.55	-0.23	1.00

DENRO	SR	RH	Т	WS	$SO_2$	NOx	O <sub>3</sub>	OC	CS
SR	1.00	-0.55	0.30	0.21	0.37	0.29	0.41	0.00	0.26
RH	-0.55	1.00	-0.45	-0.09	-0.26	-0.17	-0.42	-0.20	-0.29
Т	0.30	-0.45	1.00	0.04	0.22	0.12	0.25	0.39	0.41
WS	0.21	-0.09	0.04	1.00	-0.16	-0.12	0.53	-0.19	-0.12
SO <sub>2</sub>	0.37	-0.26	0.22	-0.16	1.00	0.80	0.01	0.31	0.62
NOx	0.29	-0.17	0.12	-0.12	0.80	1.00	-0.02	0.20	0.67
O <sub>3</sub>	0.41	-0.42	0.25	0.53	0.01	-0.02	1.00	-0.01	0.05
OC	0.00	-0.20	0.39	-0.19	0.31	0.20	-0.01	1.00	0.36
CS	0.26	-0.29	0.41	-0.12	0.62	0.67	0.05	0.36	1.00

GERRU	SR	RH	Т	WS	P	OC	SO <sub>4</sub> <sup>2-</sup>	CS
SR	1.00	-0.70	0.55	0.20	0.13	-0.07	-0.07	0.02
RH	-0.70	1.00	-0.61	-0.31	-0.12	0.08	0.10	-0.01
T	0.55	-0.61	1.00	0.01	0.11	-0.34	-0.29	-0.11
WS	0.20	-0.31	0.01	1.00	-0.24	-0.14	-0.09	-0.35
P	0.13	-0.12	0.11	-0.24	1.00	0.11	0.13	0.23
OC	-0.07	0.08	-0.34	-0.14	0.11	1.00	0.83	0.65
SO <sub>4</sub> <sup>2-</sup>	-0.07	0.10	-0.29	-0.09	0.13	0.83	1.00	0.52
CS	0.02	-0.01	-0.11	-0.35	0.23	0.65	0.52	1.00

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

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

FINRU	SR	RH	T	WS	P	$SO_2$	NOx	O <sub>3</sub>	OM	SO <sub>4</sub> <sup>2-</sup>	CS
SR	1.00	-0.67	0.50	0.11	0.11	0.00	-0.24	0.30	-0.05	-0.14	0.09
RH	-0.67	1.00	-0.56	-0.21	-0.27	-0.12	0.31	-0.55	0.00	0.17	-0.20
T	0.50	-0.56	1.00	0.01	0.03	-0.20	-0.28	-0.14	0.27	-0.20	0.28
WS	0.11	-0.21	0.01	1.00	0.17	0.11	0.13	0.35	-0.20	-0.20	-0.07
P	0.11	-0.27	0.03	0.17	1.00	0.00	-0.08	-0.08	0.34	0.12	0.19
SO <sub>2</sub>	0.00	-0.12	-0.20	0.11	0.00	1.00	0.18	0.09	NA	NA	0.21
NOx	-0.24	0.31	-0.28	0.13	-0.08	0.18	1.00	-0.24	NA	NA	0.12
O <sub>3</sub>	0.30	-0.55	-0.14	0.35	-0.08	0.09	-0.24	1.00	NA	NA	0.02
OM	-0.05	0.00	0.27	-0.20	0.34	NA	NA	NA	1.00	0.43	0.61
SO <sub>4</sub> <sup>2-</sup>	-0.14	0.17	-0.20	-0.20	0.12	NA	NA	NA	0.43	1.00	0.18
CS	0.09	-0.20	0.28	-0.07	0.19	0.21	0.12	0.02	0.61	0.18	1.00

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

FINRO	SR	RH	Т	WS	P	NOx	$O_3$	CS
SR	1.00	-0.58	0.47	0.03	0.08	0.05	0.20	0.09
RH	-0.58	1.00	-0.29	-0.05	-0.24	0.02	-0.34	0.01
Т	0.47	-0.29	1.00	-0.07	-0.02	-0.08	0.18	0.05
WS	0.03	-0.05	-0.07	1.00	-0.25	-0.29	0.41	-0.32
P	0.08	-0.24	-0.02	-0.25	1.00	0.10	-0.09	0.13
NOx	0.05	0.02	-0.08	-0.29	0.10	1.00	-0.61	0.75
O <sub>3</sub>	0.20	-0.34	0.18	0.41	-0.09	-0.61	1.00	-0.51
CS	0.09	0.01	0.05	-0.32	0.13	0.75	-0.51	1.00

SPARU	SR	RH	Т	WS	P	$SO_2$	NO2	O <sub>3</sub>	CS
SR	1.00	-0.45	0.50	0.38	0.09	0.10	-0.02	0.34	0.34
RH	-0.45	1.00	-0.29	-0.20	-0.24	-0.08	0.05	-0.48	-0.06
Т	0.50	-0.29	1.00	0.16	0.24	0.07	-0.05	0.54	0.47
WS	0.38	-0.20	0.16	1.00	-0.16	0.13	-0.02	0.25	0.10
P	0.09	-0.24	0.24	-0.16	1.00	-0.15	0.12	0.09	0.14
$SO_2$	0.10	-0.08	0.07	0.13	-0.15	1.00	0.14	0.19	0.25
NO <sub>2</sub>	-0.02	0.05	-0.05	-0.02	0.12	0.14	1.00	-0.02	0.42
O <sub>3</sub>	0.34	-0.48	0.54	0.25	0.09	0.19	-0.02	1.00	0.44
CS	0.34	-0.06	0.47	0.10	0.14	0.25	0.42	0.44	1.00

SPAUB	SR	RH	Т	WS	P	$SO_2$	NO2	$O_3$	CS
SR	1.00	-0.43	0.44	0.18	0.03	0.25	-0.09	0.32	0.00
RH	-0.43	1.00	-0.04	-0.23	-0.16	-0.12	0.10	-0.23	0.16
Т	0.44	-0.04	1.00	-0.14	0.11	0.35	-0.07	0.38	0.11
WS	0.18	-0.23	-0.14	1.00	-0.26	-0.08	-0.34	0.32	-0.43
P	0.03	-0.16	0.11	-0.26	1.00	0.13	0.15	-0.10	0.10
SO <sub>2</sub>	0.25	-0.12	0.35	-0.08	0.13	1.00	0.20	0.13	0.16
NO <sub>2</sub>	-0.09	0.10	-0.07	-0.34	0.15	0.20	1.00	-0.66	0.59
O <sub>3</sub>	0.32	-0.23	0.38	0.32	-0.10	0.13	-0.66	1.00	-0.35
CS	0.00	0.16	0.11	-0.43	0.10	0.16	0.59	-0.35	1.00

GRERU	SR	RH	Т	WS	P	NO <sub>2</sub>	O <sub>3</sub>	OC	CS
SR	1.00	-0.30	0.33	0.02	-0.11	0.36	0.19	0.09	0.18
RH	-0.30	1.00	-0.25	-0.27	0.20	-0.20	-0.12	-0.06	0.08
T	0.33	-0.25	1.00	0.00	-0.53	0.02	0.54	0.35	0.46
WS	0.02	-0.27	0.00	1.00	-0.21	-0.03	0.15	0.14	0.11
P	-0.11	0.20	-0.53	-0.21	1.00	-0.10	-0.35	-0.24	-0.09
NO <sub>2</sub>	0.36	-0.20	0.02	-0.03	-0.10	1.00	0.00	0.01	-0.02
$O_3$	0.19	-0.12	0.54	0.15	-0.35	0.00	1.00	0.50	0.62
OC	0.09	-0.06	0.35	0.14	-0.24	0.01	0.50	1.00	0.47
CS	0.18	0.08	0.46	0.11	-0.09	-0.02	0.62	0.47	1.00

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

Table S2: Slopes Gradients and R<sup>2</sup> for the relationrelationship between VOCs and NPF events
 variables.

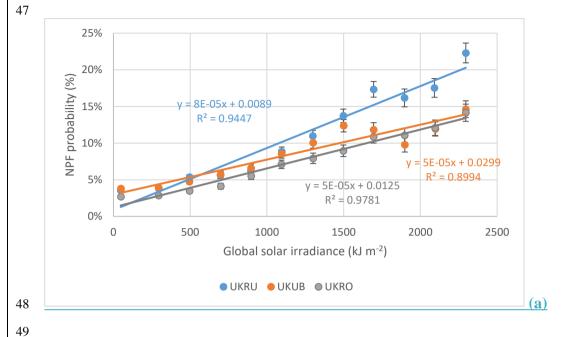
UKRU	$a_{N}$	$\mathbb{R}^2$	agr	$\mathbb{R}^2$	ал	$\mathbb{R}^2$
benzene	-3.37E-01	0.88	1.24E+00	0.16	-5.99E-03	0.07
ethane	-5.42E-02	0.88	-4.79E-01	0.26	-4.61E-03	0.77
ethene	-1.65E-01	0.83	2.64E+00	0.60	-1.70E-02	0.57
ethylbenzene	-7.01E-01	0.79	6.78E+00	0.41	-5.77E-02	0.63
iso.butane	-2.06E-01	0.75	1.41E+00	0.70	-5.62E-03	0.11
iso.octane	-5.23E-01	0.45	1.09E+01	0.80	9.32E-03	0.11
iso.pentane	-1.96E-01	0.74	2.36E+00	0.58	2.36E-02	0.72
m.p.xylene	-2.92E-01	0.86	3.21E+00	0.68	-1.98E-02	0.35
n.butane	-1.67E-01	0.79	1.04E+00	0.44	1.43E-02	0.11
n.heptane	-9.63E-01	0.80	1.36E+01	0.73	-1.46E-02	0.13
n.hexane	-1.21E+00	0.84	6.82E+00	0.67	1.33E-02	0.11
n.pentane	-3.71E-01	0.67	3.49E+00	0.64	-8.97E-03	0.06
o.xylene	-5.34E-01	0.71	8.59E+00	0.86	-1.81E-02	0.42
propane	-7.77E-02	0.76	1.97E-01	0.24	-4.28E-03	0.49
propene	-1.50E-01	0.67	-4.01E-01	0.02	6.20E-03	0.08
toluene	-1.48E-01	0.79	1.88E+00	0.81	-9.26E-03	0.43
1.2.4.trimethylbenzene	-4.36E-01	0.46	5.38E+00	0.29	-4.78E-02	0.68
1.3.butadiene	-1.17E+00	0.40	-1.68E+01	0.71	-7.55E-02	0.66
1.butene	-9.39E-02	0.03	-4.77E+00	0.25	-1.99E-02	0.07
2.methylpentane	-7.66E-01	0.77	8.49E+00	0.57	4.56E-02	0.64

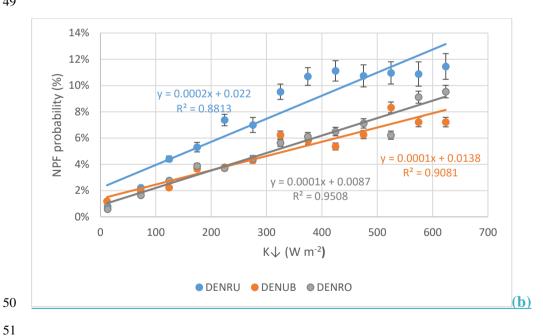
FINRU	an	$\mathbb{R}^2$	2	agr		$R^2$	ај		R
Acetaldehyde	-1.04E-01	0.05	5	-2.16E+0	0	0.69	1.23E-0	2	0.0
Aceticacid	1.19E-01	0.13	3	5.88E+00	0	0.77	7 3.33E-0	2	0.2
Acetolnitrite	-1.02E+00	0.13	3	1.33E+01	1	0.59	6.62E-0	2	0.1
Acetone	-4.63E-02	0.08	3	3.38E+00	0	0.74	5.85E-0	3	0.1
Benzene	-4.46E-01	0.11	1	-2.02E+0	1	0.83	-4.13E-0	)2	0.0
Ethanol	4.04E-02	0.06	5	1.31E+00	0	0.10	4.77E-0	3	0.1
Isoprene	-3.17E+00	0.51	1	1.59E+01	1	0.87	7 -1.50E+0	00	0.3
MEK	6.45E-01	0.34	1	-8.03E+0	0	0.36	2.95E-0	2	0.0
Methacrolein.MVK	-5.15E+00	0.45	5	3.75E+01	1	0.66	2.92E-0	2	0.0
Methanol	1.68E-02	0.05	5	1.48E+00	0	0.75	3.48E-0	3	0.1
Monoterpenes	-1.17E-01	0.38	3	2.84E+00	0	0.56	1.11E-0	3	0.0
Toluene	-4.25E+00	0.59	)	2.88E+01	1	0.80	-5.55E-0	)2	0.1
<del>UKRO</del>	₩	₽≟		<del>a</del> gr	Į	€≟	æĮ	R	2
benzene	-1.03E-01	0.68		1.36E+00	0.	.80	4.42E 02	0.7	<del>78</del>
cis.2.butene	-1.93E-01	0.59		8.33E-01	0	.02	1.70E-01	0.4	18
ethane	2.45E-02	0.53		2.99E 02	0	.06	2.28E 03	0.1	14
ethene	-4.59E-02	0.69		5.74E-01	0	.83	2.50E-02	0.9	<del>)</del>
ethylbenzene	<del>-7.13E-02</del>	0.87		1.22E+00	0	.77	3.59E-02	0.4	<del>   </del>
ethyne	-8.43E-02	0.74		1.23E+00	0	.75	4.22E-02	0.6	54
iso.butane	-4.70E-02	0.55		6.07E-01	0	.78	1.79E-02	0.9	)2
iso.octane	-7.53E-02	0.80		2.14E+00	0	.78	7.35E-02	0.6	<del>57</del>
iso.pentane	-1.10E-02	0.70		2.64E-01	0	<del>.72</del>	1.00E-02	0.8	32
isoprene	-2.75E-02	0.07		4.34E-01	0	.01	2.24E-03	0.0	<del>)()</del>
m.p.xylene	-1.99E-02	0.91		3.81E-01	0	.56	1.47E-02	0.6	54
n.butane	-2.17E-02	0.61		2.58E-01	0	.78	4.07E-03	0.1	17
n.heptane	-1.53E-01	0.75		2.51E+00	0	.80	1.15E-01	0.8	32
n.hexane	-1.10E-01	0.63		2.86E+00	0	.75	8.28E-02	0.7	74
n.octane	-2.64E-01	0.55		7.06E+00	0	<del>.72</del>	2.73E-01	0.9	<del>8</del>
n.pentane	-5.44E-02	0.53	L.	1.03E+00	0	.80	2.99E-02	0.8	36
<del>o.xylene</del>	-4.69E-02	0.88		9.58E-01	0	.65	4.37E-02	0.8	36
<del>propane</del>	-3.16E-02	0.68		1.95E-01	0	<del>.32</del>	1.01E-02	0.9	<del>)</del>
propene	-6.69E-02	0.87	١,	1.15E+00	0	.85	3.55E-02	0.7	78

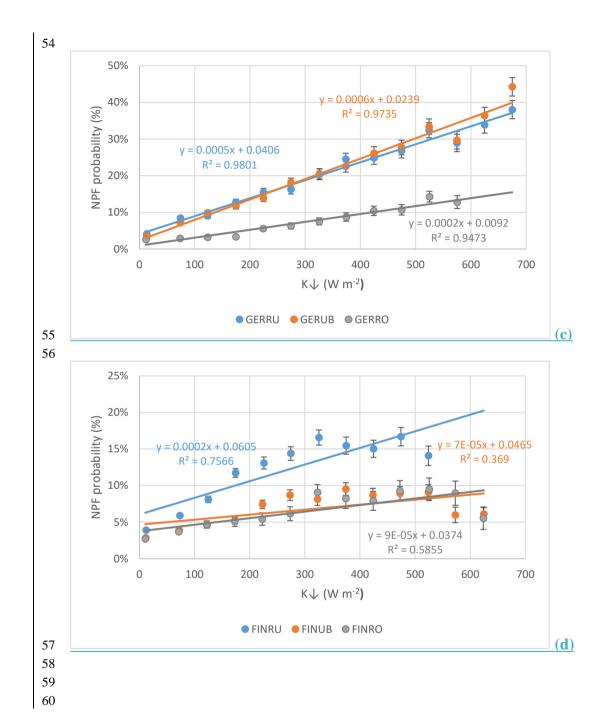
toluene	-1.22E-02	0.84	2.76E-01	0.74	1.15E-02	0.85
trans.2.butene	-2.63E-01	0.72	3.16E+00	0.35	<del>1.41E-01</del>	0.60
trans.2.pentene	<del>-1.67E-01</del>	0.73	2.69E+00	0.31	1.16E-01	0.52
1.2.3.trimethylbenzene	-1.45E-01	0.78	3.31E+00	0.66	1.28E-01	0.81
1.2.4.trimethylbenzene	4.89E 02	0.85	7.64E-01	0.43	3.26E-02	0.46
1.3.5.trimethylbenzene	-8.62E-02	0.77	1.56E+00	0.67	6.65E-02	0.64
1.3.butadiene	-1.78E-01	0.81	2.99E+00	0.44	9.04E 02	0.26
1.butene	-2.18E-01	0.38	2.51E+00	0.25	1.24F-01	0.64
1.pentene	-2.43E 01	0.52	6.92E+00	0.37	3.00E 01	0.82
2.methylpentane	-3.73E-02	0.68	8.57E-01	0.67	2.83E-02	0.80

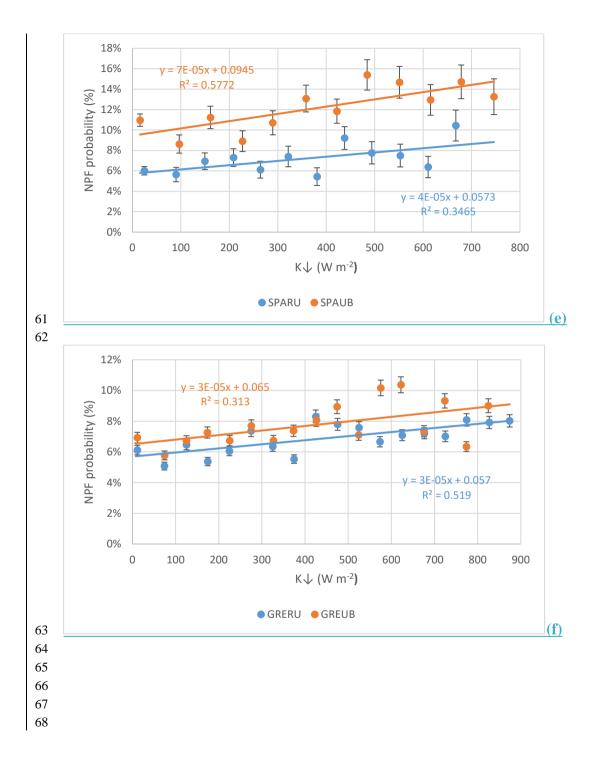
<u>UKRO</u>	<u>a</u> <u>N</u>	<u>R</u> <sup>2</sup>	<u>agr</u>	<u>R</u> <sup>2</sup>	<u>a</u> j	<u>R</u> <sup>2</sup>
<u>benzene</u>	-1.03E-01	0.68	1.36E+00	0.80	4.42E-02	0.78
<u>cis.2.butene</u>	-1.93E-01	0.59	8.33E-01	0.02	1.70E-01	0.48
<u>ethane</u>	-2.45E-02	0.53	2.99E-02	0.06	2.28E-03	0.14
<u>ethene</u>	<u>-4.59E-02</u>	0.69	<u>5.74E-01</u>	0.83	2.50E-02	0.97
ethylbenzene	<u>-7.13E-02</u>	0.87	1.22E+00	0.77	3.59E-02	0.41
ethyne	-8.43E-02	0.74	1.23E+00	0.75	4.22E-02	0.64
<u>iso.butane</u>	<u>-4.70E-02</u>	0.55	6.07E-01	0.78	1.79E-02	0.92
iso.octane	<u>-7.53E-02</u>	0.80	2.14E+00	0.78	7.35E-02	<u>0.67</u>
<u>iso.pentane</u>	-1.10E-02	0.70	2.64E-01	0.72	1.00E-02	0.82
isoprene	<u>-2.75E-02</u>	0.07	4.34E-01	0.01	2.24E-03	0.00
<u>m.p.xylene</u>	-1.99E-02	0.91	3.81E-01	0.56	1.47E-02	<u>0.64</u>
<u>n.butane</u>	<u>-2.17E-02</u>	0.61	2.58E-01	0.78	4.07E-03	0.17
<u>n.heptane</u>	<u>-1.53E-01</u>	0.75	2.51E+00	0.80	1.15E-01	0.82
<u>n.hexane</u>	<u>-1.10E-01</u>	0.63	2.86E+00	0.75	8.28E-02	0.74
<u>n.octane</u>	<u>-2.64E-01</u>	0.55	7.06E+00	0.72	2.73E-01	0.98
<u>n.pentane</u>	-5.44E-02	0.53	1.03E+00	0.80	2.99E-02	0.86
o.xylene	<u>-4.69E-02</u>	0.88	9.58E-01	0.65	4.37E-02	0.86
propane	<u>-3.16E-02</u>	0.68	1.95E-01	0.32	1.01E-02	0.90
propene	-6.69E-02	0.87	1.15E+00	0.85	3.55E-02	0.78
toluene	-1.22E-02	0.84	2.76E-01	0.74	1.15E-02	0.85
trans.2.butene	-2.63E-01	0.72	3.16E+00	0.35	1.41E-01	0.60
trans.2.pentene	<u>-1.67E-01</u>	0.73	2.69E+00	0.31	1.16E-01	0.52
1.2.3.trimethylbenzene	-1.45E-01	0.78	3.31E+00	0.66	1.28E-01	0.81
1.2.4.trimethylbenzene	-4.89E-02	0.85	7.64E-01	0.43	3.26E-02	0.46
1.3.5.trimethylbenzene	-8.62E-02	0.77	1.56E+00	0.67	6.65E-02	0.64
1.3.butadiene	-1.78E-01	0.81	2.99E+00	0.44	9.04E-02	0.26
1.butene	-2.18E-01	0.38	2.51E+00	0.25	1.24E-01	0.64
1.pentene	-2.43E-01	0.52	6.92E+00	0.37	3.00E-01	0.82
2.methylpentane	-3.73E-02	0.68	8.57E-01	0.67	2.83E-02	0.80

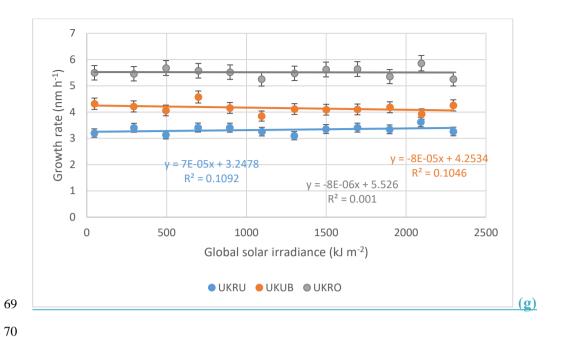
Figure S1: Relationship of solar radiation with NPF variables.

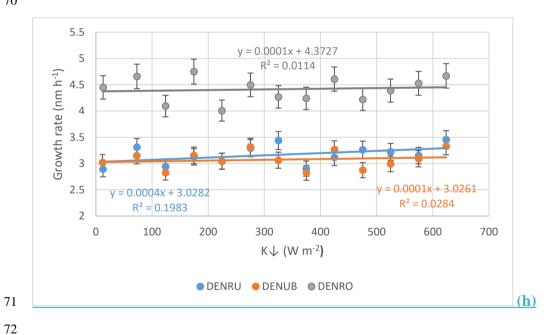


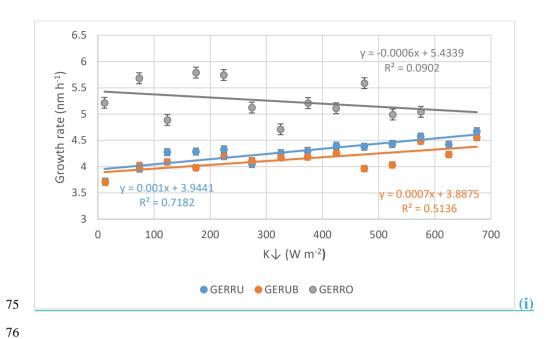


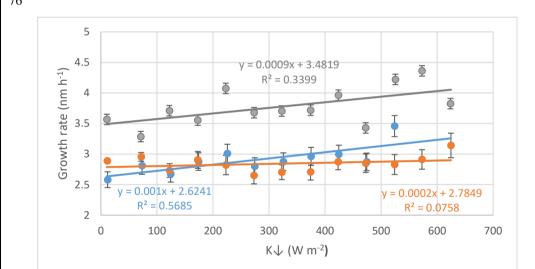










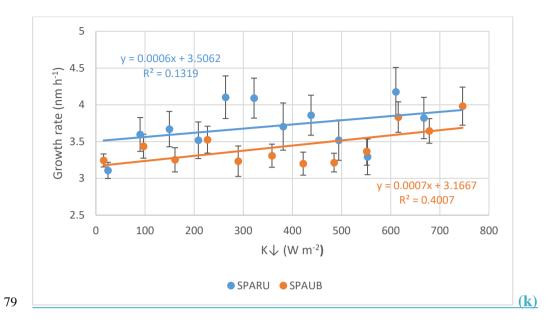


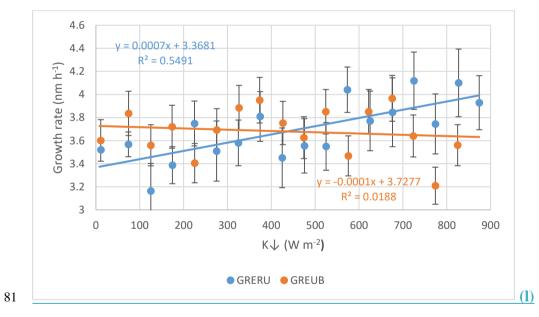
● FINRU ● FINUB ● FINRO

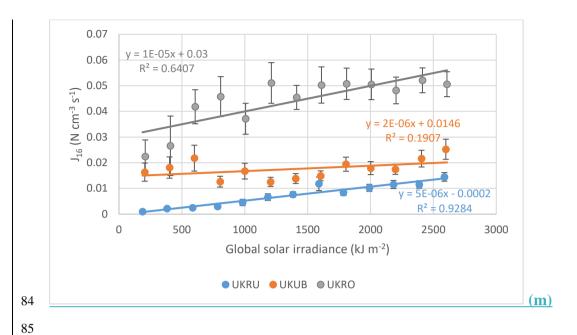
77

78

**(i)** 







0.12 0.10 y = 5E-05x + 0.0482  $R^2 = 0.4956$  y = 5E-05x + 0.0482 y = 2E-05x + 0.0093 y = 2E-05x + 0.0093y = 2E-05x + 0.0093

200

● DENRU ● DENUB ● DENRO

100

y = 2E - 05x + 0.0093  $R^{2} = 0.6385$   $R^{2} = 0.4753$   $300 \quad 400 \quad 500 \quad 600 \quad 700$   $K \downarrow \text{ (W m-2)}$ 

