



1 **Opinion: A paradigm shift in investigating the general characteristics of atmospheric new**  
2 **particle formation using field observations**

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4 Markku Kulmala<sup>1,2,3,4</sup>, Diego Aliaga<sup>1</sup>, Santeri Tuovinen<sup>1</sup>, Runlong Cai<sup>1,5</sup>, Chao Yan<sup>2,3,4</sup>, Federico  
5 Bianchi<sup>1</sup>, Yafang Cheng<sup>6</sup>, Aijun Ding<sup>3,4</sup>, Douglas R. Worsnop<sup>1</sup>, Tuukka Petäjä<sup>1,3</sup>, Katrianne  
6 Lehtipalo<sup>1</sup>, Pauli Paaasonen<sup>1</sup> and Veli-Matti Kerminen<sup>1,3</sup>

7  
8 <sup>1</sup> Institute for Atmospheric and Earth System Research/Physics, Faculty of Science, University of  
9 Helsinki, 00014 Helsinki, Finland

10 <sup>2</sup> Aerosol and Haze Laboratory, Beijing Advanced Innovation Center for Soft Matter Science and  
11 Engineering, Beijing University of Chemical Technology, 100029 Beijing, China

12 <sup>3</sup> Joint International research Laboratory of Atmospheric and Earth System Research, School of  
13 Atmospheric Sciences, Nanjing University, 210023 Nanjing, China

14 <sup>4</sup> Nanjing-Helsinki Institute in Atmospheric and Earth System Sciences, Nanjing University, Nanjing,  
15 210023, China.

16 <sup>5</sup> Shanghai Key Laboratory of Atmospheric Particle Pollution and Prevention (LAP<sup>3</sup>), Department of  
17 Environmental Science and Engineering, Jiangwan Campus, Fudan University, 200438 Shanghai,  
18 China

19 <sup>6</sup> Minerva Independent Research Group, Max Planck Institute for Chemistry, 55128 Mainz, Germany  
20

21 *Correspondence to:* Markku Kulmala (markku.kulmala@helsinki.fi)  
22

23 **Abstract**  
24

25 Atmospheric new particle formation (NPF), together with secondary production of  
26 particulate matter in the atmosphere, dominate aerosol particle number concentrations and  
27 submicron particle mass loads in many environments globally. Our recent investigations  
28 show that atmospheric NPF produces a significant amount of particles on days when no  
29 clear NPF event has been observed/identified. Furthermore, it has been observed in  
30 different environments all around the world that growth rates of nucleation mode particles  
31 vary little, usually much less than the measured concentrations of condensable vapors. It has  
32 also been observed that the local clustering, which in many cases acts as a starting point of  
33 regional new particle formation (NPF), can be described with the formation of intermediate  
34 ions at the smallest sizes. These observations, together with a recently developed ranking  
35 method, leads us to propose a paradigm shift in atmospheric NPF investigations. In this  
36 opinion paper, we will summarize the traditional approach to describe atmospheric NPF, and  
37 describe an alternative method, covering both particle formation and initial growth. The  
38 opportunities and remaining challenges offered by the new approach are discussed.  
39

40 **1. Background**  
41

42 Atmospheric new particle formation (NPF) includes the formation of molecular clusters via  
43 different chemical pathways, and the activation of some of these clusters for growth to  
44 larger sizes (Zhang et al., 2012; Kulmala et al., 2014; Lee et al., 2019; Kirkby et al., 2023).  
45 Depending on their subsequent fate in the atmosphere, essentially whether and how long  
46 they will survive from various sink processes (e.g. Kerminen et al., 2004; Pierce and Adams,  
47 2007; Kulmala et al., 2017; Cai et al., 2022), these newly formed particles will contribute to  
48 the cloud condensation nuclei in a regional and global atmosphere (Spracklen et al., 2008;  
49 Wiedensohler et al., 2009; Kerminen et al., 2012; Gordon et al., 2017; Ren et al., 2021), and



50 will act as seeds for haze particles during air pollution episodes in urban environments (e.g.  
51 Guo et al., 2014; Kulmala et al., 2022a).

