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# 1 **New Particle Formation at a peri-urban agricultural site**

2

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## 22 **Highlights**

- 23 • NPF has been observed at a mixed peri-urban agricultural site
- 24 • NPF occurred during both the daytime and the night-time
- 25 • Night-time NPF events are suggested to be related to agricultural emissions, whereas  
26 daytime events are not

## 27 **Abstract**

28 New Particle Formation (NPF) is a major source of ultrafine particles that affect both air quality  
29 and climate. Despite emissions from agricultural activities having a strong potential to lead to NPF,  
30 little is known about NPF within agricultural environments. The aim of the present study was to  
31 investigate the occurrence of NPF events at an agricultural site, and any potential relationship  
32 between agricultural emissions and NPF events. A field campaign was conducted for 3 months at  
33 the FR-Gri-ICOS site (France), at an experimental farm 25 km west of Paris city centre.

34 16 NPF events have been identified from the analysis of particle number size distributions; 8 during  
35 the daytime, and 8 during the night-time. High solar radiation and ozone mixing ratios were  
36 observed during the days NPF occurred, suggesting photochemistry plays a key role in daytime  
37 NPF. These events were also associated with higher levels of VOCs such as isoprene, methanol, or  
38 toluene compared to non-event days. However, ammonia levels were lower during daytime NPF  
39 events, contributing to the hypothesis that daytime NPF events were not related to agricultural  
40 activities.

41 On the other hand, temperature and ozone were lower during the nights when NPF events were  
42 observed, whereas relative humidity was higher. During these nights, higher concentrations of NO<sub>2</sub>  
43 and ammonia were observed. As a result, agricultural activities, in particular the spreading of  
44 fertilizer on surrounding crops, are suspected to contribute to night-time NPF events.

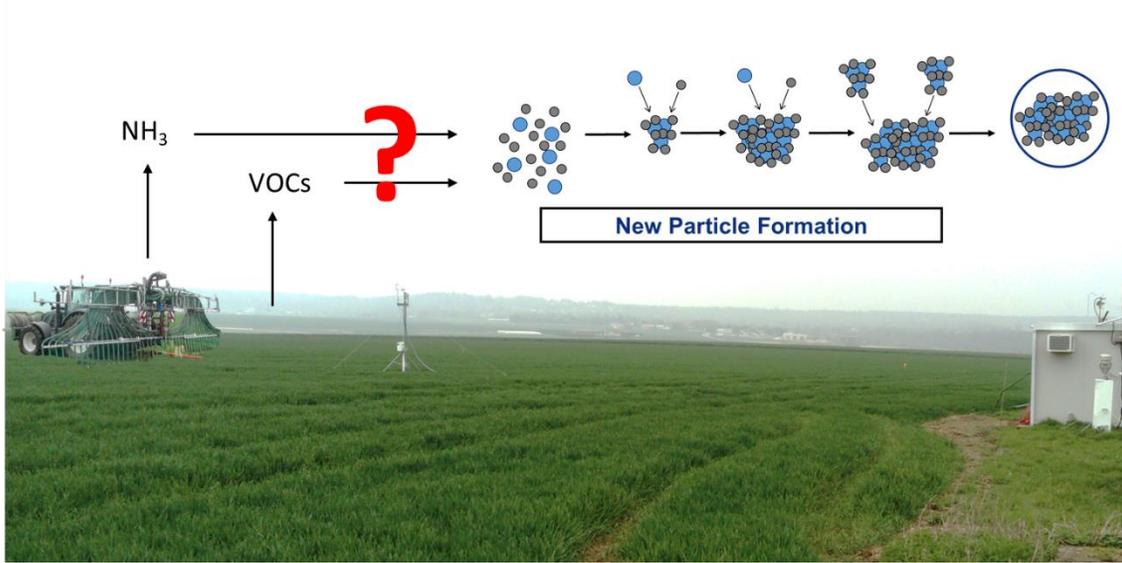
45 Finally, all the identified NPF events were also observed at SIRTAs monitoring station 20 km from  
46 the FR-Gri ICOS site, showing that both night-time and daytime NPF events were regional

47 processes. We hypothesise that night-time NPF may be related to fertilizer spreading over a regional  
48 scale, as opposed to the local activities at the farm. To our knowledge, this is the first time night-  
49 time NPF has been observed in the agricultural context.

50 **Keywords:** New Particle Formation, Agriculture, Air quality, Ammonia, VOCs

51

52 **Graphical abstract**



53 •

## 54 1. Introduction

55 Atmospheric particles are widely recognised as strongly affecting both air quality and climate. In  
56 the last AR5 report, the IPCC (International Panel for Climate Change) evaluated that interactions  
57 of atmospheric particles with solar radiation and clouds were still one of the greatest uncertainties  
58 in the evaluation of the Earth's radiative forcing budget (IPCC, 2021). New particle formation  
59 (NPF) is the secondary process by which particles are generated in the atmosphere, from the gas-  
60 particle conversion of extremely low volatility vapours. Several studies have already highlighted  
61 that NPF is a significant source of atmospheric particles, and can be a major contributor to cloud  
62 condensation nuclei (CCN) in different locations worldwide (Merikanto et al., 2009; Rose et al.,  
63 2014; Wang et al., 2017).

64 NPF generally occurs through two steps: the nucleation, which is initiated by the formation of a  
65 stable clusters from gaseous species; and the growth, the activation of these clusters followed by a  
66 subsequent growth, which implies the condensation of low volatility vapours. The first step, *i.e.*  
67 nucleation, consists of the formation of stable clusters, ranging from a few molecules to up to more  
68 than 10 molecules (Andreae, 2013). Currently, it is assumed that nucleation is mostly based on  
69 sulfuric acid clusters formed with other gaseous species, such as water, ammonia (NH<sub>3</sub>), or  
70 extremely low volatility organic compounds – ELVOCS - (Berndt et al., 2014; Kulmala et al.,  
71 2014). There is also evidence that clusters are formed from NO<sub>3</sub><sup>-</sup> ions and highly oxidised molecules  
72 (HOMs), especially at night, when sulfuric acid concentrations are lower (Bianchi et al., 2019; Rose  
73 et al., 2018). A laboratory study evidenced nucleation from the oxidation of pure biogenic volatile  
74 organic compounds (BVOCs), meaning that BVOC oxidation products are able to produce stable  
75 clusters without the presence of inorganic ions (Kirkby et al., 2016). Several field studies have  
76 reported the involvement of BVOCs in NPF when combined with other gases, but evidence of pure  
77 biogenic nucleation has only been demonstrated in laboratory experiments (Bianchi et al., 2019;  
78 Debevec et al., 2018; Kammer et al., 2020; Rose et al., 2018). The second step, the growth, is

79 associated with the coagulation of the smallest particles and the condensation of low to extremely  
80 low volatility compounds such as HOMs that are produced from the oxidation of biogenic or  
81 anthropogenic volatile organic compounds (VOCs) (Bianchi et al., 2019; Wang et al., 2017). The  
82 growth step is essential if the particles are to reach a diameter where they can act as CCN.

83 NPF has been observed in various locations worldwide, e.g. in urban environments, in the free  
84 troposphere, in remote boreal forests, tropical environments, coastal sites, etc. (Alam et al., 2003;  
85 Kanawade et al., 2022; Kerminen et al., 2018; Kulmala et al., 2004; Wang et al., 2017; Xiao et al.,  
86 2021; Young et al., 2013; Yu et al., 2020). This process is also known to occur not only locally, but  
87 also at regional levels (Kerminen et al., 2018; Kulmala et al., 2012; Wehner et al., 2007). In urban  
88 environments, NPF events are often related to environmental parameters, such as solar radiation,  
89 but also air mass history and properties, including precursor gas concentrations, and the  
90 atmospheric particle loading (Dall'Osto et al., 2013b; Dunn et al., 2004; Hakala et al., 2022;  
91 Kanawade et al., 2012; Nilsson et al., 2001; Salma et al., 2011; Stanier et al., 2004; Wonaschütz et  
92 al., 2015). The presence of pre-existing particles favours the condensation of low volatility gases,  
93 leading to a competition between condensation and nucleation processes. NPF under conditions of  
94 high particle loading has also been reported, in particular in developing countries such as China and  
95 India, due to the very high concentration of precursors in the air. This emphasizes the variety of  
96 conditions under which NPF can occur (Huang et al., 2016; Kanawade et al., 2022; Wang et al.,  
97 2017; Xiao et al., 2015; Yu et al., 2017). The balance between low volatility gases and pre-existing  
98 particles is thus one of the elements in determining the occurrence of NPF. It is clear that sulfuric  
99 acid is involved in NPF in urban environments, but it cannot explain alone the intensity of the  
100 observed NPF events. Thus, it is proposed that photochemistry of anthropogenic VOCs, especially  
101 those resulting from biomass burning, could be involved in NPF in urban environments, but this  
102 has not been clearly demonstrated so far (Wang et al., 2017). Considering the prevalence of nitrogen

103 oxides in the urban environment,  $\text{NO}_3^-$  based clusters might also play a significant role in urban  
104 NPF.

105 NPF has been mostly reported during the daytime and related to photochemical processes, notably  
106 to the formation of sulphuric acid. However, a few studies have reported night-time events, mainly  
107 in remote areas, and rarely in urban areas (Junninen et al., 2008; Kammer et al., 2018b; Kecorius  
108 et al., 2015; Lee et al., 2008; Ortega et al., 2012; Salimi et al., 2017; Svenningsson et al., 2008;  
109 Vehkamäki et al., 2004). For night-time events, the sulphuric acid based nucleation scheme is not  
110 likely to explain night-time NPF, as sulphuric acid production is related to photochemistry  
111 (Kammer et al., 2018b; Lee et al., 2008; Ortega et al., 2012). Previous studies have suggested that  
112 monoterpene ozonolysis can contribute to the initiation of nocturnal events observed at forest sites,  
113 but it has yet to be demonstrated. Overall, it is clear that our current understanding of NPF  
114 mechanisms is still incomplete.

115 A recent review regarding NPF observations noted the lack of studies detailing NPF in agricultural  
116 areas, as only one study has been recently published regarding NPF at a mixed coastal/agricultural  
117 site (Kerminen et al., 2018; Olin et al., 2022). Indeed, agriculture covers around a third of land area  
118 according to the Food and Agriculture Organization of the United Nations  
119 (<http://www.fao.org/faostat/en/#data/RL>, last access September 2022). Yet, agriculture is the main  
120 source of  $\text{NH}_3$  on a global scale (Behera et al., 2013), a molecule known to catalyse nucleation by  
121 clustering with sulphuric or nitric acids (Aksoyoglu et al., 2020; Berndt et al., 2014; Lehtipalo et  
122 al., 2018).  $\text{NH}_3$  emissions from crop fields are related to the spreading of fertilisers, such as manure  
123 or slurry. Organic fertilisers also emit a wide variety of VOCs into the atmosphere, including  
124 carboxylic acids (e.g. butyric acid, acetic acid), aromatics (e.g. phenol, methyl phenol), nitrogen  
125 containing VOCs (e.g. dimethylamine, indole) and sulphur containing VOCs (e.g. methanethiol,  
126 dimethylsulfide) (Behera et al., 2013; Feilberg et al., 2015; Liu et al., 2018; Parker et al., 2013;  
127 Potard et al., 2017; Woodbury et al., 2014). Amines emitted by the application of organic fertilisers

128 are reported to be highly efficient clustering molecules (Chee et al., 2019; Ciuraru et al., 2021;  
129 Schobesberger et al., 2013; Yao et al., 2018). A few studies also showed that sulphur containing  
130 compounds such as dimethylsulphide or methane sulphonic acid may be involved in NPF processes  
131 (Berndt et al., 2014; Schobesberger et al., 2013; Xu et al., 2020). Thus, considering the nature of  
132 the VOCs emitted during the spreading of organic fertilisers, there is clearly a strong potential for  
133 NPF formation in agricultural areas, especially during organic fertiliser spreading periods.

