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

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43 **ABSTRACT**

44 New particle formation (NPF) events occur almost everywhere in the world and can play an

45 important role as a particle source. The frequency and characteristics of NPF events vary spatially

46 and this variability is yet to be fully understood. In the present study, long term particle size

47 distribution datasets (minimum of three years) from thirteen sites of various land uses and climates

48 from across Europe were studied and NPF events, deriving from secondary formation and not

49 traffic related nucleation, were extracted and analysed. The frequency of NPF events was

50 consistently found to be higher at rural background sites, while the growth and formation rates of

51 newly formed particles were higher at roadsides (though in many cases differences between the

52 sites were small), underlining the importance of the abundance of condensable compounds of

53 anthropogenic origin found there. The growth rate was higher in summer at all rural background

54 sites studied. The urban background sites presented the highest uncertainty due to greater variability

55 compared to the other two types of site. The origin of incoming air masses and the specific

56 conditions associated with them greatly affect the characteristics of NPF events. In general, cleaner

57 air masses present higher probability for NPF events, while the more polluted ones show higher

58 growth rates. However, different patterns of NPF events were found even at sites in close proximity

59 (< 200 km) due to the different local conditions at each site. Region-wide events were also studied

60 and were found to be associated with the same conditions as local events, although some variability

61 was found which was associated with the different seasonality of the events at two neighbouring

62 sites. NPF events were responsible for an increase in the number concentration of ultrafine particles

3

63 of more than 400% at rural background sites on the day of their occurrence. The degree of

64 enhancement was less at urban sites due to the increased contribution of other sources within the

65 urban environment. It is evident that, while some variables (such as solar radiation intensity,

66 relative humidity or the concentrations of specific pollutants) appear to have a similar influence on

67 NPF events across all sites, it is impossible to predict the characteristics of NPF events at a site

68 using just these variables, due to the crucial role of local conditions.

69

72

1.   INTRODUCTION

Ultrafine particles (particles with diameter smaller than 100 nm), while not yet regulated, are

believed to have adverse effects upon air quality and public health (Atkinson et al., 2010; Politis et

al., 2008; Tobías et al., 2018), as well as having a direct or indirect effect on atmospheric properties

(Makkonen et al., 2012; Seinfeld and Pandis, 2012). The source of ultrafine particles can either be

from primary emissions (Harrison et al., 2000; Masiol et al., 2017), including delayed primary

emissions (Hietikko et al., 2018; Olin et al., 2020; Rönkkö et al., 2017), or from secondary

formation from gaseous precursors (Brean et al., 2019; Chu et al., 2019; Kerminen et al., 2018;

Kulmala et al., 2004a; Yao et al., 2018), which is considered as an important source of CCN in the

atmosphere (Dameto de España et al., 2017; Kalivitis et al., 2015; Spracklen et al., 2008). For the

latter, while the process of formation of initial clusters that subsequently lead to particle formation

has been extensively studied (Dal Maso et al., 2002; Kulmala et al., 2014; Riipinen et al., 2007;

Weber et al., 1998), there is no consistent explanation of the factors which determine the occurrence

and development of NPF events in the atmosphere. Additionally, events that resemble NPF, with

the initial particles deriving from primary emissions, especially close to traffic sources (Rönkkö et

al., 2017), have been also reported but these are out of the scope of the present study.

A large number of studies both in laboratories and in real world conditions have been conducted to

either describe or explain the mechanisms that drive NPF events. The role of meteorological

conditions, such as solar radiation intensity (Kumar et al., 2014; Shi et al., 2001; Stanier et al.,

93   2004) and relative humidity (Li et al., 2019; Park et al., 2015), are well documented, while great

94   diversity was found for the effect of other meteorological factors such as the wind speed (Charron et

95   al., 2008; Németh and Salma, 2014; Rimnácová et al., 2011) or temperature (Jeong et al., 2010;

96   Napari et al., 2002). There are also influences of atmospheric composition, with the positive role of

97   low condensation sink and concentrations of pollutants such as NO_x upon NPF event occurrence

98   being widely agreed upon (Alam et al., 2003; Cheung et al., 2013; Kerminen et al., 2004; Wang et

99   al., 2014; Wehner et al., 2007). Contrary to that, while the indirect role of SO_2 is well established in

100 the nucleation process, via the formation of new clusters of H_2SO_4 molecules (Boy et al., 2005; Iida

101 et al., 2008; Kulmala et al., 2005; Sipila et al., 2010; Xiao et al., 2015), uncertainty exists in the role

102 that different concentrations of SO_2 play in the occurrence of NPF events in real world atmospheric

103 conditions (Alam et al., 2003; Dall'Osto et al., 2018; Wonaschütz et al., 2015; Woo et al., 2001).

104 Ammonia is known to enhance the formation of initial clusters (Korhonen et al., 1999; Ortega et al.,

105 2008; Schobesberger et al., 2015), and volatile organic compounds are regarded as the main drivers

106 of the growth of the newly formed particles (Kulmala et al., 2013; Riccobono et al., 2014; Tröstl et

107 al., 2016). NPF events in different locations do not appear to follow consistent trends with the

108 concentrations of these compounds and meteorological parameters (McFiggans et al., 2019;

109 Minguillón et al., 2015; Riipinen et al., 2007), though links between NPF events and sulphuric acid

110 vapour concentrations (Petäjä et al., 2009; Weber et al., 1995) and organics (Bianchi et al., 2019;

111 Ehn et al., 2014) have been reported.

112

113 It is evident that NPF events and their development are complex, and local conditions play an

114 important role in their variability. Many studies have attempted to explain this variability by

115 analyzing multiple datasets from wider areas. Studies in the UK (Bousiotis et al., 2019; Hama et al.,

116 2017), Spain (Brines et al., 2014; Carnerero et al., 2018; Dall'Osto et al., 2013; Minguillón et al.,

117 2015), Hungary (Németh and Salma, 2014; Salma et al., 2014, 2016), Greece (Kalkavouras et al.,

118 2017; Siakavaras et al., 2016), Germany (Costabile et al., 2009; Ma and Birmili, 2015; Sun et al.,

119 2019) and China (Peng et al., 2017; Shen et al., 2018; Wang et al., 2017) have attempted to explain

120 the differences found in NPF event conditions and variability between different sites in close

121 proximity, while larger scale studies using descriptive (Brines et al., 2015; Hofman et al., 2016;

122 Jaatinen et al., 2009; Kulmala et al., 2005) or statistical methods (Dall'Osto et al., 2018; Rivas et

123 al., 2020) have provided insights into the effect of the variability of parameters that are considered

124 to play an important role in the occurrence and development of NPF events on a broader scale.

125

126 The present study, combining thirteen long term datasets (minimum of three years) from different

127 countries across Europe and combined with the results from a previous study in the UK, attempts to

128 elucidate the effect of the local conditions on NPF event characteristics (frequency of NPF events,

129 formation rate and growth rate) both for sites in close proximity (< 200 km), and by

130 intercomparison of sites on a continental scale in order to find general trends of the variables that

131 affect the characteristics and development of NPF events on a larger scale. Finally, the effect of

132 NPF events upon the ultrafine particle number concentrations was calculated, providing insight to

133 the potential of NPF events to influence the local air quality conditions in all areas studied.

134

135 **2.   DATA AND METHODS**

136 **2.1   Site Description and Data Availability**

137 In the present study, particle number size distribution data from 13 sites in Europe (Figure 1) are

138 analysed in the size range 3 nm < Dp < 1000 nm. A detailed list of the site locations and the data

139 available for each is found in Table 1 (seasonal data availability is found in Table S1). For site naming

140 the first three letters refer to the country (DEN = Denmark, GER = Germany, FIN = Finland, SPA =

141 Spain, GRE = Greece) while the next two refer to the type of site (RU = Rural background, UB =

142 Urban background, RO = Roadside). Average meteorological conditions and concentrations of

143 chemical compounds for all sites are found in Tables S2 and S3 respectively; their seasonal variation

144 is found in Table S4.

145

146 **2.2   Methods**

147 **2.2.1   NPF event selection**

148 The identification of NPF events was conducted manually using the criteria set by Dal Maso et al.

149 (2005). According to these, a NPF event is considered to occur when:

150 • a distinctly new mode of particles appears in the nucleation range,

151 • this new mode prevails for some hours,

152 • the new mode shows signs of growth.

153

154 The NPF events extracted using this method are then classified into classes I or II depending on the

155 level of confidence. Class I (high confidence) is further classified as Ia and Ib, with class Ia

156 containing the events that both present a clear formation of a new mode as well as a distinct growth

157 of this mode, while Ib includes those with a less distinct formation and development. In the present

158 study, only the events classified as Ia were used as they are considered as more suitable for study.

159 As the growth criterion is not fully defined, in the present study a minimum growth rate of 1 nm h^{-1}

160 is required for NPF events to be considered. The events found using this method should not be

161 confused with the formation and growth of particles deriving from primary emissions next to

162 pollution sources, such as traffic. While to an extent the particle formation found can be biased by

163 primary emissions (especially at roadside sites), great effort was made using additional data, such as

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

165

166 **2.2.2 Calculation of condensation sink, growth rate, formation rate, Nucleation Strength**

167 **Factor (NSF) and NPF event probability**

168 The calculation of the condensation sink was made using the method proposed by Kulmala et al.

169 (2001). The condensation sink (CS) is calculated as:

170

171 $$CS = 4\pi D_{vap} \sum \beta_M \, r \, N$$

172 where r and N are the radius and the number concentration of the particles and D_{vap} is the diffusion

173 coefficient, calculated for T = 293 K and P = 1013.25 mbar, according to Poling et al. (2001):

174

175 $$D_{vap} = 0.00143 \cdot T^{1.75} \frac{\sqrt{M_{air}^{-1} + M_{vap}^{-1}}}{P\left(D_{x,air}^{\frac{1}{3}} + D_{x,vap}^{\frac{1}{3}}\right)^2}$$

176

177 where M and D_x are the molar mass and diffusion volume for air and H_2SO_4. β_M is the Fuchs

178 correction factor calculated from Fuchs and Sutugin (1971):

179

180 $$\beta_M = \frac{1 + K_n}{1 + \left(\frac{4}{3a} + 0.377\right)K_n + \frac{4}{3a}K_n^2}$$

181

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

183

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

185

186 $$GR = \frac{D_{P_2} - D_{P_1}}{t_2 - t_1}$$

187

188   for the size range between the minimum available particle diameter up to 30 nm. For the calculation

189   of the growth rate, the time considered was from the start of the event until a) growth stopped, b)

190   GMD reached the upper limit set or c) the day ended. Due to the differences in the smallest particle

191   size available between the sites, a discrepancy would exist for the growth rate values presented

192   (sites with lower size cut would present lower values of growth rate, as the growth rate tends to

193   increase with particle size in this range (Deng et al., 2020)). As a result, a direct comparison of the

194   growth rate values found among sites with significant differences at the smallest particle size

195   available was avoided.

196

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

198

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

200

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

202

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

204

205 as proposed by Kerminen et al. (2001). $K(d_p, d'_p)$ is the coagulation coefficient of particle sizes d_p

206 and d'_p. S_{losses} accounts for the additional loss terms (i.e. chamber walls), not considered here. Initial

207 particle formation starts at about 1.5 ± 0.4 nm (Kulmala et al., 2012). The formation rate calculated

208 here refers to particles in the atmosphere that reached the diameter of 10 nm during NPF events for

209 uniformity reasons. This means that these particles were formed earlier during the day of the events,

210 survived and grew to this size later in the day. Furthermore, due to the effect of the morning rush

211 which biased the results at roadsides, the averages are calculated for the time window between 9:00

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

213 the sites in this study for consistency.

214

215 As mentioned in the methodology for NPF event selection (chapter 2.2.1) days with particle

216 formation resulting directly from traffic emissions were excluded. For those identified as NPF event

217 days though, mainly for the roadside sites, formation associated with traffic emissions still occurs. It

218 is impossible with the data available for this study to remove the traffic related particle formation in

219 the calculations included in this study, by effectively separating it from secondary particle

220 formation or calculate it. Using average conditions for comparison would lead to negative formation

221 rate values in most cases, since in order for an NPF event to occur, traffic related particles are

222 usually reduced to a greater extent compared to the formation from NPF, leading to lower particle

223 concentrations on event days as found from a previous study in Marylebone Road, London

224 (Bousiotis et al., 2019). This may result in an overestimation of the formation rates at roadside sites

225 presented in this study., The choice of a time window for which we would have the maximum effect

226 of secondary particle formation and the minimum possible effect from traffic related particle

227 formation attempts to reduce this discrepancy as much as possible.