52

53 In order to understand how atmospheric NPF influences climate and air quality, and how  
54 these influences have changed over time or will change in the future as a result of  
55 anthropogenic and natural emission changes, we need to have detailed knowledge about  
56 the following issues in different atmospheric environments: 1) what is the general  
57 characteristics of atmospheric NPF, including its frequency and intensity, 2) by which  
58 chemical mechanisms and constituents molecular clusters form and grow to larger sizes, and  
59 3) how effectively newly formed particles reach sizes relevant to climate or air quality. In this  
60 opinion paper, we will focus on the first issue, acknowledging that the synergic effects of all  
61 of them need to be considered in order to get a full understanding of atmospheric NPF. We  
62 concentrate solely on field observations, as the power of laboratory experiments and model  
63 simulations is the strongest when investigating the issues 2 and 3 mentioned above.

64

65 Traditionally, the general characteristics of atmospheric NPF have been investigated in terms  
66 of discrete NPF events, during which the formation of new particles has been so intense that  
67 a new mode of particles has been clearly observed. These events take place either locally  
68 close to emission sources of precursor compounds for this phenomenon, or regionally over  
69 distances up to 1000 km or more (Kerminen et al., 2018, and references therein; Chu et al.  
70 2019). With this approach, the particle concentrations resulting from atmospheric NPF can  
71 be quantified only for these clear NPF events, leaving little room for potentially low-intensity  
72 NPF in a regional atmosphere, and providing practically no tools to handle local NPF.  
73 Another problem with the traditional approach is that the subsequent growth of newly  
74 formed particle to sizes relevant to climate or air quality can only be estimated for a small  
75 subset of cases, essentially those when both particle formation and growth take place  
76 relatively homogeneously in the regional atmosphere. In this opinion paper, we will propose  
77 an alternative approach to investigate atmospheric NPF, covering both particle formation  
78 and initial growth on all days with suitable aerosol data. We will discuss the opportunities  
79 that the new approach will offer for future investigations, as well as the remaining  
80 challenges, noting its complementary role when compared with traditional NPF event  
81 analysis and large-scale atmospheric model simulations.

82

## 83 **2. Approaches to investigate atmospheric NPF using field observations**

84

85 In this section, we will shortly discuss the approach traditionally used to investigate  
86 atmospheric NPF, including the history leading to this approach and its weaknesses. Based  
87 on our very recent work and findings, we then propose an alternative approach to tackle the  
88 problem, which may lead to a paradigm shift in investigating the general characteristics of  
89 atmospheric new particle formation using field observations. The main features of both  
90 approaches are summarized in Figures 1 and 2, and are discussed in more detail in the  
91 following sections.

92

### 93 **2.1 Traditional approach and associated shortcomings**

94

95 Before continuous field observations, our understanding of atmospheric NPF relied entirely  
96 on theories and laboratory experiments. The first steps of NPF were described using classical



97 nucleation theories which predict a very high dependence of the particle formation rate, or  
98 the nucleation rate, on the concentrations of vapors participating in NPF (e.g. Doyle et al.,  
99 1961; Jaecker-Voirol and Mirabel, 1989; Kulmala et al., 1991; Vehkamäki et al., 2002; Gaman  
100 et al., 2005). For many years, in conducted laboratory experiments, it has been assumed the  
101 binary water-sulfuric acid nucleation to be the only atmospherically relevant NPF pathway,  
102 and early experiments on this system supported the high sensitivity of the nucleation rate to  
103 the gas-phase sulfuric acid concentration (e.g. Wyslouzil et al., 1991; Viisanen et al., 1997).  
104 As a consequence, atmospheric NPF was essentially thought to be an on/off phenomenon  
105 that occurred sporadically under specific atmospheric conditions, essentially at high sulfuric  
106 acid concentrations.

107  
108 The first field measurements of atmospheric NPF were made for specific types of plumes,  
109 including power plant plumes, in which NPF did not reach a regional extent (see Kerminen et  
110 al., 2018, and references therein). Such measurements were campaign-based, and thus lack  
111 of a statistical view on how frequent and intense NPF was or whether the newly formed  
112 particles were able to grow into sizes relevant to climate or air quality. Later field  
113 observations, based either on campaign-wise or more continuous measurements at fixed  
114 locations, made it possible to identify and characterize regional NPF (Mäkelä et al., 1997;  
115 Kulmala et al., 2004; Kerminen et al., 2018). While such observations have dramatically  
116 enhanced our understanding of atmospheric NPF, they suffered from instrumental  
117 limitations and the non-homogenous nature of air masses that reach the measurement  
118 sites. As a result, it became a common practice to characterize regional NPF by first  
119 estimating the NPF event frequency at the measurement site (i.e. fraction of days showing  
120 clear NPF) using some NPF event classification criteria (e.g. Dal Maso et al., 2005; Kulmala et  
121 al., 2012; Dada et al., 2018), and then determining particle formation and growth rates for a  
122 relatively small sub-set of days (Fig. 1), essentially those being strong and homogenous  
123 enough to permit determination of these quantities (e.g. Nieminen et al., 2018; Chu et al.,  
124 2019; Kanawade et al., 2022).