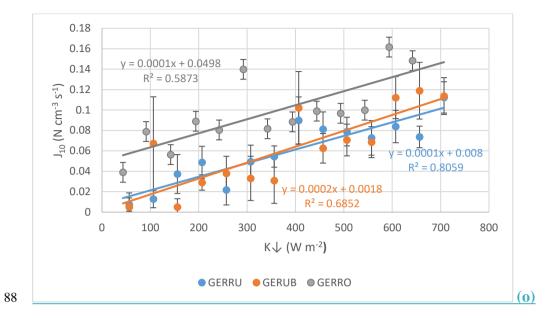
**(n)** 

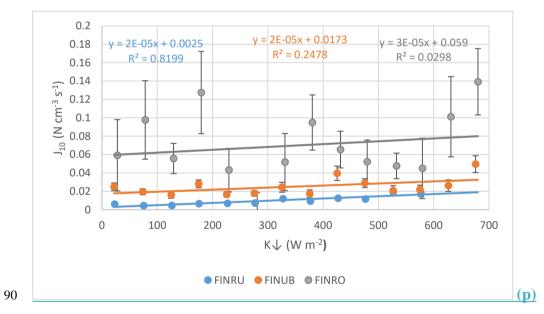
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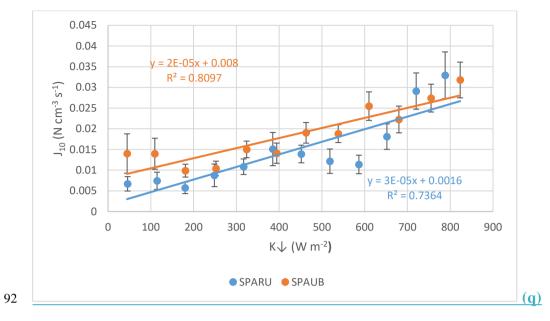
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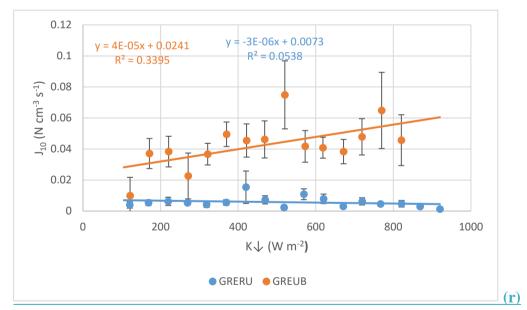
0.02

0.00

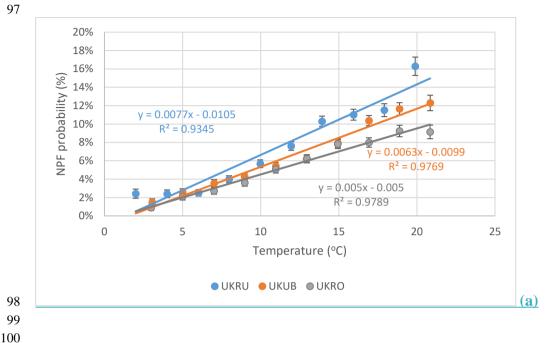


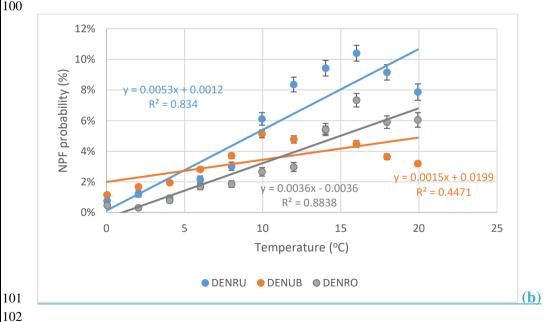


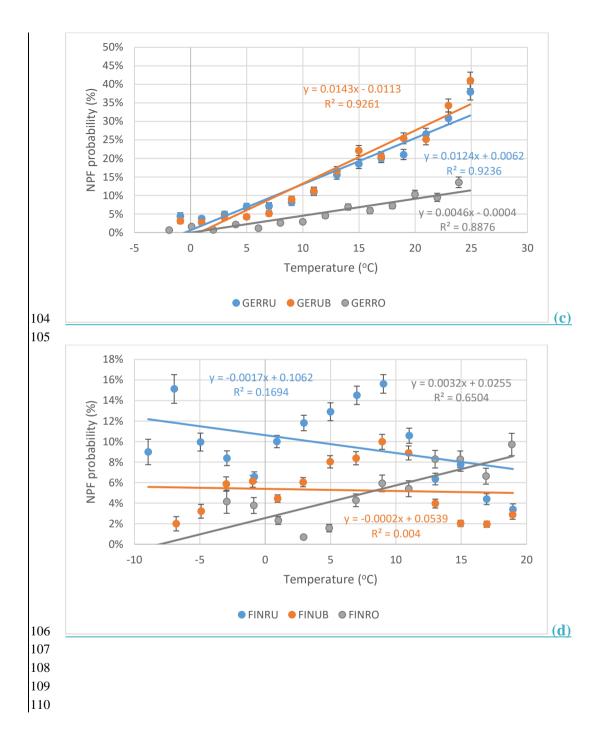


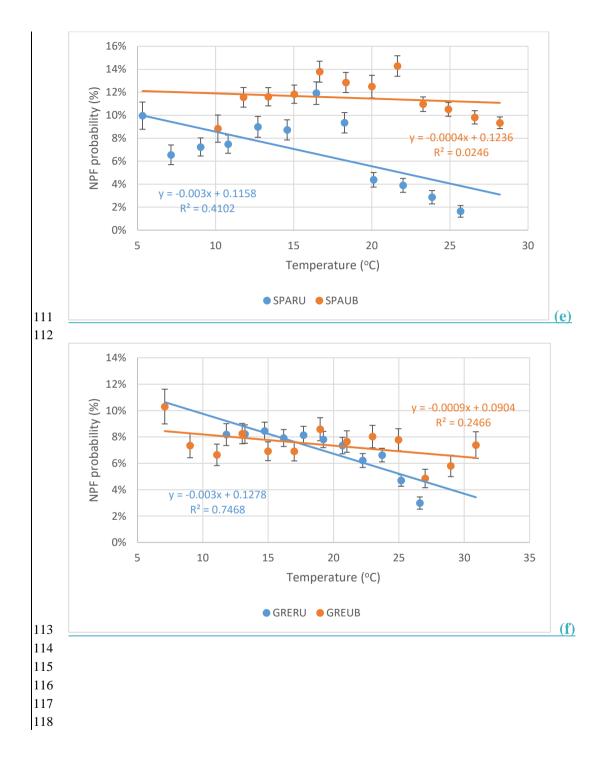


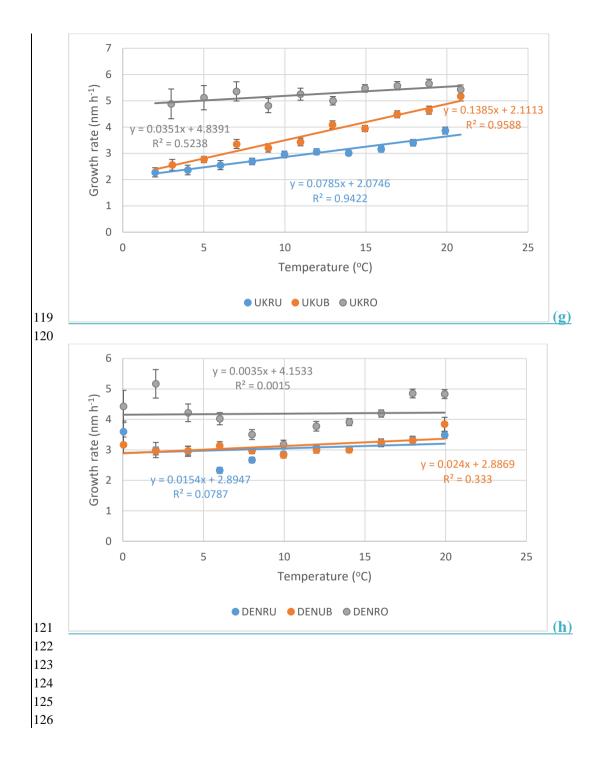


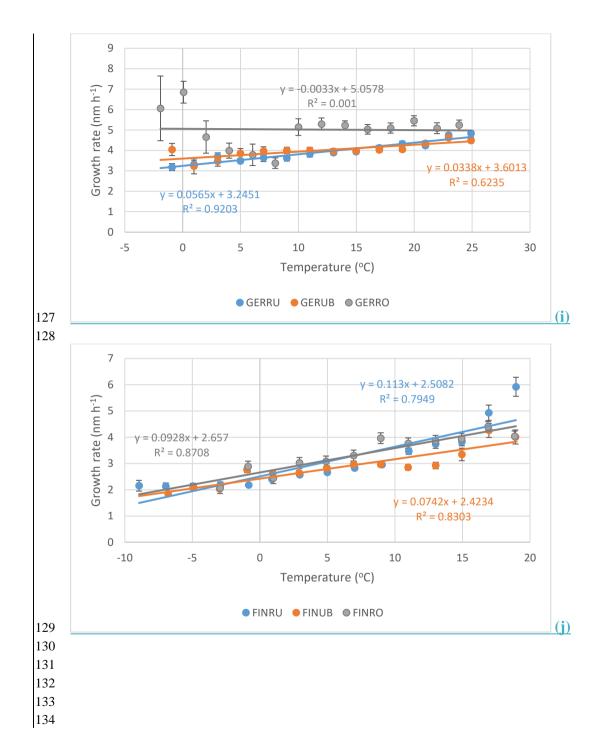


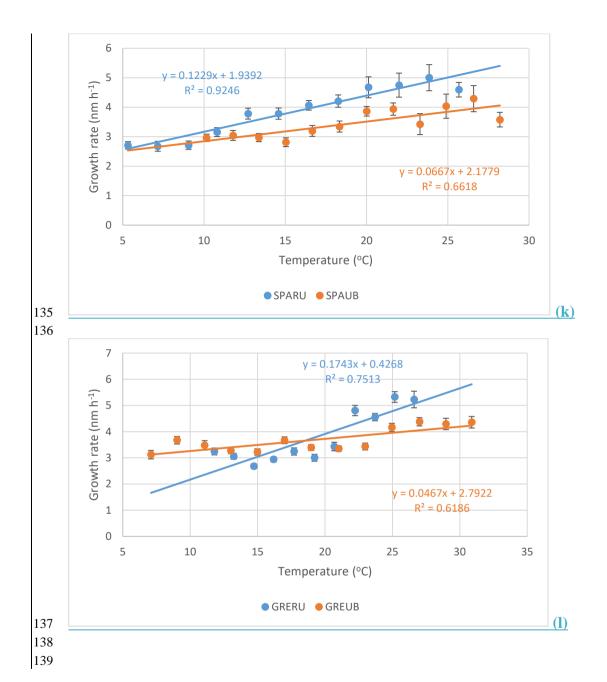


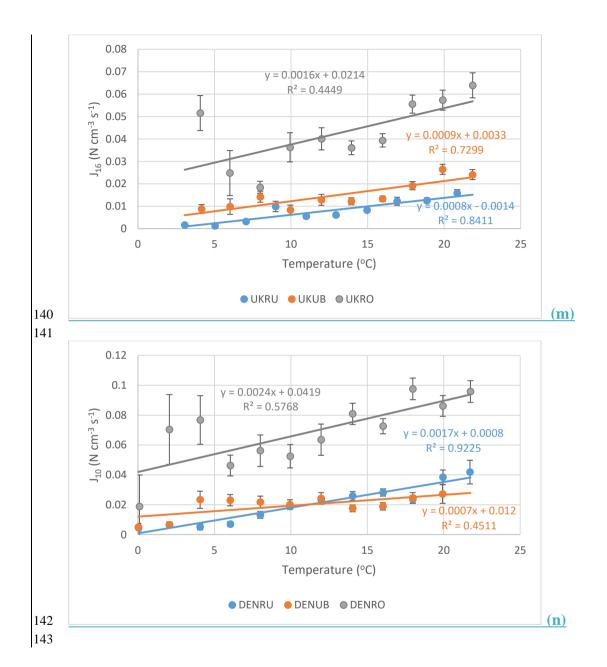


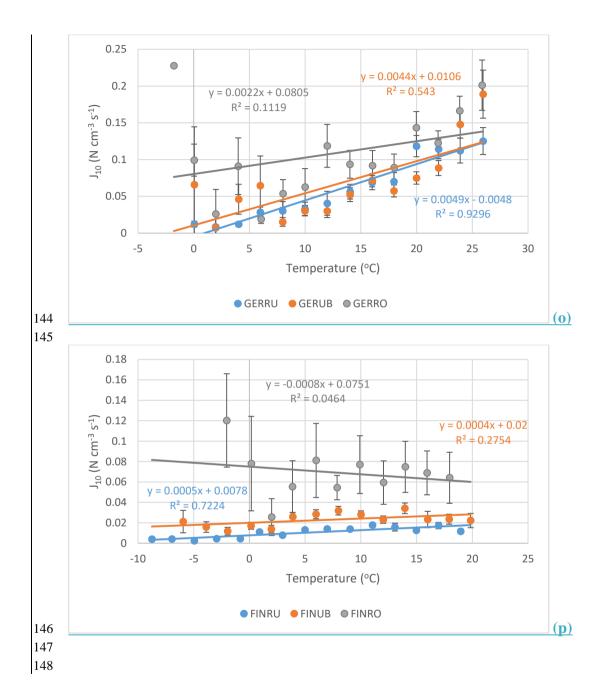


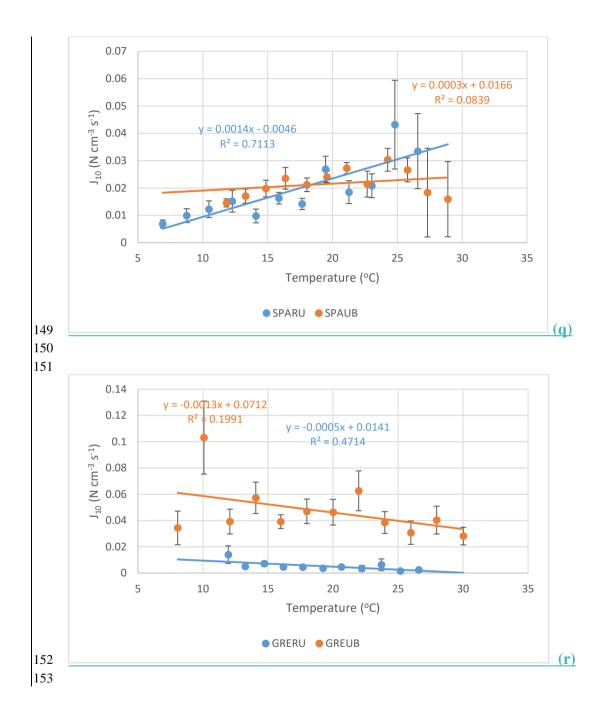




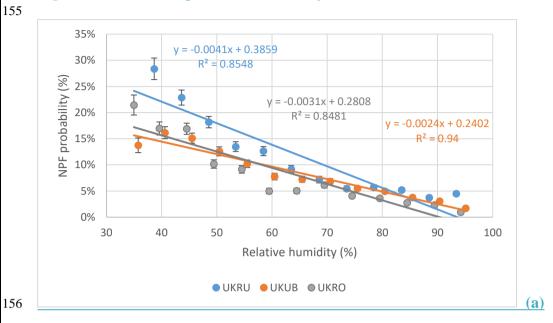


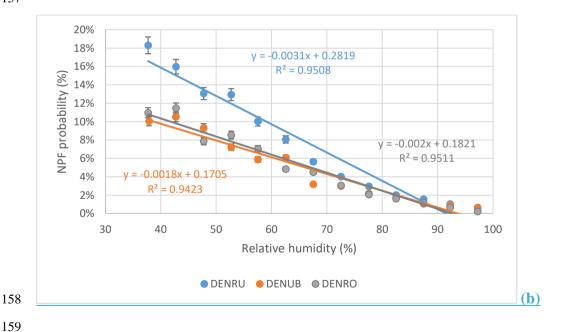




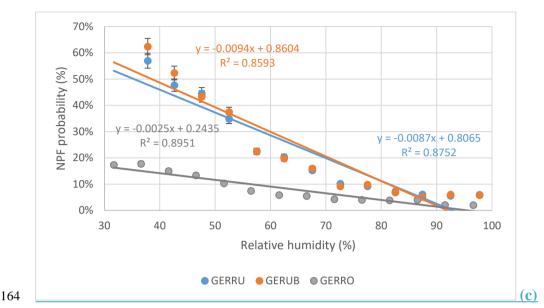


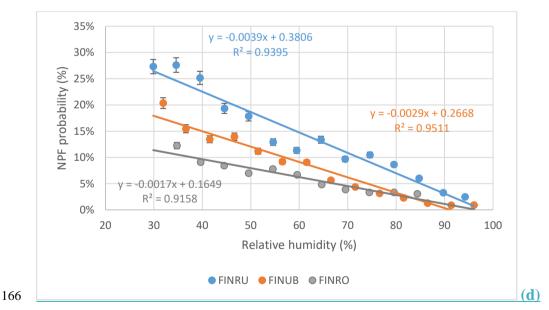
**Figure S3:** Relationship of relative humidity with NPF variables.

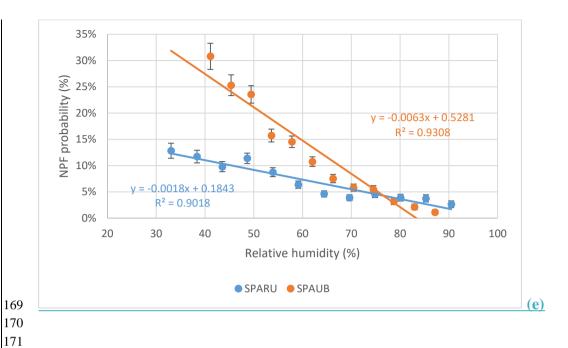


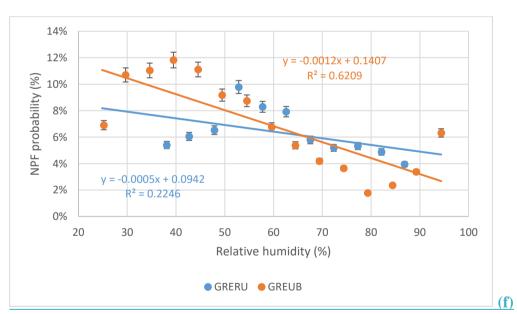


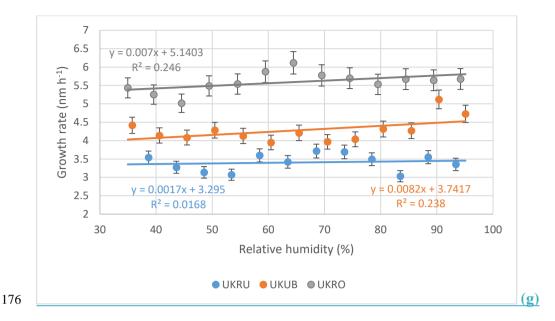




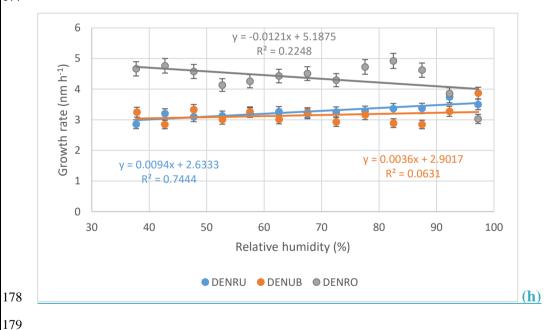


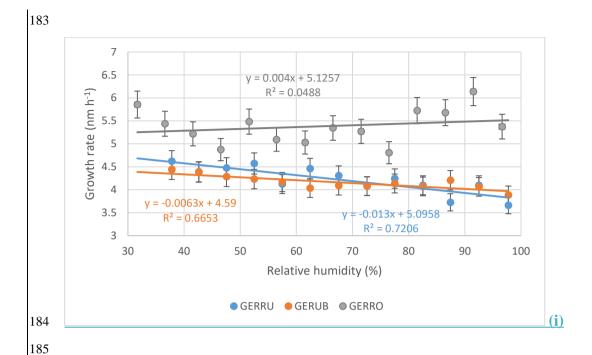


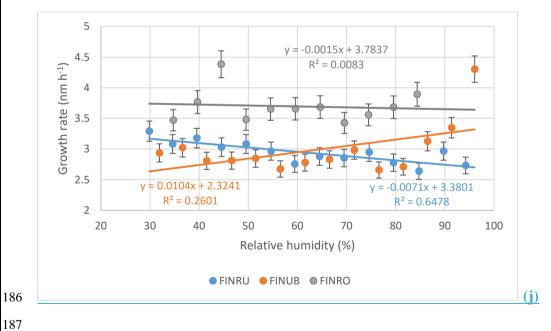


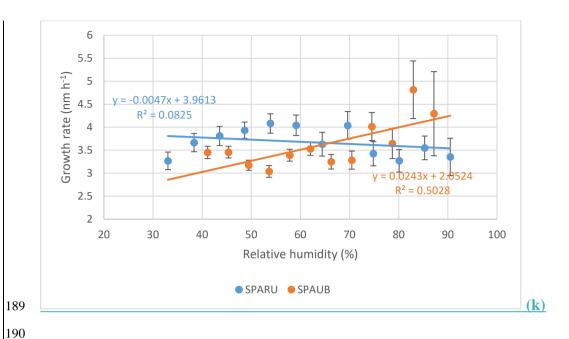


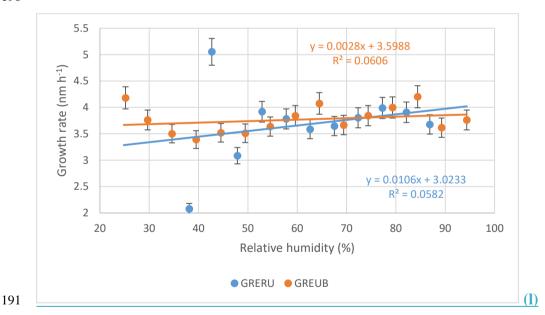


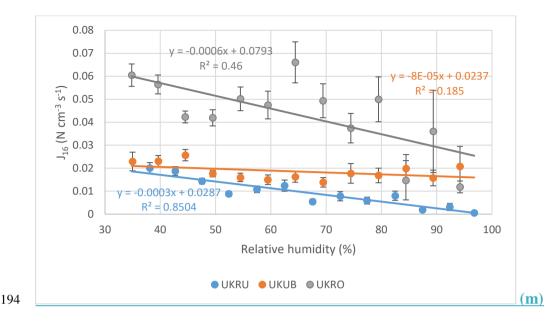


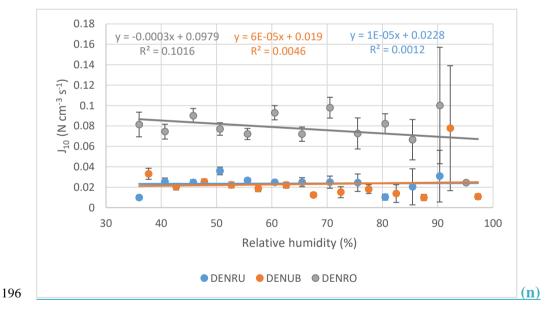


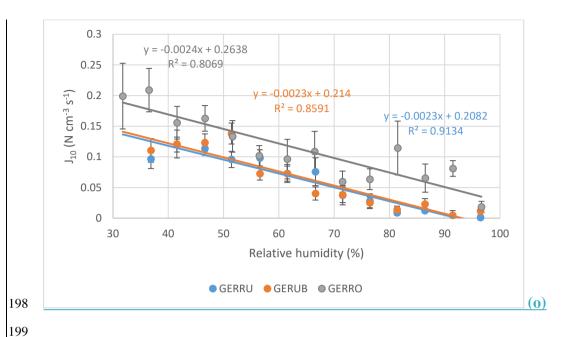


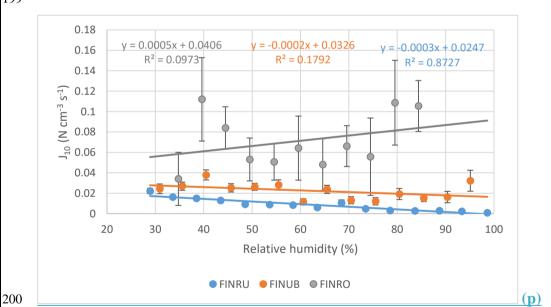












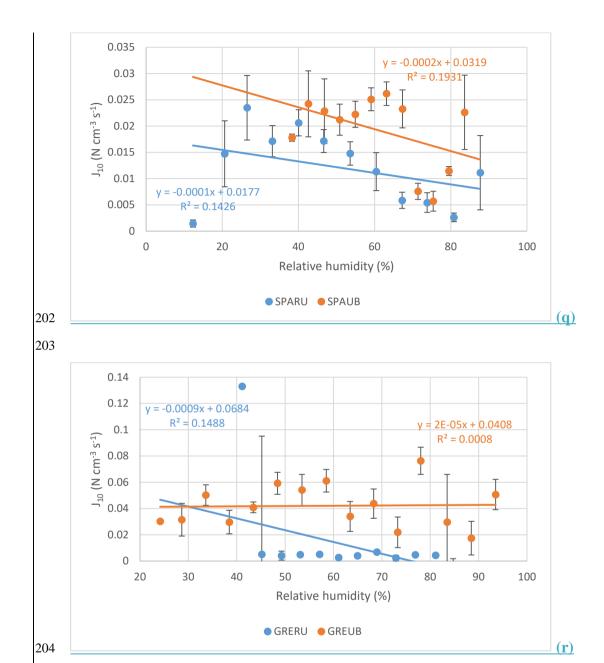
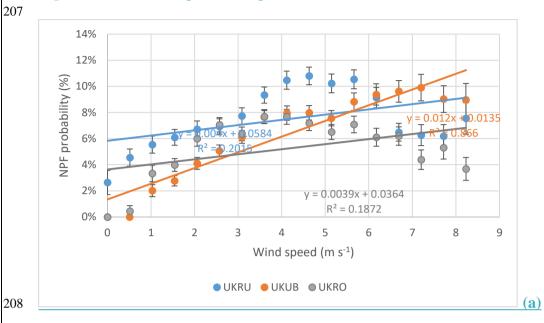
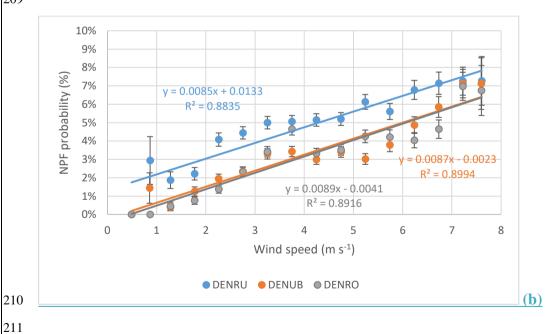
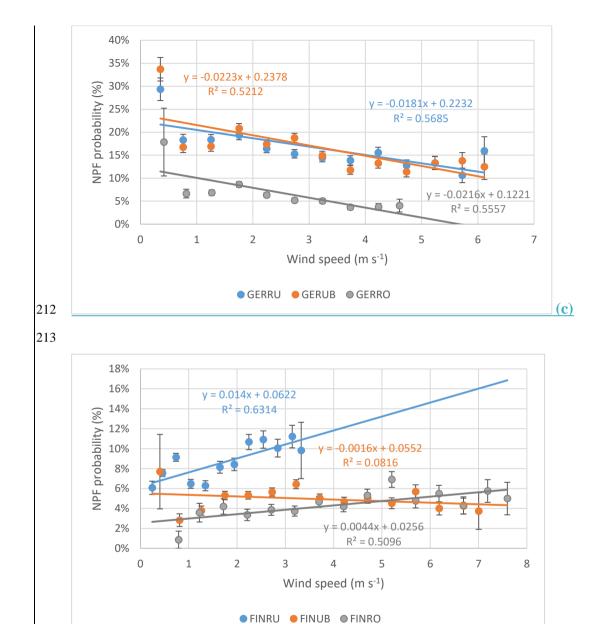


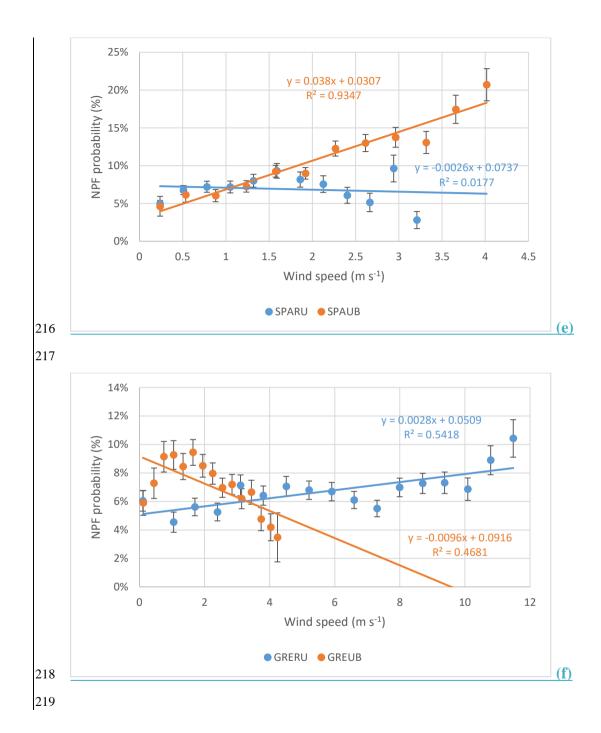
Figure S4: Relationship of wind speed with NPF variables.