228

229 The Nucleation Strength Factor (NSF) proposed by Nemeth and Salma (2014) is a measure of the

230 effect of NPF events on ultrafine particle concentration. It can either refer to the effect of NPF

231 events on the day of their occurrence, calculated by:

232

233
$$NSF_{NUC} = \frac{\left(\frac{N_{smallest\ size\ available-100nm}}{N_{100nm\ -largest\ size\ available}}\right)_{nucleation\ days}}{\left(\frac{N_{smallest\ size\ available-100nm}}{N_{100nm-largest\ size\ available}}\right)_{non-nucleation\ days}}$$

234

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

236

237
$$NSF_{GEN} = \frac{\left(\frac{N_{smallest\ size\ available-100nm}}{N_{100nm-largest\ size\ available}}\right)_{all\ days}}{\left(\frac{N_{smallest\ size\ available-100nm}}{N_{100nm-largest\ size\ available}}\right)_{non-nucleation\ days}}$$

238

239 The NPF event probability is a simple metric of the probability of NPF events calculated by the

240 number of NPF event days divided by the number of days with available data for the given group

241 (temporal, wind direction etc.). Finally, it should be mentioned that all the results presented are

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

243 $NPF_{probability} = \dfrac{N_{NPF\ event\ days\ for\ group\ of\ days\ X}}{N_{days\ with\ available\ data\ for\ group\ of\ days\ X}}$

244

245 **3.   RESULTS**

246 The seasonal NPF probability for all sites is found in Table S5. The annual number of NPF events,

247 growth rate and formation rate for all the sites is found in Table S6, for which no clear interannual

248 trend is found for any of the sites in this study. This may be due to the relatively short period of

249 time studied for such variations to be observed.

250

251 **3.1   Frequency and Seasonality of NPF Events**

252 In Denmark, NPF events occurred at all three sites with a similar frequency for the urban sites

253 (5.4% for DENRO and 5.8% for DENUB) and higher for the rural DENRU site (7.9%). The

254 seasonal variation favoured summer at DENRU and DENRO, while at DENUB a similar frequency

255 for spring and summer was found (Figure 2). The within-week variation of the events favours

256 weekends compared to weekdays going from the rural background site to the roadside site (Figure

257 3). Interesting is the increased frequency of NPF events found in all Danish sites on Thursday

258 among the weekdays. This trend though does not have a plausible explanation and is probably

259 coincidental.

260

14

261 A higher frequency of events for all types of environments is found for the German sites compared

262 to all other countries in this study. The background sites had NPF events for more than 17% of the

263 days, while the roadside had a lower frequency of about 9%, with a seasonal variability favouring

264 summer at all sites. It should be noted though that, due to the lack of spring and summer data for the

265 first two years at the German roadside site, the frequency of events is probably a lot higher, and the

266 seasonal variation should further favour these seasons. No substantial within-week variation was

267 found for any of the sites in this country, a feature that is expected mainly at background sites. For

268 GERRO, this may be due to not being as polluted as other sites of the same type, having an average

269 condensation sink comparable to that of urban background sites in this study.

270

271 NPF events at the sites in Finland presented the most diverse seasonal variation, peaking at the

272 background sites in spring and at the roadside site in summer (while the spring data availability is

273 somewhat reduced for the Finnish roadside site, the general trend remains the same if all seasons

274 had the same data availability). The frequency of NPF events at FINRU was higher (8.66%)

275 compared to the urban sites (4.97% at FINUB and 5.20% at FINRO). Strong within-week variation

276 favouring weekends is found for the roadside site, while no clear variation was found for the

277 background sites. This may be due to either the higher condensation sink during weekdays that

278 supresses the events, or the dominant impact of the traffic emissions which could make the

279 detection of NPF events harder.

280

281   For Spain, data was available for an urban and a rural background site in the greater area of

282   Barcelona. NPF events were rather frequent, occurring on about 12% of the days at the rural

283   background site and 13.1% at the urban site. Though the sites are in close proximity (about 50 km),

284   the seasonality of NPF events was different between them, peaking in spring at SPARU and autumn

285   at SPAUB. The frequency of NPF events in winter was relatively high compared to the sites in

286   central and northern Europe and higher than summer for both sites. For both sites a higher NPF

287   probability was found on weekends compared to weekdays, though this trend is stronger at SPAUB.

288   Finally, for Greece data are available for two background sites, though not in close proximity (the

289   distance between the sites is about 350 km). While in Greece meteorological conditions are

290   favourable in general for NPF events, with high solar radiation and low relative humidity, their

291   frequency was only 8.5% for the urban background site in Athens and 6.5% for the rural

292   background site in Finokalia, similar to the frequency of Class I events reported in the study by

293   Kalivitis et al. (2019). Most NPF events occurred in spring at both sites, peaking in April. It is

294   interesting that the sites in southern Europe (in Spain and Greece) have a considerable number of

295   NPF events during winter, which might be due to the specific meteorological conditions found in

296   this area, where winter is a lot warmer than at the sites in northern and central Europe, and

297   insolation is higher.

298

299

300

3.2 The Formation and Growth Rates

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

Similar to the frequency of NPF events, the German sites also had higher growth rates compared to sites of the same type in other areas of this study, with GERRU having 4.34 ± 1.73 nm h^{-1}, GERUB 4.24 ± 1.69 nm h^{-1} and GERRO 5.17 ± 2.20 nm h^{-1} (Figure 3). While the difference between GERRU and GERUB is not statistically significant, there is a significant difference with GERRO ($p <$ 0.005). Higher growth rates were found in summer compared to spring for all sites (Figure 5). Specifically, for the roadside though, the highest average growth rates were found in autumn, which may be either a site-specific feature or an artefact of the limited number of events in that season (total of 11 NPF events in autumn). Similarly, J_{10} at the German sites was also the highest among the sites of this study, increasing from the GERRU to GERRO. It was found to be higher in summer for the background sites and in autumn for GERRO.

17

321   For the Finnish sites, growth rates were similar at the background sites (2.91±1.68 nm h^{-1} at FINRU

322   and 2.87±1.33 nm h^{-1} at FINUB), peaking in the summer months, similar to the findings of Yli-Juuti

323   et al. (2011), while the peak for FINRO (growth rate at 3.74±1.48 nm h^{-1}) was found in spring,

324   though the differences between the seasons for this site were rather small. The formation rate was

325   the highest at FINRO, peaking in autumn for both urban sites (with small differences with spring),

326   while FINRU presented the highest J_{10} in summer.

327

328   At the Spanish sites, the growth rate was similar for the two sites, being 3.62 ± 1.86 nm h^{-1} at

329   SPARU and 3.38 ± 1.53 nm h^{-1} at SPAUB, again being higher in autumn for the urban site (which

330   appears to be a feature of more polluted sites), while the rural site follows the general trend of rural

331   background sites, peaking in summer. The formation rate at SPAUB is comparable to the other

332   urban background sites (apart from GERUB) and peaked in spring, while once again the peak at

333   SPARU was found in summer, similar to the other rural sites of this study apart from the Greek. At

334   the urban site both the growth and formation rates were higher on weekdays compared to weekends

335   (both $p < 0.001$). While the higher growth rate during weekdays may be associated with the

336   increased presence of condensable species from anthropogenic activities, the higher formation rate

337   might be affected by the increased emissions during these days, which bias to an extent its value.

338   Finally, the growth rate of particles was found to be similar at both Greek sites (3.68±1.41 nm h^{-1}

339   for GREUB and 3.78±2.01 nm h^{-1} for GRERU) and was higher in summer compared to the other

340   seasons, having a similar trend with the temperature and particulate organic carbon concentrations

18

341   in the area. The formation rate presented a unique trend, having high averages in winter for both

342   sites. Interestingly, contrary to most background sites in this study, the lowest average J_{10} was found

343   for summer at both sites.

344

345   **3.3   Conditions Affecting NPF Events**

346   The average and NPF event day conditions are presented in tables S2 and S3 (for meteorological

347   conditions and atmospheric composition respectively). A number of variables present consistent

348   behaviour on NPF days. For all the sites in this study the solar radiation intensity was higher on

349   NPF days compared to the average conditions, while the relative humidity was lower. Additionally,

350   all the chemical compounds with available data present either lower or similar concentrations. This

351   is consistent even for the chemical compounds which are associated with the NPF process (such as

352   the SO_2). This probably indicates that they are in sufficient concentrations for not being a limiting

353   factor in the occurrence of the events, while higher concentrations are associated with increased

354   pollution conditions which may suppress their occurrence. The exceptions found are SPARU and

355   GRERU for NO_2 and FINRU for SO_2. In these sites the concentrations of these gaseous components

356   are very low in general (being rural background sites) and were found to be only marginally higher

357   on NPF event days. These differences indicate that the variability of these compounds is not playing

358   a significant role in the occurrence of the events and thus should not be considered as an important

359   factor. The ozone concentration though, was found to be consistently higher on event days

360   compared to the average conditions at all sites regardless of their geographical location and type. As

361 the ozone concentration variability is directly associated with the solar radiation intensity, it is

362 unknown whether it plays a direct role in the occurrence of the events or it is the result of its

363 covariance with the solar radiation intensity.

364

365 Following that, differences were found in the variability of some of the meteorological conditions,

366 as well as local conditions (either meteorological or specific pollution sources), which played a

367 significant role in the occurrence and the metrics of NPF events across the sites of this study. These

368 will be further explored in the following sections.

369

370 **3.3.1   Denmark**

371 The meteorological conditions that prevailed on NPF event days followed the general trend

372 mentioned earlier, while wind speed and temperature were higher than average (consistently at all

373 sites, meteorological condition variability was significant for all ($p < 0.001$) except the wind speed).

374 As meteorological data were available from the urban background site (the variation between the

375 rural and urban sites should not be great since they are about 25 km away from each other), the

376 average conditions for the three sites are almost the same, with the only variability being the data

377 availability among the sites. Thus, the more common wind directions in the area are southwesterly;

378 for all sites though the majority of NPF events are associated with direct westerly and northwesterly

379 winds, similar to the findings of Wang et al. (2013) for the same site, which are those with the

380 lowest concentrations of pollutants and condensation sink for all sites (Table S7), probably being of

381   marine origin as elemental concentrations showed an increased presence of Na, Cl and Mg (results

382   not included). The wind directions with the highest probability for NPF events presented low

383   growth rates and vice versa (Table S4), though it was proposed by Kristensson et al. (2008) that

384   there is a possibility for events observed at the nearby Vavihill site in Sweden with northwesterly

385   winds to be associated to the emissions of specific ship lanes that pass from that area. Wind

386   direction sectors with higher concentrations of OC coincide with higher growth rates at DENRO,

387   while this variability is not found at DENRU possibly showing that different compounds and

388   mechanisms take part in the growth process of the newly formed particles (Kulmala et al., 2004b).

389

390   As mentioned earlier, DENUB although close to the DENRO site has different seasonal variation of

391   NPF events, with a marginally lower frequency in summer compared to the other two Danish sites,

392   which have almost the same seasonal variation of NPF events. At DENUB, a strong presence of

393   particles in the size range of about 50 – 60 nm is observed (Figure S1), especially during summer

394   months, increasing the condensation sink in the area (this enhanced mode of particles is visible at

395   DENRO as well, but its effect is dampened due to the elevated particle number concentrations in

396   the other modes). This mode is probably part of the urban particle background. The strongest source

397   though at DENUB appears to be from the east and consistently appears at both urban sites; this

398   sector is where both elevated pollutant concentrations and condensation sink are found. In this

399   sector, there are two possible local sources, either the port located 2 km to the east or the power

400   plant located at a similar distance (or both). In general, both stations are located only a few

401 kilometres away from the Øresund strait, a major shipping route. Studying the SMPS plots it can be

402 seen that NPF events at DENUB, especially in summer, tend to start but are either suppressed after

403 the start or have a lifetime of a couple of hours before the new particles are scavenged or evaporate.