125  
126 The traditional approach to investigate and characterize atmospheric NPF based on event  
127 classification has obvious drawbacks. First, there are days when NPF is either relatively weak  
128 or affected by air mass non-homogeneities or changing weather conditions. Such days are  
129 often classified as undefined days (e.g. Dal Maso et al., 2005; Kulmala et al., 2012),  
130 sometimes further divided into a small number of sub-categories (e.g. Buenrostro Mazon et  
131 al., 2009, Dada et al., 2018), or incorrectly classified as non-event days (Kulmala et al.,  
132 2022b). Long-term measurements indicate that these specious days tend to constitute a  
133 large or even a dominant fraction of all the days (Asmi et al., 2011; Kyrö et al., 2014; Dada et  
134 al., 2017; Wang et al., 2017; Kalivitis et al., 2019; Salma and Nemeth, 2019). Further analyses  
135 are thus difficult for such days. Second, the traditional approach is limited in providing  
136 information about the spatial and temporal variability of regional NPF, especially what it  
137 comes to regional intensity. Third, this approach is practically unable to capture sub-regional  
138 scale NPF.

139  
140 Over the years, the expected on/off behavior of atmospheric NPF has not been borne out by  
141 observations. Since the first simultaneous measurements of NPF and gas-phase sulfuric acid  
142 concentration (Weber et al., 1995, 1996), it has become clear that the observed formation  
143 rate of new particles in the atmosphere often scales between the first and second power of



144 the sulfuric acid concentration (Sihto et al., 2006; Kuang et al., 2008), a much weaker  
145 dependency than predicted by classical nucleation theories discussed above. Such relatively  
146 weak dependence of the particle formation rate on gas-phase concentrations of compounds  
147 participating in NPF have been observed for practically all the NPF pathways identified as  
148 relevant for the atmosphere (Paasonen et al., 2010; Lehtipalo et al., 2018; Yao, et al., 2018;  
149 Brean et al., 2020; He et al., 2021; Yan et al., 2021; Kirkby et al., 2023). Combining these field  
150 and laboratory observations strongly indicates that atmospheric NPF requires additional  
151 vapors beyond sulfuric acid and occurs over much larger concentration ranges of its  
152 precursor compounds than that has been thought before. Kulmala et al. (2022b) developed  
153 a method by which one could detect and even quantify NPF intensity on days traditionally  
154 classified as non-event days. They showed that NPF indeed occurs on such days, and that the  
155 overall contribution of this “quiet NPF” appears to be non-negligible.

156  
157 Investigations on the growth of newly formed particles to larger sizes are usually based on a  
158 relatively small subset of days on which NPF clearly occurs at a measurement site (Fig. 1).  
159 The main reason for this is that the most commonly used methods for determining particle  
160 growth rates (GR) are applicable only for days during which there is a clear new mode of  
161 particles present in the particle size distribution that can be followed for several hours, and  
162 that the growth is minimally affected by changes in measured air masses (e.g. Dal Maso et  
163 al., 2005; Kulmala et al., 2012). There is a danger that this approach gives a biased view on  
164 GR associated with atmospheric NPF, including the average level, variability and particle size  
165 dependency of GR. Another, very common assumption is that the GR is determined almost  
166 solely by condensation of low-volatile vapors onto newly formed particles. This view is  
167 challenged by the relatively low observed variability of GR in general (e.g. Kerminen et al.,  
168 2018; Nieminen et al., 2018) and, even more importantly, by the weak dependence of the  
169 observed GR on concentrations of “known” low-volatility vapors (Kulmala et al., 2022b;  
170 2023a). Acknowledging that aerosol physical processes (coagulation, cluster collisions with  
171 growing particles) have usually minor influences on GR (Stolzenburg et al., 2023), this  
172 “growth mystery” has two apparent explanations: either 1) our understanding of the mixture  
173 of vapors effectively condensing onto small particles, or the associated thermodynamics, is  
174 fundamentally incomplete, or 2) more volatile atmospheric vapors contribute to GR, e.g. via  
175 chemical reactions on particle surfaces.