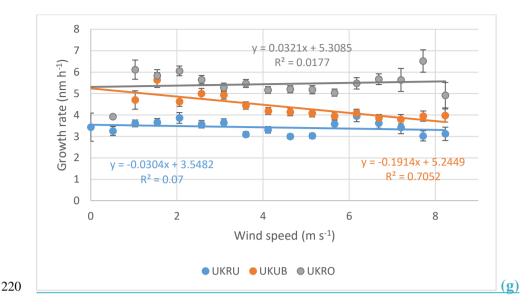




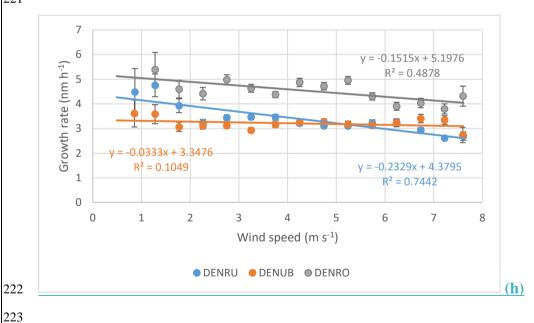


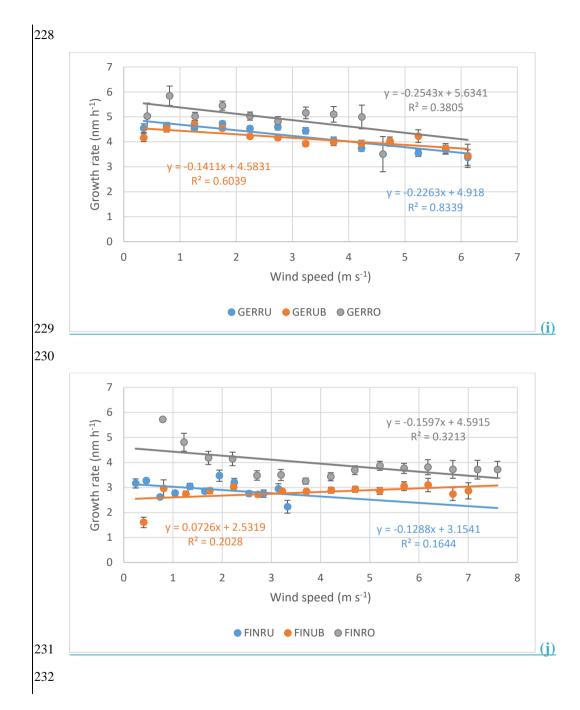
**(d)** 

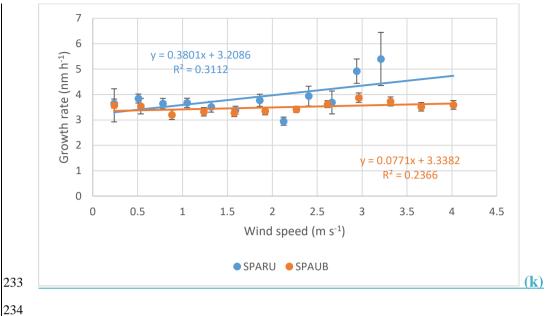




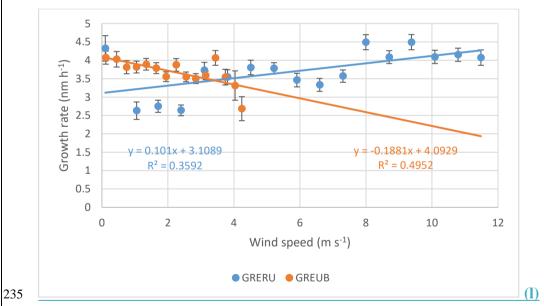


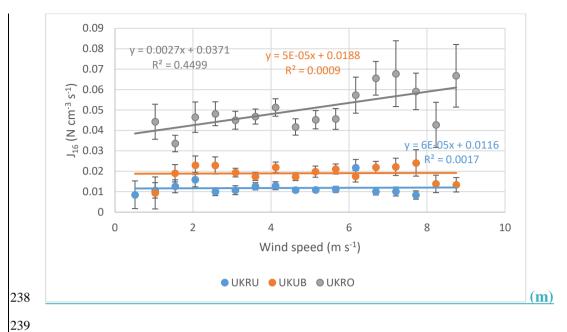


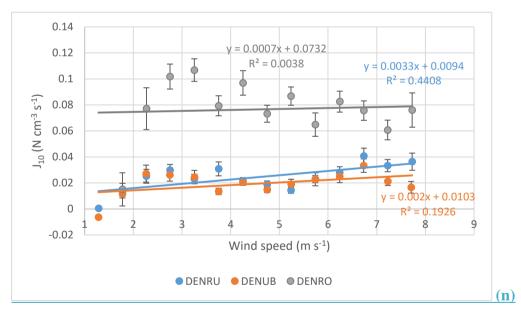


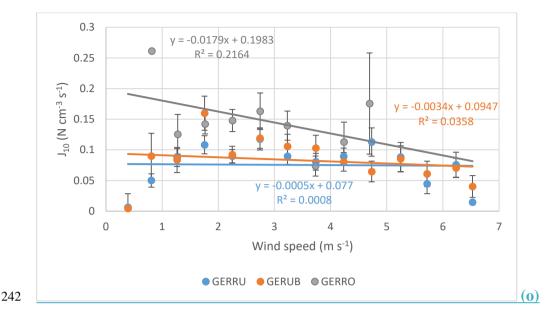




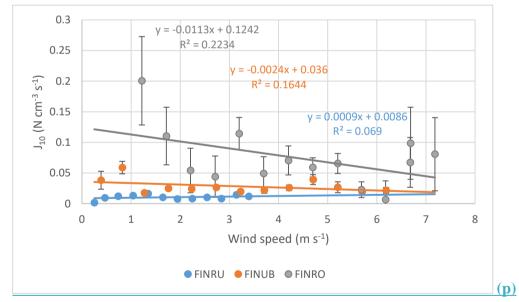


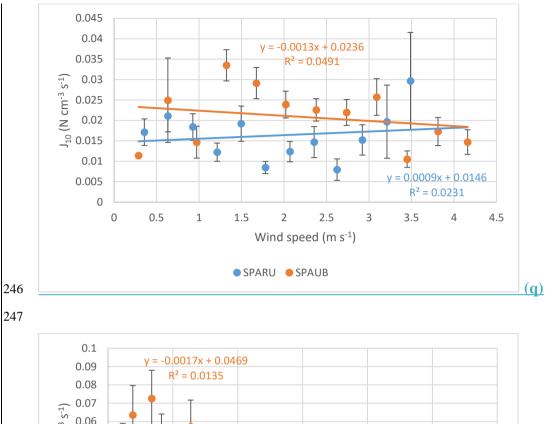








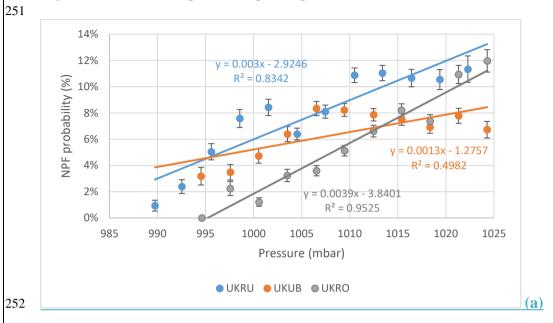


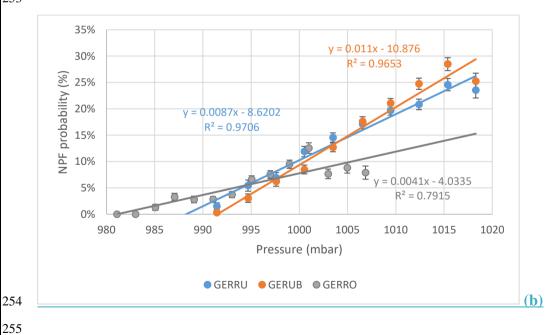


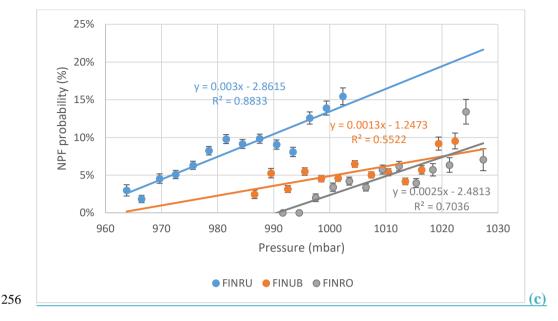
O.07 O.06 O.05 O.04 O.03 y = 9E-06x + 0.0043 $R^2 = 0.0009$ 0.02 0.01 0 6 8 0 2 4 10 12 Wind speed (m s<sup>-1</sup>) ● GRERU ● GREUB <u>(r)</u>

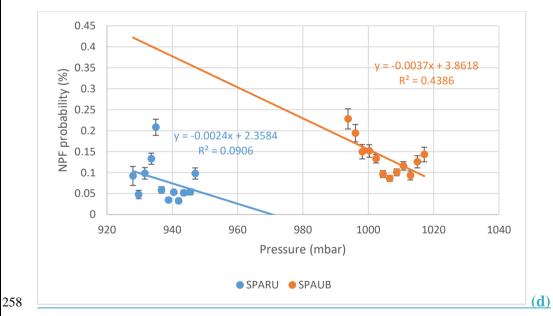
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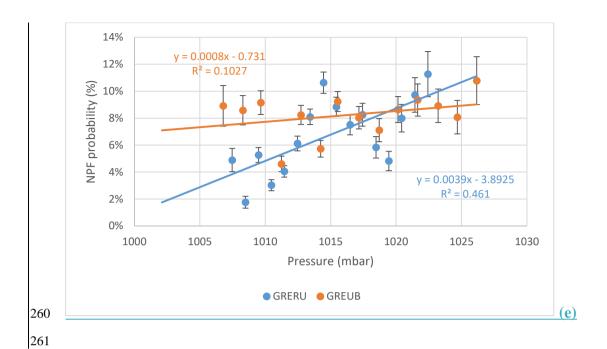
Figure S5: Relationship of atmospheric pressure with NPF variables.

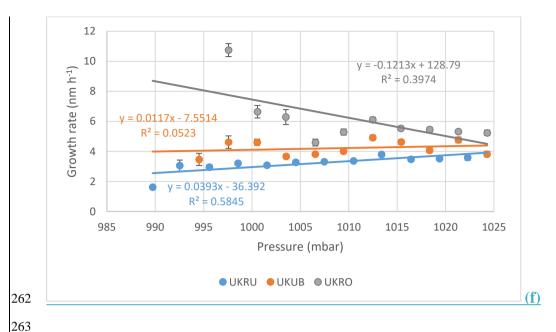




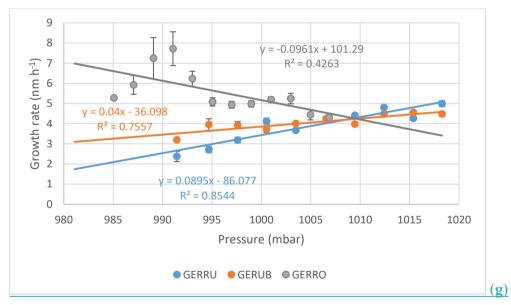


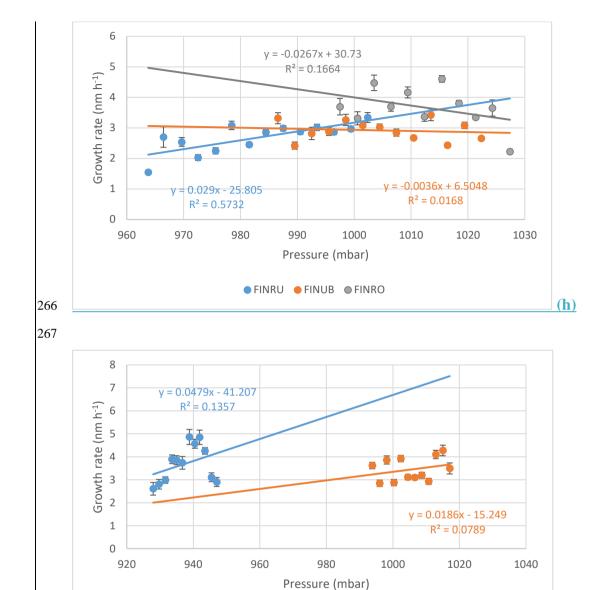










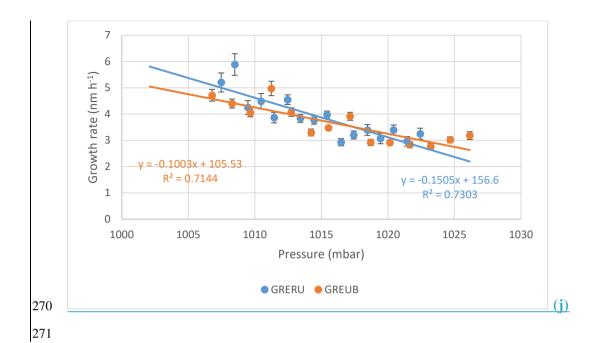


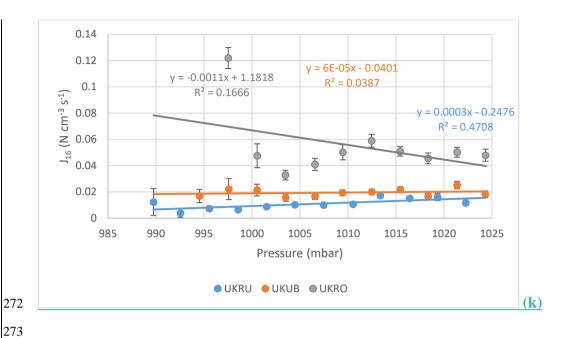
● SPARU ● SPAUB

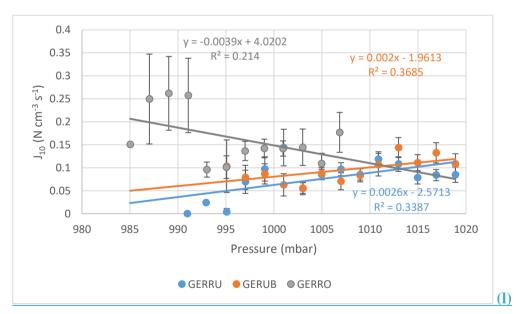
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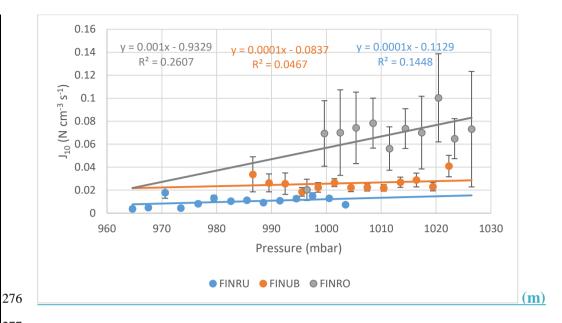
269

**(i)** 

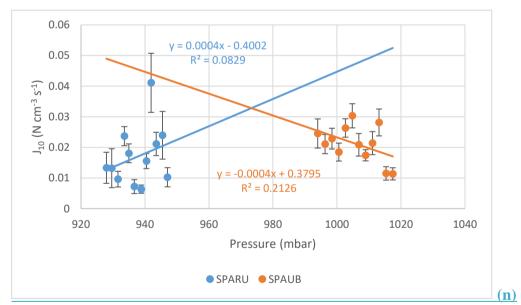


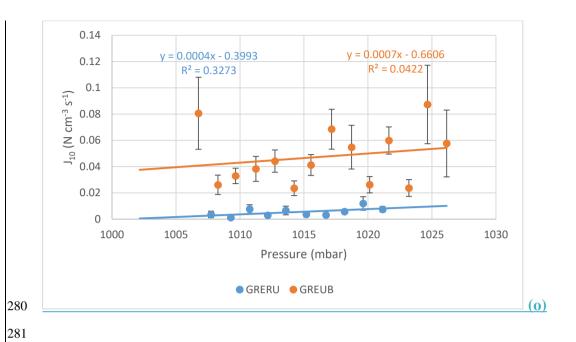




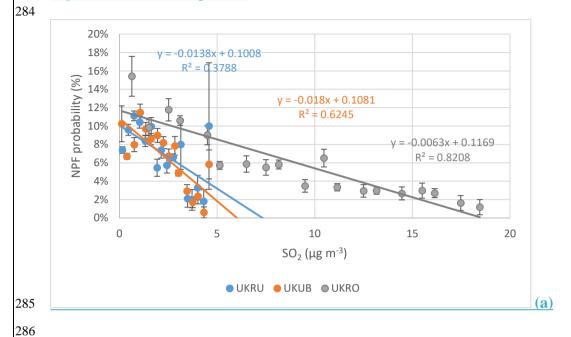




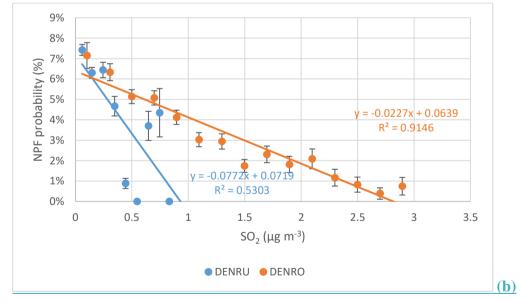


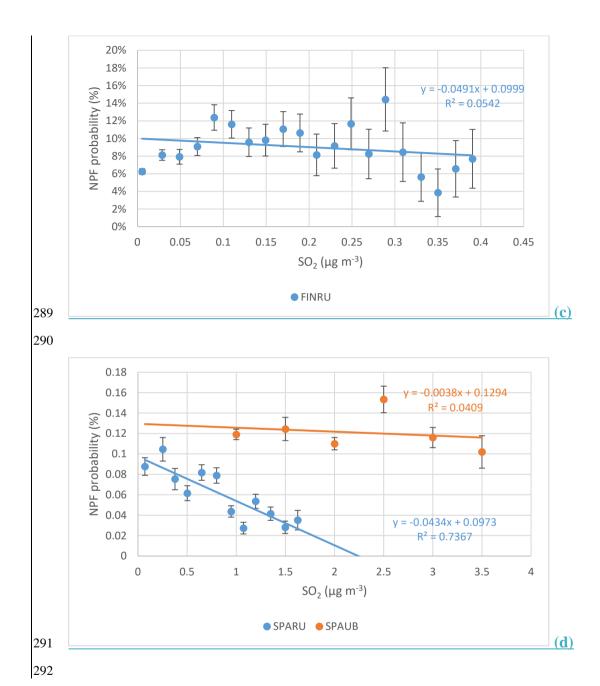


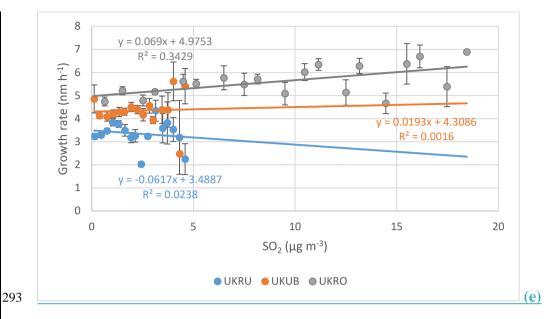
**Figure S6:** Relationship of SO<sub>2</sub> concentration with NPF variables.



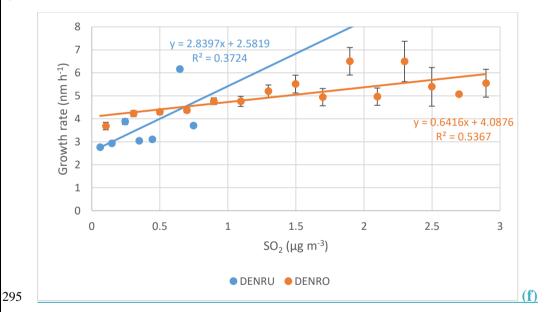


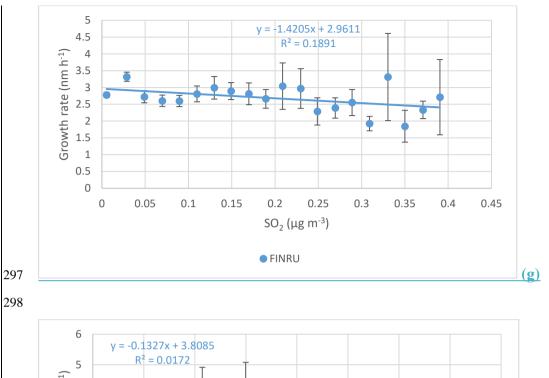


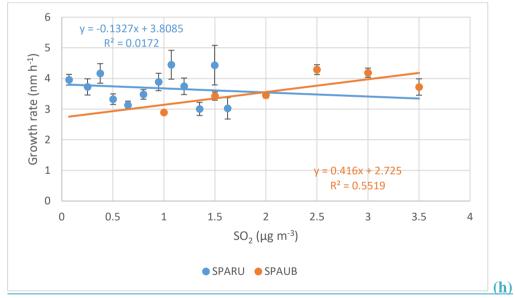


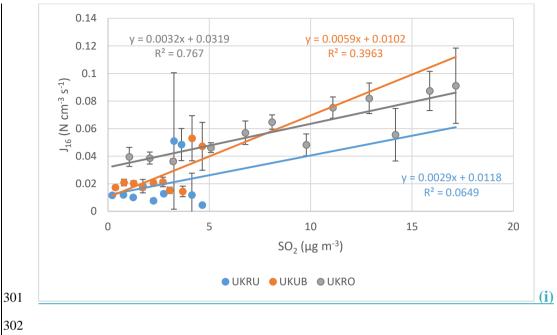


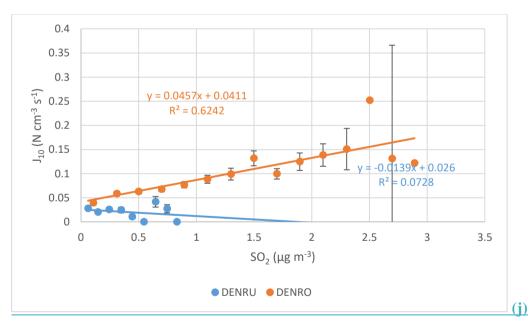


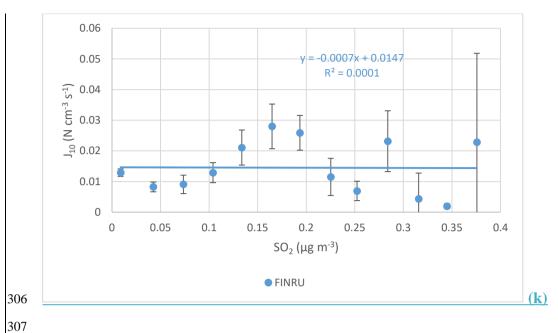


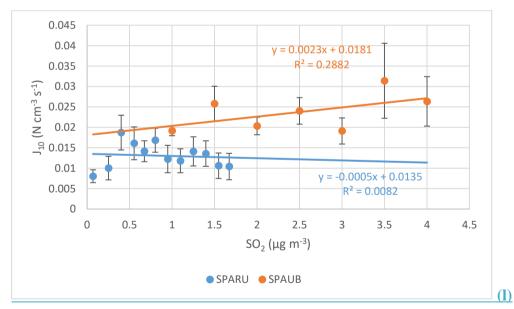




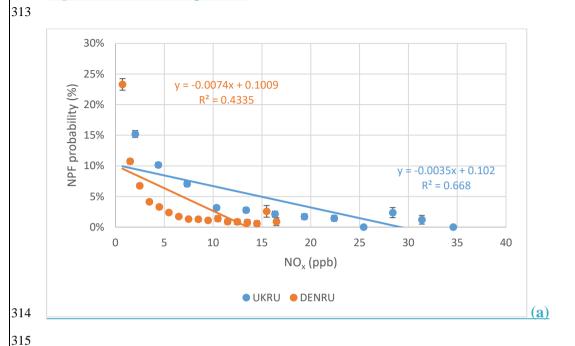




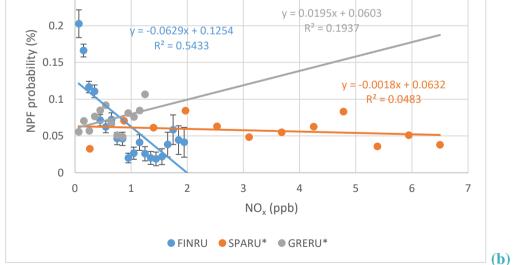




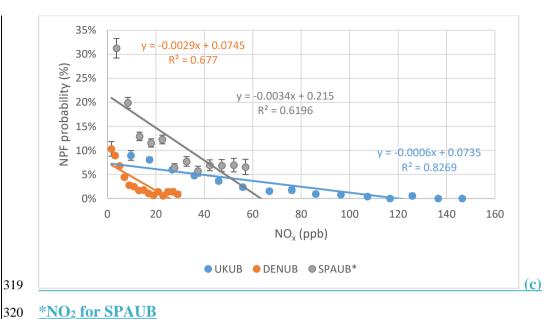
**Figure S7:** Relationship of NO<sub>2</sub> / NO<sub>x</sub> concentration with NPF variables.