404 While this might explain to an extent the frequency and variability of NPF events at this site, the

405 balance between the condensation sink and the concentration of condensable compounds is

406 highlighted. While at DENRO the condensation sink is considerably higher than at DENUB and the

407 effect of the aforementioned mode of particles is present at both, the occurrence and development of

408 NPF events at DENRO are more pronounced in the data, due to the higher concentrations of

409 condensable compounds.

410

411 **3.3.2   Germany**

412 Compared to the average conditions, a higher temperature was found on NPF event days, while

413 wind speed was lower at all German sites. The condensation sink was also higher on event days

414 compared to the average, though this may be the result of the high formation rates found for the

415 German sites. The wind profile is different between the urban and the rural sites, with mainly

416 northeasterly and southwesterly winds at the rural site and a more balanced profile for the urban

417 sites. This difference is probably due to differences in the local topography. For the urban sites the

418 majority of NPF events are associated with easterly winds (to a lesser extent westerly as well for

419 GERRO). At GERUB, along with the increased frequency of NPF events, the highest average

420 growth rate is also found with easterly wind directions (though the differences are rather small). At

421 GERRO the frequency and growth rate appear to be affected by the topography of the site.

422 Eisenbahnstraße is a road with an axis at almost $90^o – 270^o$ and although the H/W ratio

423 (surrounding building height to width ratio) is not high, the effect of a street canyon vortex is

424 observed (Voigtländer et al., 2006). Possibly as a consequence of this, the probability of NPF events

425 is low for direct northerly and southerly winds, although there are high growth rates of the newly

426 formed particles (highest growth rates observed with southerly winds, associated with cleaner air).

427

428 At GERRU an increased probability of NPF events and growth rate are also found for wind

429 directions from the easterly sector, although these are not very frequent for this site. For this site

430 chemical composition data for $PM_{2.5}$ and PM_{10} are available, and it is found that the generally low

431 (on average) concentrations of pollutants (such as elemental carbon, nitrate and sulphate), in general

432 are elevated for wind directions from that sector. This is also reported for the Melpitz site (GERRU)

433 by Jaatinen et al. (2009) and probably indicates that in a relatively clean area, the presence of low

434 concentrations of pollutants may be favourable in the occurrence and development of NPF events,

435 as in general pollutant concentrations are lower on NPF event days compared to average conditions.

436 Another interesting point is the concentration of organic carbon at the site (average of 2.18 µg m^{-3}

437 in $PM_{2.5}$), having the highest average concentration among the rural background sites studied. As

438 other pollutant concentrations are relatively low at this site, it is possible that a portion of this

439 organic carbon is of biogenic origin, considering also that the area is largely surrounded by forests

440 and green areas, with a minimal effect of marine air masses (as indicated by the low marine

441 component concentrations – data not included) and possibly pointing to increased presence of

442 BVOCs. The increased presence of organic species at GERRU may explain to some extent the

443 increased frequency of NPF events as well as the highest growth and formation rates found among

444 the sites of this study.

445

446 ### 3.3.3   Finland

447 At the background sites in Finland, temperature was lower on NPF event days compared to the

448 average conditions, whereas it was higher for FINRO associated with the different seasonality of

449 the events. No significant differences were found for the wind speed on NPF events for all sites.

450 There are though some significant differences in the wind conditions for NPF events compared to

451 average conditions. At FINRU, NPF events were more common with northerly wind directions, as

452 was also found by Nieminen et al. (2014) and Nilsson et al. (2001). This is probably due to the

453 lower condensation sink which can be associated with the lower relative humidity also found for

454 incoming winds from that sector and explains the lower temperatures found with NPF events at this

455 site. Similarly, at FINUB NPF events were favoured by wind directions from the northerly sector,

456 while there is almost a complete lack of NPF on southerly winds. This is due to its position at the

457 north of both the city centre and the harbour, though winds from that sector are not common in

458 general for that site. Finally, the wind profile for NPF events at FINRO also favours northerly winds

459 with an almost complete absence of NPF in southerly winds, probably due to the elevated pollutant

460 concentrations and condensation sink associated with them.

461 At all sites, NPF event days had a lower condensation sink compared to the average for the site. The

462 seasonal variation of NPF events in Finland favouring spring, was explained by earlier work as the

463 result of the seasonal variation of H_2SO_4 concentrations (Nieminen et al., 2014), which in the area

464 peak in spring. The variation of H_2SO_4 concentrations is directly associated with SO_2 concentrations

465 in the area, which follow a similar trend. The seasonal variation of NPF events at FINRO though

466 cannot be explained by the variation of H_2SO_4 in the area. SO_2 concentrations, which were available

467 only for the nearby urban background site at Kalio (about 3 km away from FINRO) and may

468 provide information upon the trends of SO_2 in the greater area, peak during January (probably due

469 to increased heating in winter and the limited oxidation processes due to lower incoming solar

470 radiation) and are higher during spring months compared to summer. In general, the variation of

471 pollutant concentrations and the condensation sink is not great for the spring and summer seasons.

472 The only variable out of the ones considered that may explain to an extent the seasonality of NPF

473 events at the site is the increased concentrations of PM_{10} found for spring months, which might be

474 associated with road sanding and salting that takes place in Scandinavian countries during the

475 colder months (Kupiainen et al., 2016) with emissions to the ambient air during spring months

476 (Stojiljkovic et al., 2019). The source of these particles though is uncertain, as no major differences

477 in the wind roses are found between the two seasons. Another study by Sarnela et al. (2015) at a

478 different site in southern Finland attributed the seasonality of NPF events in Finland to the absence

479 of H_2SO_4 clusters during summer months due to a possible lack of stabilizing agents (e.g.

480 ammonia). This could explain the limited number of small particles (smaller than 10 nm) at the

481   background sites during summer. In the more polluted environment at a roadside site these agents

482   may exist, but such data was unfortunately not available.

483

484   Finally, a feature mentioned by Hao et al. (2018) in their study at the site of Hyytiälä, in which late

485   particle growth is observed was also found in this study. This happened on about 20% of NPF days

486   at FINRU (and a number of non-event days) and in most cases in early spring (before mid-April) or

487   late autumn (after mid-September). New particles were formed and either did not grow or grew very

488   slowly until later in the day when growth rates increased (Figure S2). In all these cases, growth

489   started when solar radiation was very low or zero, which probably associates the growth of particles

490   with nighttime chemistry leading to the formation of organonitrates (as found by the same study). A

491   similar behaviour was also occasionally found at FINUB. Particle growth at late hours is not a

492   unique feature for the Finnish sites, as it was found at all sites studied. What is different in the

493   specific events is the lack or very slow growth during the daytime. Lower temperature (-0.81°C),

494   incoming solar radiation (112 Wm^{-2}) and higher relative humidity (68.4%) occurred on event days

495   with later growth, while no clear wind association was found. Lower concentrations of organic

496   matter and nitrate were found throughout the days with later growth compared to the rest of the

497   NPF days. The very high average particle number concentration in the smaller size bins is due to

498   particles, though not growing to larger sizes for some time, persisting in the local atmosphere for

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

500

3.3.4 Spain

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

While NPF events with subsequent growth of the particles were rare during summer, cases of bursts of particles in the smallest size range available were found to occur frequently, especially in August and July (the month with the fewest NPF events, despite the favourable meteorological conditions). In such cases, a new mode of particles appears in the smallest size available, persisting for many hours though without clear growth (brief or no growth is only observed), as reported by Dall'Osto

521   et al. (2012). Due to the lack of growth of the particles these burst events do not qualify as NPF

522   events using the criteria set in the present study. These burst events are associated with southerly

523   winds (known as Garbí-southwest and Migjorn-south in Catalan, which are common during the

524   summer in the area) that bring a large number of particles smaller than 30 nm to the site from the

525   nearby airport (located about 15 km to the southwest) and port (7 km south), as well as Saharan

526   dust, increasing the concentrations of PM (Rodríguez et al., 2001) and thus suppressing NPF events

527   due to the increased condensation sink.

528

529   Finally, the wind direction profile at SPARU appears to have a daily trend, with almost exclusively

530   stronger southeasterly winds at about midday (Figure S3), probably due to a local mesoscale

531   circulation caused by the increased solar activity during that time (which results in different heating

532   patterns of the various land types in the greater area). These incoming southeast winds are more

533   polluted and have a higher condensation sink (being affected by the city of Barcelona), and almost

534   consistently bring larger particles at the site during the midday period. This may explain to an

535   extent the lowest probability for NPF events from that sector, despite the very high concentrations

536   of O_3 associated with them, with some extreme values well above 100 $\mu g\ m^{-3}$ (Querol et al., 2017).

537   The highest average growth rates are also found from that direction.

538

539

540

3.3.5   Greece

Temperature and wind speed are found to be lower on NPF event days at the Greek sites, though the differences are minimal and are associated with the seasonal variability of the events. The wind rose in GREUB mainly consists of northeasterly and southwesterly winds. Due to its position, the site is heavily affected by emissions in Athens city centre with westerly winds, resulting in increased particle number concentrations and condensation sink. Despite this, the highest NPF probability and growth rates were found with a northwesterly wind directions. This may be due to them being associated with the highest solar radiation (probably the result of seasonal and diurnal variation), temperature and the lowest relative humidity, along with the highest condensation sink and particle number concentrations of almost all sizes. Chemical composition data was not available for GREUB, though SO_2 concentrations are rather low in Athens and kept declining after the economic crisis (Vrekoussis et al., 2013). The seasonality of SO_2 concentration in Athens favoured winter months and was at its lowest during summer for the period studied (ΥΠΕΚΑ, 2012) (this trend changed later as SO_2 concentrations further declined), which may also be a factor in the seasonality of NPF events, though this will be further discussed later.

At the GRERU site, the wind profile is mainly westerly, and though it coincides with the most important source of pollutants in the area, the city of Herakleio, its effect while observable is not significant due to the topography in the area. The wind profile for NPF events is similar to the average with significantly higher wind speeds ($p < 0.001$). In general, GRERU has very low

561 pollutant concentrations, with an average NO of 0.073 µg m^{-3}, NO$_2$ of 0.52 µg m^{-3} and SO$_2$ in

562 concentrations below 1 ppb (Kouvarakis et al., 2002). Due to this, the differences in the chemical

563 composition in the atmosphere are also minimal. For the specific site two different patterns of

564 development of NPF events were found. In one case, NPF events occurred in a rather clear

565 background, while in the other one they were accompanied with an increase in number

566 concentrations of larger particles or a new mode appearing at larger sizes (about a third of the

567 events). No differences were found in the seasonal variation between the two groups; increased

568 gaseous pollutant and particulate organic carbon concentrations were found for the second group

569 (though the differences were rather small) and a wind rose that favoured southwesterly winds

570 (originating from mainland Crete) instead of the northwesterly (originating from the sea) ones for

571 the first group. The growth rate for the two groups was found to be 3.56 nm h^{-1} for the first group

572 and 4.17 nm h^{-1} for the second, which might be due to the increased presence of condensable

573 compounds. As the dataset starts from the particle size of 8.77 nm, the possibility that these

574 particles were advected from nearby areas should not be overlooked, though they persisted and

575 grew at the site. Other than that, no significant differences were found for the different wind

576 directions.

577

578 As mentioned earlier, both sites had a very low frequency of events and J$_{10}$ in summer similar to

579 previous studies also reporting few or no events during summer (Vratolis et al., 2019; Ždímal et al.,

580 2011), though the incoming solar radiation is the highest and relative humidity is the lowest during

581 that season. This variation was also observed by Kalivitis et al. (2012) who associated the seasonal

582 variation of NPF events at GRERU with the concentrations of atmospheric ions. The effect of the

583 Etesian winds (known as Meltemia in Greek), which dominate the southern Aegean region during

584 the summer months though should not be overlooked. These result in very strong winds with an

585 average wind speed of 8.15 m s^{-1} during summer at the Finokalia site, and increased turbulence

586 found in all years with available data, affecting both sites of this study. During this period, $N_{<30nm}$

587 drops to half or less compared to other seasons at both sites, while $N_{>100nm}$ is at its maximum due to

588 particle aging (Kalkavouras et al., 2017), increasing the condensation sink, especially in GRERU

589 (the effect in GREUB is less visible due to both the wind profile, blowing from east which is a less

590 polluted area, as well as the reduction of urban activities during summer months in Athens). Both

591 the increased condensation sink and turbulence are possible factors for the reduced number of NPF

592 events found at both sites in summer. Another possible factor is the effect of high temperatures in

593 destabilising the molecular clusters critical to new particle formation.