## 176 177 **2.2 New, alternative approach and new opportunities**

178  
179 Instead of separately estimating the frequency of NPF and its intensity for a sub-set of days,  
180 often comprising only a small fraction of all days at any individual site, we propose that  
181 these two quantities will be combined into a probability distribution of the intensity of NPF  
182 that in practice covers all the days (Fig. 2). The intensity of NPF would in this case mean the  
183 formation rates of new particles,  $J_c$ , at some fixed particle diameter,  $d_c$ . Provided that the  
184 particle growth rate at sizes close to or slightly above  $d_c$  are known, or can be estimated, the  
185 values of  $J_c$  can be determined in the same way as in the traditional NPF event analysis (e.g.  
186 Kulmala et al., 2012), and then integrated over a desired period of time (daily, sub-daily or  
187 instantaneous values). To get the best benefit of the derived distribution of  $J_c$  for subsequent  
188 application purposes, the chosen value of  $d_c$  should be large enough so that the complicated  
189 and poorly understood processes that determine survival probabilities of growing clusters  
and particles would mainly be restricted to the sizes below  $d_c$  (see, e.g. Kulmala et al., 2017).



191 According to our current understanding in this respect (Cai et al., 2022; Tuovinen et al.,  
192 2022),  $d_c$  should be 3 nm in minimum, and preferably somewhat larger under heavily  
193 polluted conditions. The upper limit of  $d_c$  should be selected so that the calculated values of  
194  $J_c$  would be minimally affected by primary particle sources. While this is of little concern in  
195 remote or most rural areas, fresh primary particles, e.g., from traffic emission, are known to  
196 extend size well below 10 nm (e.g. Rönkkö et al., 2017).

197

198 An immediate question is how to determine the probability distribution of the particle  
199 formation rate, especially at the lower end of this distribution that represents weak to  
200 moderate NPF. Kulmala et al. (2022b) demonstrated that by averaging and suitably scaling  
201 over a large number of measurement days, it is possible to estimate particle formation rates  
202 on days previously classified as non-event days using the traditional NPF event classification  
203 methods. However, this is not the only available option. Previous analyses have shown that  
204 atmospheric NPF is strongly associated with concentrations of intermediate ions, i.e., ions in  
205 the size range from 2 to a few nm (e.g. Tammet et al., 2014; Leino et al., 2016). Motivated by  
206 this finding, we recently investigated the sensitivity of NPF to the total particle number  
207 concentration in a similar size range using long-term measurement data from the SMEAR II  
208 station in Finland (Aliaga et al., 2023). We found that the days with higher (suitably scaled)  
209 2.5-5 nm particle concentrations (high ranking) showed, on average, both higher particle  
210 formation rates and, in terms of traditional NPF event classification, higher probability of a  
211 NPF event to occur (Aliaga et al., 2023). Such a ranking method appears a promising  
212 candidate for creating a probability distribution of particle formation rates; however, its  
213 performance in different environments needs to be carefully tested. So far, besides the  
214 SMEAR II station, the preliminary results from a mountain site (Chacaltaya in Bolivia) and  
215 polluted sites (Beijing in China, Po Valley in Italy) are promising.

216

217 Concerning particle growth, we propose that rather than determining GR for only a small  
218 subset of days, as usually done when analyzing field measurements, one should aim to find a  
219 relation between GR and the prevailing chemical environment. By a chemical environment  
220 we mean the presence (concentrations) of vapors that potentially contribute to GR, and the  
221 activity of processes (condensation, heterogeneous reactions) that link these vapors to GR.  
222 Important to keep in mind when doing all this that not only the least volatile vapors, but also  
223 more volatile vapors capable of producing non-volatile vapors via heterogeneous reactions  
224 in and/or on particles, may have a significant contribution to GR. To a first approximation,  
225 regionally representative values of GR and its variability could be derived using the largest  
226 sub-set of high rankings that display particle growth; even without detailed information  
227 needed to tie GR with the chemical environment. This approach can be justified by the fact  
228 that GR is determined by the prevailing chemical environment rather than the intensity of  
229 NPF, so that losing information from days with low-intensity NPF does not cause a serious  
230 bias in GR estimates (Kulmala et al., 2022b). It is also possible to improve the  
231 representativity of the GR for individual days, depending on the length of the observation  
232 data set, by investigating the typical GR in different seasons, under different meteorological  
233 conditions or under otherwise varying situations in the chemical environment.