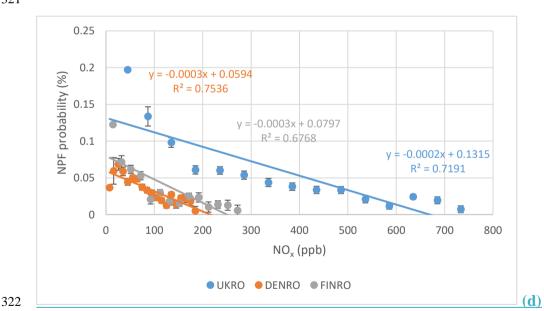


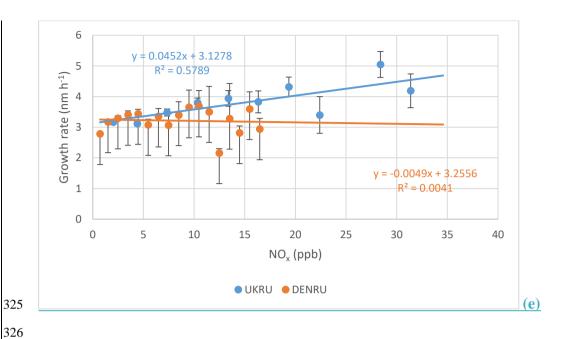


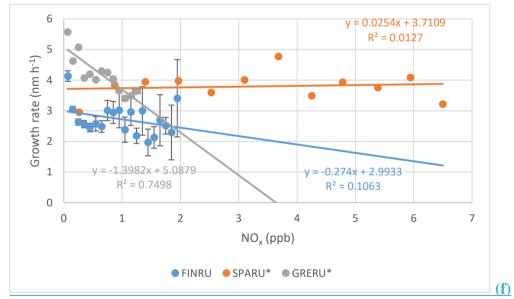
\*NO2 for SPARU and GRERU



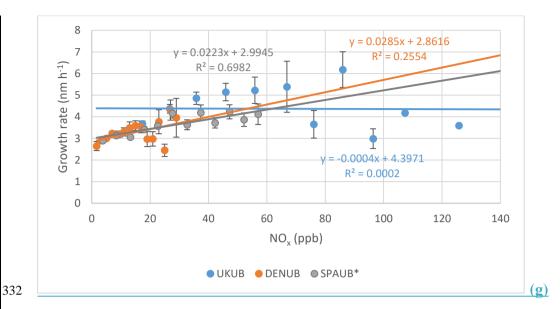
#### \*NO2 for SPAUB



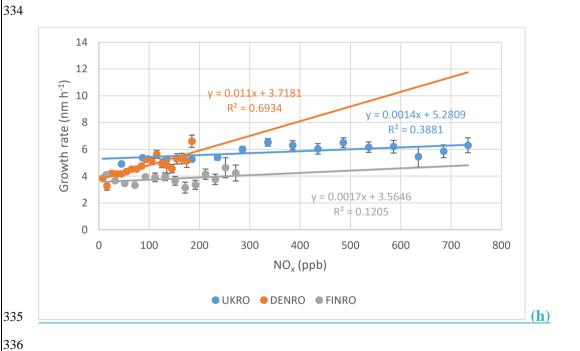


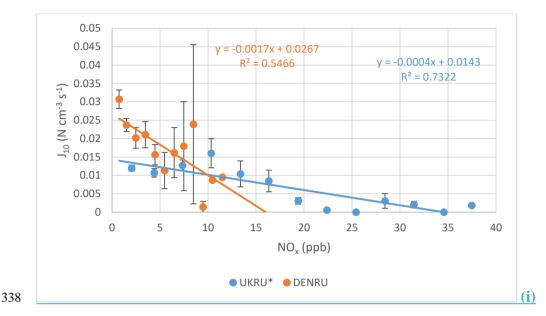


\*NO<sub>2</sub> for SPARU and GRERU

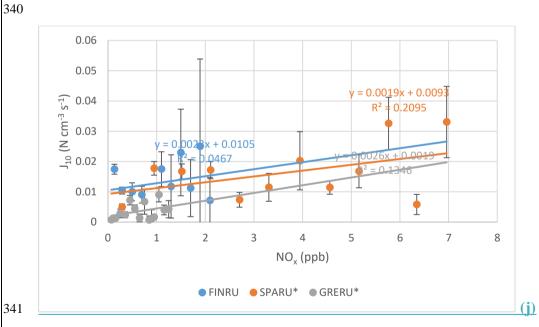


## \*NO<sub>2</sub> for SPAUB

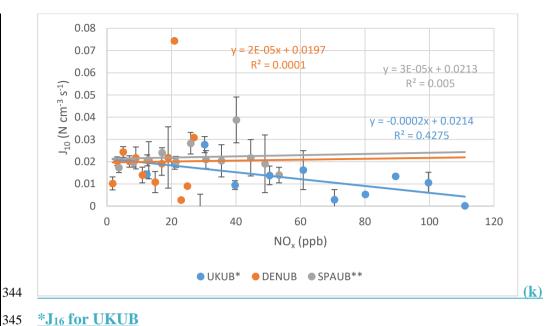




## \*J<sub>16</sub> for UKRU

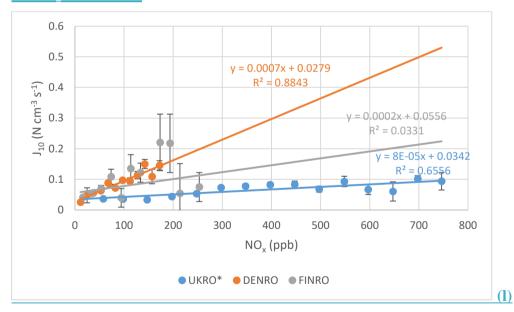


\*NO2 for SPARU and GRERU



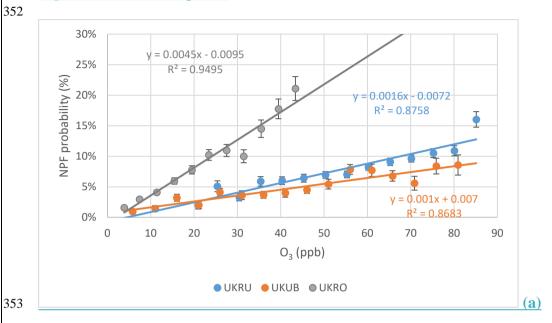
# \*J<sub>16</sub> for UKUB

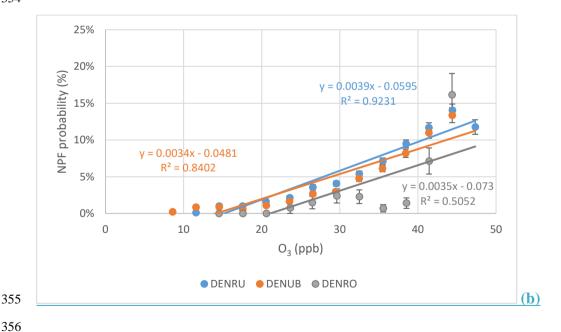
#### \*\* NO2 for SPAUB



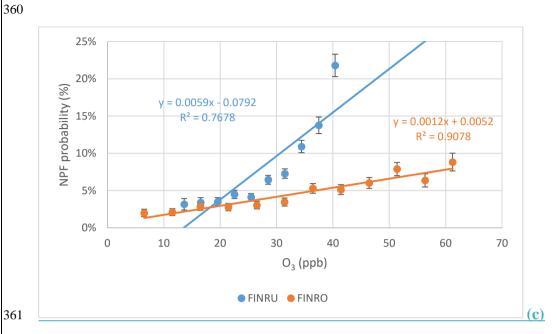
## \*J<sub>16</sub> for UKRO

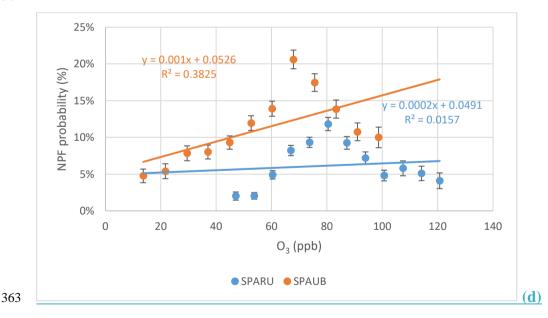
**Figure S8:** Relationship of O<sub>3</sub> concentration with NPF variables.

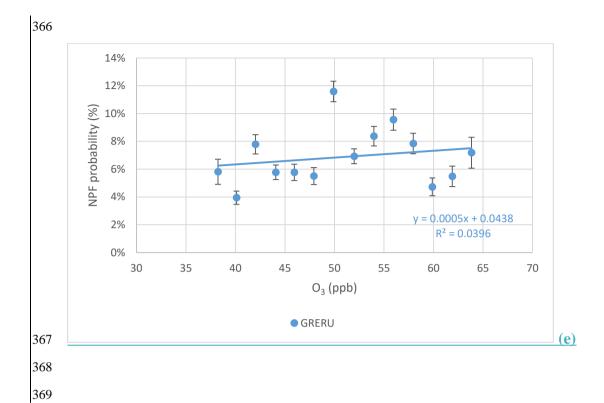


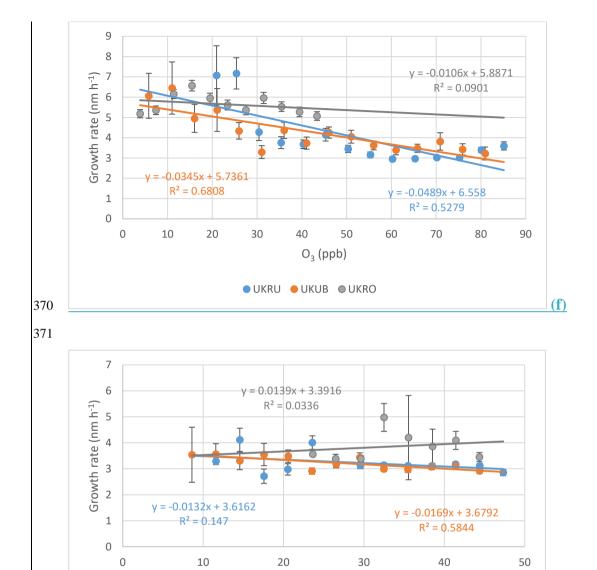












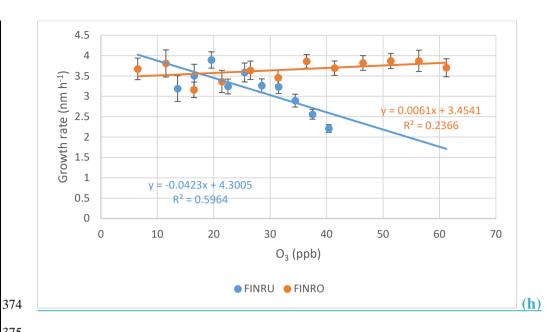
O<sub>3</sub> (ppb)

● DENRU ● DENUB ● DENRO

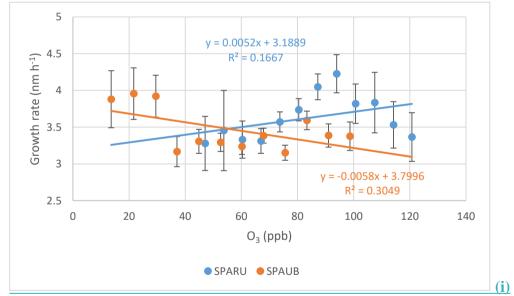
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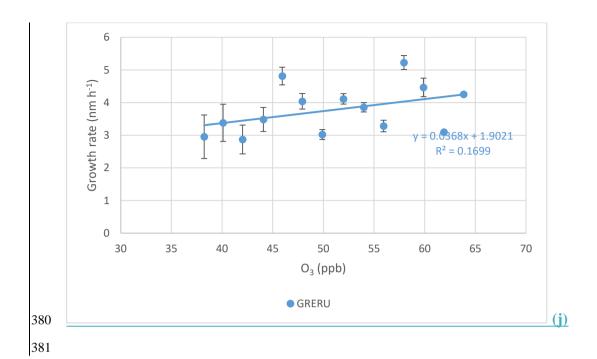
373

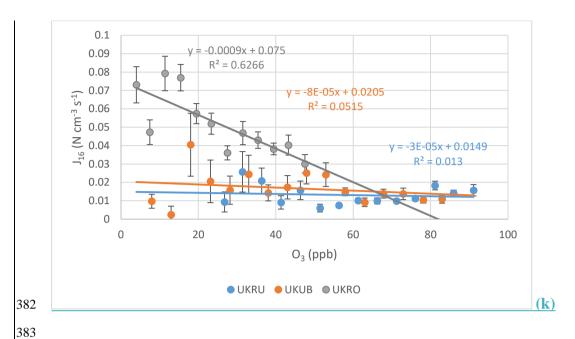
**(g)** 

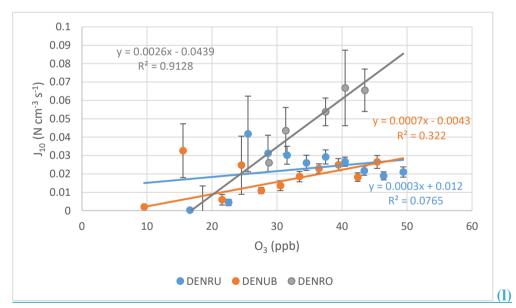


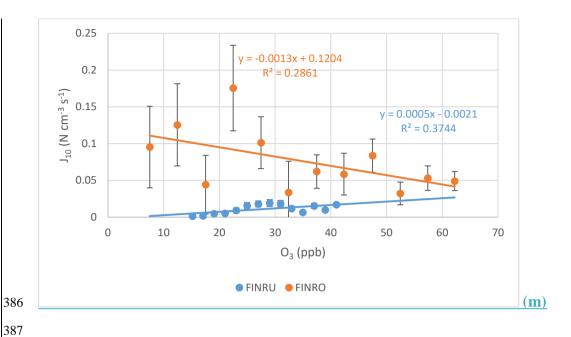


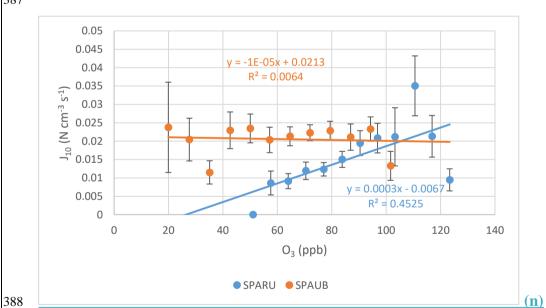












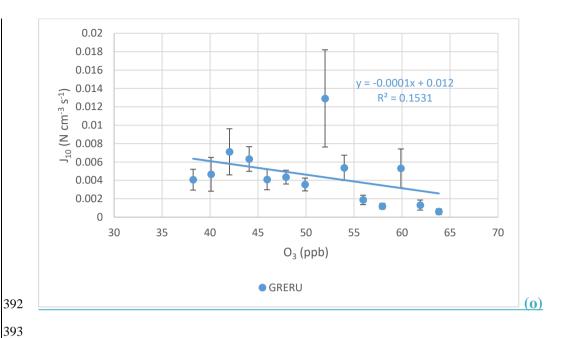
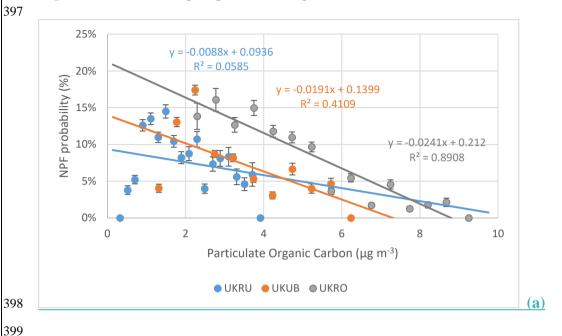
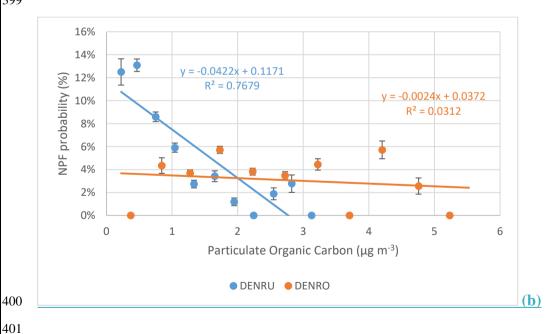
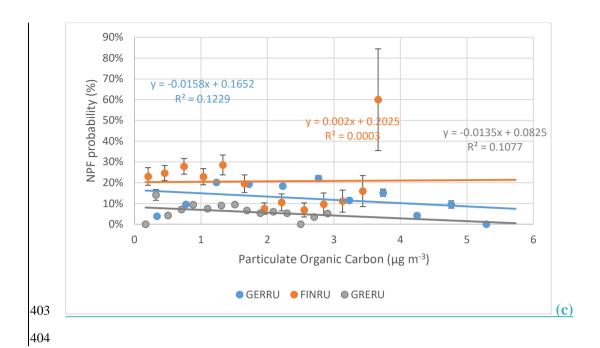
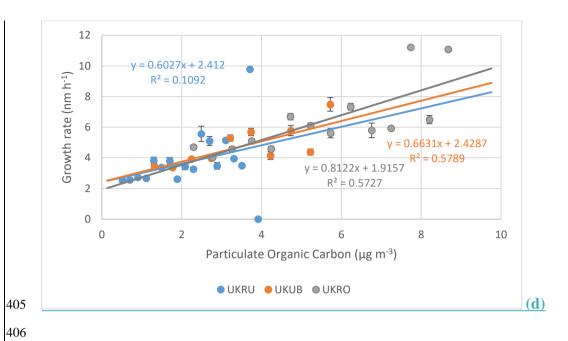


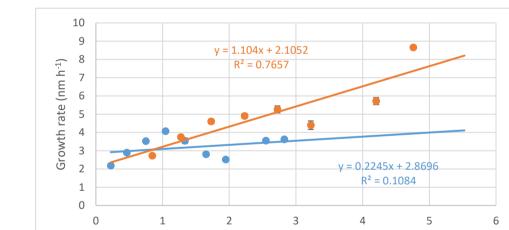
Figure S9: Relationship of particulate organic carbon concentration with NPF variables.











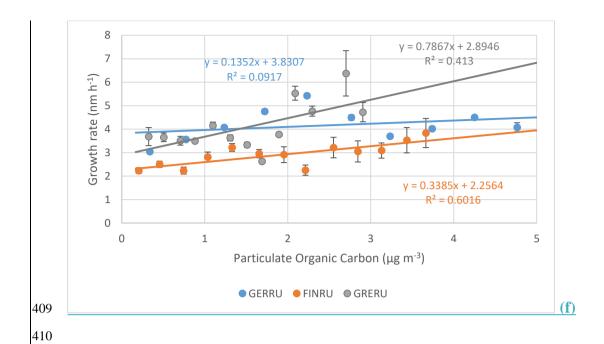
Particulate Organic Carbon (μg m<sup>-3</sup>)

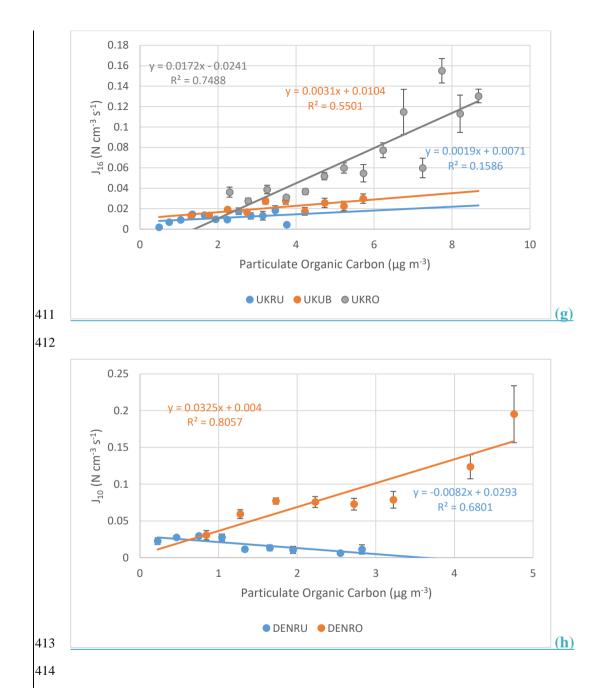
DENRUDENRO

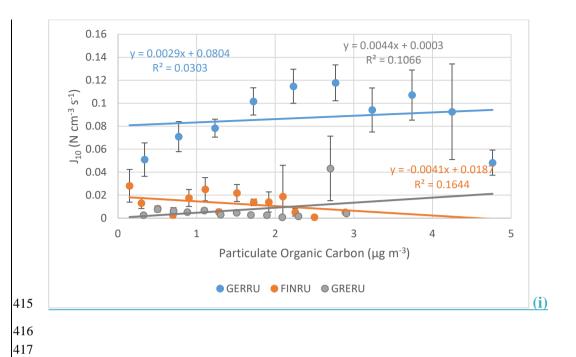
407

408

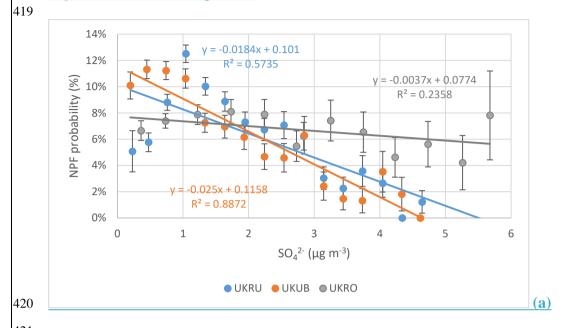
**(e)** 



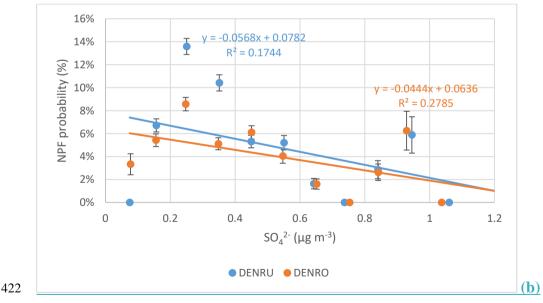


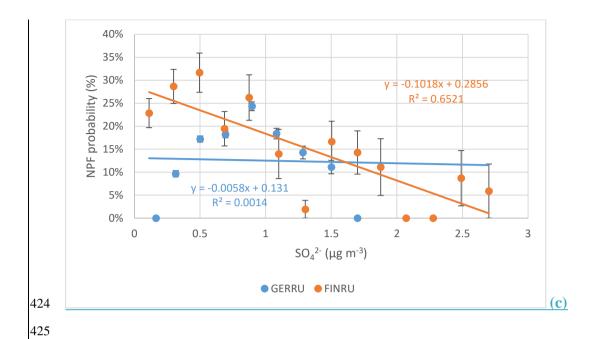


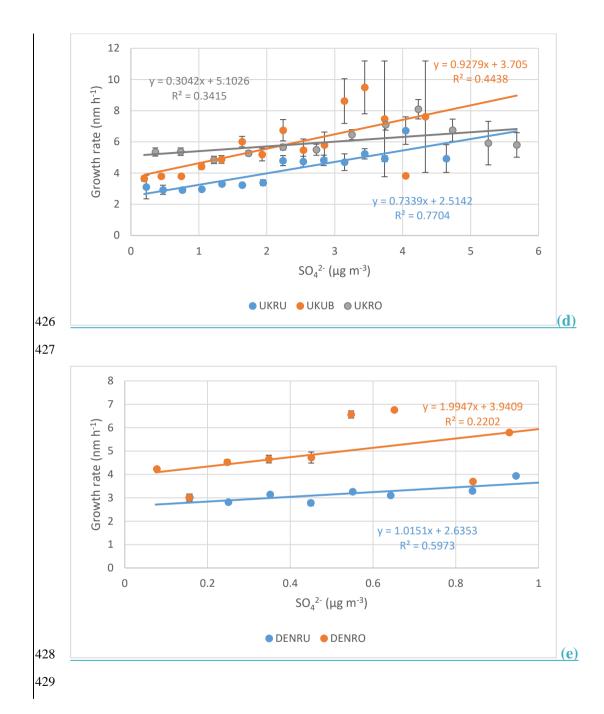
**Figure S10:** Relationship of SO<sub>4</sub><sup>2-</sup> concentration with NPF variables.

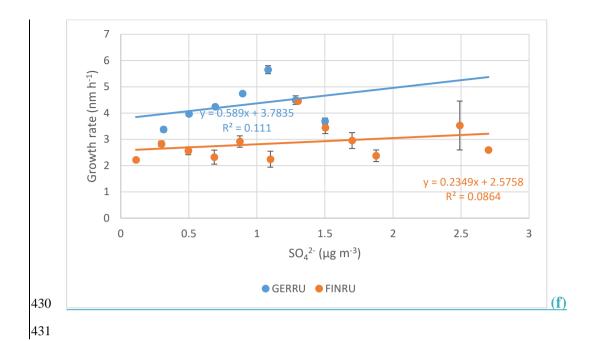


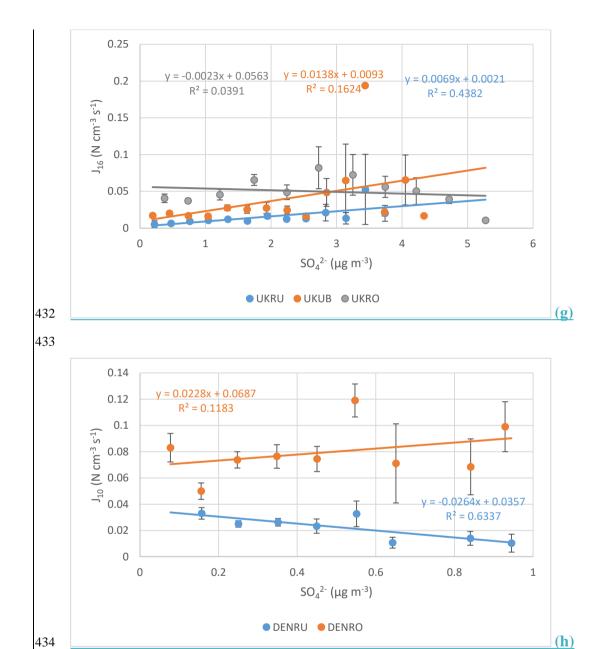












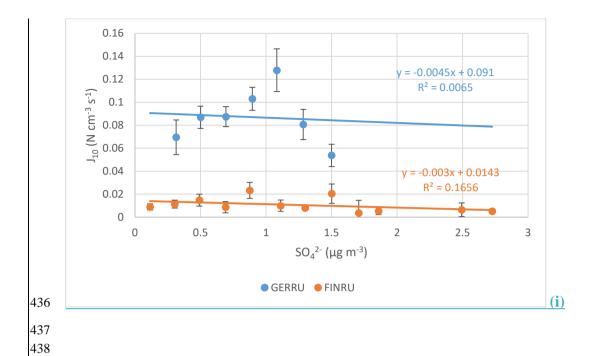
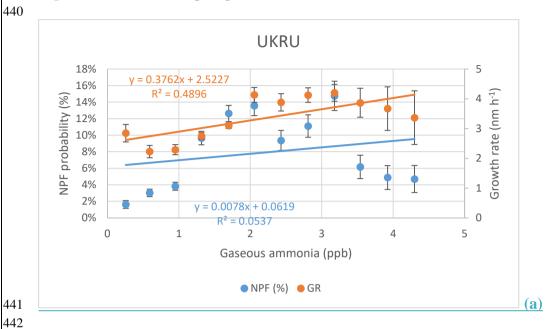
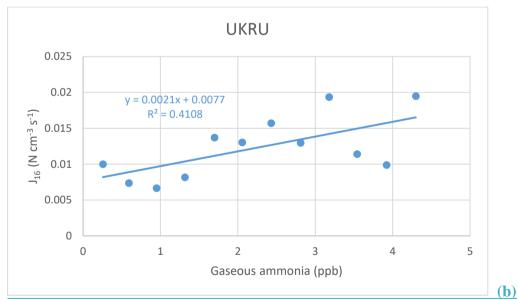
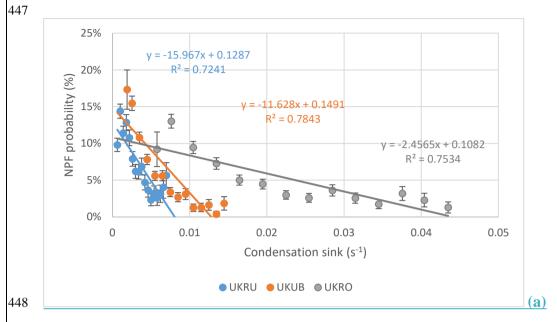


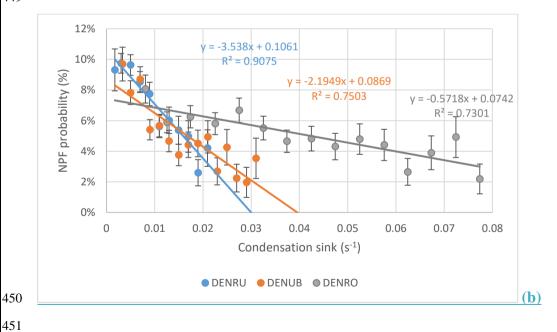
Figure S11: Relationship of gaseous ammonia concentration with NPF variables.