134 In this context, the present study is aimed at investigating NPF in relation to agricultural activities  
135 at a peri-urban agricultural site. It consisted of *i*) identifying the presence (or absence) of NPF, *ii*)  
136 highlighting the meteorological and physico-chemical conditions favouring NPF, *iii*) evaluating a  
137 potential relationship between agricultural emissions and NPF, and *iv*) an assessment of whether  
138 NPF is a local or regional process.

## 139 **2. Materials and methods**

### 140 **2.1. Experimental set-up**

141 The field campaign was part of the AgriMultiPol programme funded by the French Environment  
142 and Energy Management Agency (ADEME n°17-03 C0012). The study was conducted from  
143 March 13<sup>th</sup> 2018 to May 9<sup>th</sup> 2018, at the FR-Gri ICOS field site (48.84 N, 1.95 E, 125 m above  
144 mean sea level), in the Grignon experimental farm, which combines crops and livestock  
145 (<https://www.fermedegrignon.fr/>). The field campaign was conducted on a 20 ha crop field of  
146 winter wheat at its growth stage, on a soil that can be classified as luvisol. The site is located 25 km  
147 west of Paris city centre, and near the city of Plaisir (*ca.* 31 000 inhabitants), which is situated to  
148 the south of the site, heavily trafficked roads surround the east and south sides of the farm (**Figure**  
149 **S1 and S2**). Dairy and sheep buildings are positioned to the southwest of the farm (**Figure S1**).  
150 VOC and NH<sub>3</sub> emissions as well as the composition of particles from the farm have been  
151 characterised in a previous study (Kammer et al., 2019). The prevailing air masses is expected to  
152 come from the west-south-west, originating from the Atlantic Ocean. The field site is part of the

153 European Research Infrastructure ICOS (FR-Gri-ICOS, Integrated Carbon Observation System)  
154 and has been previously described in detail elsewhere (Loubet et al., 2012, 2011; Stella et al., 2013).  
155 Several agricultural practices were carried out during the field campaign: mineral fertilizer was  
156 spread on March 16<sup>th</sup> 2018, liquid manure on March 29<sup>th</sup> 2018, a fungicide on April 17<sup>th</sup> 2018, and  
157 an herbicide on the contour of the field on May 5<sup>th</sup> 2018.

## 158 **2.2. Sampling and instrumentation**

159 To investigate NPF, particle number size distribution (PNSD) was measured using a scanning  
160 mobility particle sizer (SMPS, TSI, model 3080). This instrument has been used in numerous  
161 studies and already fully described elsewhere (Kammer et al., 2018b; Kulmala et al., 2012). Briefly,  
162 the instrument consists of a long differential mobility analyser (DMA, TSI model 3082) and a  
163 water-based condensation particle counter (CPC, TSI model 3768). Ambient air was sampled at 0.6  
164 L min<sup>-1</sup> through a 1.8 m long conductive silicon tubing (4.5 mm inner diameter), above the shelter  
165 at 3 m above ground level. In this configuration, the instrument was able to characterise the PNSD  
166 with an electrical mobility diameter between 4.6 nm and 156.8 nm. The sampled air was not dried,  
167 to avoid any modification to its size, due to evaporation of semi volatile material and water.  
168 Condensation in the line and the instrument cannot be completely ruled out, but was not expected  
169 as the SMPS, was located in a temperature-controlled shed, and the residence time in the line was  
170 very short (0.9 s). In addition, the sample relative humidity measured by the instrument never  
171 reached more than 66 % (sample relative humidity at SMPS inlet). More, RH was below 90 % most  
172 of the time (over 93 % of the time during the campaign), and 94 % of temperatures were below 20  
173 °C, while the inside temperature was maintained at least at 20 °C during the campaign. In addition,  
174 high temperatures coincided with low relative humidity, leading to the conclusion that condensation  
175 was unlikely to happen. Leak checks were performed twice a week throughout the campaign using  
176 a high efficiency particle filter (HEPA filter, TSI) placed at the inlet of the instrument. Particle  
177 losses inside the tubing have been estimated based on the methodology proposed by Willeke and

178 Baron (1993). The highest loss rates were found to be around 45 % for the finest size ranges  
179 measured by the SMPS (*i.e.* particles with an electrical mobility diameter of 4.6 nm). The PNSD  
180 measured by the SMPS has thus been corrected to take these losses into account.

181 Meteorological parameters were measured from a mast located at the centre of the crop field, as  
182 part of the ICOS network (ICOS level 2 station). Wind speed and directions were recorded using a  
183 2-dimensional sonic anemometer (Wind Sonic, GILL), and air temperature and relative humidity  
184 using a probe (HMP155 Vaisala). Solar radiation was measured as photon photosynthetic flux  
185 density (PPFD) using a pyranometer (CMP22, Kipp & Zonen). Friction velocity ( $u^*$  in  $\text{m s}^{-1}$ ) can  
186 be used as proxy to represent the intensity of turbulence in the atmosphere, which was derived from  
187 3D-sonic anemometer measurement (Wind Sonic, GILL). All the meteorological measurements  
188 were monitored at 5 m above ground level and were recorded at 10 min time intervals using a data  
189 logger (CR1000, Campbell scientific).

190  $\text{NH}_3$  concentrations have been measured using a laser-based photoacoustic detector (LSE Monitors  
191 B.V.). The air was sampled using 1.2 m long Teflon tubing ( $\frac{1}{8}$  inches diameter) at a flow rate of  
192  $0.040 \text{ L min}^{-1}$ . The main inlet was heated at  $40/45 \text{ }^\circ\text{C}$  to prevent condensation, and a Teflon filter  
193 was used to avoid particles penetrating into the instrument. The  $\text{NH}_3$  analyser was calibrated before  
194 the campaign and had a detection limit of 1 ppb (1 standard deviation), one data point was recorded  
195 every minute.

196 VOCs were also measured using a proton transfer reaction – quadrupole ion guide – time of flight  
197 – mass spectrometer (PTR-Qi-TOF-MS, Ionicon), that has been extensively described previously  
198 (Abis et al., 2018; Gonzaga-Gomez et al., 2019; Loubet et al., 2021). The air was sampled at 1.8 m  
199 above ground level at  $50 \text{ L min}^{-1}$  through 30 m Teflon tubing ( $\frac{1}{2}$  inch diameter) using a Busch SV-  
200 1010 pump (Busch, SW), and was regulated using a mass flow controller (Bronkhorst). The air was  
201 sub-sampled by the PTR-Qi-TOF-MS at  $0.3 \text{ L min}^{-1}$ , using a 1 m long PFA tube ( $\frac{1}{8}$  inches  
202 diameter). The pressure in the drift tube was regulated at  $4 \pm 0.0001 \text{ mbar}$  at a temperature of  $80 \pm 0.06$

203 °C, and a drift voltage of  $996.5 \pm 0.5$  V. These conditions in the drift tube ensured an E/N ratio  
204 (where E is the electric field strength and N the gas number density) of  $132 \pm 1$  Td ( $1 \text{ Td} = 10^{17} \text{ V}$   
205  $\text{cm}^{-2}$ ), which has been reported to be a good compromise to between sensitivity variations due  
206 ambient humidity changes, and high fragmentation (Pang, 2015; Tani et al., 2003). Blanks were  
207 frequently performed using zero air (Alphagaz 1, zero air: 80% nitrogen, 20% oxygen, purity:  
208 99.9999%, Air Liquide), and the instrument sensitivity during the campaign was evaluated using a  
209 toluene gas standard ( $102 \pm 10$  ppb, Messer), following the methodology described in Kammer et al.  
210 (2019).

### 211 **2.3. Measurements at SIRTA monitoring station**

212 PNSD were measured simultaneously with another SMPS at the SIRTA (Site Instrumental de  
213 Recherche par Télédétection Atmosphérique, <http://sirta.ipsl.fr>) monitoring station, which is part  
214 of the EU ACTRIS (Aerosols, Clouds, and Traces gases Research InfraStructure Network,  
215 <http://www.actris.net>) research infrastructure. SIRTA is a monitoring station located 20 km south  
216 of the Paris metropolitan area ( $48.71^\circ\text{N}$ ,  $2.15^\circ\text{E}$ , , 150 m above sea level), and 20 km to the south  
217 east of the Grignon field site (see **Figure S2**), and has been described extensively in numerous  
218 previous studies (see for example, Haeffelin et al. (2005)). At SIRTA, the PNSD was recorded with  
219 an SMPS (GRIMM, model 5416), measuring electrical mobility diameters from 10 to 500 nm. As  
220 at the FR-Gri-ICOS site, the sample was not dried, and leak checks were frequently performed  
221 throughout the campaign using a HEPA filter (TSI).

## 222 **3. Methodology**

### 223 **3.1. NPF event detection and classification**

224 NPF events were identified through visually inspection of the PNSD following the method  
225 described by Dal Maso et al. (2005): an NPF event was defined as a clear increase in the particle  
226 concentration in the nucleation mode, followed by a growth during at least two hours producing the

227 well-known “banana shape” (Dal Maso et al., 2005; Heintzenberg et al., 2007). Then, to analyse  
228 the environmental factors driving NPF at the FR-Gri-ICOS site, we classified days according to the  
229 presence or absence of NPF (event days or non-event days). A third category of weak event and/or  
230 growth days was also defined, when NPF was suspected but the banana shape could not be clearly  
231 differentiated from the growth of pre-existing particles, or when the PNSD was too heavily loaded  
232 to clearly identify the banana shape. These days were termed “undefined” days. Days were  
233 classified as:

- 234 • NPF event days: a clear NPF was identified
- 235 • Undefined days: a weak NPF and/or a growth was identified
- 236 • Non-event days: no NPF was identified

237 The same classification has been performed for nights, as NPF events were recorded during both  
238 nights and days during the field campaign. In this study, days are defined as periods when solar  
239 radiation (*i.e.* the PPFD in this study) is over  $50 \mu\text{mol m}^{-2} \text{s}^{-1}$  (and nights when it is below this  
240 threshold). This corresponds to daytime period typically from 7:00 to 17:00 UTC (8:00 to 18:00  
241 local time) for the first days of the campaign, and typically from 5:00 to 18:00 UTC (7:00 to 20:00  
242 local time) at the end of the campaign. Night-time periods are thus typically from 17:00 to  
243 7:00 UTC (18:00 to 8:00 local time) for the first days of the campaign, and typically from 18:00  
244 pm to 5:00 UTC (20:00 to 7:00 local time) at the end of the campaign.

### 245 **3.2.Characterisation of NPF events**

246 The potential loss of semi volatile vapours on existing particles could be estimated by calculating  
247 the condensation sink (CS) as in (Dal Maso et al., 2005; Kulmala et al., 2001):

$$248 \quad CS = 4\pi D \sum_i \beta_{Mi} r_i N_i \quad (1)$$

249 Where  $CS$  is the condensation sink (in  $s^{-1}$ ),  $D$  is the diffusion coefficient of the condensing vapour  
 250 (assimilated here to sulphuric acid,  $D$  is  $0.14 \text{ cm}^2 \text{ s}^{-1}$ ),  $\beta_{Mi}$  is the transitional correction factor,  $r_i$  is  
 251 the mobility radius of particles (in  $\text{cm}$ ) taken from the measured PNSD by the SMPS, and  $N_i$  is the  
 252 particle concentration (in  $\text{particles cm}^{-3}$ ) in the size bin  $i$ . The transitional correction factor was  
 253 computed as:

$$254 \quad \beta_{Mi} = \frac{Kn+1}{0.377 + 1 + \frac{4}{3}\alpha^{-1}Kn^2 + \frac{4}{3}\alpha^{-1}Kn} \quad (2)$$

255 Where  $\alpha$  is the sticking coefficient (assumed to be 1, as recommended by Kulmala et al. (2001))  
 256 and  $Kn$  (unitless) is the Knudsen number computed as:

$$257 \quad Kn = \frac{\lambda_v}{r_i} \quad (3)$$

258 Where  $\lambda_v$  (in  $\text{cm}$ ) is the mean free path.

259 NPF events could be characterised by the growth rate (the speed at which particles grow during the  
 260 event) and the nucleation rate of particles. The growth rates (GR, in  $\text{nm h}^{-1}$ ) have been calculated  
 261 at each time step following the maximum concentration method described by Kulmala et al. (2012):

$$262 \quad GR = \frac{dD_p}{dt} \quad (4)$$

263 Where  $D_p$  is the geometrical mean diameter of the nucleation mode (in  $\text{nm}$ ), and  $dt$  is the  
 264 measurement time step (in  $\text{s}$ ).  $D_p$  was obtained for each PNSD event, by identifying the diameter  
 265 corresponding to the maximum concentration of particles in the nucleation range (particles with an  
 266 electrical mobility diameter  $< 25 \text{ nm}$ ) (Kulmala et al., 2012).