594

595 **3.4   Region-Wide Events**

596 Region-wide events are NPF events which occur over large-scale areas, that may cover hundreds of

597 kilometres (Shen et al., 2018). In the present study, NPF events that took place on the same day at

598 both background sites (urban background and rural) are considered as regional and their conditions

599 are studied (Table S8). The background sites in Greece were not considered due to the great

600 distance between them (about 350 km). There is also uncertainty for the background sites in

601 Finland, where the distance is about 190 km, though a large number of days were found when NPF

602 events occurred on the same day. The number of region-wide events per season (or the fraction of

603 region-wide events to total NPF events) is found in Figure 8 and it appears as if they are more

604 probable in spring at all the sites of the present study (apart from Finland, though the number of

605 events in winter was low), despite the differences found in absolute numbers.

606

607 In Denmark, about 20% of NPF events in DENRU were regional (the percentage is higher for

608 DENUB due to the smaller number of events, at 29%). The relatively low frequency of region-wide

609 NPF events can be explained by the different seasonal dependence of NPF events (region-wide NPF

610 events were more frequent in spring compared to the average due to the seasonality of NPF events

611 in DENUB). Compared to local NPF event conditions, higher wind speed and solar radiation, as

612 well as O_3 and marine compound concentrations (results not included) were found, while the

613 concentrations of all pollutants (such as NO, NO_x, sulphate, elemental and organic carbon) were

614 lower. These cleaner atmospheric conditions are also confirmed by the lower CS associated with

615 region-wide events, which is probably one of the most important factors in the occurrence of these

616 large-scale events. The exceptions found at DENRU (increased relative humidity and less incoming

617 solar radiation) are probably due to the different seasonality between local and region-wide NPF

618 events at the site, though region-wide events rarely present similar characteristics at different sites

619 even in the same country due to the differences in the initial meteorological and local conditions

620 (Hussein et al., 2009). The growth rates of region-wide events were found to be lower than those of

32

621   local events at both sites, which is probably associated with the limited concentrations of

622   condensable compounds due to the cleaner air masses of marine origin (as confirmed by the higher

623   concentrations of marine compounds).

624

625   In Germany, the majority of NPF events of this study were region-wide (about 60%). Compared to

626   the average, the meteorological conditions found for NPF event days compared to average

627   conditions were more distinct for the region-wide events, with even lower wind speed and relative

628   humidity and higher temperature and solar radiation, and all of these differences were significant (p

629   < 0.001). At GERRU where chemical composition data was available, higher concentrations of

630   particulate organic carbon and sulphate and lower nitrate concentrations were found. The

631   differences are significant ($p < 0.001$) and may explain the higher growth rates found in region-wide

632   events at both sites compared to the average, which is a unique feature. It should be noted that as

633   the majority of NPF events at the German sites are associated with easterly winds, it is expected that

634   in most cases the region-wide events will be associated with these, carrying the characteristics that

635   come along with them (increased growth rates and concentrations of organic carbon, as discussed in

636   Section 3.2).

637

638   In Finland, about a quarter of the NPF event days at FINRU (26%) occurred on the same day as at

639   FINUB (the frequency is a lot higher for FINUB, at 39%). As in Germany, the meteorological

640   conditions found on NPF event days compared to average conditions were more distinct during

641   region-wide events. Thus, for both sites temperature and relative humidity were lower while solar

642   radiation was higher. The different trend found for the wind speed at the two sites (being higher on

643   average NPF days at FINRU and lower at FINUB compared to average conditions) was enhanced

644   as well at the two sites for region-wide events. At FINRU where chemical composition data was

645   available, NO_x and SO_2 had similar concentrations on region-wide event days compared to the

646   averages on total event days, while O_3 was significantly higher ($p < 0.001$). As at most other sites,

647   the growth rate was found lower on region-wide event days compared to the average at both sites.

648

649   Finally, in Spain the datasets of the two sites did not overlap greatly, having only 322 common

650   days. Among these days, 13 days presented with NPF events that took place simultaneously at both

651   sites, with smaller growth rates on average compared to local events (43% of the events at SPARU

652   and 36% of the events at SPAUB in the period 8/2012 to 1/2013 and 2014 when data for both sites

653   were available). Due to the small number of common events the results are quite mixed with the

654   only consistent result being the lower relative humidity and higher O_3 concentrations for regional

655   events at both sites, though none of these differences is significant. The wind profile at SPAUB

656   seems to further favour the cleaner sector, with the majority of incoming winds being from the NW

657   and even higher wind speeds (though with low significance). The result is similar at SPARU,

658   though less clear and with lower wind speeds.

659

34

660 These results are in general in agreement with those found in the UK in a previous study, where

661 meteorological conditions were more distinct on region-wide event days compared to local NPF

662 events; pollutant concentrations were lower as well as the growth rates of the newly formed

663 particles (Bousiotis et al., 2019).

664

665 Common events were also found between either of the background sites and the roadside, but they

666 were always fewer in number, due to the difference in their temporal variability compared to the

667 background sites, resulting from the effect of roadside pollution.

668

669 **3.5   The Effect of NPF Events on the Ultrafine Particle Concentrations**

670 The NSF is a metric of the effect of NPF events upon particle concentrations on either the days of

671 the events or over a larger timescale. Both the NSF_{NUC} and NSF_{GEN} were calculated for all sites of

672 this study and the results are presented in Figure 9. For almost all rural background sites NSF_{NUC},

673 which indicates the effect of NPF on ultrafine particle concentrations on the day of the event, was

674 found to be greater than 2 (the only exception was GERRU), which means that NPF events more

675 than double the number of ultrafine particles (particles with diameter smaller than 100 nm) at the

676 site on the days of the events, as NPF events are one of the main sources of ultrafine particles in this

677 type of sites, especially below 30 nm. This reaches up to 4.18 found at FINRU (418% more

678 ultrafine particles on the day of the events – 100% being the average), showing the great effect NPF

679 events have on rather clean areas. The long-term effect was smaller, and it was found that at FINRU

680 NPF events increase the number of ultrafine particles by an additional 130% in general. The effect

681 of NPF events was a lot smaller at the urban sites, though still significant at urban background sites

682 (reaching up 240% at FINUB on the days of events), while roadsides had the smallest NSF

683 compared to their respective background sites. This is because of the increased effect of local

684 sources such as traffic or heating, and the associated increased condensation sink found within these

685 sites, which cause the new particles to be scavenged by the more polluted background.

686

687 The calculation of NSF at the sites around Europe showed a weakness of the specific metric, which

688 points to the need for more careful interpretation of the results of this metric, especially at roadside

689 sites. At FINRO, the NSF_{NUC} provided a value smaller than 1, which translates as ultrafine particles

690 are lost instead of formed on NPF event days. This though is the result of both the sharp reduction

691 in particle number concentrations at all modes that are required for NPF events to occur at a busy

692 roadside (much lower condensation sink), as well as a difference in the ratio between smaller to

693 larger particles (smaller or larger than 100 nm) on NPF event days (favouring the larger particles) at

694 the specific site. Similarly, the long-term effect of NPF events at the site was found to be 1, which

695 means that NPF events appear to cause no changes in the number concentration of ultrafine

696 particles.

697

698

699

4.   DISCUSSION

4.1   Variability of the Frequency and Seasonality of the Events

700

701

702 A higher frequency of NPF events at the rural background sites compared to roadsides was found

703 for all countries with available data for both types of site. This pattern comes in contrast with what

704 was found for the more polluted Asian cities (Peng et al., 2017; Wang et al., 2017), where NPF

705 events were more frequent at the urban sites. This is probably associated with the even greater

706 abundance of condensable species (which further enhances the growth of the particles, thus

707 increasing their chance of survival), deriving from anthropogenic emissions, found in Asian

708 megacities compared to European ones and results in a greater frequency of NPF events in Asian

709 cities, even compared to the most polluted cities in Europe. This contrast emphasises the differences

710 in the occurrence of NPF events between the polluted cities in Europe and Asia, which are

711 associated with the level of pollution found in them, as well as the influence that the level of

712 pollution has on the occurrence of NPF events in general.

713

714 The type of site dependence found in Europe together with the average conditions found on NPF

715 event days compared to the average for each site, underline the importance of clear atmospheric

716 conditions (high solar radiation and low relative humidity and pollutant concentrations) at all types

717 of sites in Europe, especially for region-wide events. The temperature and wind speed presented

718 more diverse results which in many cases are associated with local conditions. The origin of the

719 incoming air masses though, appears to have a more important influence upon the NPF events.

720 Cleaner air masses tend to have higher probability for NPF events, a result which was consistent

721 among the sites of this study regardless of their type.

722

723 The frequency of NPF events at roadsides peaked in summer in all three countries with available

724 data. Greater variability in the seasonality of NPF events was found at the background sites. The

725 urban background sites presented more diverse results, for both the occurrence and development of

726 NPF events, especially compared to rural background sites. The within-week variation of NPF

727 events was found to favour weekends in most cases, as the pollution levels decrease, due to the

728 weekly cycle, especially at the roadsides. As background sites have smaller variations between

729 weekdays and weekends, the within-week variation of NPF events is smaller at the urban

730 background sites and almost non-existent at the rural background sites. Finally, it should be noted

731 that no clear interannual trend was found in the frequency of the events for any site, even for those

732 with longer datasets.

733

734 **4.2   Variability and Seasonality of the Formation and Growth Rate**

735 The growth rate of the newly formed particles was found to be higher at all the roadsides compared

736 to their respective rural and urban background. The picture is similar for J_{10}, (the rate of formed

737 particles associated with NPF events that reached 10 nm diameter), for which urban background

738 sites were between their respective rural background sites and the roadsides with the sole exception

739 of DENUB (the difference with DENRU is rather small though). The growth and formation rate at

740   the rural background sites (apart from the Greek site) were found to be higher in summer compared

741   to the other seasons. On the other hand, the seasonality of the growth rate at the roadsides is not

742   clear but the formation rate peaks in the autumn at all three roadside sites. While the trend at the

743   rural sites is probably associated with the enhanced photochemistry and increased concentrations of

744   BVOCs during summer, the seasonality of the growth rate at the roadside sites is more difficult to

745   explain and probably shows the smaller importance of the BVOCs compared to the compounds of

746   anthropogenic origin (which are in less abundance in summer) in this type of environment. In

747   general, higher temperatures were associated with higher growth rates. This though applies only for

748   the specific conditions at each site and cannot be used as a general rule for the expected growth rate

749   at a site, as locations with higher temperatures did not present higher growth rates. Additionally, the

750   origin of the incoming air masses appears to have an effect on the growth of the particles as well. In

751   most of the sites in this study, incoming air masses from directions associated with higher

752   concentrations of pollutants presented higher growth rates of the newly formed particles. The effect

753   of the different wind directions upon the formation rate was more complex and a definitive

754   conclusion cannot be made. Finally, as with the frequency of the events, no significant interannual

755   trend was found in the variation of the formation or the growth rate across the sites studied.

756

757   **4.3   Effect of Local Conditions in the Occurrence and Development of NPF Events**

758   Apart from the general meteorological and atmospheric conditions that affect the occurrence and

759   the metrics of NPF events, conditions with a more local character were found to play a significant

760 role as well. These include synoptic systems, such as the one occurring during the summer at the

761 Greek sites, affecting the frequency and seasonality of the events. As a result, sites or seasons with

762 conditions that favoured NPF presented decreased frequency of events and unexpected seasonality,

763 due to the increased turbulence caused by such pressure systems. Additionally, local sources of

764 pollution can also have a significant impact in the temporal trends and metrics of the events, even

765 for sites of very close proximity. One such example was the urban sites in Denmark, which despite

766 being affected by the same source of pollution (the nearby port) and being only a few kilometres

767 away from each other, presented different outcomes in the occurrence of the events. This was due to

768 the different atmospheric composition found between them, being a background and a roadside site,

769 which led to a different response in that local variable. In this case, the effect of the specific source

770 was more prominent at the urban background site compared to the roadside, resulting in fewer NPF

771 events, as the newly formed particles were more effectively supressed at the urban background site,

772 due to their slower growth.