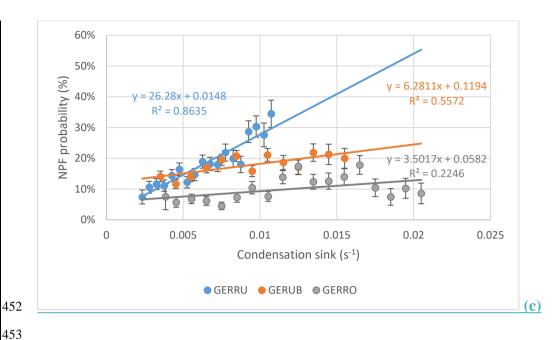


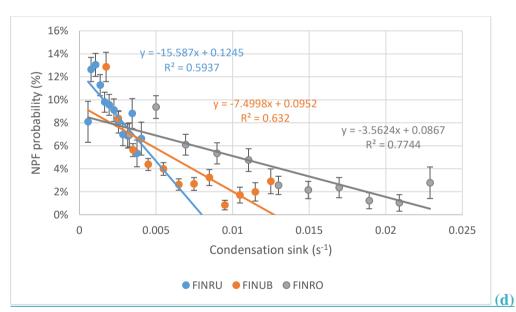


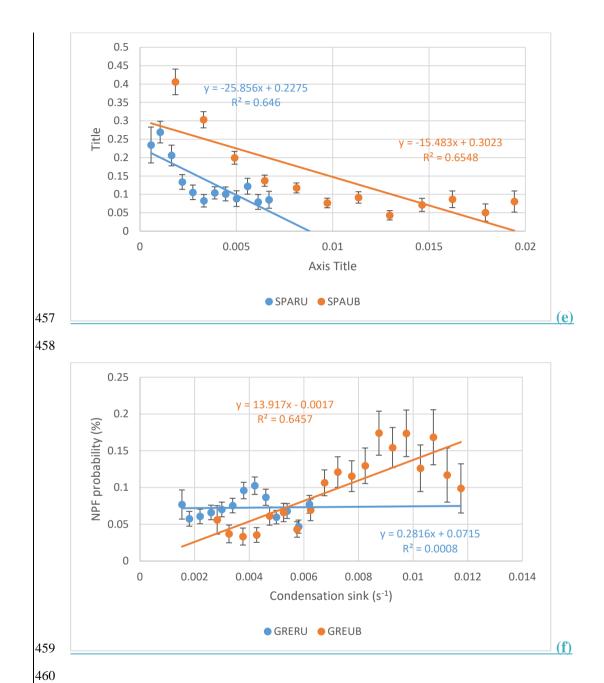
**Figure S12:** Relationship of the condensation sink with NPF variables.

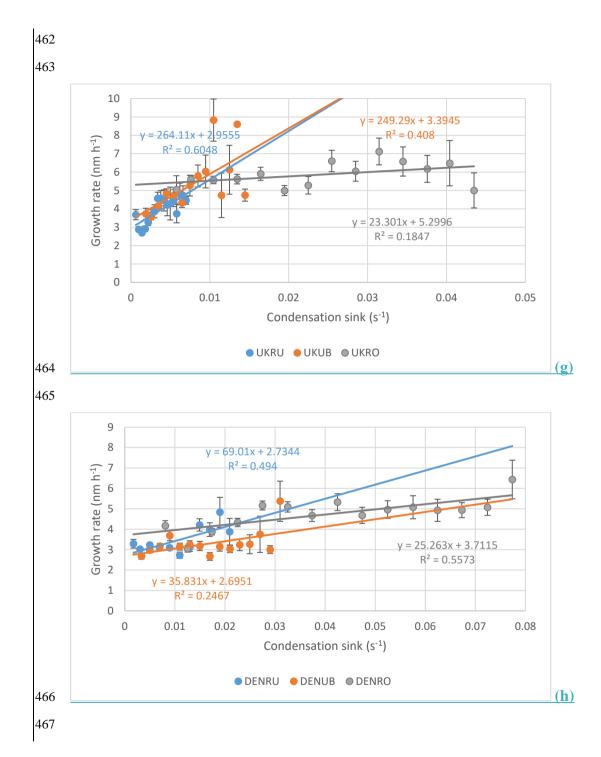


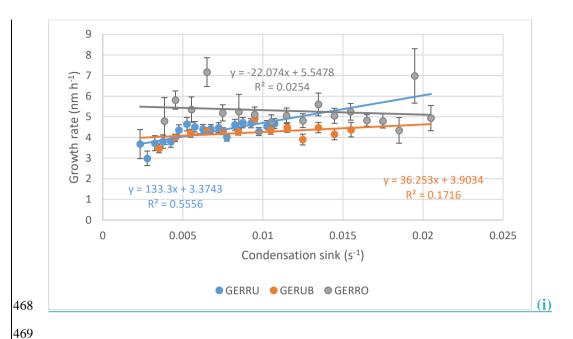


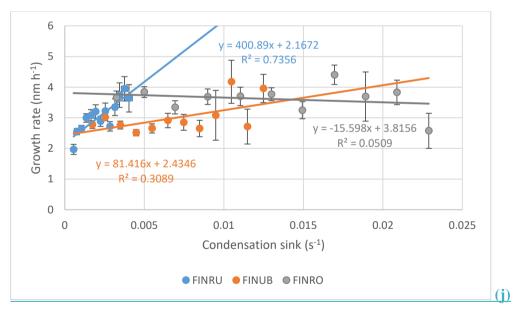


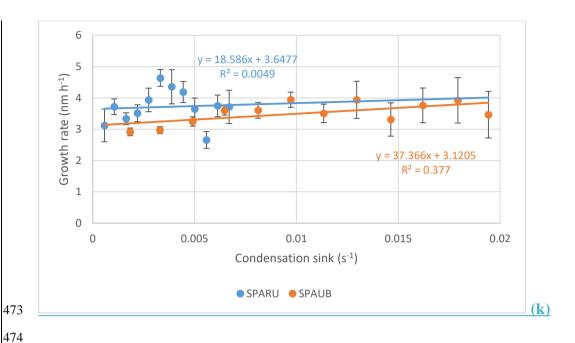


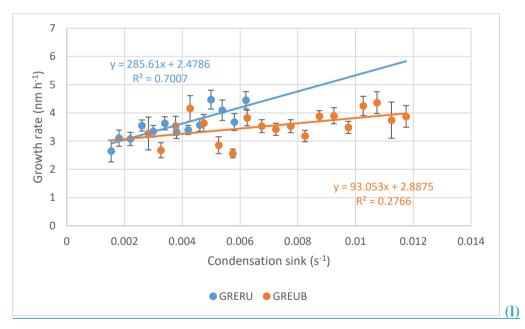


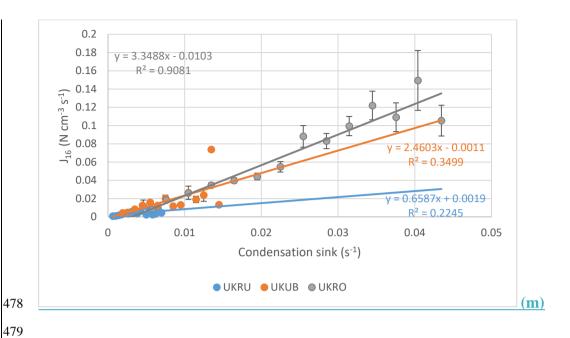


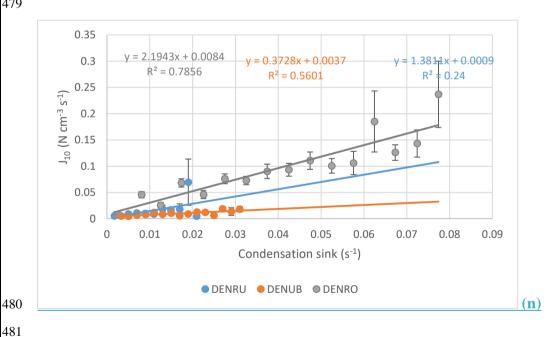


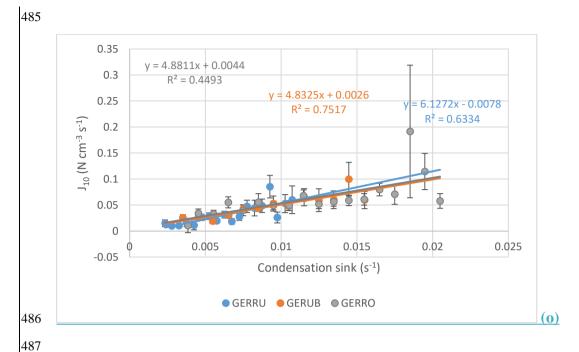


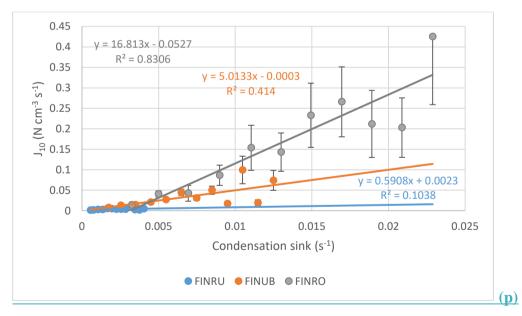












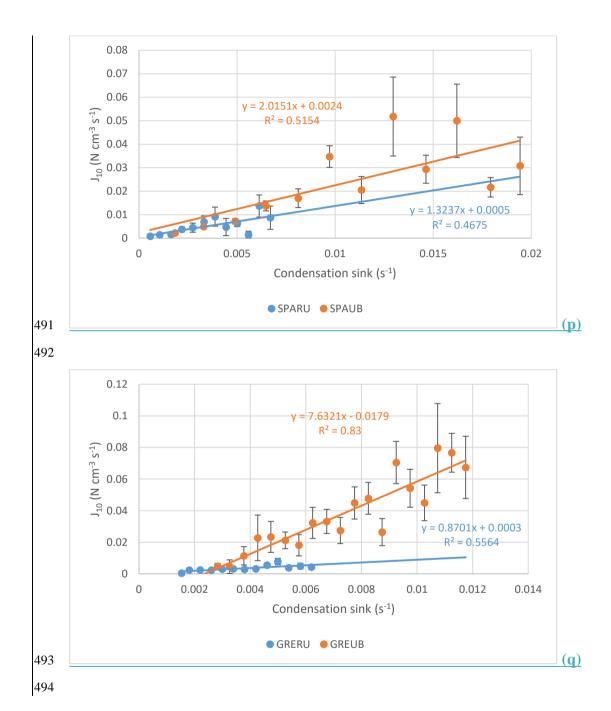


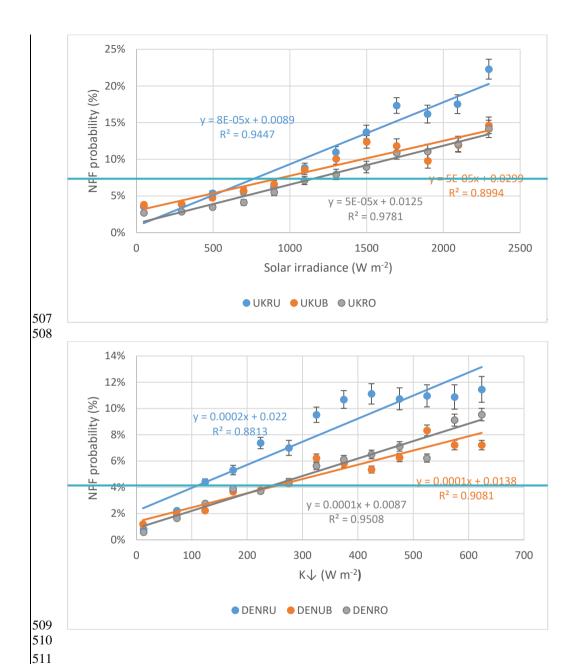
Figure S13: Relationship of average temperature and normalised gradients a<sub>N</sub>\* for all but the Finnish 495 496 sites. 497 0.15 -UKRU UKUB **UKRO** DENRU **DENUB** Normalised gradient  $a_N^*$  ( $^{\circ}C^{-1}$ ) 0.10 **SPARU SPAUB** GRERU 0.05  $R^2 = 0.617$ GREUB 0.00 -0.05 10 14 12 16 18 20 8 Average T (°C) 498 499

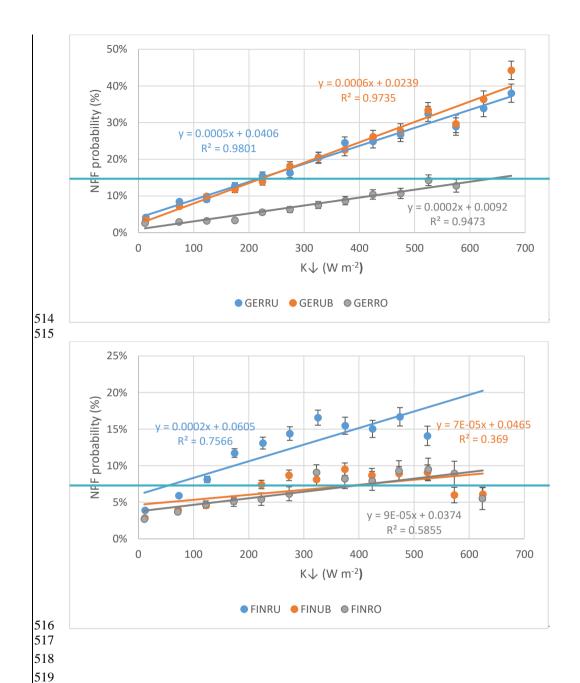
## Figure S14: Seasonal variation of NPF events

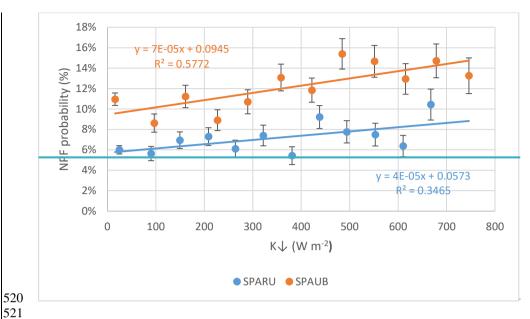
Fraction of NPF events (%) SPARIJAJB JRRI JRJB JRRO DETRU DETUB DETRO GERRY GERIE FITRE FITTIBED GRERU GREUB

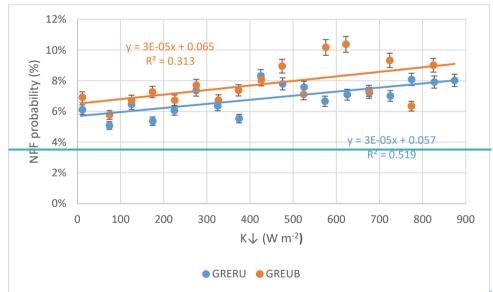
**Figure S1:** Relation of meteorological and atmospheric variables with NPF variables for all sites of the present study.