267 The nucleation rate ( $J_{nuc}$  in  $\text{particle cm}^{-3} \text{ s}^{-1}$ ) is defined as the flux of newly formed particles in the  
 268 nucleation size range, here taken as particles with a diameter less than  $25 \text{ nm}$  (electrical mobility  
 269 diameter) (Hussein et al., 2005; Jaatinen et al., 2009; Seinfeld and Pandis, 2006). The estimation of

270 the nucleation rate during NPF events is based on the description of the evolution of the particle  
 271 population, as a function of its sources and sinks:

$$272 \quad J_{nuc} = \frac{dN}{dt} + F_{coag} + F_{growth} \quad (5)$$

273 Where  $\frac{dN}{dt}$  is the evolution of particle concentration in the nucleation mode (particles with an  
 274 electrical mobility diameter  $< 25$  nm) as a function of time (assuming that the only source of  
 275 particles is from nucleation, in particle  $\text{cm}^{-3} \text{s}^{-1}$ ),  $F_{coag}$  is the loss of particles due to coagulation in  
 276 the nucleation size range (in particle  $\text{cm}^{-3} \text{s}^{-1}$ ) and  $F_{growth}$  the growth of particles to sizes larger  
 277 than the nucleation range (in particle  $\text{cm}^{-3} \text{s}^{-1}$ ). The  $\frac{dN}{dt}$  term in the equation can be directly estimated  
 278 from SMPS measurements:

$$279 \quad \frac{dN}{dt} = \int_{D_{p_{min}}}^{25} n_N(D_p) \cdot dD'_p \quad (6)$$

280 With  $D_{p_{min}}$  the minimum diameter measured by the SMPS (electrical mobility diameter 4.6 nm)  
 281 and  $n_N(D_p)$  is the measured PNSD in the nucleation range.  $F_{growth}$  has been estimated following  
 282 the methodology described by Kulmala et al. (2012):

$$283 \quad F_{growth} = GR(D_p) \cdot n_N(D_p) \quad (7)$$

284 where GR is the growth rate of particle over the nucleation range estimated based on equation 4.

285 Then,  $F_{coag}$  is estimated as (Hussein et al., 2008; Kulmala et al., 2012) :

$$286 \quad F_{coag} = CoagS_{nuc} N_{nuc} \quad (8)$$

287 Where  $CoagS_{nuc}$  is the coagulation sink (in  $\text{cm}^3 \text{s}^{-1}$ ) over the nucleation range expressed as:

$$288 \quad CoagS_{nuc} = \int_{D'_{p1}}^{D'_{p2}} K(D_p D'_p) \cdot n_N(D_p) \cdot dD'_p \quad (9)$$

289 Where  $K(D_p D'_p)$  (in  $\text{cm}^3 \text{s}^{-1}$ ) is the coagulation coefficient of 2 particles of diameters  $D_p$  and  $D'_p$ ,  
 290 calculated following Fuchs (1964).

## 4. Results

### 4.1. General conditions of the campaign

During the field campaign, the average air temperature was 10.8 °C, with a minimum temperature of -2.4 °C and a maximum of 26.7°C, and relative humidity varied between 100 % and 24 %, covering a wide range of atmospheric conditions. The climate was largely under the influence of the Atlantic Ocean, as about half of the air masses originated from west-south-west. Air masses from the east-northeast were downwind from the Paris urban area and central European continent, and generally presented higher concentrations of NO<sub>x</sub>, as illustrated by the NO<sub>2</sub> pollution rose (**Figure 1.a**). This is supported by NO<sub>2</sub> maps of the Ile de France region that have been generated through modelling or from satellite measurements, clearly marking the Paris area and roads as NO<sub>2</sub> hotspots (**Figure S2**). During the campaign, NO and NO<sub>2</sub> were on average 1.1 ppb and 5.6 ppb, respectively, but reached values up to 48 ppb and 34 ppb for NO and NO<sub>2</sub>, respectively. High NO concentrations could be associated road traffic, due to the proximity of roads, especially to the east of the field site, which can be subject to heavy traffic loads during morning and evening rush hours (**Figure S2**).

The site was located inside the vicinity of the Grignon experimental farm, which combined crops and livestock (dairy and sheep). VOCs, particles and NH<sub>3</sub> emissions from the farm buildings have been characterised in previous studies (Kammer et al., 2019; Loubet et al., 2012). More generally, the measurement site was in an agricultural region dominated by crops with little livestock. The influence of agricultural activities on the gas phase chemical composition have been evaluated through the measurement of NH<sub>3</sub>. Concentrations of NH<sub>3</sub> were on average 13.4 ppb and ranged from the detection limit of the instrument (1 ppb) up to 42.8 ppb. The highest levels of NH<sub>3</sub> were recorded during the first half of the campaign that usually corresponded to a period of fertilizer application on crops on a regional level. As the atmospheric lifetime of NH<sub>3</sub> is rather short (from a few hours to a day, depending on conditions), its atmospheric concentration is mainly driven by

emissions. Mineral and organic (bovine sludge) fertilizers were applied to the measurement site on March 17<sup>th</sup> 2018 and March 29<sup>th</sup> 2018 respectively. Both spreading induced an increase in NH<sub>3</sub> concentrations but did not correspond to the measured maximum of NH<sub>3</sub> (**Figure S3**). The highest concentration of NH<sub>3</sub> was observed on the April 1<sup>st</sup> 2018, four days after the spreading of bovine sludge on the field site. The delay can be explained by i) rain events following the application and ii) sources other than the local spread fertilizers, such as activities at the surrounding fields. More, a long trend increase of NH<sub>3</sub> was observed from March 20<sup>th</sup> 2018 until April 10<sup>th</sup> 2018, where the concentration remained above 10 ppb, suggesting a higher NH<sub>3</sub> background concentration. This increase over a long time period suggests that in addition to local applications, regional sources affected the ambient NH<sub>3</sub> levels at the measurement site. This is confirmed by the NH<sub>3</sub> pollution rose showing contributions from all wind directions at both low and high wind speeds (**Figure 1**). In particular, the direction of the farm does not necessarily correspond to the highest concentration of NH<sub>3</sub>. More, NH<sub>3</sub> did not seem to be favoured neither by high nor low wind speed (**Figure 1**). As a conclusion, sources responsible for high NH<sub>3</sub> levels cannot be solely attributed to the agricultural activities at the farm. The contribution of other surrounding fields where fertilizer had been spread, or even at larger scale probably contributed to the observed increase in NH<sub>3</sub> during the first half of the campaign.

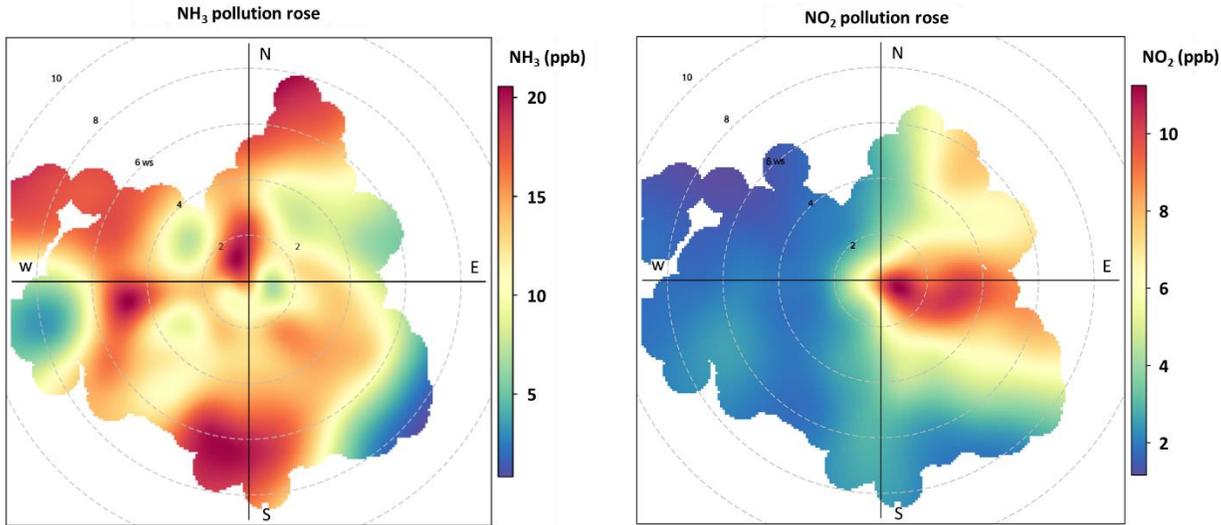


Figure 1: Pollution roses of NH<sub>3</sub> (in ppb, left panel) and NO<sub>2</sub> (in ppb, right panel).

333 PNSD was measured during the campaign at FR-Gri-ICOS and simultaneously at SIRTA station,  
334 20 km away. The location of the 2 sites regarding Paris area can be seen on **Figure S2**. The PNSD  
335 at both sites present many similarities despite the instruments measuring slightly different particle  
336 size ranges (**Figure S3**). At both sites, the particle modes were generally centred between 50 nm  
337 and 100 nm. The PNSD from FR-Gri-ICOS site was more loaded than the SIRTA one, probably  
338 due to its proximity to the particle sources, in particular traffic roads nearby the site. However, it  
339 seems that many events observed from the PNSD measurements at one site can also be observed at  
340 the other one. A more in-depth comparison of the PNSD and NPF events at the 2 sites is provided  
341 in section 4.5.

## 342 **4.2.NPF events**

343 During the field campaign, a total of 16 NPF events were identified according to the classification  
344 described in **section 3.1**. All the identified NPF events and several parameters characterising the  
345 NPF are listed in **Table 1**. Two types of NPF events were observed. The first type of event occurred  
346 during the daytime: the nucleation phase typically started in the morning, then particles grew during  
347 the following daylight hours and lasted until the end of the day, or even into the hours of darkness  
348 for a few NPF events (events 9, 11, 16). These events correspond to typical regional daytime events  
349 frequently reported in the literature in remote as well as urban areas (Dal Maso et al., 2005;  
350 Kerminen et al., 2018; Kulmala et al., 2004; Lee et al., 2019). For the second type of event NPF  
351 was initiated in the evening, around sunset (just before or just after), and the following growth  
352 period occurred during the night and ended before sunrise. In the following parts of the manuscript,  
353 these second type of events will be referred as night-time events as they mostly took place during  
354 the hours of darkness. A typical example of each type (daytime and night-time) of event is provided  
355 in **Figure 2**, and examples of undefined and non-event days are provided in **Figure S4**. Among the  
356 16 events, 8 occurred during daytime and 8 have been classified as night-time events (**Table 1**).