773

774 **5.   CONCLUSIONS**

775 There are different ways to assess the occurrence of new particle formation (NPF) events. In this

776 study, the frequency of NPF events, the formation and growth rate of the particles associated with

777 secondary formation of particles and not primary emissions, at 13 sites from five countries in

778 Europe are considered. NPF is a complicated process, affected by many meteorological and

779 environmental variables. The seasonality of these variables, which varies throughout Europe, results

780   in the different temporal trends found for the metrics studied in this paper. Apart from

781   meteorological conditions though, some of which have a uniform effect (such as the solar radiation

782   intensity and relative humidity), many local variables can also have a positive or negative effect in

783   the occurrence of these events. Sites with less anthropogenic influence seem to have temporal

784   trends dependant on the seasonality of synoptic conditions and general atmospheric composition.

785   The urban sites though and especially those with significant sources of pollution in close proximity,

786   present more complex trends as the NPF occurrence depends less upon favourable meteorological

787   conditions and more upon the local atmospheric conditions, including composition. As NPF event

788   occurrence is based on the balance between the rapid growth of the newly formed particles and their

789   loss from processes, such as the evaporation or coagulation of the particles, the importance of

790   significant particle formation, fast growth (which is enhanced by the increased presence of

791   condensable compounds from anthropogenic activities found in urban environments) and low

792   condensation sink is increased within such environments, also affecting the temporal trends of the

793   events, making them more probable during periods with smaller pollution loads (e.g. summer,

794   weekends). This explains the smaller frequency of NPF events at roadside sites compared to their

795   respective background sites, despite the greater formation and growth rates observed in them.

796   Consequently, NPF events have a smaller influence on the ultrafine particle load at the urban sites

797   compared to background sites, due to both the increased presence of ultrafine particles from

798   anthropogenic emissions as well as the smaller probability of ultrafine particles to survive in such

799   environments.

800 Nevertheless, NPF events are an important source of ultrafine particles in the atmosphere for all

801 types of environment and are an important factor in the air quality of a given area. The present

802 study underlines the importance of both the synoptic and local conditions on NPF events, the mix of

803 which not only affects their development but can also influence their occurrence even in areas of

804 very close proximity. NPF is a complex process, affected by numerous variables, making it

805 extremely difficult to predict any of its metrics without considering multiple factors. Since the

806 mechanisms and general trends in NPF events are yet to be fully explained and understood, more

807 laboratory and field studies are needed to generate greater clarity and predictive capability.

808

809 **DATA ACCESSIBILITY**

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

811 https://doi.org/10.25500/edata.bham.00000467

812

813 **AUTHOR CONTRIBUTIONS**

814 The study was conceived and planned by MDO and RMH who also contributed to the final

815 manuscript. The data analysis was carried out by DB who also prepared the first draft of the

816 manuscript. AM, JKN, CN, JVN, HP, NP, AA, GK, SV and KE have provided with the data for the

817 analysis. FDP, XQ, DCB and TP provided advice on the analysis.

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820 **COMPETING INTERESTS**

821 The authors have no conflict of interests.

822

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1390 **Table 1:** Location and data availability (seasonal data availability is found in Table S4) of the sites in the present study. In the studies referenced an extended description of the sites can be found.

Site	Location	Available data	Meteorological data location	Data availability	Reference
DENRU	Lille Valby, 25 km W of Copenhagen, (55º 41' 41" N; 12º 7' 7" E) (2008 – 6/2010) Risø, 7 km north of Lille Valby, (55° 38' 40" N; 12° 5' 19" E) (7/2010 – 2017)	DMPS and CPC (5.8 - 700 nm, 65.4% availability), NO, NO_x, SO_2, O_3, minerals, OC, EC, NO_3^-, SO_4^{2-}, NH_4^+	Ørsted – Institute station	2008 – 2017	Ketzel et al., 2004
DENUB	Ørsted - Institude, 2 km NE of the city centre, Copenhagen, Denmark (55º 42' 1" N; 12º 33' 41" E)	DMPS and CPC (5.8 - 700 nm, 59.0% availability), NO, NO_x, O_3, minerals, EC	On site	2008 – 2017	Wang et al., 2010
DENRO	H.C. Andersens Boulevard, Copenhagen, Denmark (55º 40' 28" N; 12º 34' 16" E)	DMPS and CPC (5.8 - 700 nm, 65.0% availability), NO, NO_x, SO_2, O_3, minerals, OC, EC, NO_3^-, SO_4^{2-}, NH_4^+	Ørsted – Institute station	2008 – 2017	Wang et al., 2010
GERRU	Melpitz, 40 km NE of Leipzig, Germany (51º 31' 31.85" N; 12º 26' 40.30" E)	TDMPS with CPC (4.8 - 800 nm, 87.1% availability), OC, NO_3^-, SO_4^{2-}, NH_4^+, Cl^-	On site	2008 – 2011	Birmili et al., 2016
GERUB	Tropos, 3 km NE from the city centre of Leipzig, Germany (51º 21' 9.1" N; 12º 26' 5.1" E)	TDMPS with CPC (3 - 800 nm,88.0% availability)	On site	2008 – 2011	Birmili et al., 2016
GERRO	Eisenbahnstraße, Leipzig, Germany (51º 20' 43.80" N; 12º 24' 28.35" E)	TDMPS with CPC (4 - 800 nm, 64.4% availability)	Tropos station	2008 – 2011	Birmili et al., 2016
FINRU	Hyytiälä, 250 km N of Helsinki, Finland (61º 50' 50.70" N; 24º 17' 41.20" E)	TDMPS with CPC (3 – 1000 nm, 98.7% availability), NO, NO_x, SO_2, O_3, CO, CH_4, VOCs, H_2SO_4	On site	2008 – 2011 & 2015 – 2018	Aalto et al., 2001
FINUB	Kumpula Campus 4 km N of the city centre, Helsinki, Finland (60º 12' 10.52" N; 24º 57' 40.20" E)	TDMPS with CPC (3.4 - 1000 nm, 94.0% availability)	On site	2008 – 2011 & 2015 – 2018	Järvi et al., 2009
FINRO	Mäkelänkatu street, Helsinki, Finland (60º 11' 47.57" N; 24º 57' 6.01" E)	DMPS (6 - 800 nm, 90.0% availability), NO, NO_2, NO_x, O_3, BC and SO_2 from Kalio Station	Pasila station and on site	2015 – 2018	Hietikko et al., 2018
SPARU	Montseny, 50 km NNE from Barcelona, Spain (41º 46' 45" N; 2º 21' 29" E)	SMPS (9 – 856 nm, 47.7% availability), NO, NO_2, SO_2, O_3, CO, OM, SO_4^{2-}	On site	2012 - 2015	Dall'Osto et al., 2013
SPAUB	Palau Reial, Barcelona, Spain (41º 23' 14" N; 2º 6' 56" E)	SMPS (10.9 – 478 nm, 64.2% availability), NO, NO_2, SO_2, O_3, CO, BC, OM, SO_4^{2-}, $PM_{2.5}$, PM_{10}	On site	2012 – 2015	Dall'Osto et al., 2012

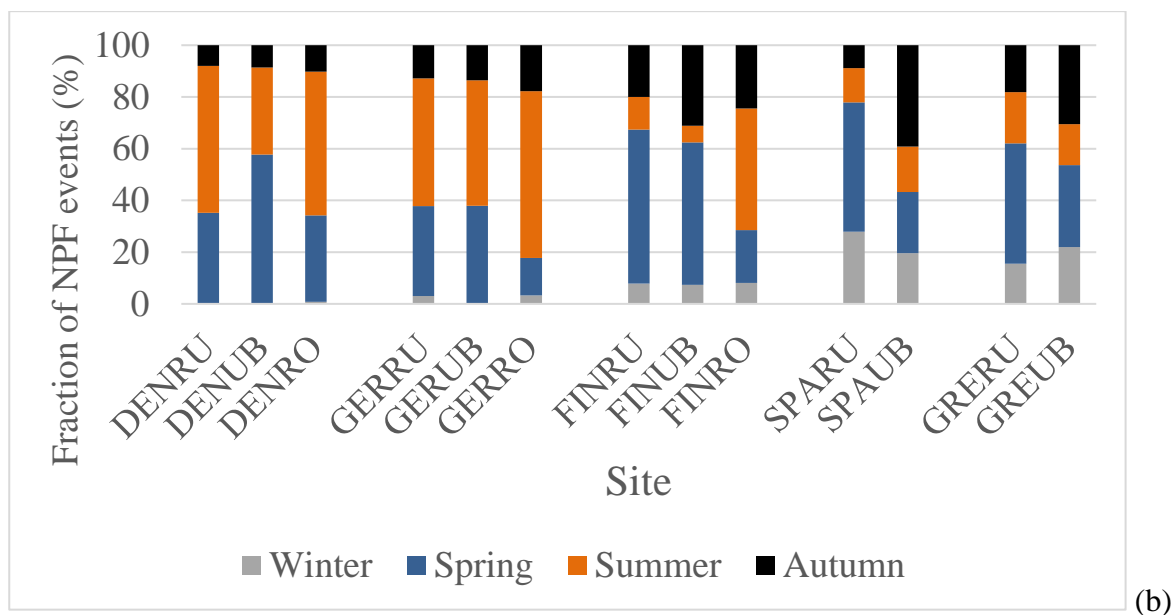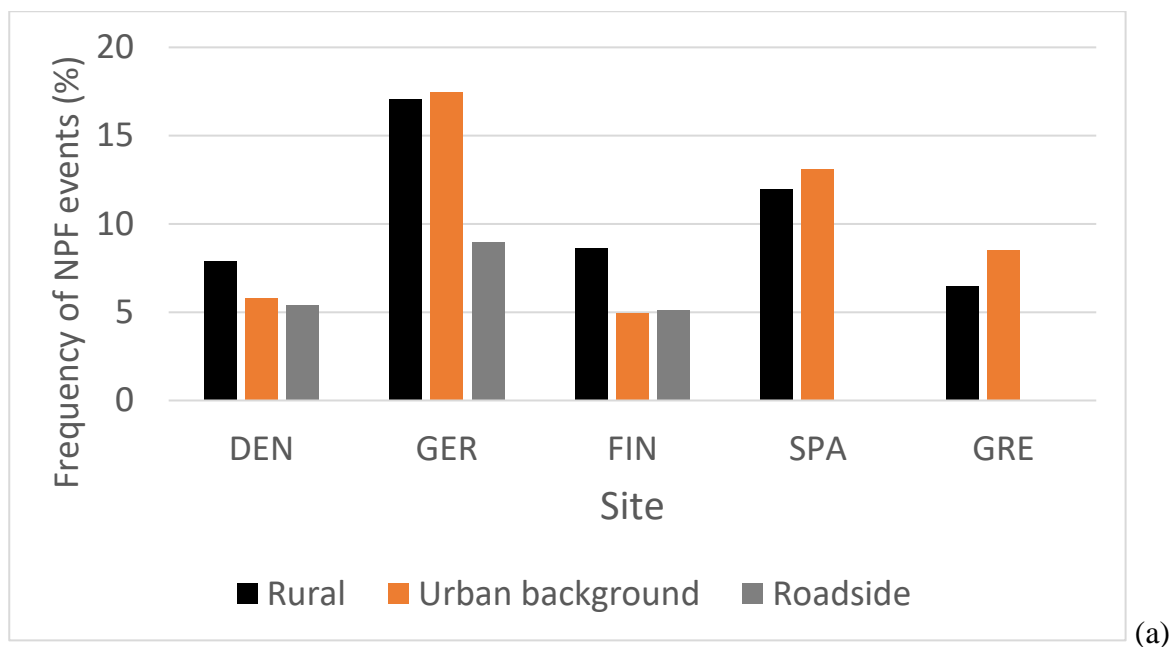|					
GRERU	Finokalia, 70 km E of Heraklion, Greece (35º 20' 16.8" N; 25º 40' 8.4" E)	SMPS (8.77 - 849 nm, 92.4% availability), NO, NO_2, O_3, OC, EC	On site	2012 – 2018	Kalkavouras et al., 2017
GREUB	"Demokritos", 12 km NE from the city centre, Athens, Greece (37º 59' 41.96" N; 23º 48' 57.56" E)	SMPS (10 – 550 nm, 77.2% availability)	On site	2015 – 2018	Vassilakos et al., 2005
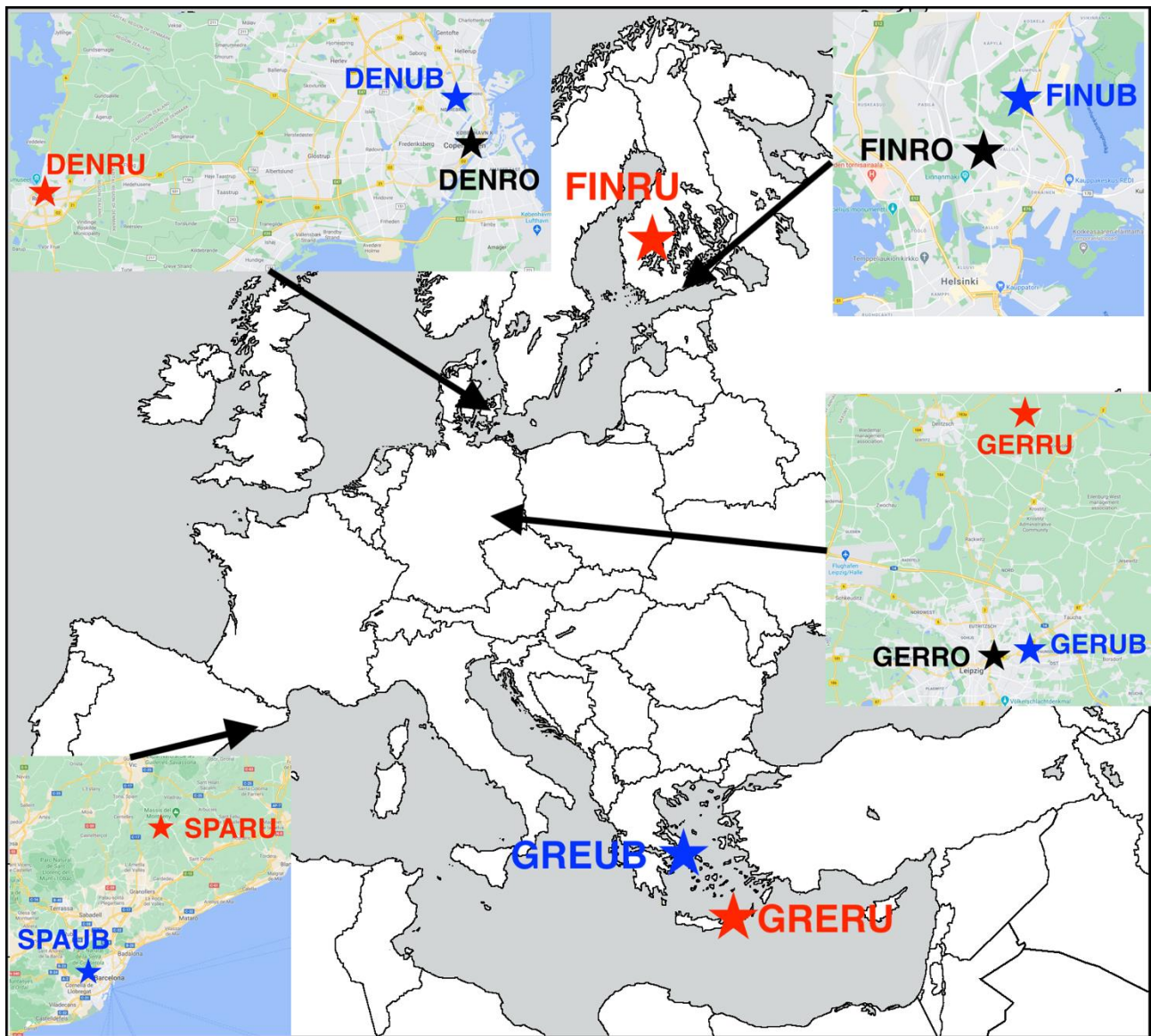
1395 **Figure 1:** Map of the areas of study.