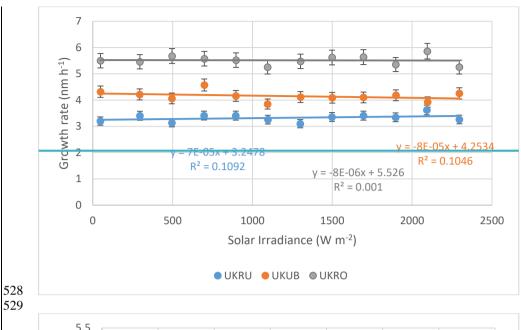
■Winter ■Spring ■Summer ■Autumn

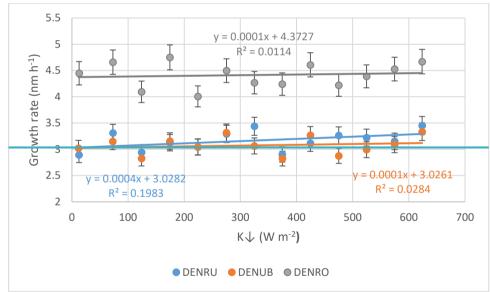


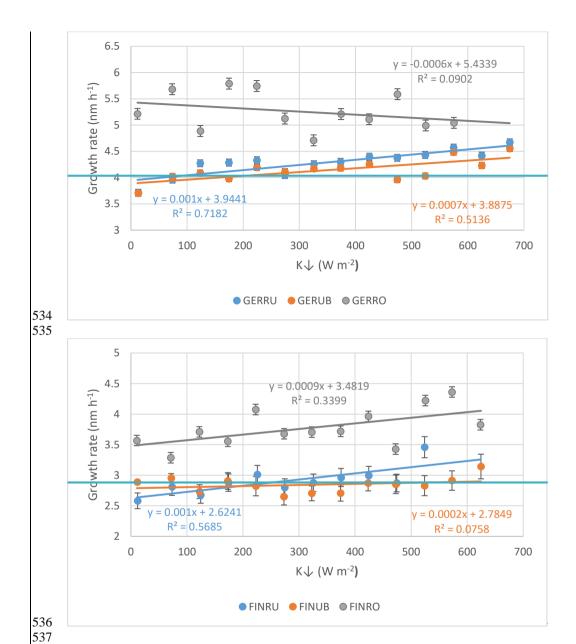


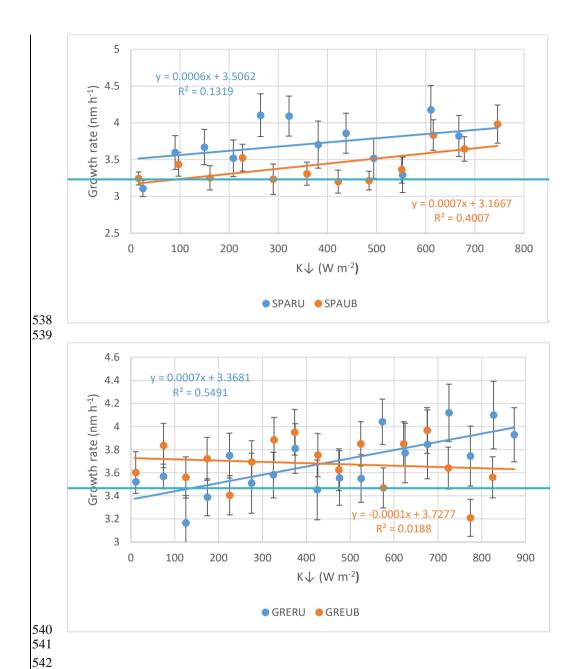


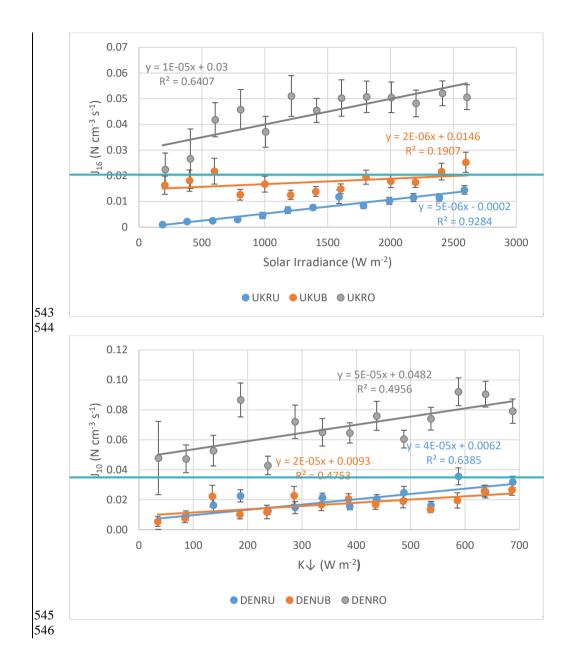


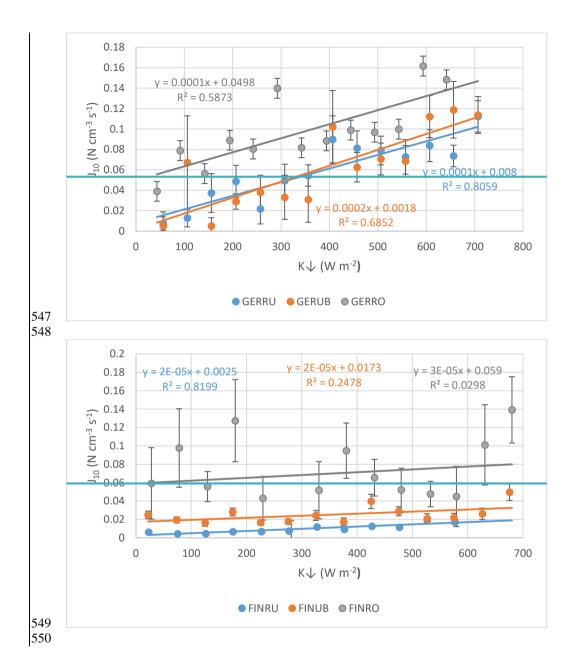


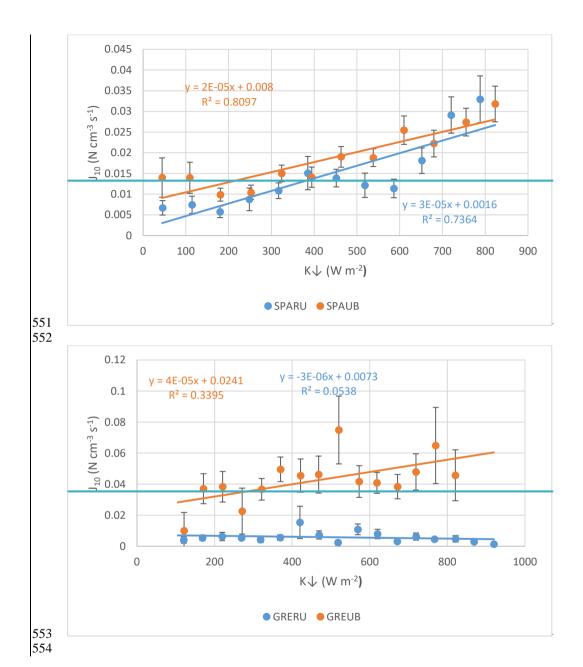


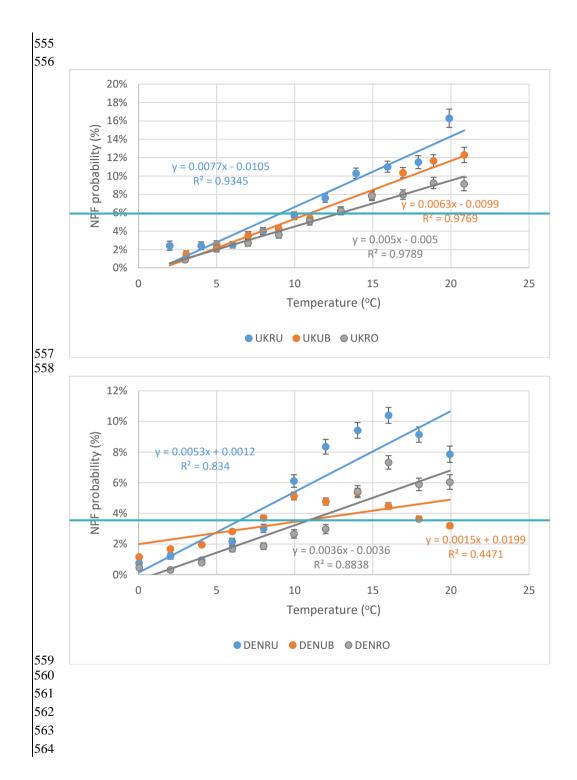


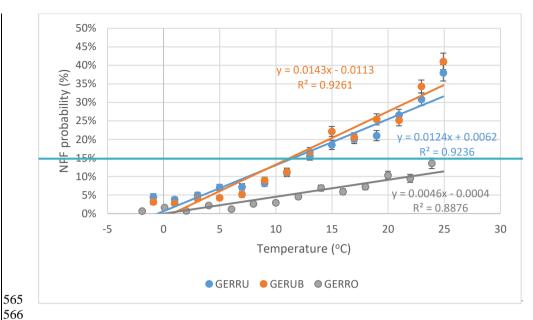


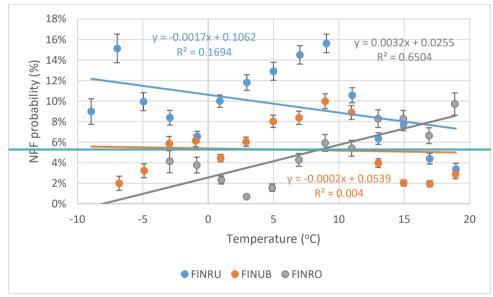


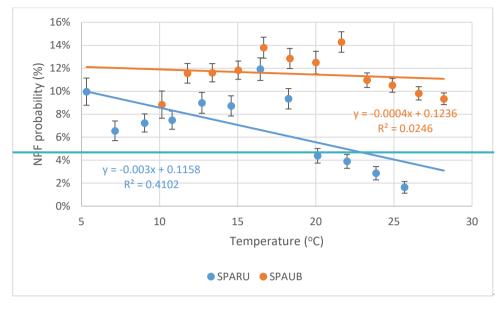


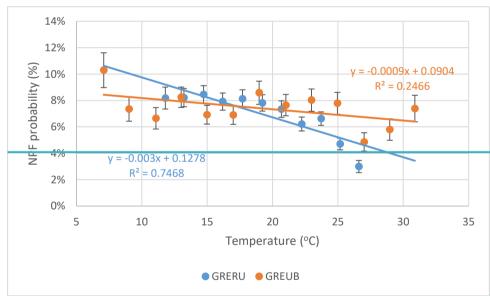


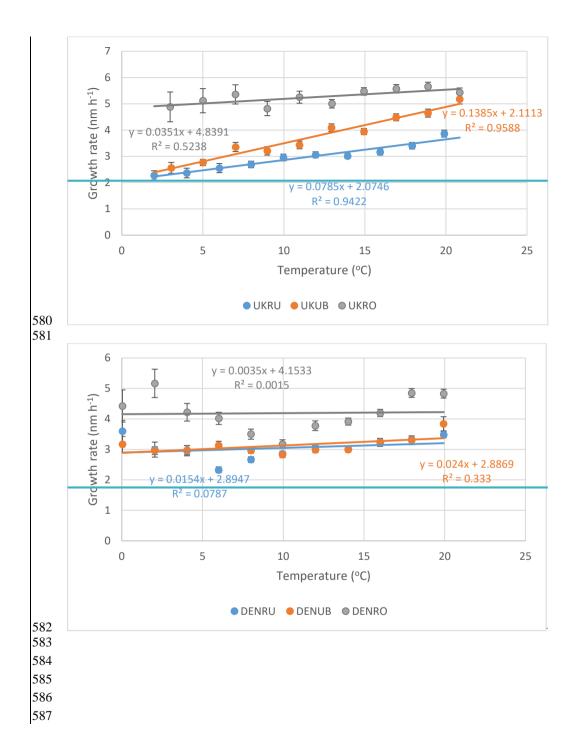


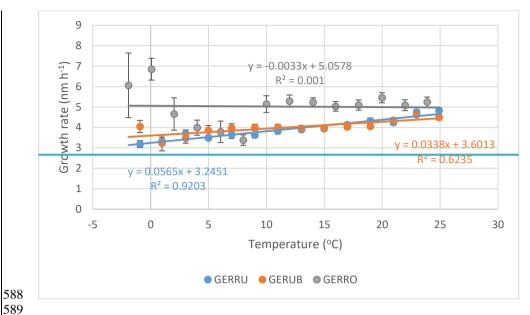


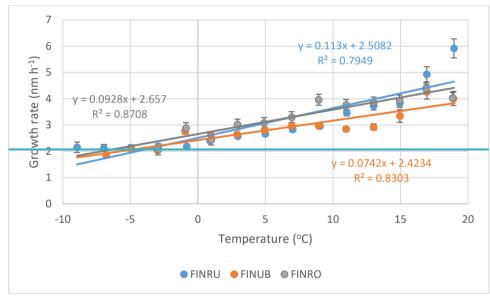


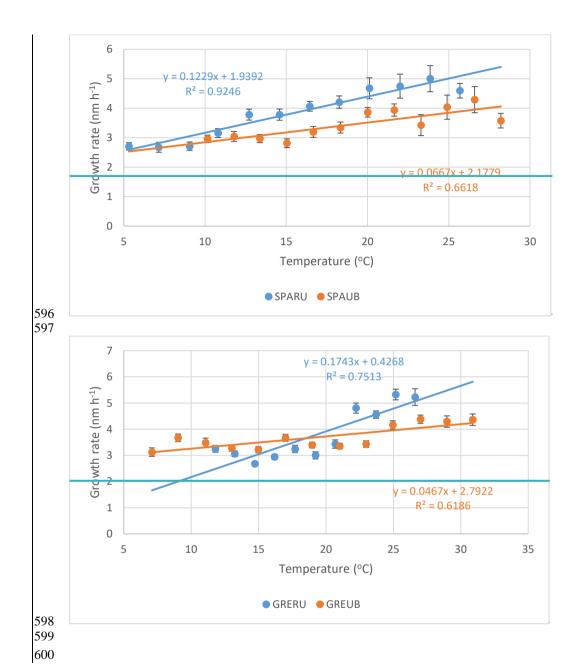


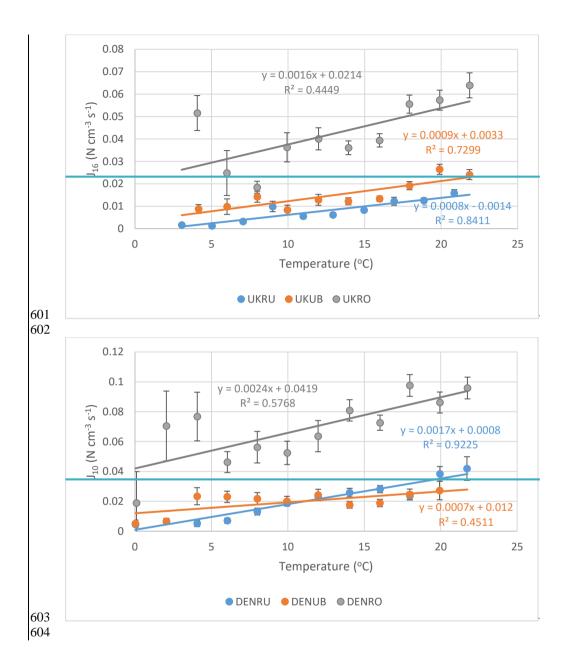


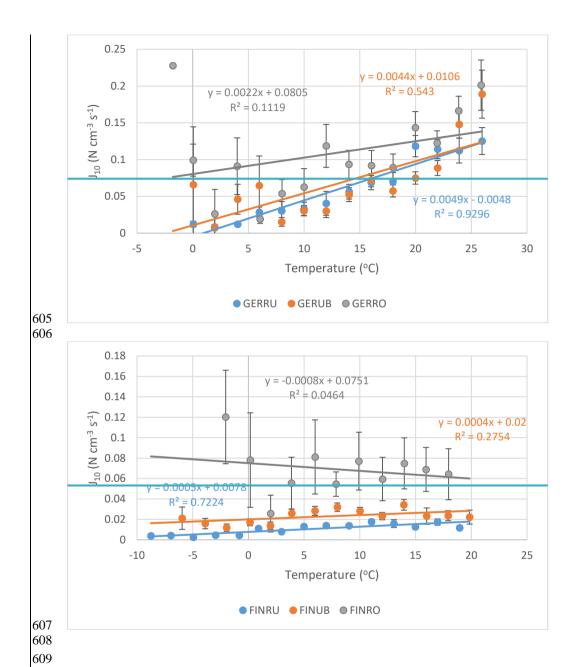


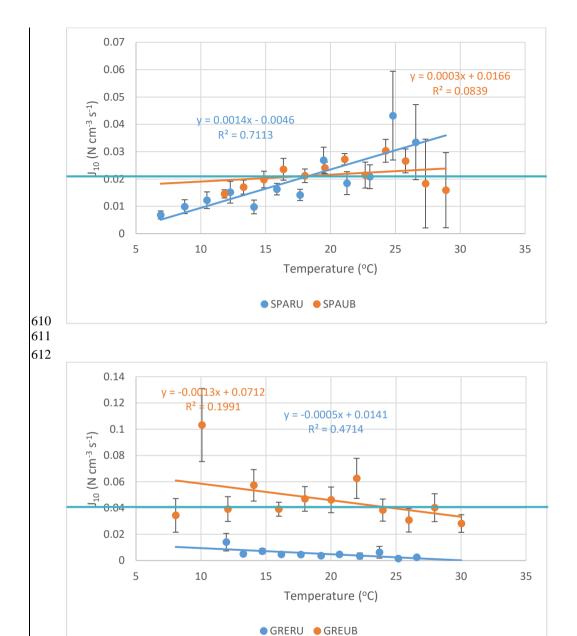


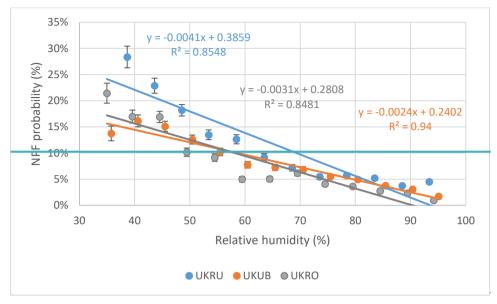


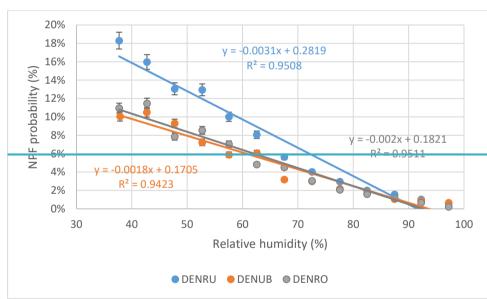


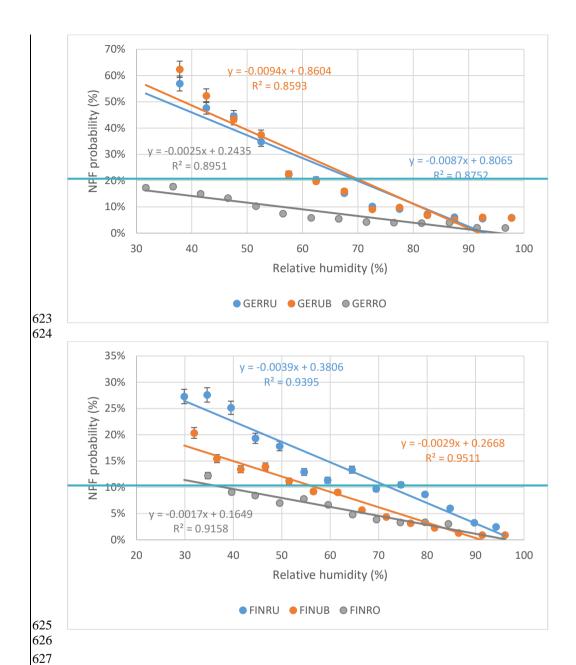


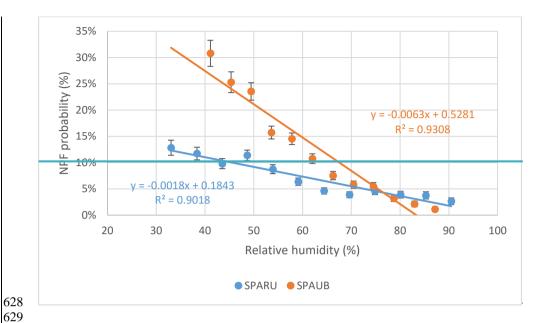


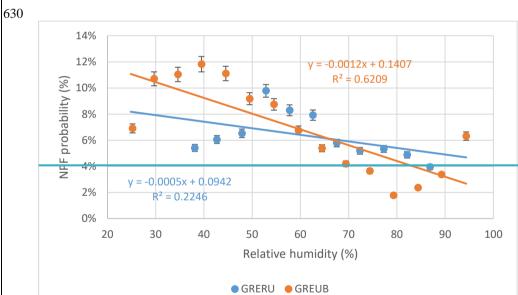


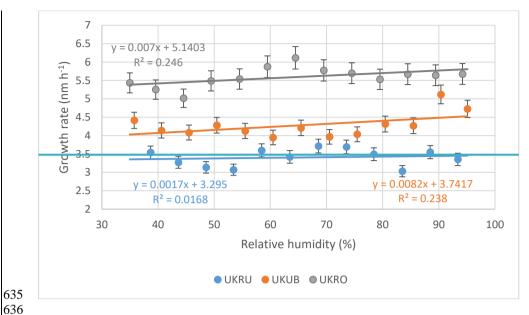


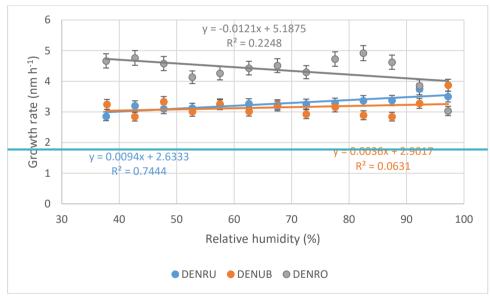


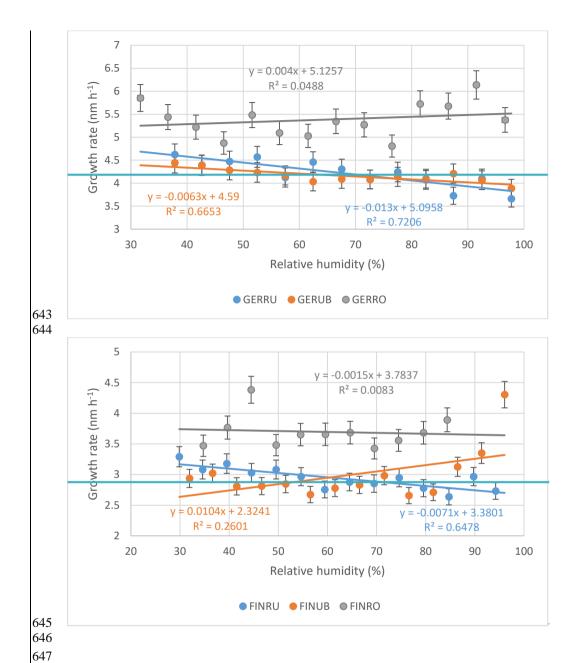


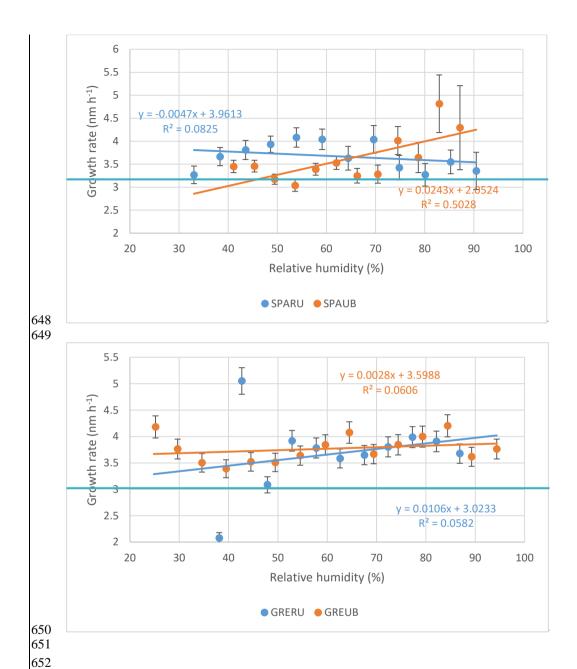


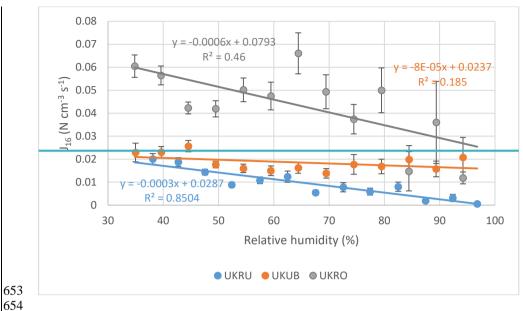


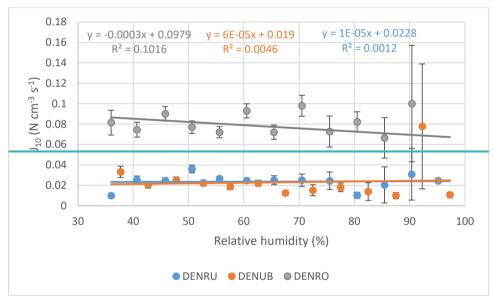


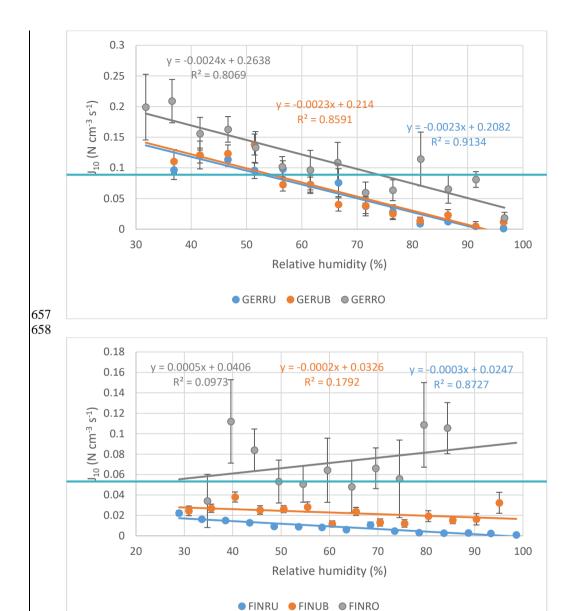


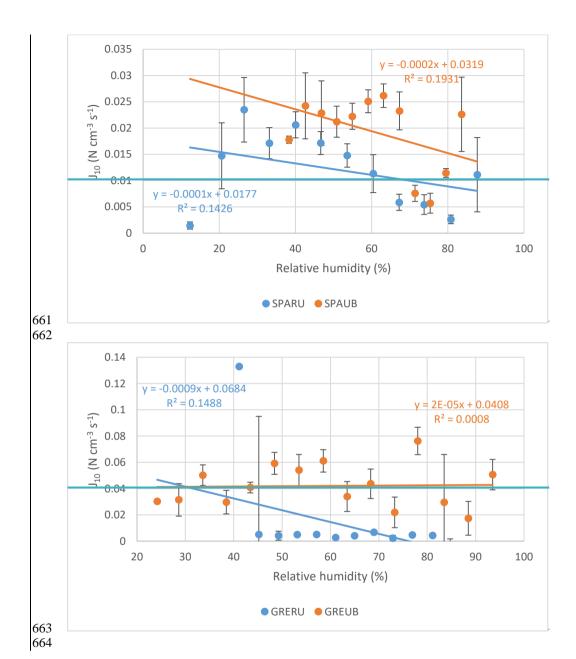


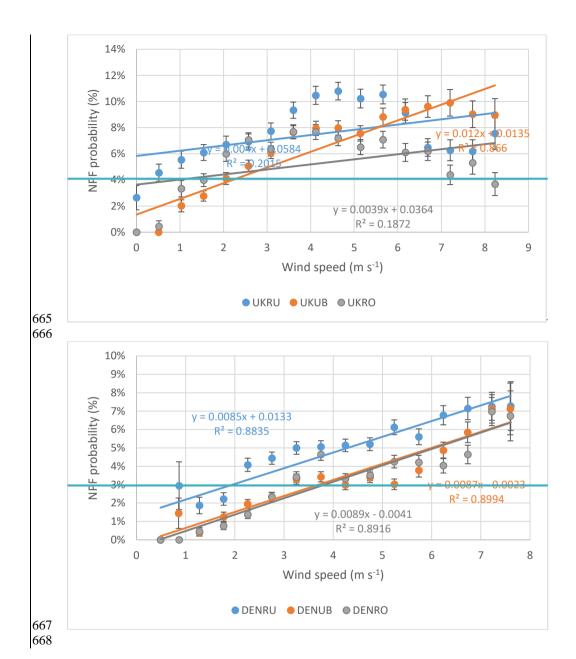


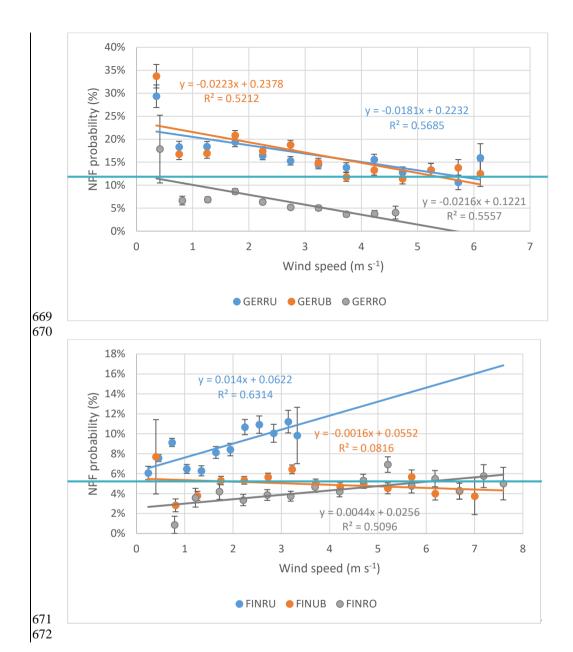


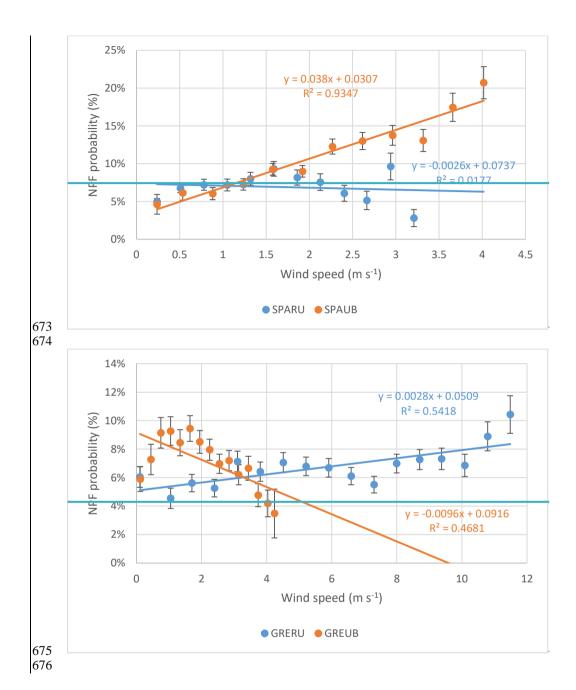


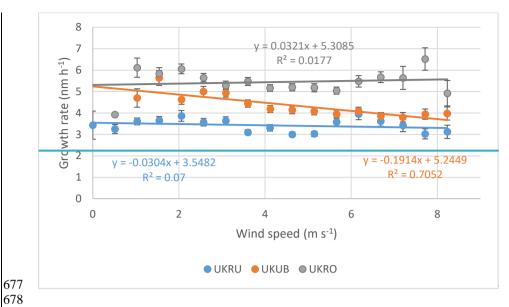


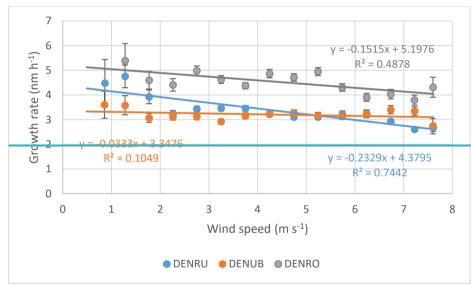


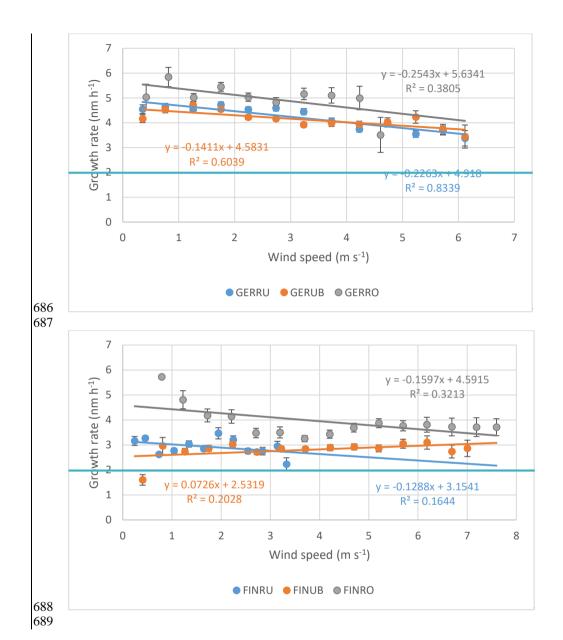


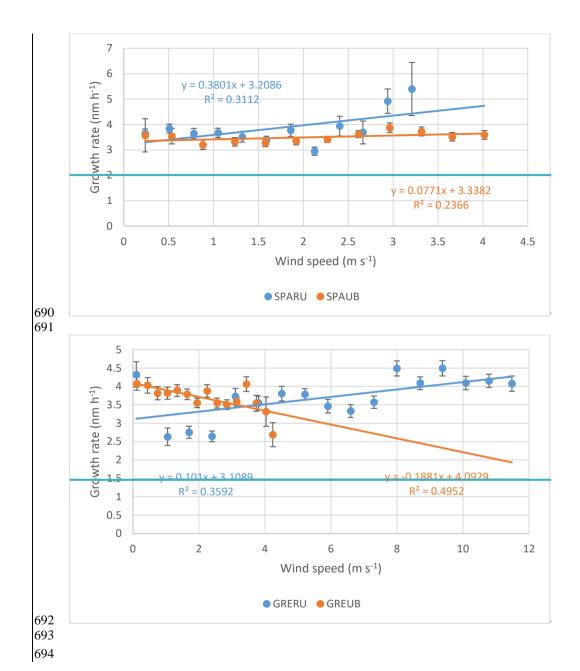


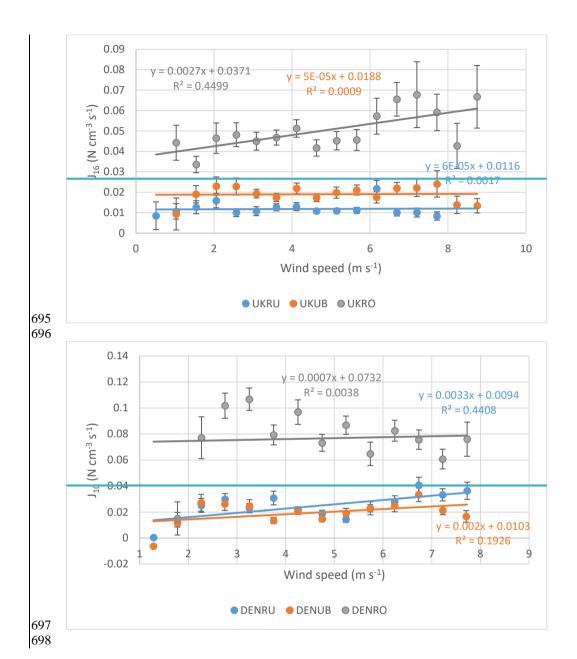