357 The occurrence of nocturnal events is not common as most studies report only daytime events,  
358 mostly related to photochemistry (Lee et al., 2019; Lehtipalo et al., 2018). The few studies reporting  
359 night-time events were in remote forested sites such as an Eucalyptus forest in Australia (Suni et  
360 al., 2008) or a pine forest in France (Kammer et al., 2018b). Only one study has reported nocturnal  
361 NPF events at an urban site (Salimi et al., 2017). **Figure 2** illustrates a clear difference between  
362 daytime and night-time events, particularly in the shape of the NPF plots. During the initiation of  
363 NPF events, the increase in the particle concentration at the lowest particle size range (between 5  
364 and 10 nm electrical mobility diameter) was shorter in duration for night-time compared to daytime  
365 events. This could be the result of a lower particle formation rate for night-time NPF events due to  
366 specific conditions at night (higher humidity, less wind, ...). Another possibility is that NPF was  
367 initiated earlier in the air mass and had been transported to the FR-Gri-ICOS site later, meaning  
368 that the initial steps of NPF were missed. The sharp increase in the lowest particle mode was also  
369 reported for night-time events in a previous study, but this cannot be generalised to all NPF night-  
370 time events, as only a few are reported in the literature (Kammer et al., 2018a).

371 During the campaign, daytime events occurred mainly during the second half of the campaign,  
372 whereas night-time events were mainly observed during the first half of the campaign (**Table 1**).

373 GR were generally higher for night-time (average GR of 5.3 nm h<sup>-1</sup>) than for daytime NPF events  
374 (average GR of 3.4 nm h<sup>-1</sup>). This is illustrated from the growth of particles to a larger diameter  
375 during the night-time event represented in **Figure 2**. The calculated GR are in line with previous  
376 studies conducted in suburban areas where GR are typically in the range 1-10 nm h<sup>-1</sup> (Chu et al.,  
377 2019; Kulmala et al., 2004; Salimi et al., 2017; Zhu et al., 2013). The GR of night-time events were  
378 slightly lower than those reported in forested areas, but are in agreement with GR reported for night-  
379 time events in Brisbane, Australia by Salimi et al. (2017).

380 Average nucleation rates were in the same range for both daytime and night-time events, with  
381 average values of  $63 \text{ particles cm}^{-3} \text{ s}^{-1}$  and  $52 \text{ particles cm}^{-3} \text{ s}^{-1}$ , respectively. However, nucleation  
382 rates for night-time events seemed to be more variable than for daytime events. (**Table 1**). The  
383 range of observed nucleation rates are comparable to previous studies (when calculated using the  
384 same size range), even if it seems slightly higher than some studies in comparable peri-urban  
385 environments (Chu et al., 2019; Hussein et al., 2008; Jaatinen et al., 2009; Wu et al., 2007).  
386 NPF events were reported for both continental and oceanic air masses, as well as different wind  
387 directions including from the east, which corresponds to Paris area (**Figure S2 and S11**).

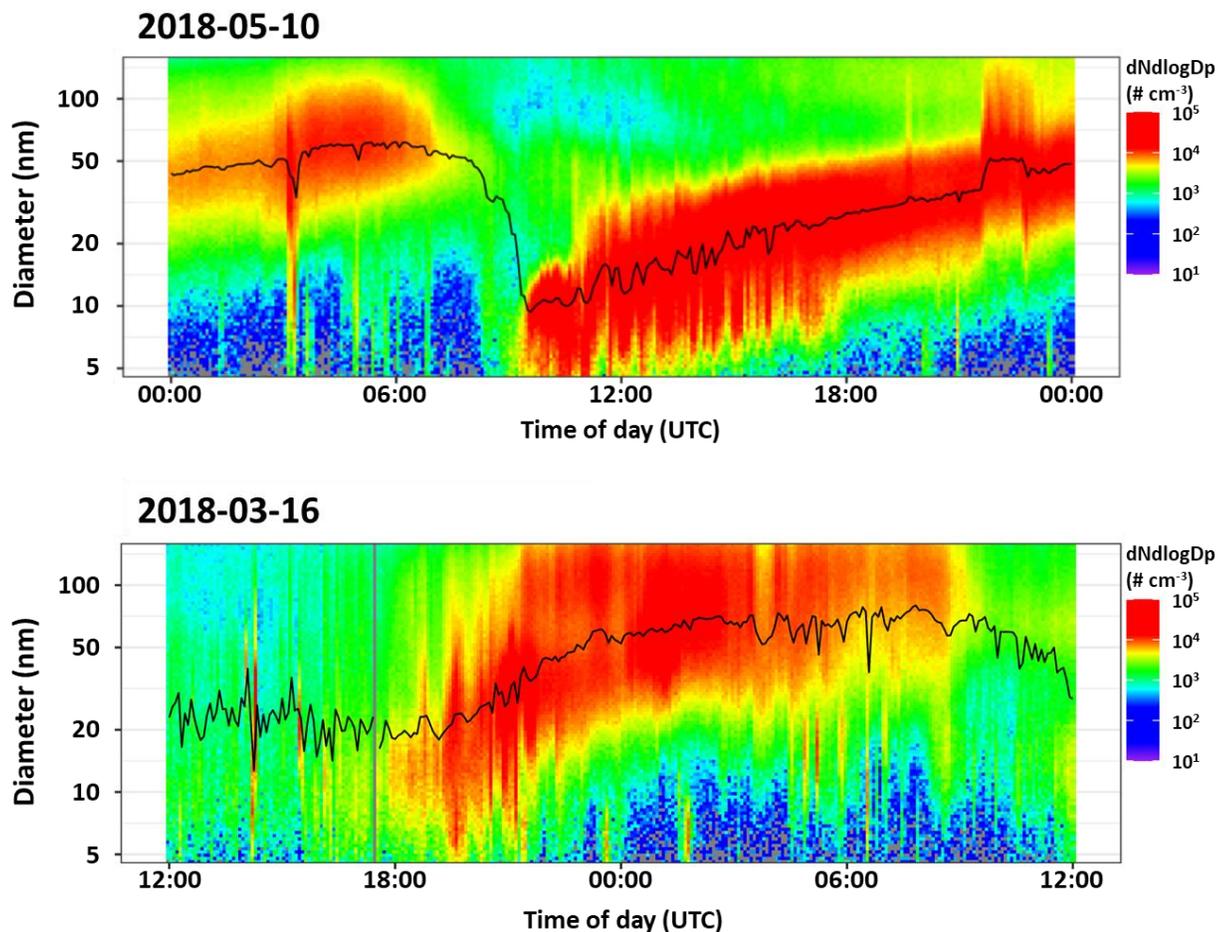


Figure 2 : Typical example of new particle formation events observe at the FR-Gri ICOS measurement station. The top panel represents a daytime NPF event, whereas the bottom panel refers to a night-time NPF event. The grey bar on the night-time panel (just before 18:00) corresponds to missing data due to a leak check.. The black line represents the median mode diameter of the particles After March 25<sup>th</sup> 2018, local time was switched from UTC+1hto UTC +2 (due to the switch from French winter to summer time).

Table 1: List of NPF events start and end times, type of event (gold = Daytime, blue = Night-time), and the corresponding mean growth rate (GR) and nucleation rate (J). The condensation sink (CS), the air temperature (T), the relative humidity (RH), the wind speed (WS) and the friction velocity ( $u^*$ ) are reported for NPF start time. After March 25<sup>th</sup> 2018, local time was switched from UTC+1h, to UTC +2 (due to the switch from French winter to summer time).

Event n°	Start date (UT)	End Date (UT)	CS ( $\times 10^{-3} \text{ s}^{-1}$ )	GR ( $\text{Nm h}^{-1}$ )	$\frac{\text{CS} \cdot 10^4}{\text{GR}}$	J ( $\text{cm}^{-3} \text{ s}^{-1}$ )	PPFD $\mu\text{mol m}^{-2} \text{ s}^{-1}$	T (°C)	RH (%)	WS ( $\text{m s}^{-1}$ )	$u^*$ ( $\text{m s}^{-1}$ )
N1	2018-03-13 17:00	2018-03-14 04:00	7.2	4.4	16	120	65	10.5	64	3.3	NA
N2	2018-03-15 17:00	2018-03-16 01:00	5.0	4.4	11	9	140	10.1	78	4.0	NA
N3	2018-03-16 18:00	2018-03-17 08:00	10.1	6.7	15	37	8.5	8.8	70	3.3	NA
N4	2018-03-24 19:00	2018-03-25 07:00	33.6	4.1	82	63	0	10.7	55	0.9	NA
N5	2018-03-30 16:00	2018-03-31 03:00	2.2	5.1	4	18	413	8.3	67	4.0	0.37
N6	2018-04-03 17:00	2018-04-04 01:00	4.9	4.1	12	12	267	10.4	78	3.5	0.36
N7	2018-04-05 17:30	2018-04-06 02:00	14.6	4.6	32	145	139	9.2	49	1.13	0.15
N8	2018-04-08 18:30	2018-04-09 03:00	12.0	9.0	13	8	66	17.0	47	3.5	0.29
<b>Average</b>	<b>17:30</b>	<b>03:30</b>	<b>11.2</b>	<b>5.3</b>	<b>23.1</b>	<b>52</b>	<b>137.3</b>	<b>10.6</b>	<b>63.5</b>	<b>3.0</b>	<b>0.3</b>
D1	2018-04-04 09:00	2018-04-04 19:00	3.1	2.2	14	54	880	11.4	68	4.4	0.36
D2	2018-04-17 10:00	2018-04-18 03:00	8.4	3.8	22	48	1357	16.2	49	3.4	0.36
D3	2018-04-22 11:00	2018-04-22 17:00	9.8	3.4	29	19	1527	23.3	42	3.8	0.16
D4	2018-04-26 08:00	2018-04-27 04:00	4.4	2.1	21	48	758	9.5	71	4.3	0.33
D5	2018-04-28 10:30	2018-04-28 16:00	5.2	2.7	19	34	809	12.8	56	4.8	NA
D6	2018-05-07 11:00	2018-05-07 16:00	18.0	6.0	30	41	1717	23.4	31	3.6	0.34
D7	2018-05-09 09:00	2018-05-09 13:00	10.4	4.3	24	160	1283	14.6	69	2.6	NA
D8	2018-05-10 09:00	2018-05-11 08:00	4.0	2.3	17	100	659	11.9	66	4.8	NA
<b>Average</b>	<b>09:40</b>	<b>00:00</b>	<b>7.9</b>	<b>3.4</b>	<b>22.0</b>	<b>63.0</b>	<b>1123.8</b>	<b>15.4</b>	<b>56.5</b>	<b>4.0</b>	<b>0.3</b>

NA: Not-Available.

389 Thus, air mass origin has no effect on the occurrence of either daytime or night-time NPF, contrary  
390 to what was observed elsewhere (Kim et al., 2019, 2016; Salimi et al., 2017; Wonaschütz et al.,  
391 2015). This result is surprising because NPF was not expected to occur when air masses were  
392 affected by emissions from Paris urban area, as NPF is known to be favoured by clean air masses.  
393 The plume from the Paris urban area usually had a higher particle loading (compared to air masses  
394 of oceanic origin), leading to a higher condensation sink (CS) competing with NPF processes. This  
395 is confirmed by the pollution rose of the CS, showing that highest levels of CS were observed to an  
396 easterly direction, similar to the NO<sub>2</sub> pollution rose, and also for lower wind speeds suggesting  
397 local sources of particles (**Figures 1 and S5**). The relation between NO<sub>2</sub> and CS is clearly visible  
398 in **Figure S6**.

399 NPF in polluted air masses has already been observed in urban areas, especially in Chinese  
400 megacities (Chu et al., 2019; Wang et al., 2017). It is worth noting that even if CS values were  
401 higher when associated with the Paris plume, they remained lower than what has been reported for  
402 Chinese megacities (Chu et al., 2019). As a result, NPF events occurred in polluted plumes as well  
403 as clean oceanic air masse, indicating that one or several factors other than CS were limiting NPF  
404 at the FR-Gri-ICOS site. The ratio of GR to CS can be used as a parameter to estimate the  
405 occurrence of NPF (Chu et al., 2019; McMurry et al., 2005). Except for some studies in highly  
406 polluted environments, it has been shown that NPF does not occur when GR/CS is higher than 50  
407 (Chu et al., 2019). The results presented here mostly conform to NPF occurring when CS/GR is  
408 lower than 50, even for night-time events, as only one exception was found on the night from March  
409 24<sup>th</sup> 2018 to Marche 25<sup>th</sup> 2018(**Table 1**).