1400

(a)

(b)

Figure 2: Frequency (a) and seasonal variation (b) of New Particle Formation events (Winter – DJF;
1405 Spring – MAM; Summer – JJA; Autumn – SON).
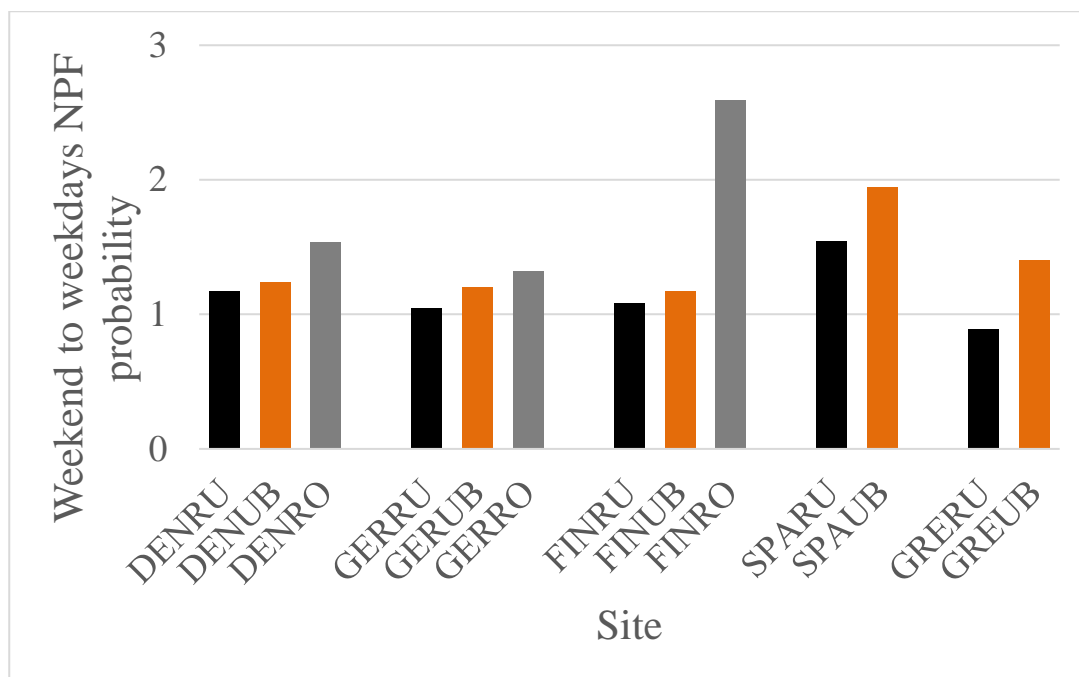
Figure 3: Ratio of New Particle Formation event probability between weekends to weekdays. The greater the ratio the more probable it is for an event to take place during weekends compared to weekdays.
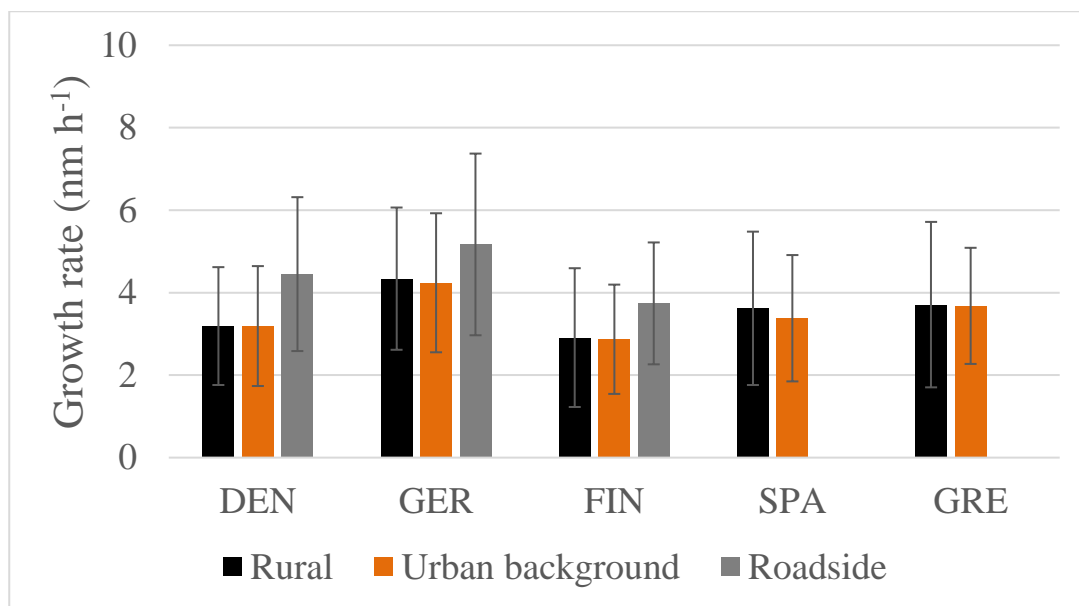
Figure 4: Growth rate of particles up to 30 nm (with standard deviations) during New Particle Formation events at all sites.

1415

(a)

(b)