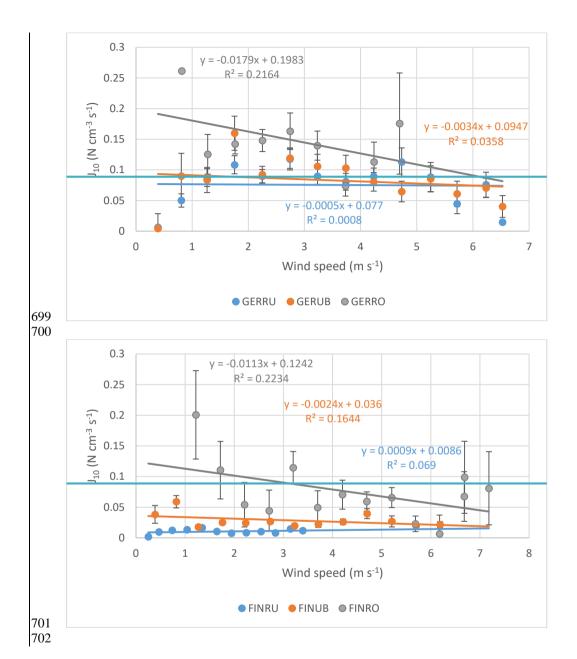


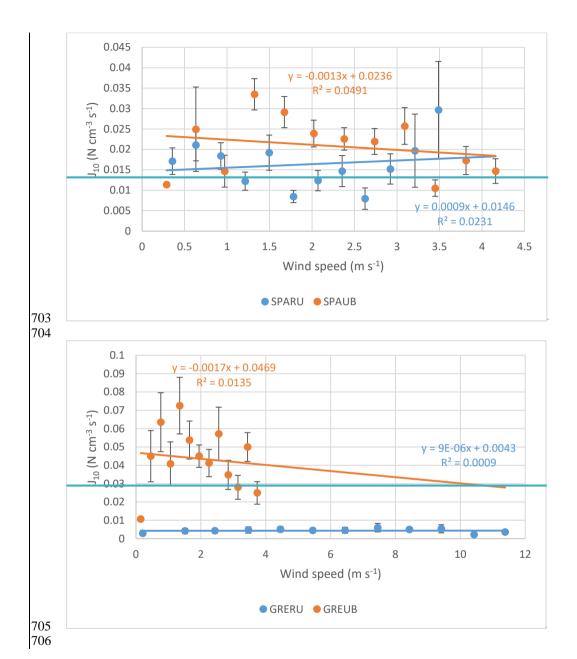


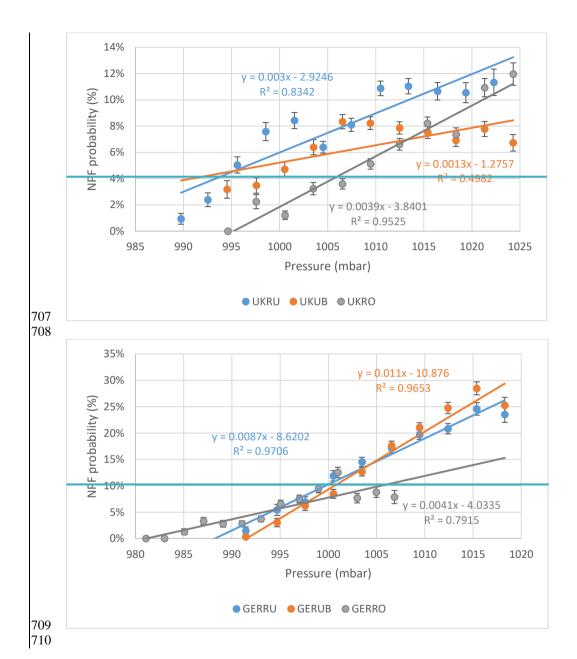


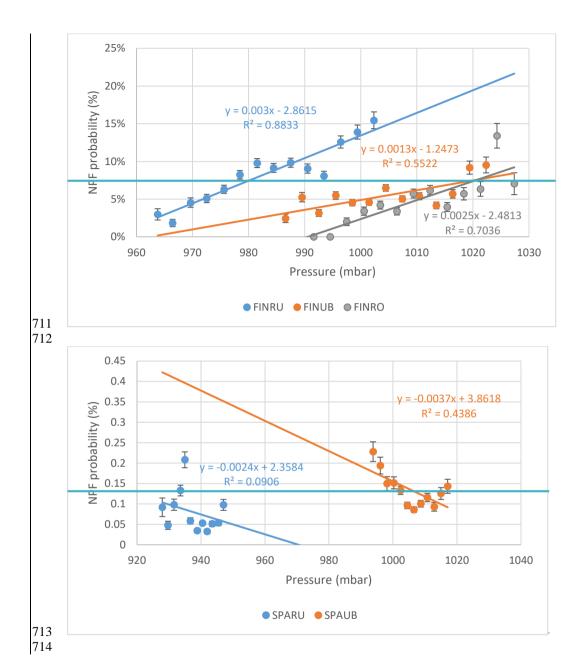


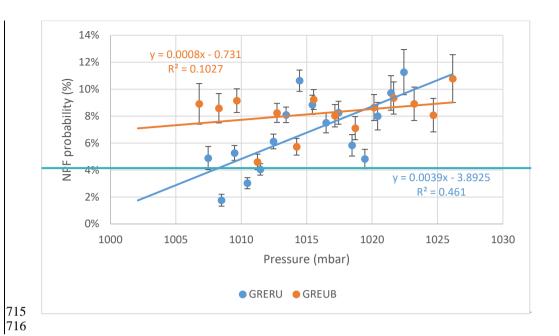


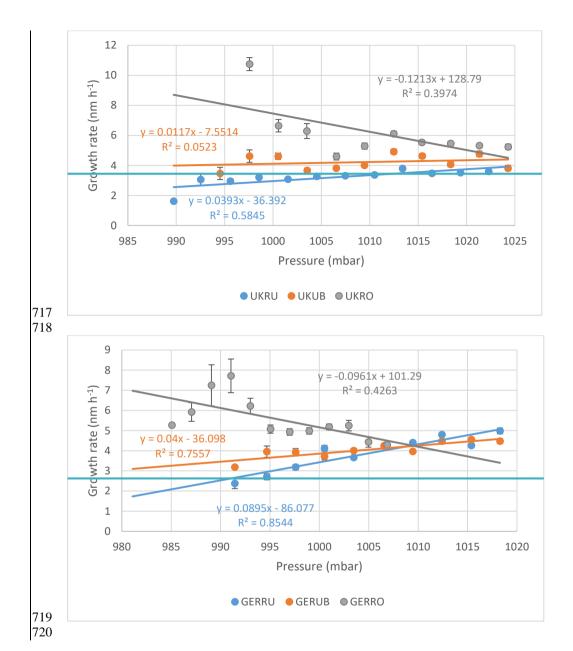


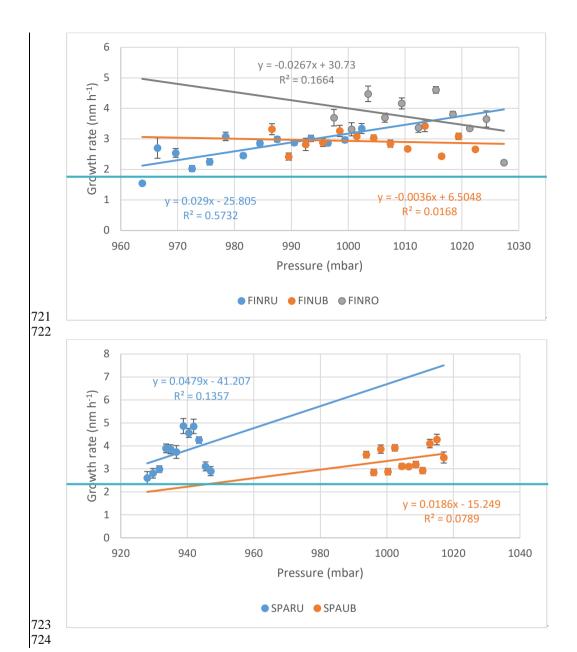


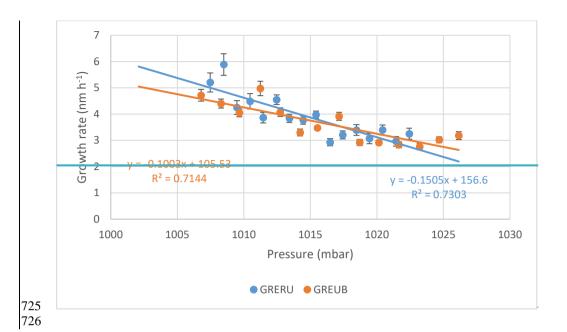


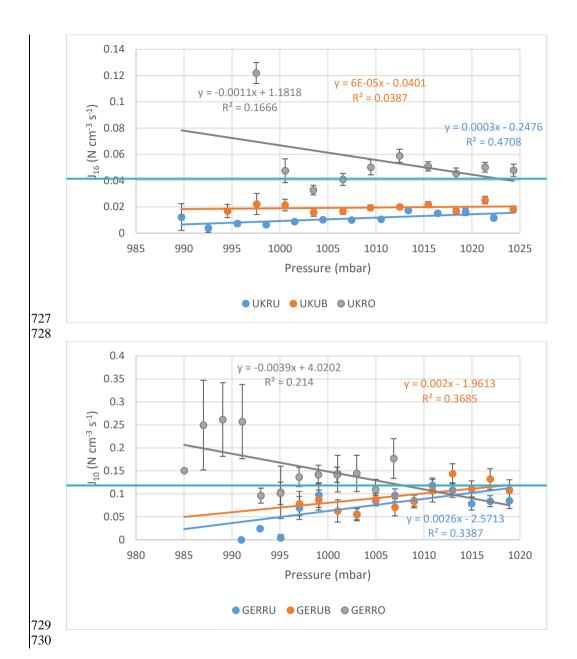


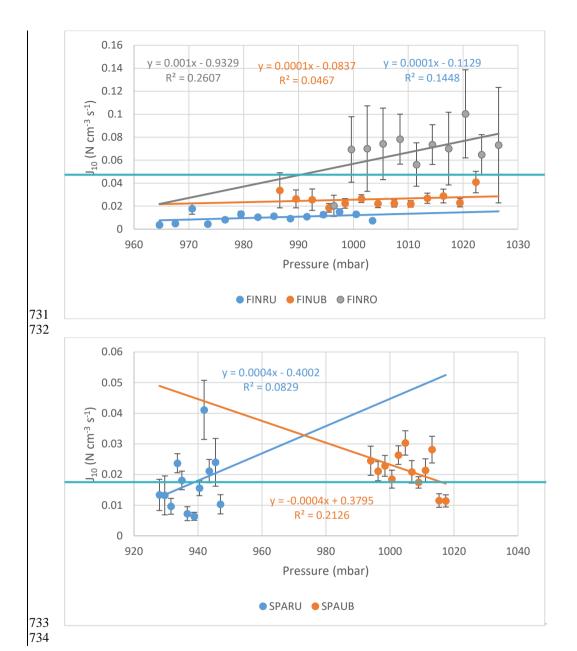


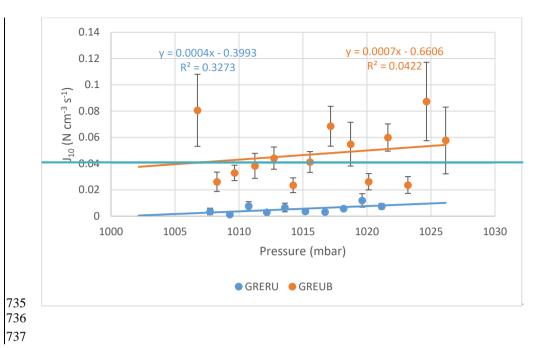


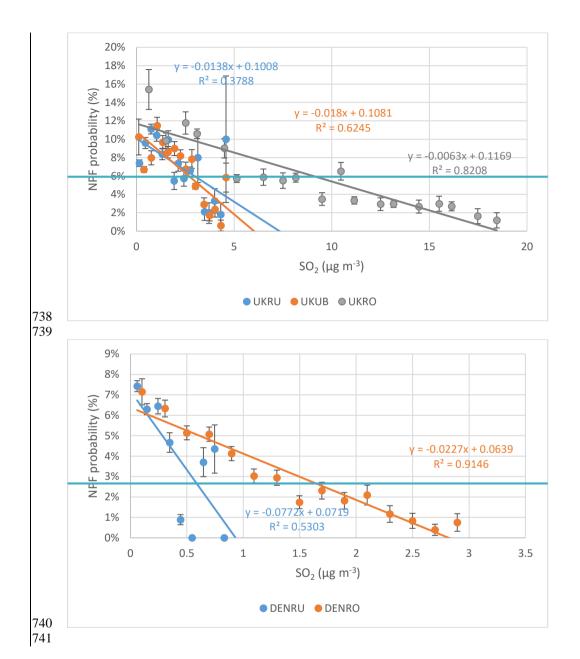


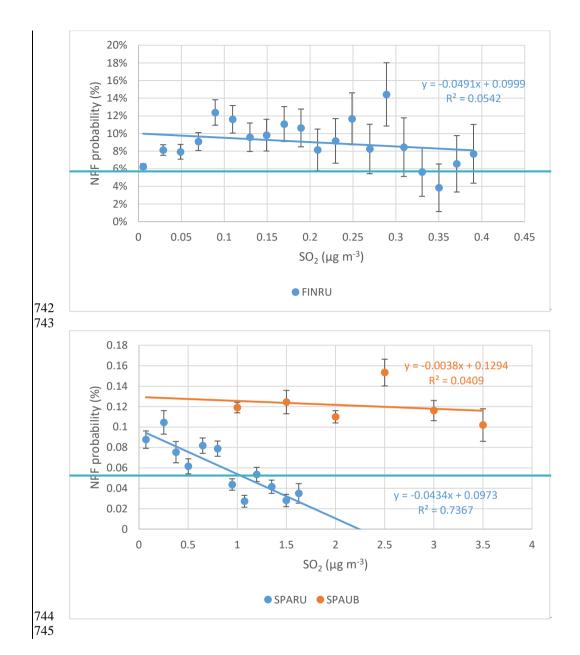


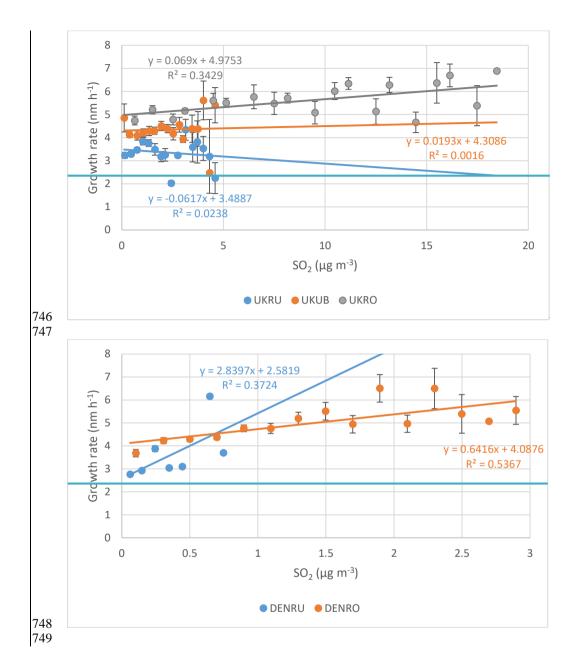


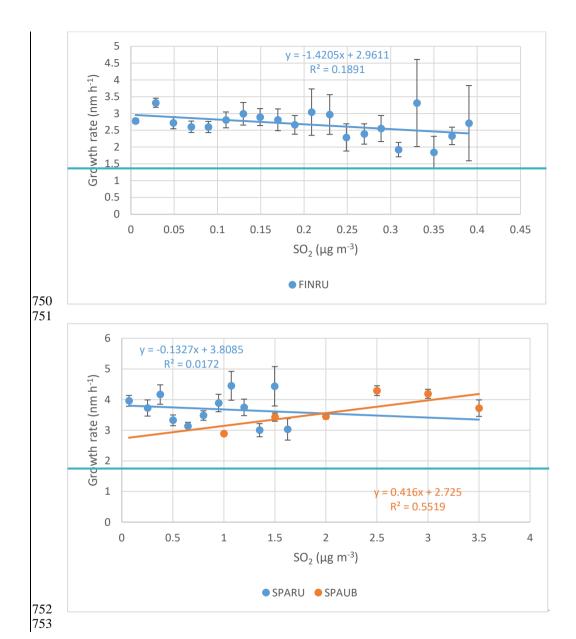


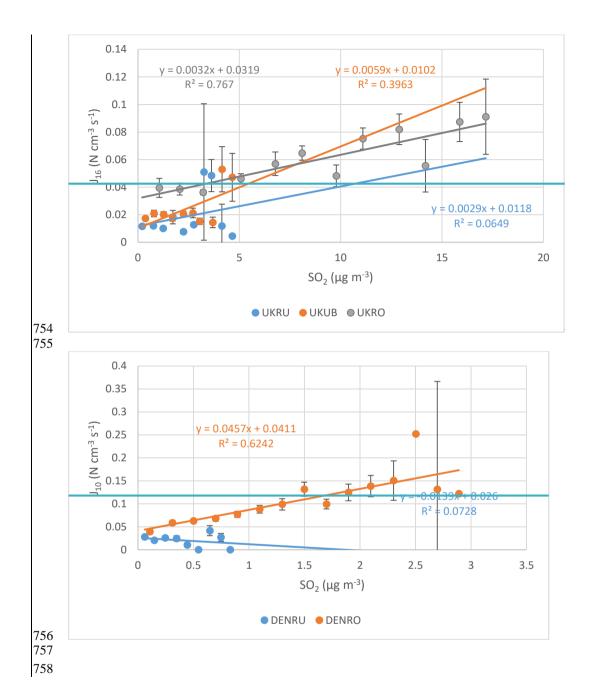


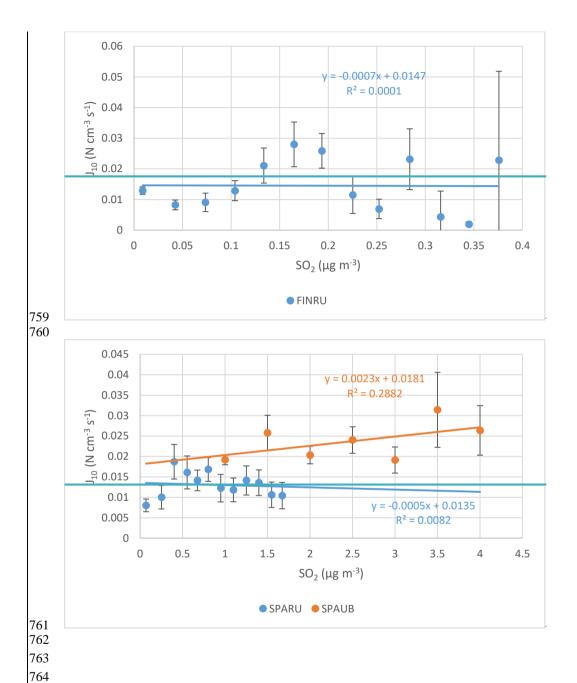


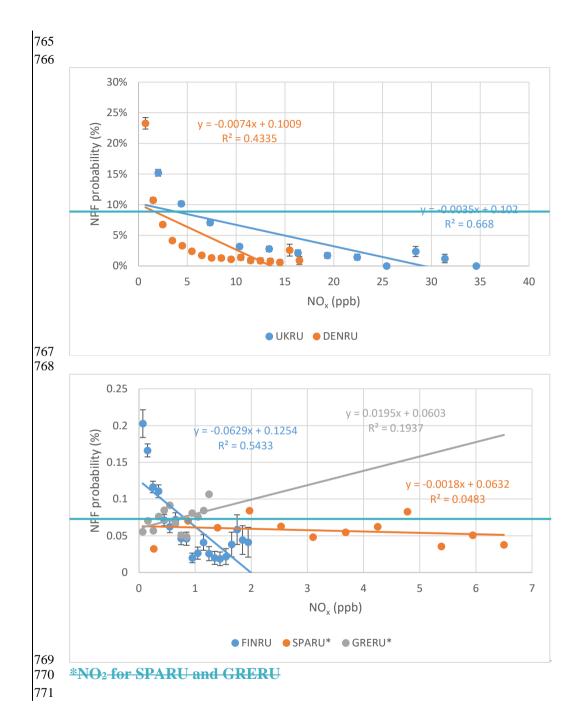


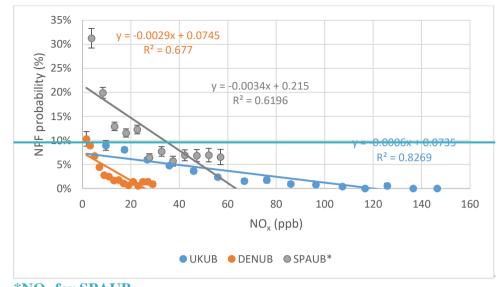




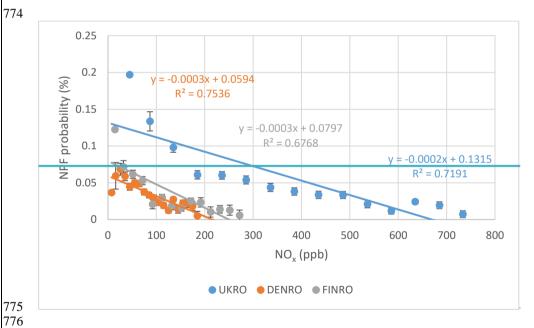


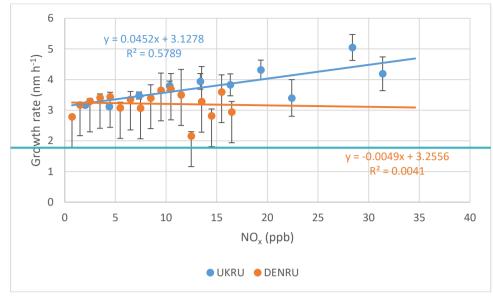


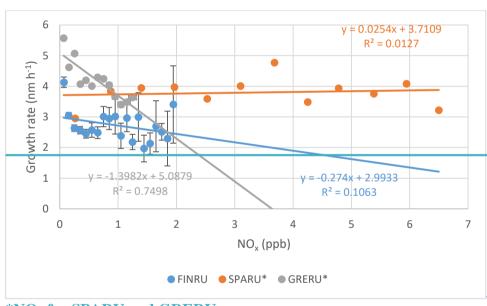




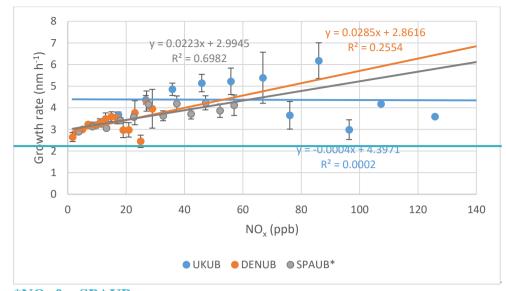
## \*NO2 for SPAUB



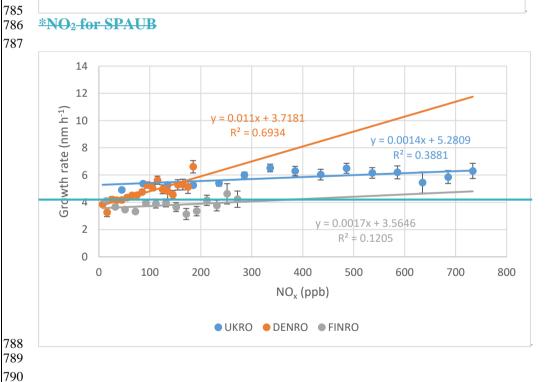


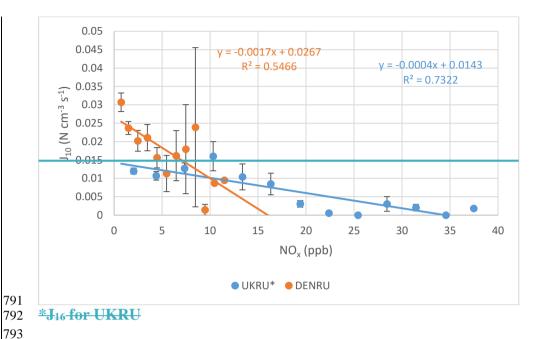


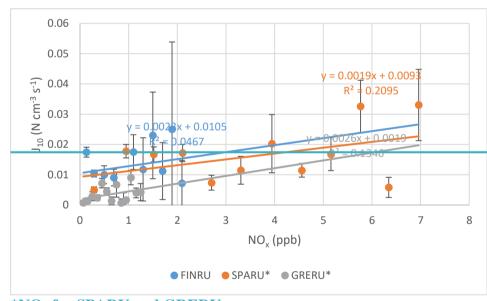
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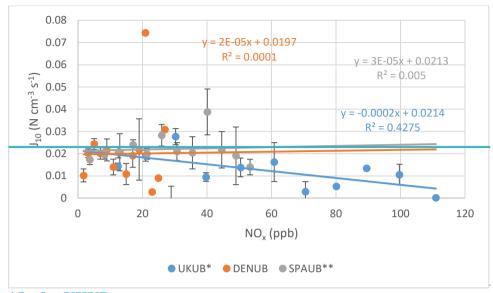
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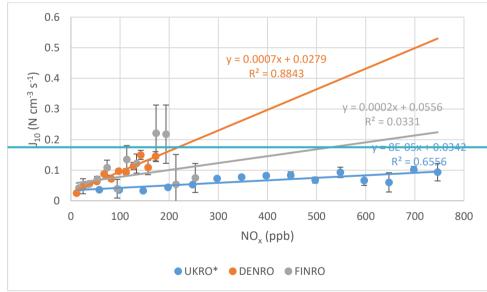


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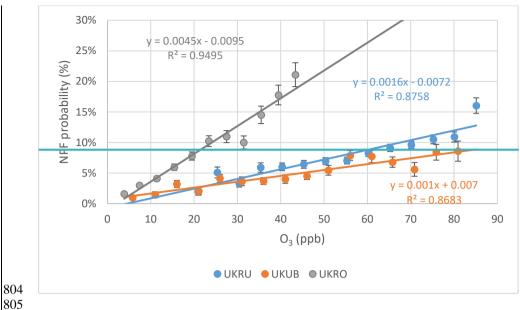


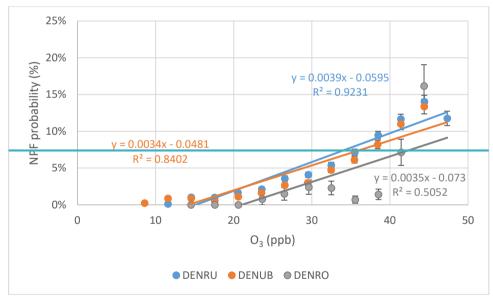
798 \*J<sub>16</sub> for UKUB

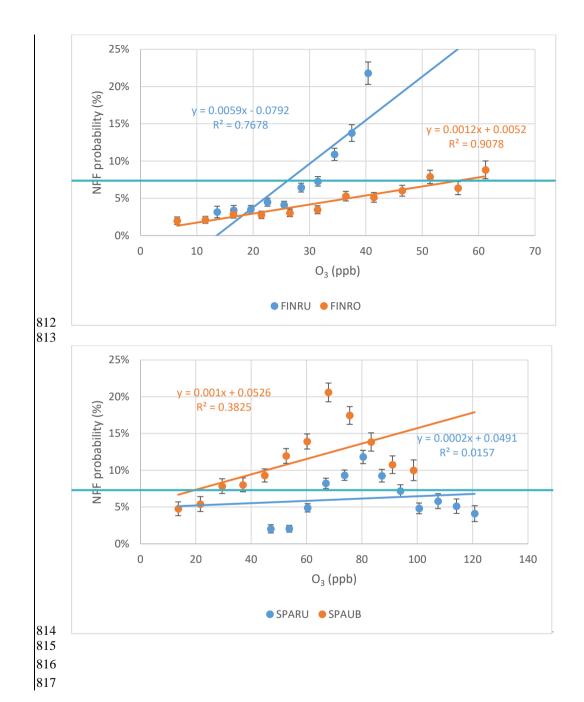


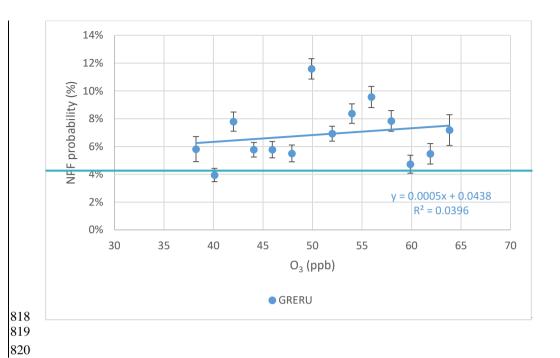


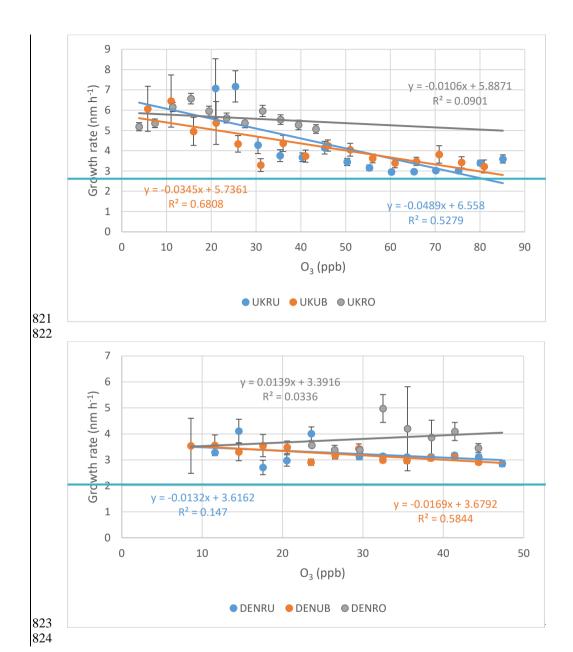
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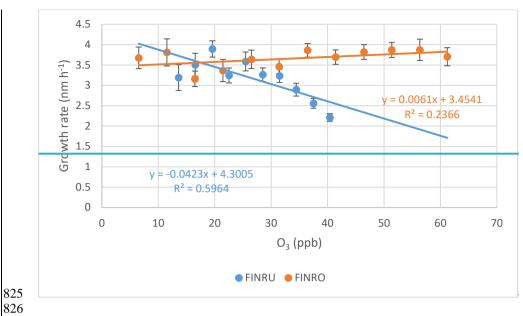