### 410 **4.3. Daytime NPF and photochemistry**

411 To investigate the driving parameters of daytime NPF, the diurnal trends resulting from the  
412 classification detailed in **section 3.1** are presented in **Figure 3**. First, as expected, the CS was lower  
413 for category 1 (corresponding to NPF days) at the NPF start time (from 08:00 to 11:00). The CS

414 then increased during the day due to the formation and growth of the particles (**Figure 3**). The  
415 difference between clear and undefined days was quite small because, as explained above, NPF  
416 sometimes occurred with high values of CS when the air mass was affected by Paris urban area  
417 emissions, and undefined days corresponded to situations when NPF was not clearly defined and  
418 potentially situations where many diameters of the PNSD were highly concentrated.

419 Daytime NPF events were clearly characterised by higher temperatures and higher solar radiation,  
420 in agreement with previous studies (Kulmala et al., 2004). This supports the theory that nucleation  
421 is triggered by sulphuric acid cluster formation, as sulphuric acid production is directly dependent  
422 on photochemistry (Andreae, 2013; Kulmala, 2003). In addition, more photochemistry results in  
423 higher concentrations of oxidants (mostly OH radicals and O<sub>3</sub> during daytime), and greater  
424 production of a wide variety of species that can be involved in NPF such as extremely low volatility  
425 oxidation products or nitric acid (Bianchi et al., 2016; Chee et al., 2019; Hakala et al., 2019).  
426 Photochemically induced NPF explains why daytime NPF events were observed during the second  
427 half of the campaign, as solar radiation increased as the campaign progressed (**Figure S2** and  
428 **Table 1**).

429 Interestingly, the time of NPF initiation varied quite a lot (**Table 1**). We noted that late start events  
430 were associated with higher values of CS or lower solar radiation (**Table 1**). This is even clearer in  
431 **Figure S7**, which represents the event start date as a function of solar radiation and CS. When CS  
432 was high, NPF initiation was delayed because of the competition between condensation and  
433 nucleation. Hence, later in the day due to the enhanced photochemistry, more condensable vapours  
434 were produced and nucleation became more favourable despite the higher CS. Similarly, when  
435 PPFD was lower, the photochemistry was less intense, and the time for key species such as  
436 sulphuric acid to reach the supersaturation concentration was longer.

437

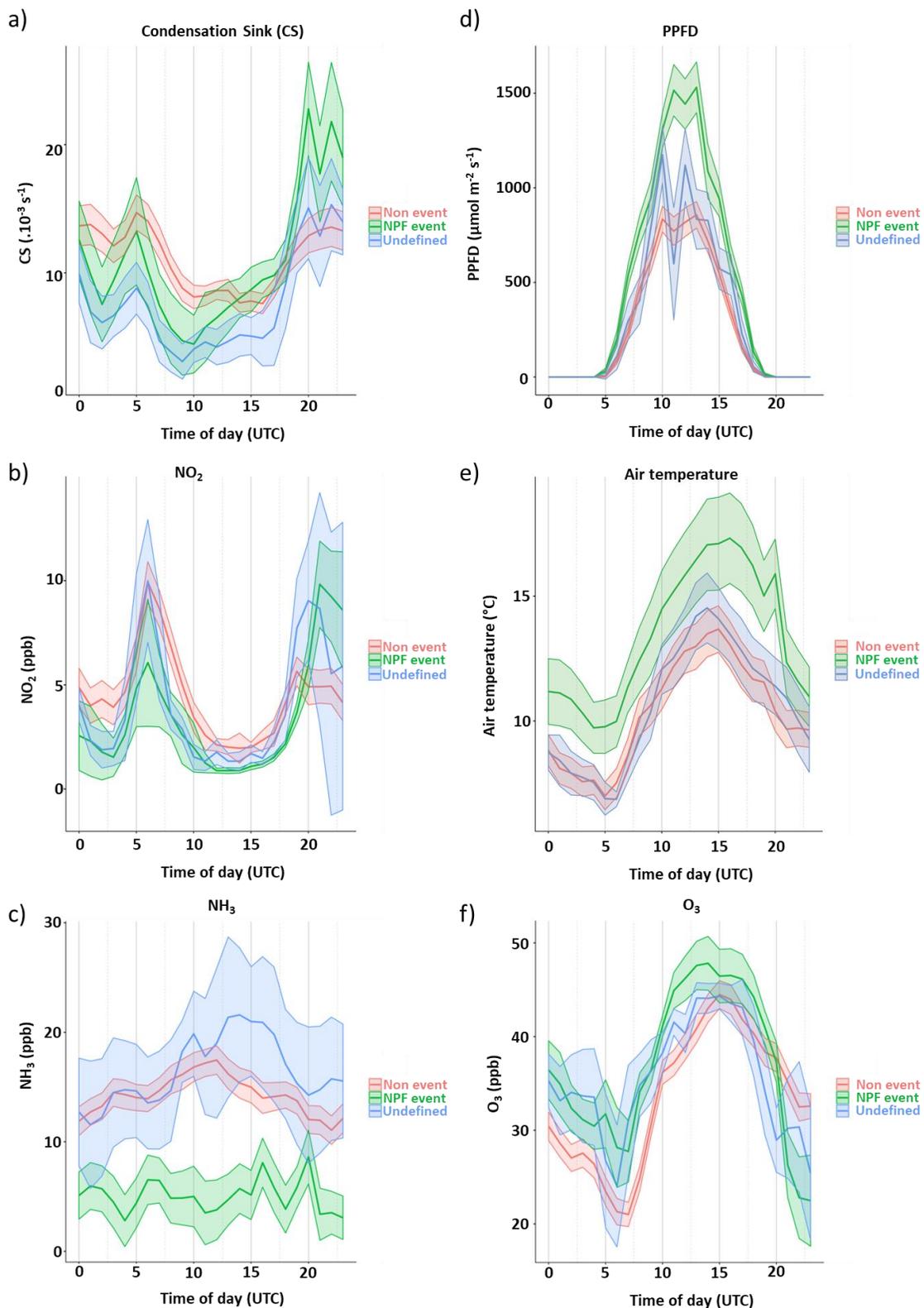


Figure 3: Diurnal profiles of a) condensation sink (CS, in  $\text{s}^{-1}$ ); b)  $\text{NO}_2$  (in ppb), c)  $\text{NH}_3$ , d) Photosynthetic Photon Flux Density (PPFD, in  $\text{W m}^{-2}$ ), e) air temperature (in  $^{\circ}\text{C}$ ) and f)  $\text{O}_3$  (in ppb) as a function of NPF categories. For each graph, the colored line and shaded area indicate mean and standard error, respectively, for NPF (green), non-event (red) and undefined (blue). After March 25<sup>th</sup> 2018, local time was switched from UTC+1h, to UTC +2 (due to the switch from French winter to summer time).

439 NO<sub>2</sub> levels were also lower during NPF days, there are 2 main reasons for this. First, the observation  
440 of low NO<sub>2</sub> during NPF days means that polluted air masses did not favour NPF. Thus, although  
441 analysis of air masses performed above showed that NPF events sometimes occurred in air masses  
442 influenced by Paris urban area emissions, it seems that they only happened when the plume was  
443 not highly polluted (highly loaded with NO<sub>2</sub> and particles). The second reason is the higher solar  
444 radiation observed during NPF days lead to higher photolysis of NO<sub>2</sub>, causing its concentration to  
445 decrease. In conclusion, the lower NO<sub>2</sub> observed during NPF days can be explained by lower  
446 emissions and higher photolysis. Regarding NPF, it supports the importance of photochemistry and  
447 the weak contribution of anthropogenic emissions to daytime NPF observed at FR-Gri-ICOS site.

448 Ozone concentrations were also higher on the days NPF occurred, strengthening the hypothesis that  
449 daytime NPF was induced by photochemistry, in agreement with higher NO<sub>2</sub> photolysis (**Figure**  
450 **3**). The consequence of stronger photochemistry is that more oxidation products can be produced  
451 due the reaction of organic species with atmospheric oxidants, such as ozone or OH radicals. To  
452 check this hypothesis, we analysed the concentrations of gaseous precursors (i.e., VOCs).

453 Methanol (CH<sub>4</sub>O, measured at m/z 33.033) and isoprene (C<sub>5</sub>H<sub>8</sub>, measured at m/z 69.070) are  
454 typically associated with biogenic emissions (Gonzaga-Gomez et al., 2019; Kesselmeier and Staudt,  
455 1999). The isoprene concentration was pretty low, with an average concentration of 0.16±0.1 ppb.  
456 These generally low isoprene levels could be a favourable condition for NPF as a high level of  
457 isoprene has been reported to suppress NPF in certain conditions (Kanawade et al., 2011; Kiendler-  
458 Scharr et al., 2009). In this study, both isoprene and methanol were more concentrated during NPF  
459 days compared to non-NPF days (**Figure 4**), potentially meaning there were more oxidation  
460 products during NPF days (note that even if methanol is not a precursor of low volatility products,  
461 it is used here as tracer for biogenic emissions). However, isoprene and methanol presented an  
462 unusual diurnal trend. Isoprene is recognised as mainly being emitted by trees, shrubs and grass,  
463 while methanol is the main VOC emitted by crops (including the soil) (Bachy et al., 2020; Gonzaga

464 Gomez et al., 2019; J Kesselmeier and Staudt, 1999; Laothawornkitkul et al., 2009). Considering  
465 the context of the field site (peri-urban agricultural site), methanol can be reasonably attributed to  
466 the surrounding crops, while forests are likely sources of isoprene, but whether these species  
467 originated from local or regional sources depends on atmospheric transport (Bachy et al., 2020;  
468 Loubet et al., 2021). As biogenic emissions are related to solar radiation and temperature which  
469 peak around midday, methanol and isoprene emissions reach their maximum in most rural areas at  
470 this time (Cai et al., 2021; Gentner et al., 2014; Graus et al., 2013). In our study, isoprene and  
471 methanol presented unusual night-time maxima, increasing in the evening around 20:00 UTC, this  
472 trend was even more pronounced during NPF event days (**Figure 4**). We hypothesise that these  
473 daily patterns resulted from the combination of a ground-based emissions during the night and the  
474 building of the stable nocturnal boundary layer (reducing the height of the atmospheric mixing  
475 layer), as already observed in forested areas for VOCs emitted during dark hours (Kammer et al.,  
476 2018b). In addition, their pollution roses are quite similar and pointed to 2 sources; a regional one  
477 to the east, in addition to a strong local contribution, more visible on the isoprene pollution rose  
478 (**Figure S8**). More, methanol and to a lesser extent isoprene show an exponential relation with air  
479 temperature (**Figure S9**), but no relation to solar radiation (not shown). Such behaviour has been  
480 observed for soil emissions at agricultural sites, and can indicate that the local source of methanol  
481 might be the soil of the FR-Gri-ICOS site and surrounding crop stands (Bachy et al., 2018).

482 More, both methanol and isoprene have similarities with the NO<sub>x</sub> diurnal cycle (*i.e.*; increase at  
483 evening around 20:00 UTC) and pollution rose, to the relation is stronger with NO<sub>2</sub> than with NO  
484 (**Figures 3, 4 and S8**), suggesting a contribution of anthropogenic sources such as residential solid  
485 fuel burning or traffic. This is supported by the similar trend (evening increase) and pollution rose  
486 of toluene, also pointing in an easterly direction (**Figures 4 and S8**), usually attributed to  
487 anthropogenic sources (Bruns et al., 2017; Languille et al., 2020). More precisely, the diurnal cycle  
488 is typical of field sites strongly influenced by biomass burning, with a large evening peak, then

489 decreasing, with a short peak appearing in the evening (Chazeau et al., 2021; Dall'Osto et al.,  
490 2013a). A noticeable pattern was the spike observed in methanol mixing ratios in the morning that  
491 may be attributed to methanol evaporation with dew (Bachy et al. 2018) although it could also be  
492 linked with traffic. The second option may be the most plausible as the same morning peak is  
493 observed for the NO<sub>2</sub> diurnal trend (**Figure 3**).