1420

(c)
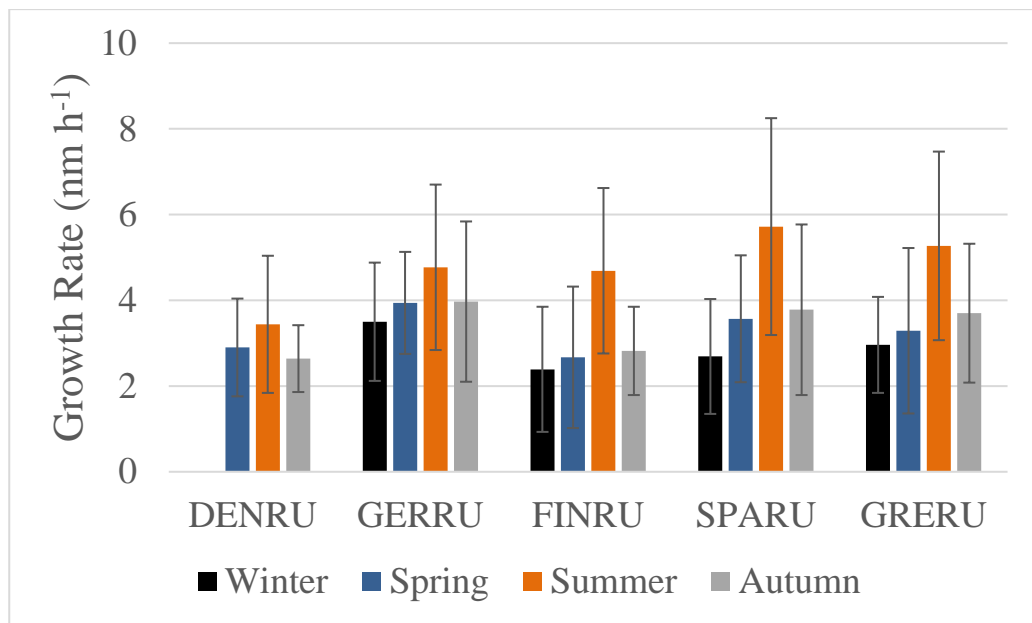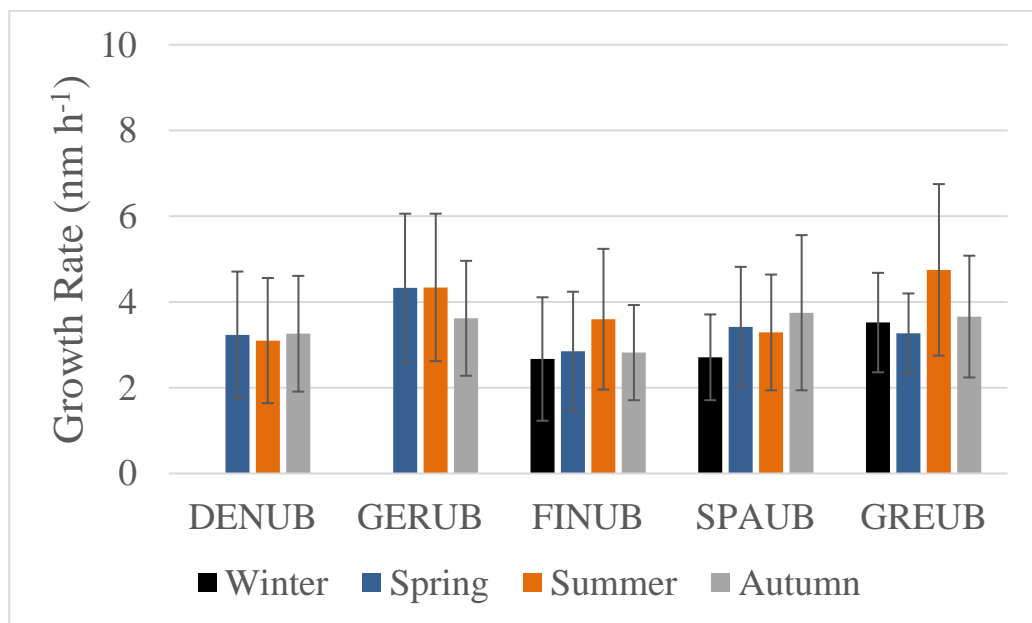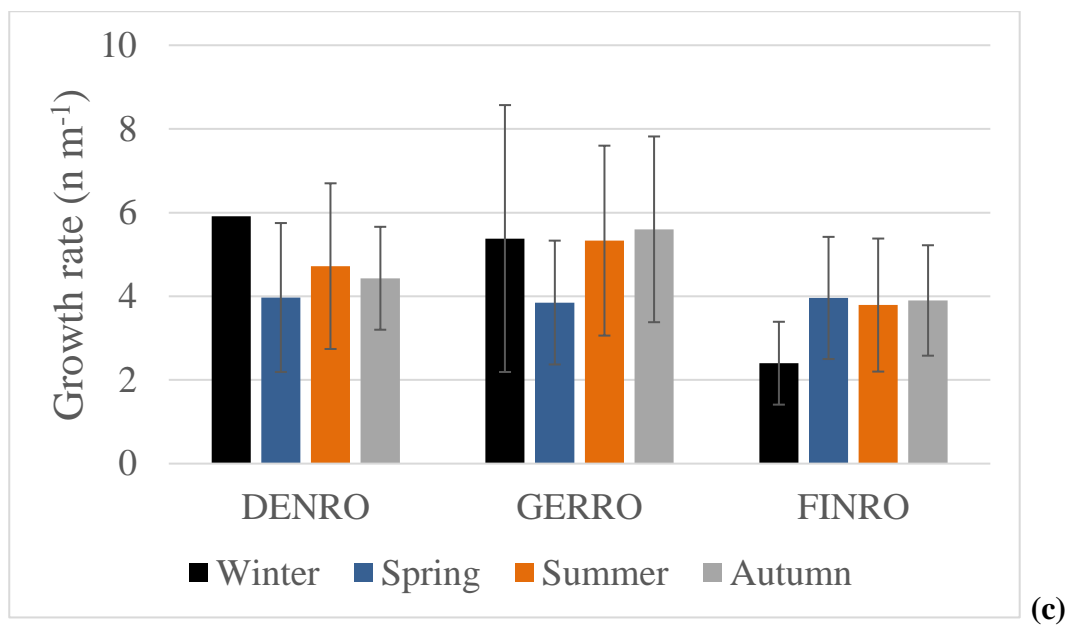
Figure 5: Seasonal variation of growth rate of particles up to 30 nm on New Particle Formation at (a) the rural background, (b) urban background and (c) roadside sites.
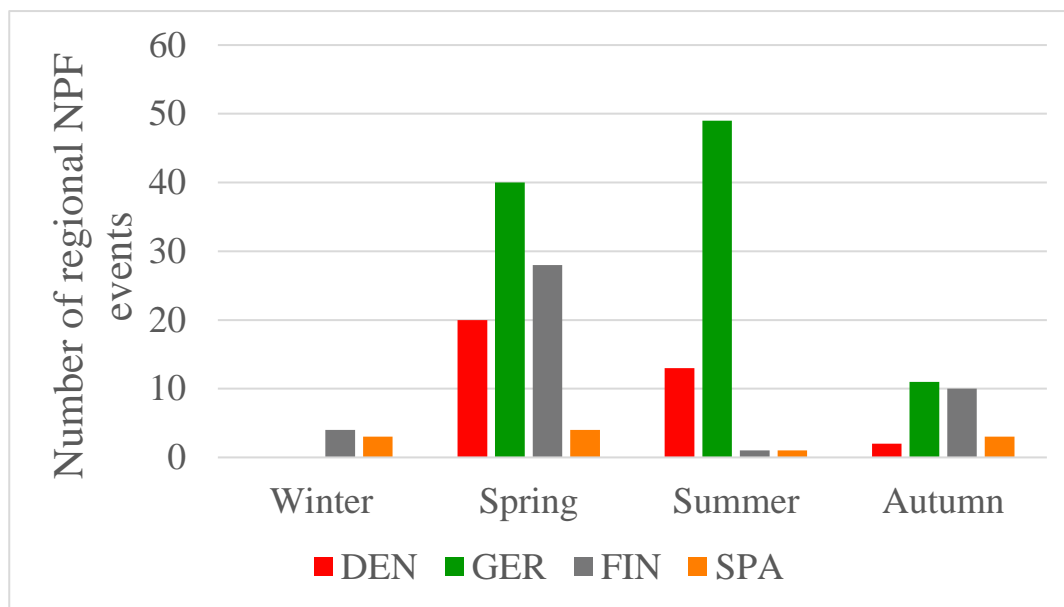
1430
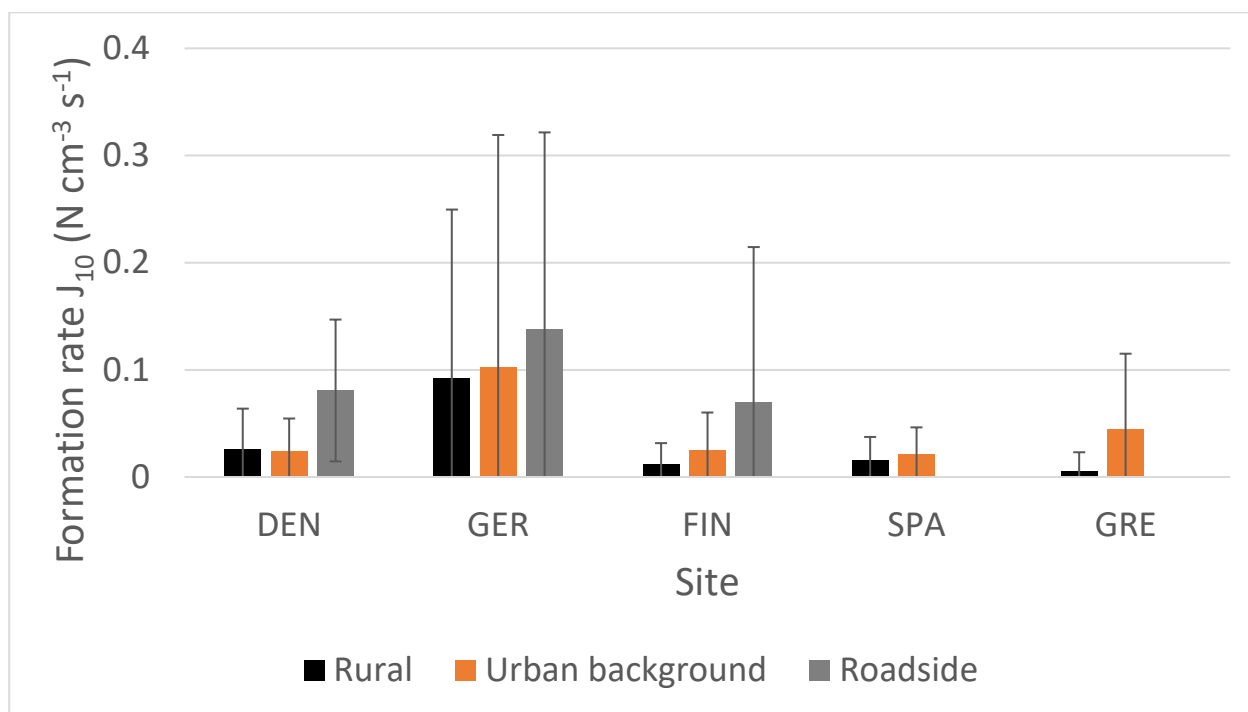
Figure 6: Formation rate of 10 nm particles (J_{10}) (with standard deviations) during New Particle Formation events at all sites.

1435

(a)

(b)