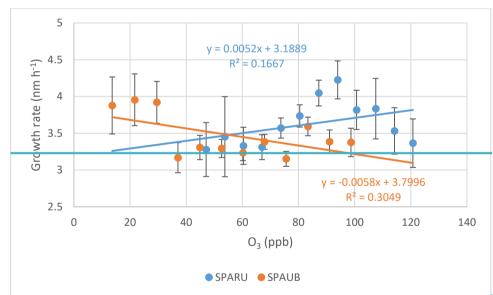


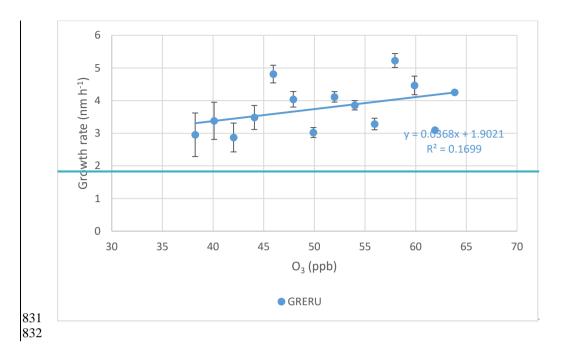


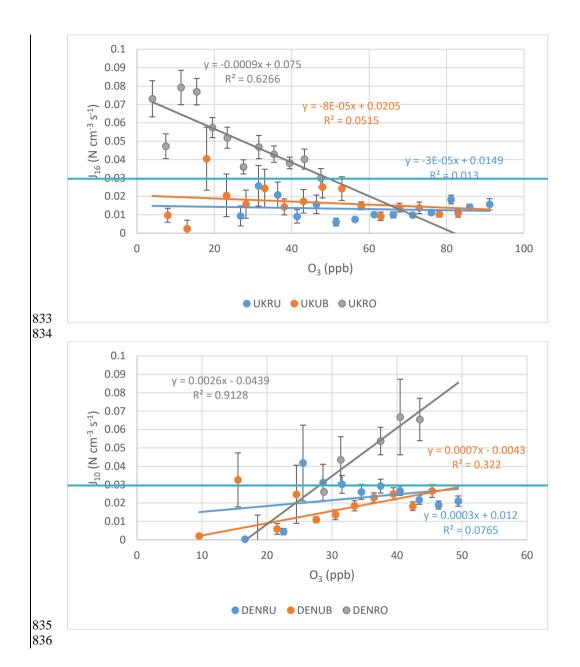


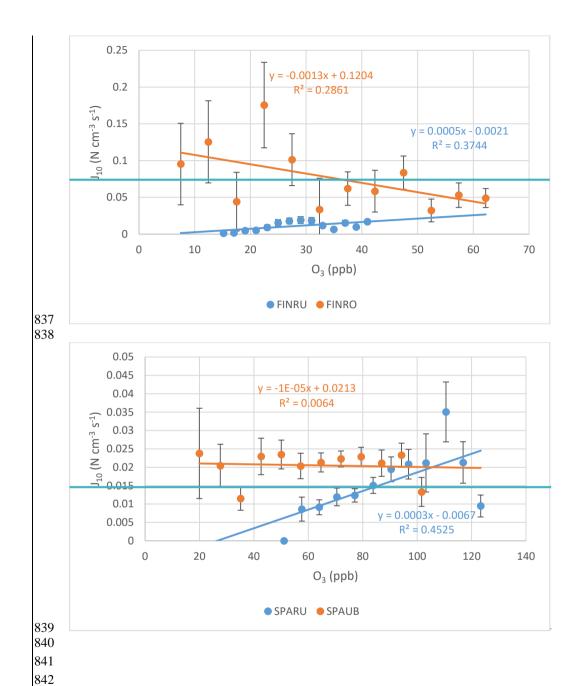


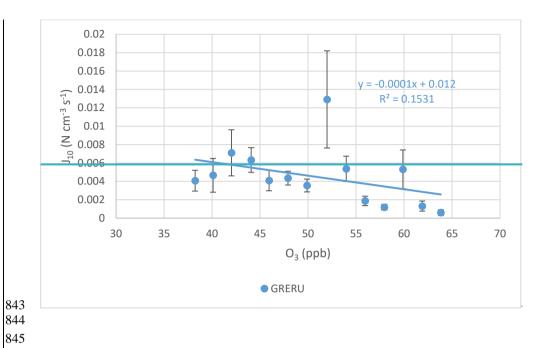


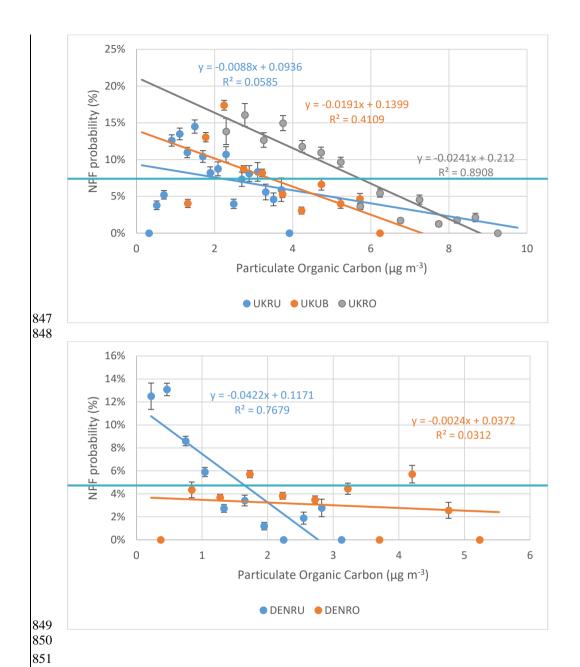


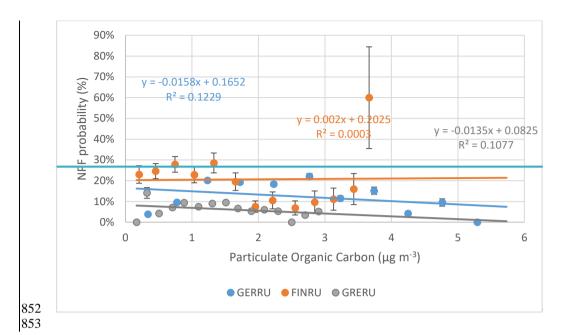


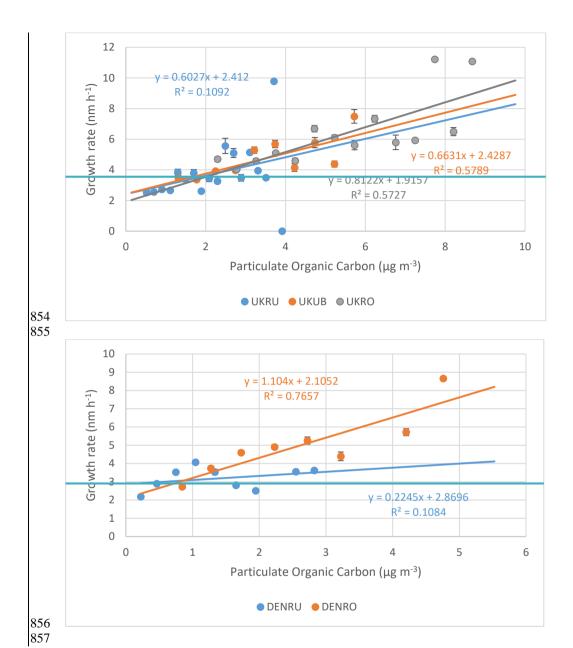


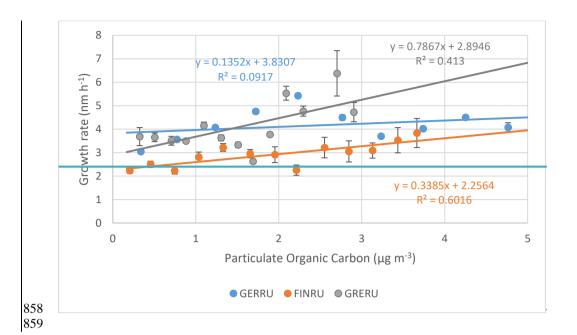


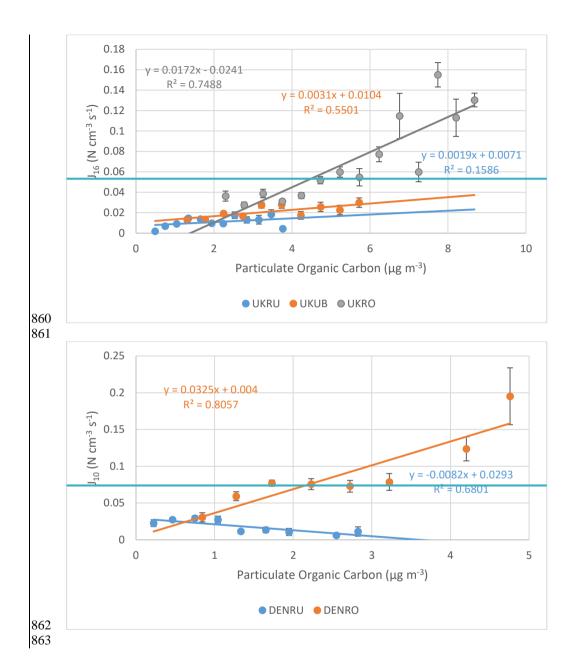


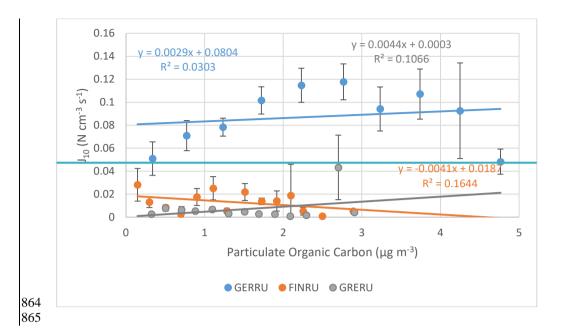


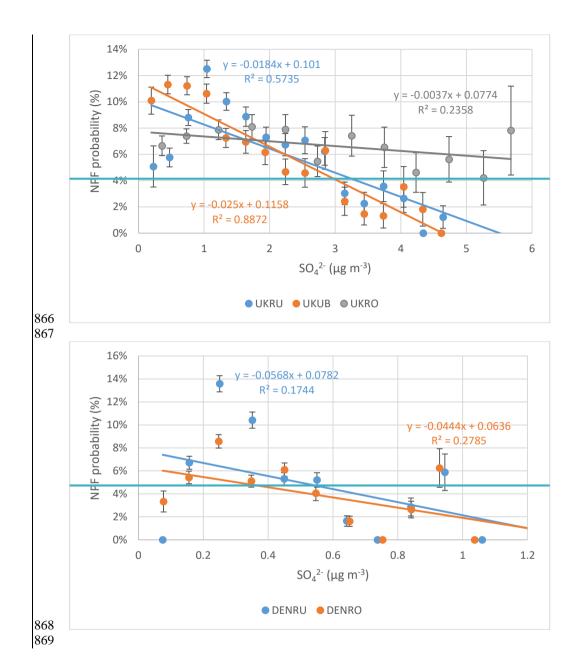


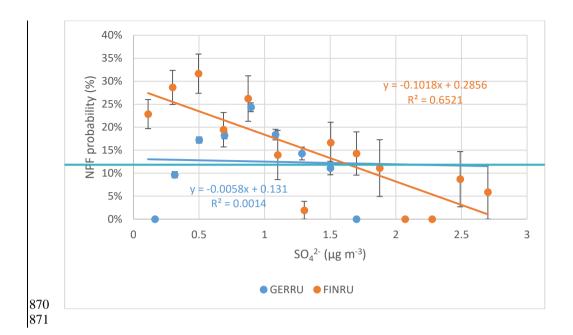


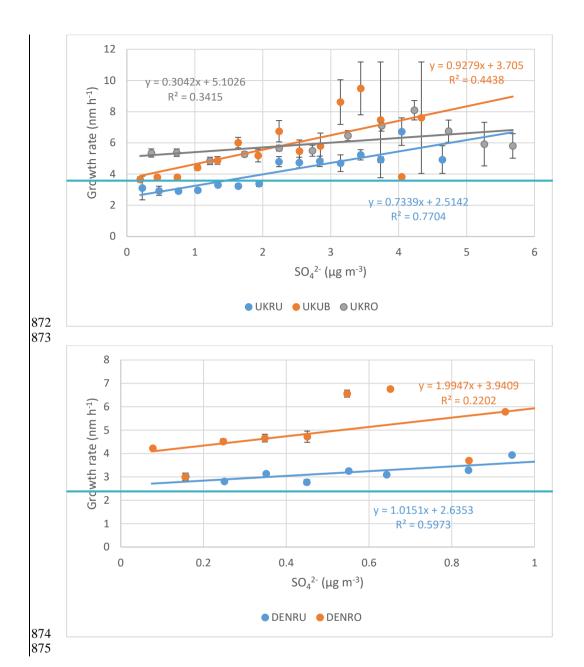


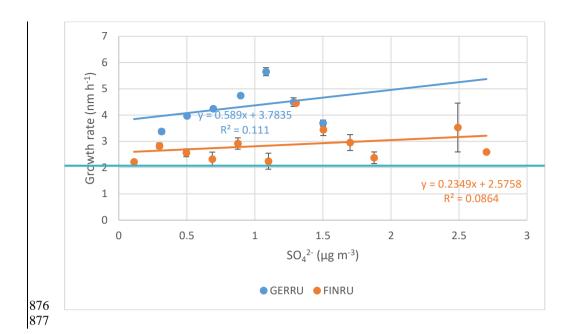


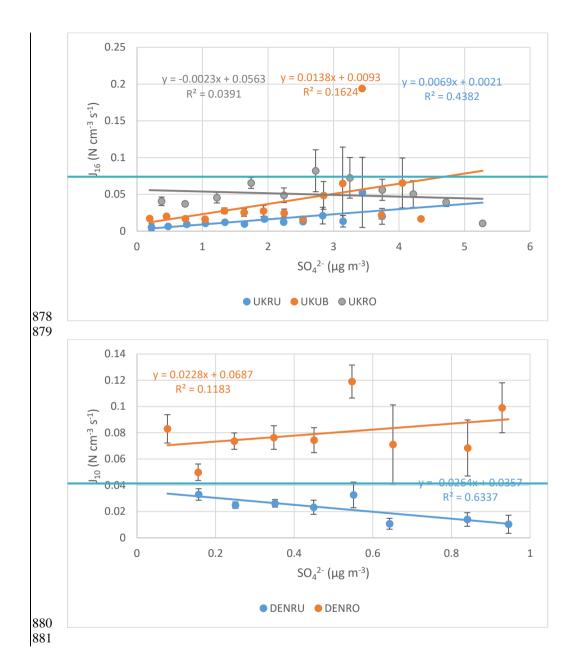


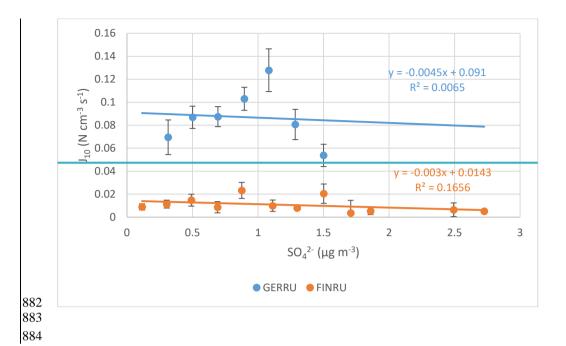


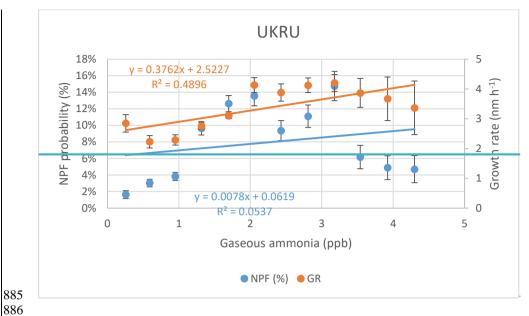


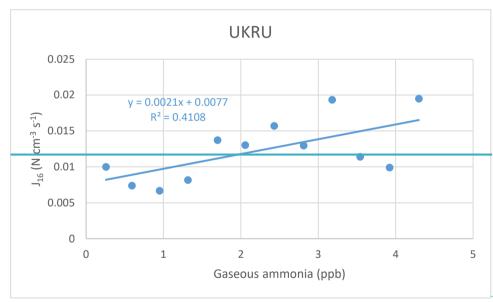


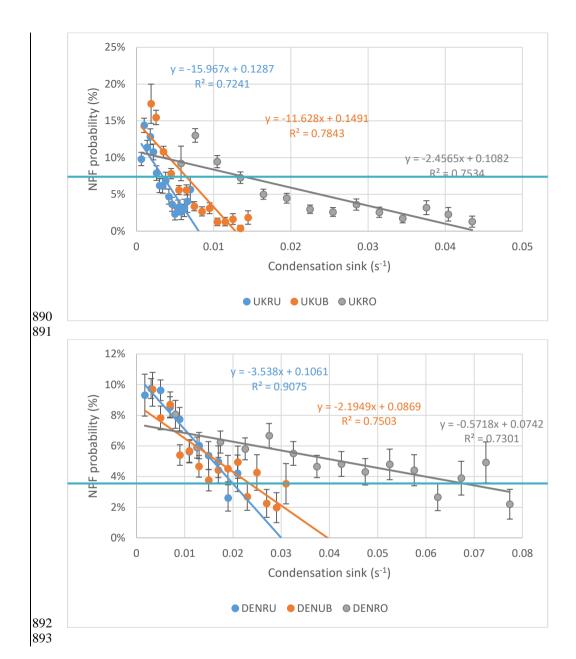


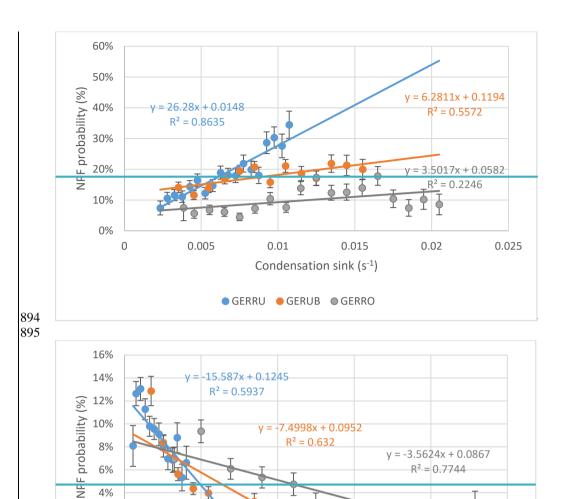












0.01

Condensation sink (s<sup>-1</sup>)

● FINRU ● FINUB ● FINRO

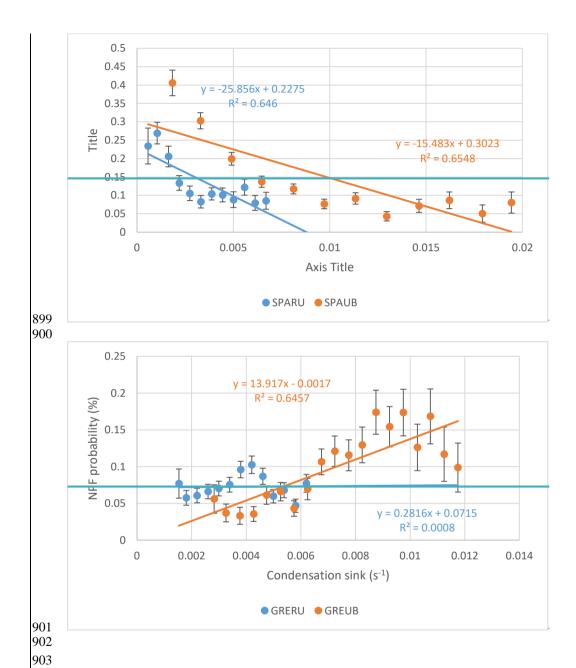
2% 0%

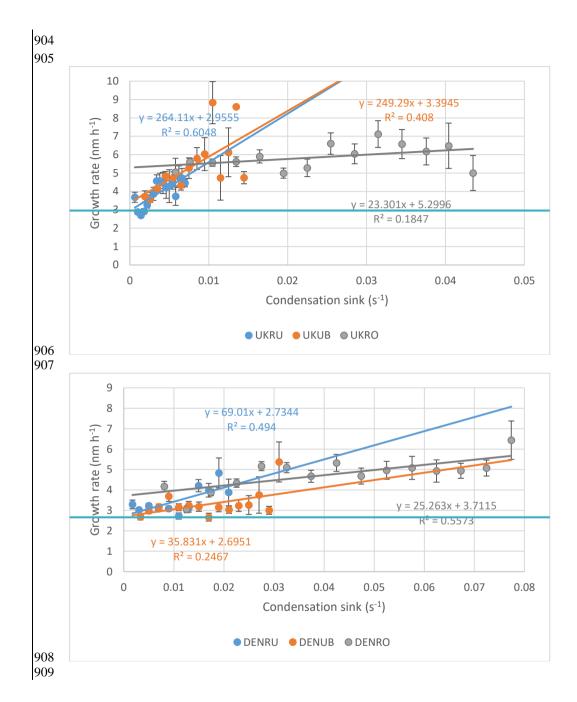
896 897 898 0.005

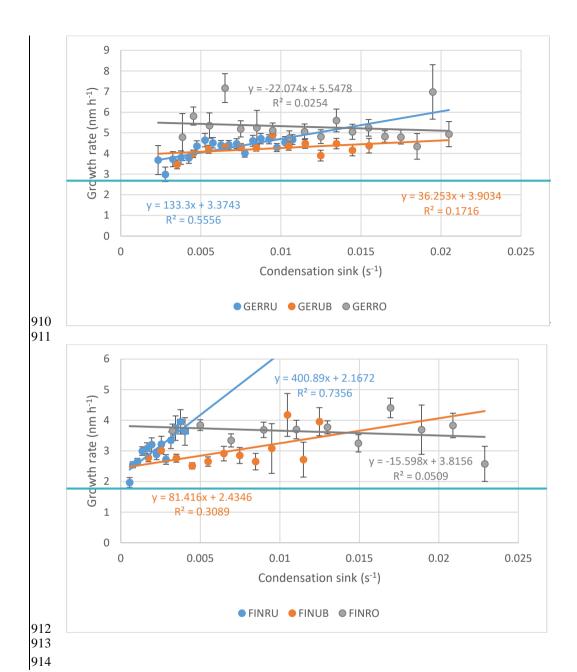
0.015

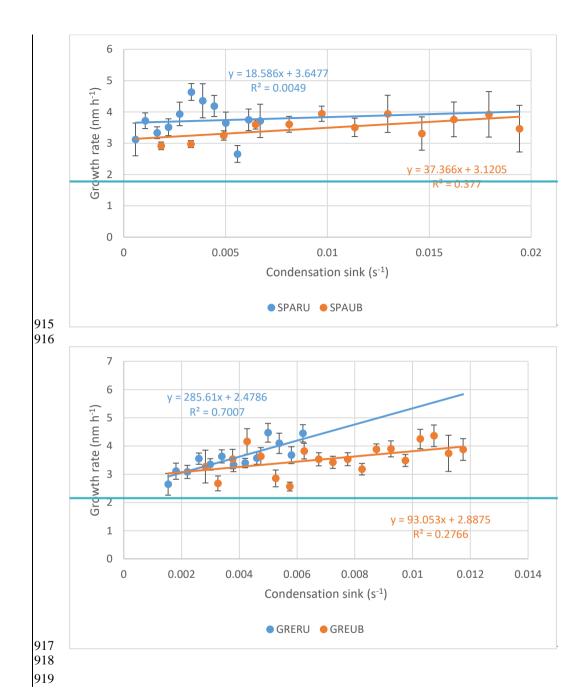
0.02

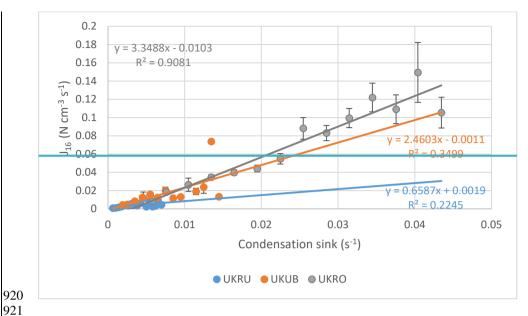
0.025

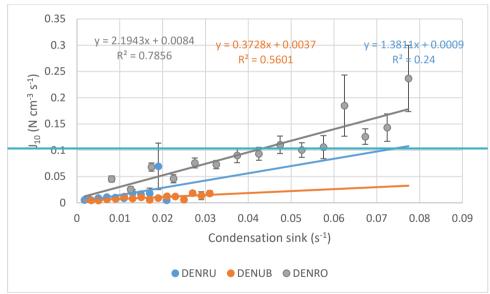


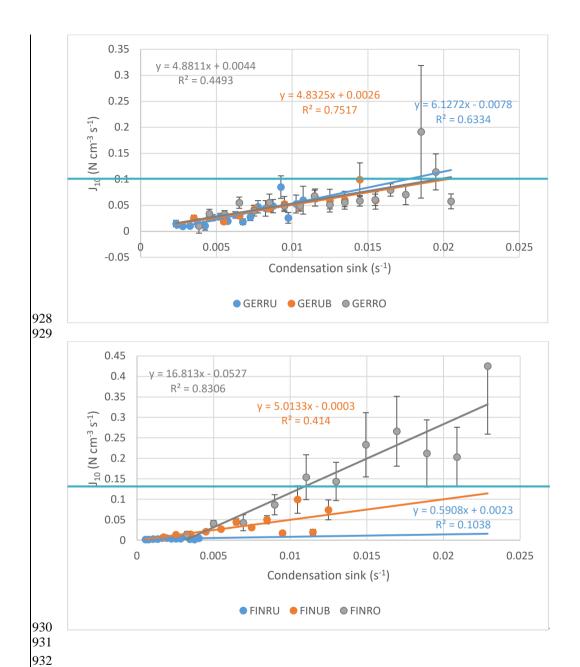












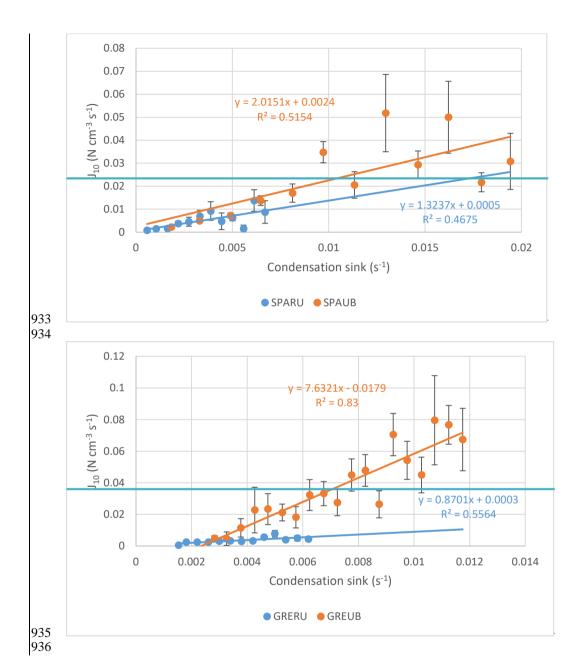


Figure S2: Relation of average temperature and normalised slopes a<sub>N</sub>\* for all but the Finnish sites. 938 0.15 UKRUUKUBUKRODENRU DENUBDENRO 0.10 -Normalised slope  $a_N^* (^{\circ}C^{-1})$ **GERRU** GERUB
GERRO
SPARU
SPAUB
GRERU 0.05  $R^2 = 0.617$ GREUB 0.00 -0.05 14 10 12 16 18 20 8 Average T (°C) 939 940