494 Methyl vinyl ketone (MVK, C<sub>4</sub>H<sub>6</sub>O) and methacrolein (MACR, C<sub>4</sub>H<sub>6</sub>O) are two gas phase  
495 oxidation products resulting from isoprene photooxidation, and are used as markers of  
496 photochemical activity (Paulot et al., 2009). As these compounds are isomers, they were both  
497 detected at m/z 71.084 by the PTR-Qi-TOF-MS (de Gouw and Warneke, 2007). The concentration  
498 of MVK+MACR was rather low (below ppb level) but was higher in the mornings of NPF daytime  
499 events (i.e. when NPF is initiated). One can suppose that this result supports the importance of  
500 photochemistry to daytime NPF. However, their daily pattern was expected to present higher  
501 concentrations during the daytime, in relation to photochemistry. In our case, especially during NPF  
502 days, the concentrations of the sum MVK+MACR peaked in the evening and during early morning,  
503 suggesting that photochemical production of MVK+MACR is unlikely. We thus propose that  
504 another isomer was probably contributing to the signal at this m/z, or another source of these  
505 compounds contributed to their signal.

506 More advanced analysis (such as positive matrix factorization, PMF) of the various source  
507 contributions to ambient VOC levels would be helpful to understand the dynamics of VOC  
508 concentrations at the measurement site, but identification and quantification of VOC sources is  
509 beyond the scope of the present paper.

510 Finally, a potential relation to agriculture was investigated using NH<sub>3</sub> as a tracer for agricultural  
511 emissions. **Figure 3** shows that the levels of NH<sub>3</sub> were clearly lower on NPF days compared to  
512 non-NPF days. This was expected considering that daytime NPF mostly occurred during the second  
513 half of the campaign, whereas the period corresponding to organic fertiliser spreading was the first

514 half of the field campaign. As a result, we can reasonably hypothesise that observed daytime NPF  
515 events at the FR-Gri-ICOS site were not associated with high  $\text{NH}_3$  concentrations and agricultural  
516 emissions, even this cannot be completely ruled out. We suggest that daytime NPF was rather linked  
517 to photochemistry, in agreement with previous studies.

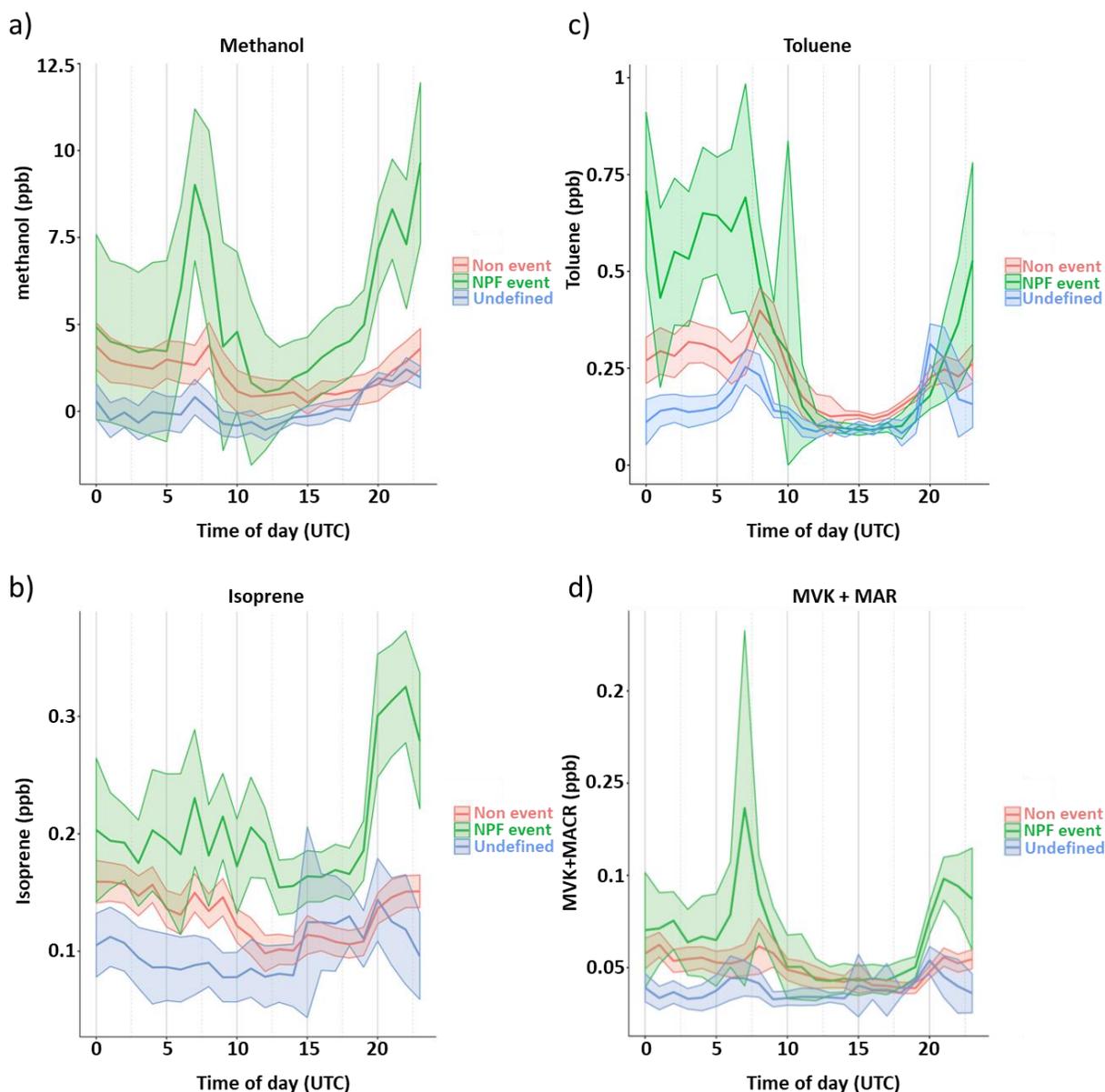


Figure 4: Diurnal profiles of a) methanol ( $m/z$  33.033, in ppb); b) isoprene ( $m/z$  69.070, in ppb), c) toluene ( $m/z$  93.070, in ppb) and d) the sum of methyl vinyl ketone and methacrolein (MVK+MACR,  $m/z$  71.048, in ppb) as a function of NPF categories. For each graph, the colored line and shaded area indicate mean and standard error, respectively, for NPF (green), non-event (red) and undefined (blue). After March 25<sup>th</sup> 2018, local time was switched from UTC+1h, to UTC +2 (due to the switch from French winter to summer time).

#### 4.4. Parameters driving night-time NPF

As explained in section 3.1, night-time NPF events differed from daytime events in several ways. Similar to the analysis performed for daytime events, nocturnal trends of the different physico-chemical and meteorological parameters have been investigated in relation to the presence (or absence) of night-time NPF events (**Figure 5**). First, we observed that on the nights when NPF events occurred, the CS was lower at NPF start time (between 16:00 and 19:00 UTC, **Figure 5**). Then, CS increased through the night due to the formation of particles and their growth throughout the night, as was observed for daytime NPF events. The CS was slightly higher for night-time undefined events (at NPF start time), meaning that during these nights there was competition between NPF and condensation processes. As expected, air temperature was lower for NPF nights (**Figure 5**), because they were identified during the first part of the field campaign, and logically, relative humidity was thus higher during NPF nights (**Figure S10**). Surprisingly, wind speed was not a meaningful factor for night-time NPF (**Figure 5**), contrary to what was reported in previous night-time NPF studies (Kammer et al., 2018). The values of  $u^*$  (**Table 1**) support the hypothesis that the building of a night-time stable boundary layer was not a prerequisite for NPF (nocturnal profiles are not represented due to a significant amount of missing data, especially during NPF nights), despite some data being missing. This is also strengthened by investigating the Monin–Obukhov length (a key parameter for atmospheric stability, Irwin and Binkowski, 1981), which indicates that the stable nocturnal layer was not a necessity for night-time NPF. Previous studies on night-time NPF evidenced that a stable atmosphere (induced by lower wind speed) is required to trigger NPF (Kammer et al., 2018b; Kecorius et al., 2015; Man et al., 2015). The building of the stable nocturnal boundary layer coincides with a drop in wind speed and, as a result, a lower dilution of ground-based emissions (that are concentrated within a smaller surface layer). Thus, the resulting increase in concentration of low volatility gases helps them to reach their super saturation ratio. In our case, wind speed reached up to  $4 \text{ m s}^{-1}$  when night-time NPF started (Table 1) and was always

544 higher than  $0.9 \text{ m s}^{-1}$  (**Table 1**). This is supported by the analysis of the friction velocity. Thus, the  
545 precursor gases present at the initiation of night-time NPF did not require a reduced boundary layer  
546 to reach supersaturation. This result suggests that night-time NPF at FR-Gri-ICOS was not related  
547 to ground-based emissions.

548  $\text{NO}_2$  concentrations were high on the nights when clear and undefined NPF occurred (category 1  
549 and 2) compared to non-event nights (**Figure 5**). High  $\text{NO}_2$  concentrations are associated with high  
550 CS values, especially for night-time conditions (**Figure 5**). This explains why  $\text{NO}_2$  was also high  
551 for clear and undefined nights, as the undefined NPF events might have corresponded to the nights  
552 when a high CS prevented a clear banana shape event. As  $\text{NO}_2$  is usually associated with  
553 anthropogenic emissions, its higher concentration might indicate that anthropogenic emissions were  
554 involved in night-time NPF. We therefore, examined the nocturnal trend of the BTEX (Benzene,  
555 Toluene, Ethylbenzene and xylenes), recognised as tracers of anthropogenic sources (Jiang et al.,  
556 2017). The toluene nocturnal trend is represented in **Figure 6** as an example of trend for BTEX  
557 species. The toluene concentration was not higher at the time of night-time NPF initiation but was  
558 higher after midnight. It means that anthropogenic VOCs might be involved in the growth steps of  
559 night-time NPF, as precursors of condensable vapours. Higher  $\text{NO}_2$  levels during NPF and  
560 undefined nights could alternatively mean higher concentrations of  $\text{NO}_3$  radicals as the oxidation  
561 of  $\text{NO}_2$  is the main source of  $\text{NO}_3$  radicals (especially due to the reaction with  $\text{O}_3$ ). On one hand,  
562 higher  $\text{NO}_3$  levels means more oxidation of VOC precursors leading to higher concentrations of  
563 low volatility species that can be involved in NPF. On the other hand, it can also lead to the  
564 formation of ammonium nitrate ( $\text{NH}_4\text{NO}_3$ ) in the particle phase. For example, Man et al. (2015)  
565 showed in a field study in Korea that  $\text{NH}_4\text{NO}_3$  was involved in the growth stage of NPF. Gas to  
566 particle conversion of  $\text{NH}_4\text{NO}_3$  has been observed by Nemitz et al. (2009) after fertilisation over  
567 grassland, but has not been associated with NPF events. In summary, high concentrations of  $\text{NO}_2$   
568 during NPF nights may suggest that two processes are involved in night-time NPF: gas to particle

569 conversion of  $\text{NH}_4\text{NO}_3$  and condensation of low volatile gases resulting from VOC oxidation by  
570  $\text{NO}_3$  radicals.