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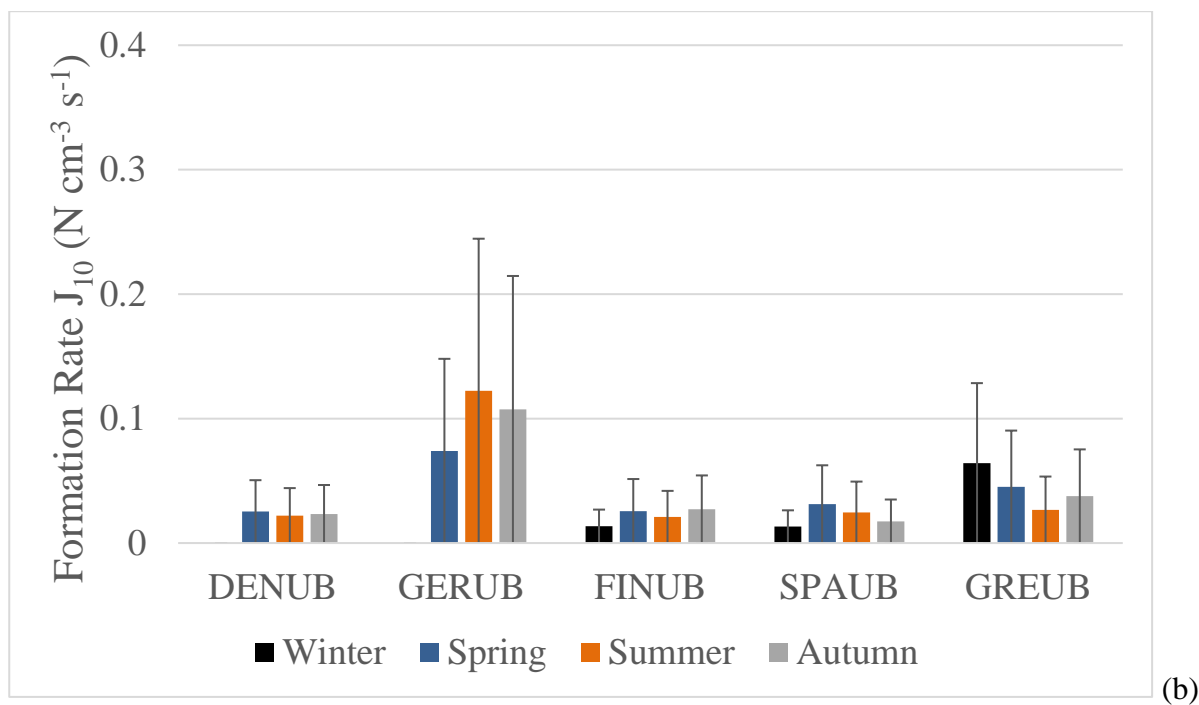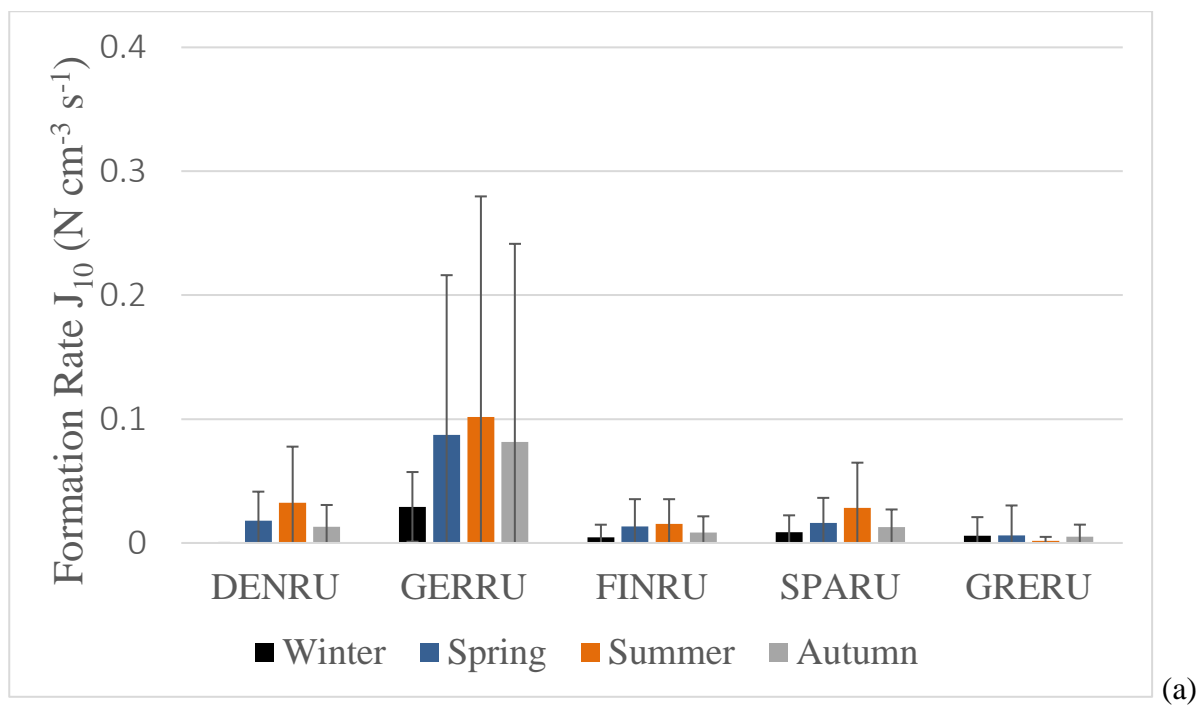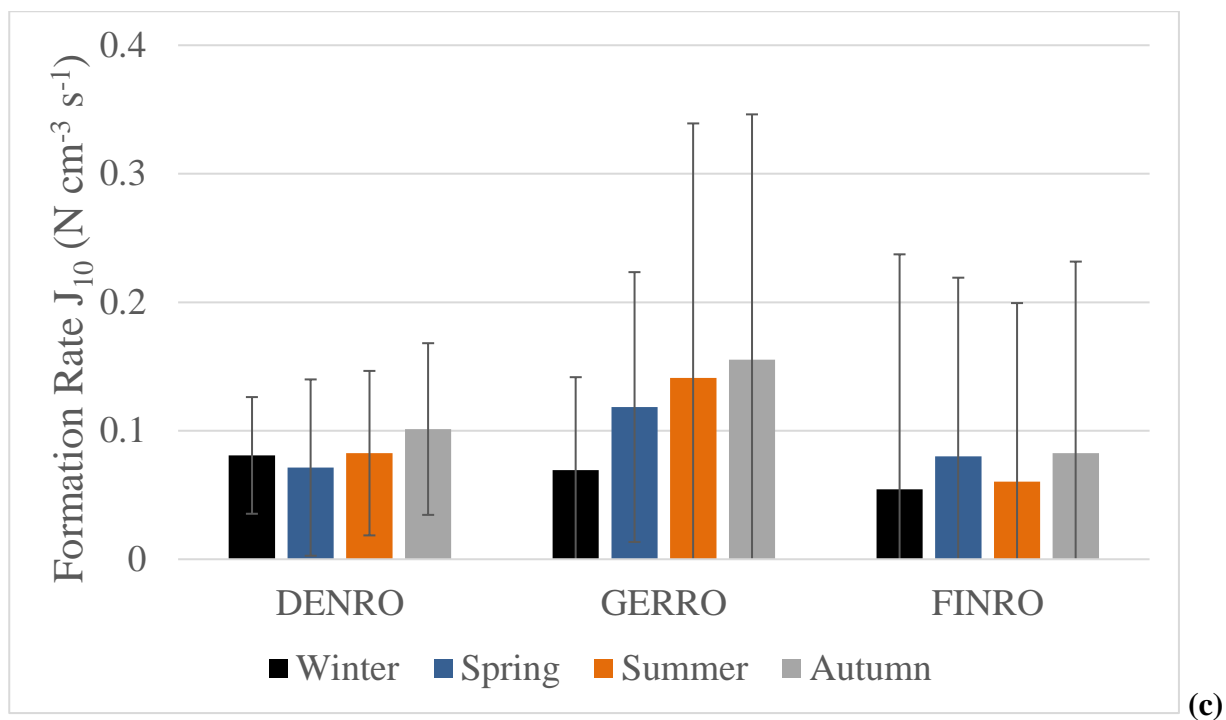
Figure 7: Seasonal variation of formation rate of 10 nm particles (J_{10}) (with standard deviations) from New Particle Formation events at (a) the rural background, (b) urban background and (c) roadside sites.
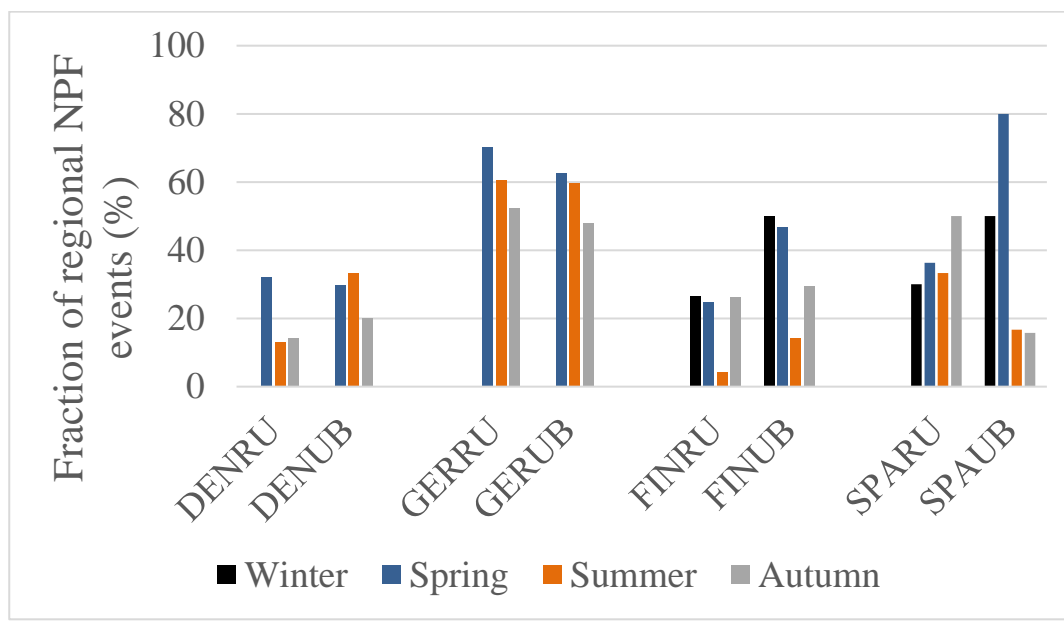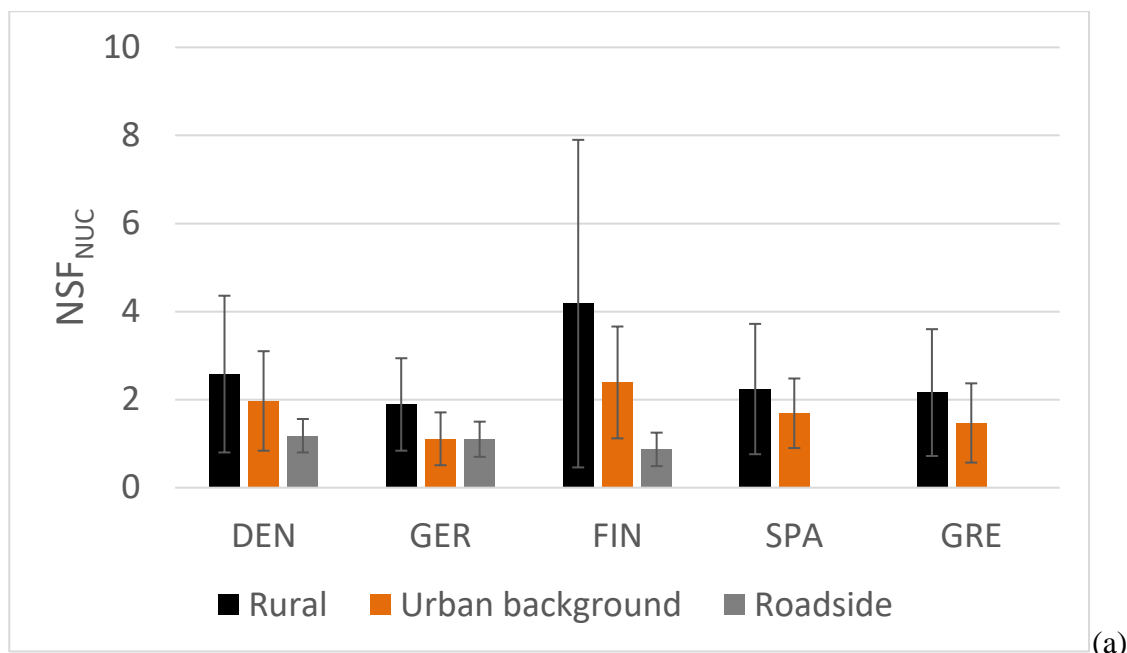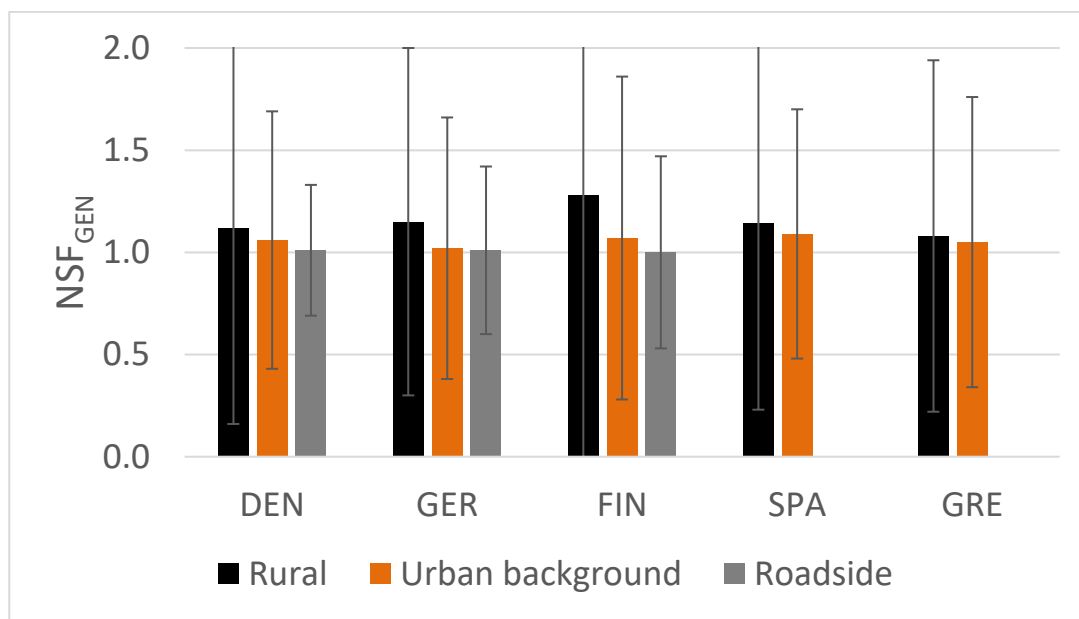
(a)

(b)

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

Figure 9: (a) NSF_{NUC} (average relative increase of ultrafine particles – particles of diameter up to 100 nm) due to New Particle Formation events on event days) and (b) NSF_{GEN} (average annual relative increase of ultrafine particles due to New Particle Formation events) at all sites.