571 As explained  $\text{O}_3$  could be involved in  $\text{NO}_3$  formation through the oxidation of  $\text{NO}_2$ . We observed  
572 that  $\text{O}_3$  concentrations were lower during NPF nights (**Figure 5**). As  $\text{O}_3$  is a secondary compound  
573 from photooxidation, it is reasonable to consider that there is no source of  $\text{O}_3$  during dark hours.  
574 Thus, the lower values of  $\text{O}_3$  might be an indication of higher consumption, either due to physical  
575 (*i.e.* deposition) or chemical sinks. As ozone surface deposition is mostly driven by turbulence  
576 (Kammer et al., 2018a; Potier et al., 2015), it cannot explain the lower  $\text{O}_3$  concentrations observed  
577 during NPF nights, as there was no difference in wind speed during these nights. We finally assume  
578 that lower  $\text{O}_3$ , during NPF nights, implies a higher consumption by chemical reactions, potentially  
579 with  $\text{NO}_2$  that would support the importance of  $\text{NO}_3$  in NPF.

580 The nocturnal trend of  $\text{NH}_3$  by category of nights supports the hypothesis of the importance of  
581  $\text{NH}_4\text{NO}_3$  in night-time NPF, as  $\text{NH}_3$  levels were higher during the nights when NPF events were  
582 identified (**Figure 5**). This is in line with the period when night-time NPF occurred (first half of the  
583 campaign), that typically corresponded to a period of intensive fertiliser spreading in the Ile de  
584 France region (Fortems-Cheiney et al., 2020). The higher humidity and lower temperatures during  
585 NPF nights are 2 environmental factors favouring gas-particle partitioning of  $\text{NH}_4\text{NO}_3$  toward the  
586 particle phase.  $\text{NH}_4\text{NO}_3$  has been shown to efficiently participate in aerosol growth after grassland  
587 fertilisation (Nemitz et al., 2009). In addition to the formation of  $\text{NH}_4\text{NO}_3$ ,  $\text{NH}_3$  is also reported to  
588 catalyse NPF by stabilising clusters in the very first steps of NPF (*i.e.* nucleation) (Almeida et al.,  
589 2013; Schobesberger et al., 2013). Thus, the role of  $\text{NH}_3$  in NPF could be both through the formation  
590 of  $\text{NH}_4\text{NO}_3$  and the formation of stable clusters. The role of  $\text{NH}_3$  has already been highlighted in  
591 the field by different studies, but is always associated with daytime events and ternary nucleation  
592 of  $\text{NH}_3/\text{H}_2\text{SO}_4/\text{H}_2\text{O}$  (Birmili and Berresheim, 2003; Größ et al., 2018; Xiao et al., 2015).

593

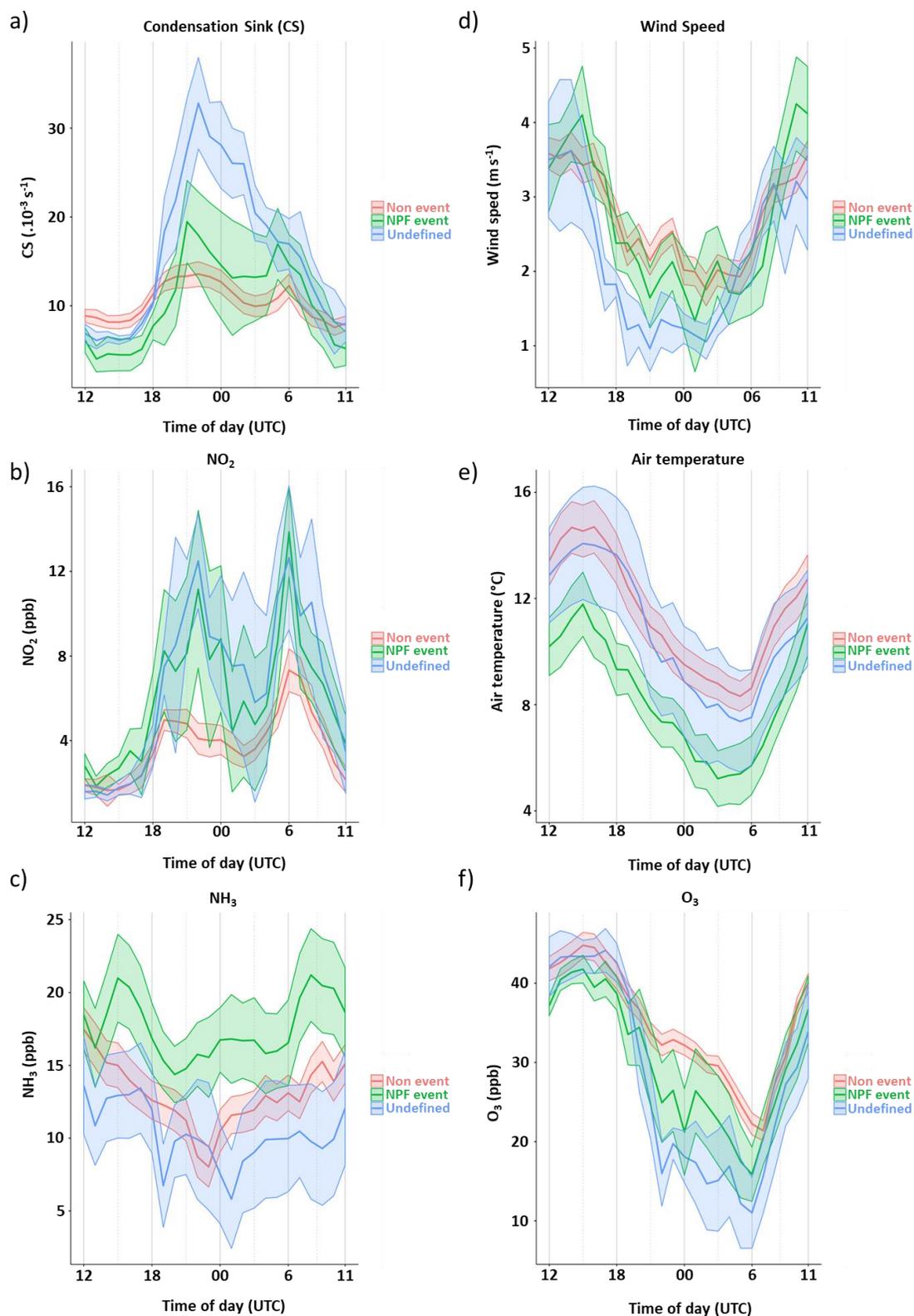


Figure 5: Diurnal profiles of a) condensation sink; b)  $NO_2$ , c)  $NH_3$ , d) wind speed, e) air temperature and f)  $O_3$  as a function of night-time NPF categories. For each graph, the colored line and shaded area indicate mean and standard error, respectively, for NPF (green), non-event (red) and undefined (blue). After March 25<sup>th</sup> 2018, local time was switched from UTC+1h, to UTC +2 (due to the switch from French winter to summer time).

595 In this case, it is very unlikely that the classical ternary nucleation involving  $\text{H}_2\text{SO}_4$  triggered the  
596 observed NPF, as  $\text{H}_2\text{SO}_4$  formation is mostly related to photochemical reactions. One possibility is  
597 that other acids, such as organic acids that can be emitted by fertilisation activities, participate in  
598 the nucleation steps. Then, the condensation of  $\text{NH}_4\text{NO}_3$  can explain the growth, as Nemitz et al.  
599 (2009) estimated that it can induce growth rates of 11 nm diameter particles from 1 to 7 nm  $\text{h}^{-1}$  over  
600 a grassland in Germany, which is typically in the range of the GR measured at FR-Gri-ICOS site.  
601 In the end, the mechanism by which  $\text{NH}_3$  promotes NPF in our study is still unclear and requires  
602 further investigation. Finally, considering the agricultural character of the field site and the period  
603 of night-time NPF, we can attribute the higher levels of  $\text{NH}_3$  to the agricultural spreading of organic  
604 fertilizer. Thus, we can reasonably hypothesise that the occurrence of night-time NPF in spring at  
605 FR-Gri-ICOS was related to fertiliser spreading.

606 As NPF night-time events were observed during the first half of the campaign, it is expected that  
607 they were not related to higher biogenic emissions, contrary to what was observed for daytime NPF  
608 events. This is confirmed by investigating the nocturnal cycle of biogenic VOCs, such as methanol  
609 that was clearly lower during NPF nights (**Figure 6**). VOCs typical of agricultural activities and  
610 organic fertiliser spreading were also investigated, even if their emissions are not well characterised  
611 thus far. Most of VOCs were less concentrated on the nights we identified NPF events. For example,  
612 trimethylamine ( $\text{C}_3\text{H}_9\text{N}$ ,  $m/z$  60.081) is known to be emitted from fertilizer spreading (Liu et al.,  
613 2018; Potard et al., 2017; Woodbury et al., 2014) and to catalyse NPF (Duporté et al., 2016; Yao et  
614 al., 2018), but was not higher during NPF nights (**Figure 6**).

615 The only VOC that was clearly higher during NPF nights (compared to non NPF nights) was acetic  
616 acid, that is also reported as an organic fertiliser emission (**Figure 6**) (Liu et al., 2018; Potard et al.,  
617 2017; Woodbury et al., 2014). Its concentrations were also high during undefined nights. Thus,  
618 acetic acid might be one of the contributors to night-time NPF events, among other parameters such  
619 as pre-existing particle loading (*i.e.* condensation sink) in determining whether NPF happened or

620 not. Acetic acid has been reported to enhance nucleation through the stabilisation of clusters, similar  
 621 to  $\text{NH}_3$  (Nadykto and Yu, 2007; Zhu et al., 2014). In addition, this acid can undergo reactions with  
 622 different bases including  $\text{NH}_3$ . A higher acetic acid concentration could thus favour the  
 623 condensation of  $\text{NH}_3$ . The contribution of acetic acid would imply agricultural emissions played a  
 624 role in night-time NPF, but a more comprehensive study regarding the chemical composition of  
 625 aerosols at the molecular scale will be necessary.

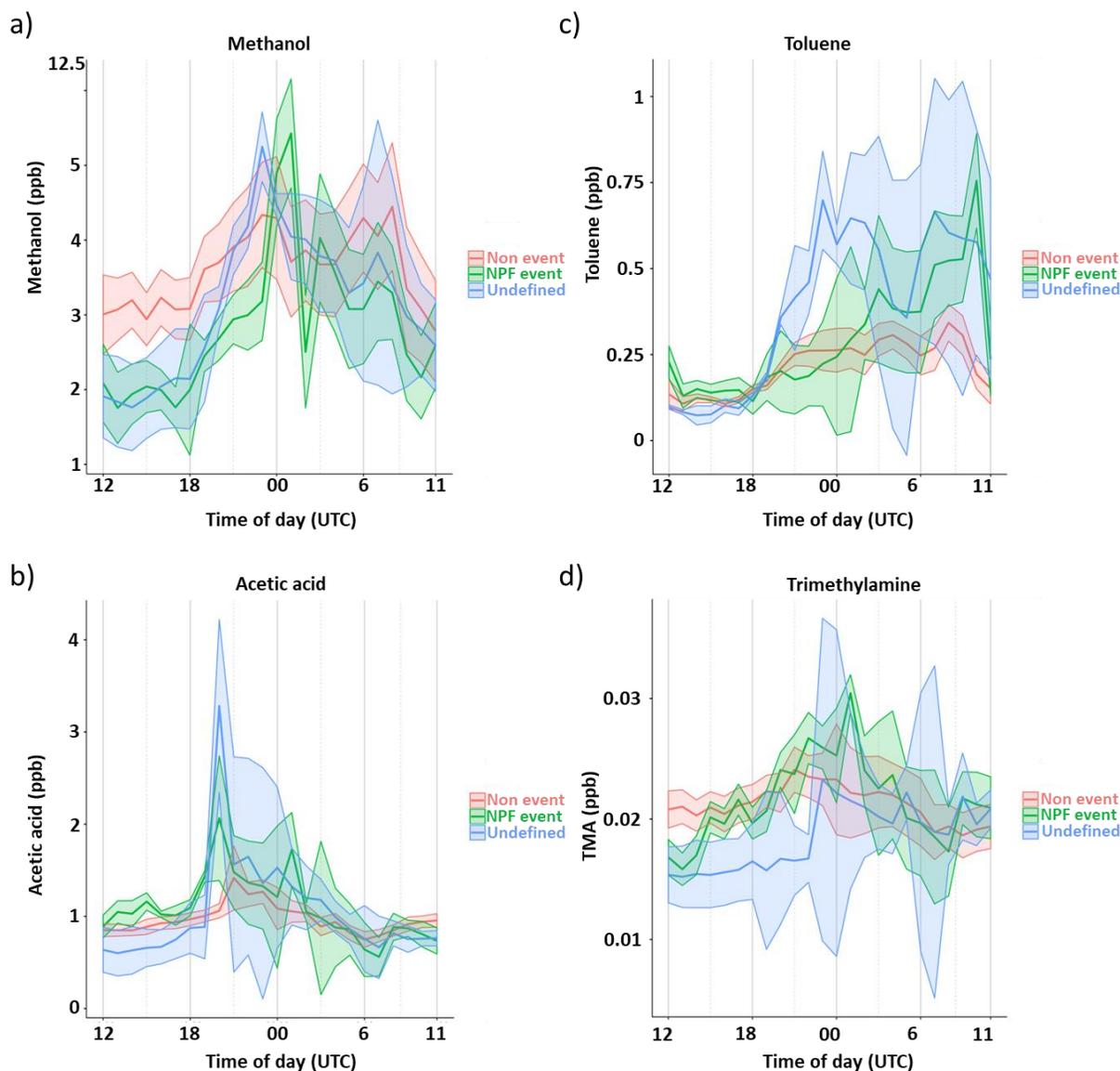


Figure 6 : Diurnal profiles of a) methanol ( $m/z$  33.033, in ppb), b) acetic acid ( $m/z$  61.028, in ppb), c) toluene ( $m/z$  93.070, in ppb) and d) trimethylamine ( $m/z$  60.081, in ppb) as a function of night-time NPF categories. For each graph, the colored line and shaded area indicate mean and standard error, respectively, for NPF (green), non-event (red) and undefined (blue). After March 25<sup>th</sup> 2018, local time was switched from UTC+1h, to UTC +2 (due to the switch from French winter to summer time).

#### 4.5. Are night-time NPF events regional?

We also investigated whether the observed NPF events were regional phenomena by comparing the PNSD obtained at the FR-Gri-ICOS site to the one from the SIRTA site, 20 km away. The methodology described in **section 3.1** was also applied to identify NPF events from PNSD measurements at the SIRTA station. First, all the NPF events observed at FR-Gri-ICOS were also observed at the SIRTA site. This result was expected for daytime NPF events, that are clearly defined as a regional process based on the numerous studies available in the literature (Kerminen et al., 2018; Kulmala et al., 2004). However, less is known about night-time NPF events, and we show here that night-time events also occurred on a regional scale. It means that the parameters driving night-time NPF were probably more regional than local. As a result, if agricultural emissions were involved in the night-time NPF observed, it was related to the fact that the Ile de France region, and more precisely the footprint area of NPF; has a high density of crops, and that farmers usually spread fertilisers during the same time period. Then, even if farmers do not spread the same types of fertilizers on the same days, a large scale spreading over typically a 1- or 2-week period creates a large source of  $\text{NH}_3$  that may last for several weeks after the initial spreading. This is supported by the high  $\text{NH}_3$  background concentrations during the first half of the field campaign, and the pollution rose of  $\text{NH}_3$  showing contributions from all wind directions (**Figures 1 and S3**). This phenomenon can explain both the regional phenomena of night-time NPF and the link with agriculture. However, this theory must be confirmed through further investigations. The importance of meteorological parameters is strengthened by the fact night-time NPF is regional, as they are generally similar at a regional scale. In the case of night-time NPF, we highlighted the importance of both temperature and relative humidity, while it is still hard to disentangle the effect of each parameter, respectively.

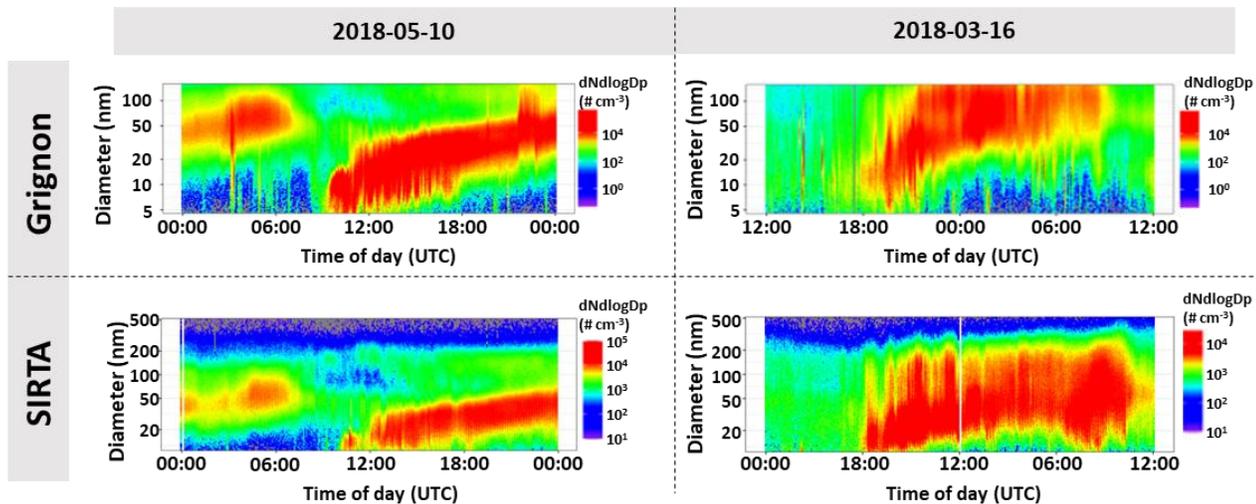


Figure 7: Typical example of new particle formation events observed at the FR-Gri ICOS measurement station and at the same time at the SIRTA station. The top panels represent events at FR-Gri ICOS and bottom panels at SIRTA. Left panels represent a daytime NPF event, whereas right panels show a night-time NPF event. Note that Y scales are different for top (Grignon) and bottom (SIRTA) plots, due to different SMPS configurations for measuring PNSD. After March 25<sup>th</sup> 2018, local time was switched from UTC+1h, to UTC +2 (due to the switch from French winter to summer time).

649 We then compared the start time of each event at each site. Generally, no lag time was observed for  
650 night-time NPF events despite the difference in the lower diameter of the two instruments, and the  
651 distance between the two sites. In contrast, the start times of daytime events were not the same for  
652 each daytime NPF event. This lag time was not constant and varied from half an hour up to 2 hours  
653 (**Table 2**). The onset of NPF events appeared always earlier at FR-Gri-ICOS, except for 1 event  
654 (event n° 13, **Table 2**). Wind speed values at NPF start time, reported in **Table 2**, were used to  
655 evaluate if the simple displacement of the air mass from one site to the other could explain the  
656 observed lag time for daytime NPF. No relation can be observed between the wind speed and the  
657 lag time. In addition, daily air mass backward trajectories have been computed for both sites for the  
658 entire campaign (**Figure S11**). Analysis of the resulting backward trajectories first showed that the  
659 same air mass was sampled at both FR-Gri-ICOS and SIRTA sites. Moreover, it seemed clear that  
660 there was no relation between the origin of the air mass and the lag time of NPF events. Even for  
661 the only event when NPF started earlier at SIRTA compared to FR-Gri-ICOS, no specific air mass

662 origin was observed (**Figure S11**). More studies are thus needed to fully understand the factors  
 663 driving NPF events observed at both sites, and more generally in peri-urban agricultural areas.

Table 2: List of NPF events start times at both FR-Gri-ICOS and SIRTA. Wind speed (WS) at FR-Gri-ICOS NPF event start time are provided to evaluate the potential relation between the evolution of the air mass and the lag time in NPF start time (UTC) between the 2 sites.

Event n°	Date	Time start at FR-Gri-ICOS (UTC)	Time start at SIRTA (UTC)	WS (m s <sup>-1</sup> )
1	2018-03-13	17:00	17:00	3.3
2	2018-03-15	17:00	17:00	4.0
3	2018-03-16	18:00	18:00	3.3
4	2018-03-24	19:00	19:00	0.9
5	2018-03-30	16:00	16:00	4.0
6	2018-04-03	17:00	17:00	3.5
7	2018-04-04	09:00	11:00	4.4
8	2018-04-05	17:30	17:30	1.13
9	2018-04-08	18:30	18:30	3.0
10	2018-04-17	10:00	11:00	3.4
11	2018-04-22	11:00	12:00	3.8
12	2018-04-26	08:00	09:00	4.3
13	2018-04-28	10:30	10:00	4.8
14	2018-05-07	11:00	12:00	3.6
15	2018-05-09	09:00	10:00	2.6
16	2018-05-10	09:00	10:00	4.8

## 667 **5. Conclusion**

668 A field campaign has been conducted within the Grignon experimental farm, which is a peri-urban  
 669 agricultural site located at 25 km west of Paris city centre. 16 NPF events have been identified, 8  
 670 NPF events during the daytime and 8 during the night-time. GR and nucleation rates were in the  
 671 range of reported values, for both daytime and night-time events. Daytime events were observed  
 672 during the second part of the campaign, when solar radiation and ozone concentrations were higher.  
 673 They are thus proposed to be related to photochemical processes in agreement with previous

674 studies. As  $\text{NH}_3$  concentrations were lower on the days NPF was observed, we supposed that  
675 daytime NPF was not related with agricultural activities.

676 In contrast, night-time NPF events occurred during the first half of the campaign and were  
677 associated with low temperatures and high relative humidity. They were associated with higher  
678  $\text{NH}_3$  levels suggesting that they might be related to agricultural emissions, and more specifically to  
679 fertiliser spreading. We hypothesised that high  $\text{NH}_3$  levels were due to both local and regional  
680 sources and cannot be solely explained by the agricultural practices that happened on the field site  
681 during the campaign. The processes at the initiation of night-time NPF were thus probably not local,  
682 but rather regional suggesting that meteorological conditions were important NPF drivers.

683 The NPF events observed at the FR-Gri-ICOS site were also detected at a the SIRTA monitoring  
684 station located 20 km to the southeast. This confirms that daytime NPF events were regional  
685 processes, as previously reported in different studies, and showed for that night-time NPF events  
686 were also regional. In conclusion, there appears to be a potential relation between night-time NPF  
687 and agriculture, due to regional agricultural activities rather than local fertiliser spreading. We  
688 suggest that the spreading of fertilisers by numerous farmers in the Ile de France region in the same  
689 time period could explain the relation between agriculture and night-time NPF. However, a more  
690 extensive study is needed to fully elucidate this. In addition, more studies detailing NPF in other  
691 regions in the world with intense agriculture would be necessary to verify whether the results  
692 obtained in this study can be extended to other agricultural areas.

693

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701 **Competing interests:** The authors declare no competing of interest.

702 **Data and materials availability:**

703 All the data used in this article are publicly available at the following address  
704 <https://doi.org/10.57745/XMPAVS> (last access September 2022) and can be identified using the  
705 corresponding doi 10.57745/XMPAVS.

706 **Author contribution**

707 JK designed and performed field measurement, analysed the data and wrote the manuscript. LS  
708 analysed SMPS data and revised the manuscript. RC, JEP, FL, PB, SB, BH, SC, OF and FT  
709 participated in the field campaign and revised the manuscript. VG and BL created the  
710 AGRIMULTIPOL project, designed the field campaign, participated in field campaign and revised  
711 the manuscript.

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