234

235 Analysis of both ions and particles in the 2-5 nm size range might provide a tool to combine  
236 regional and sub-regional NPF into the same framework. By using measurement data from  
237 the SMEAR II station and performing a theoretical analysis, it was demonstrated that the



238 concentration of negative ions in a narrow size range of 2.0-2.3 nm could be related to the  
239 intensity of NPF averaged over a spatial scale of the order 1 km surrounding the  
240 measurement site (Kulmala et al., 2023b; Tuovinen et al., 2023). If this is more generally  
241 applicable, including other sites with differing molecular particle forming mechanisms,  
242 targeted measurements of 2.0-2.3 nm ions could thereby be applied for identifying, and  
243 possibly quantifying, how effectively a specific (local) environment will produce new  
244 particles into the atmosphere (Fig. 2). Although such measurements say nothing about the  
245 subsequent fate of these particles, to a first approximation we may assume that they will  
246 grow essentially in the same manner as any newly formed in the same regional atmosphere  
247 or, more specifically, in the same prevailing chemical environment mentioned above. We  
248 cannot go to sizes smaller than 2 nm, since concentrations and dynamics of smaller ions are  
249 determined by processes which have very little to do with NPF (e.g. Tammet et al., 2014).

250

### 251 **3. Paradigm shift and remaining challenges**

252

253 Based on these recent results and the new reasoning presented above, we suggest the  
254 following paradigm shift when investigating the general characteristics of atmospheric NPF  
255 using field measurements (see also Fig. 2):

256

- 257 1) Instead of making binary (event, non-event) classification of NPF, we will utilize all  
258 days in the analysis and use a more continuous approach, such as the ranking  
259 method, for statistical information on the intensity of NPF.
- 260 2) We use particle and ion number concentrations in the smallest possible size regimes:
  - 261 a. total particles (2.5-5 nm) or intermediate ions (2-7 nm) to study regional NPF;
  - 262 b. ions at diameters as close to 2 nm as possible to study local NPF.
- 263 3) We use the regionally representative particle growth rates, derived from the largest  
264 possibly subset of data, to calculate:
  - 265 a. regional values of particle formation rates at selected sizes (3-5 nm) and  
266 integrated over desired time periods (instantaneous to daily);
  - 267 b. local particle formation rates over selected areas, and their relative  
268 contributions to regional NPF.
- 269 4) The particle formation rates can be determined for all days, and its distribution can  
270 be given as a continuous function of different parameters.

271

272 The main advance over the traditional method is that the new paradigm provides estimates  
273 of particle formation rates for all measurement days, and in principle even continuous values  
274 as a function of time. But it remains to be investigated what the best time resolution is for  
275 doing this analysis in practice. This, together with regionally representative particle growth  
276 rates, provides us with a tool to quantify the contributions of both local and regional NPF to  
277 total particle number concentrations in a regional atmosphere.

278

279 Despite its highly promising potential to investigate atmospheric NPF, the new paradigm  
280 faces apparent challenges as well. For example, while continuous aerosol size distribution  
281 measurements are being conducted in tens of locations worldwide (e.g. Rose et al., 2020), a  
282 dominant fraction of these sites do not currently have proper instrumentation (e.g. NAIS,  
283 PSM, nano-DMPS) for measuring the sub-5 nm size range needed for applying the





284 alternative approach introduced here. The lack of available measurement is even more  
285 severe for ion measurements necessary for determining local or sub-regional NPF.

286

287 Dealing with primary emissions has been found to be difficult when investigating regional  
288 NPF in polluted environments (e.g. Woo et al., 2001; Ahlm et al., 2012; Nemeth et al., 2018;  
289 Pushpawela et al., 2018; Zhou et al., 2020; Kanawade et al., 2022), and the new, alternative  
290 approach introduced here is not expected to be free from the influences of primary  
291 emissions. Recently, methods have been developed to estimate, and potentially exclude,  
292 primary emissions originating from emissions, e.g., traffic (Okuljar et al., 2021; Chen et al.,  
293 2023), but the suitability of these methods for this proposed new approach remains to be  
294 investigated.

295

296 Particle survival probabilities are sensitive to the combined effect of the degree of pollution  
297 and particle growth rate, and there are large uncertainties in predicting this quantity in the  
298 sub-3 to 5 nm size range, especially in polluted environments (e.g. Kulmala et al., 2017; Cai  
299 et al., 2022; Tuovinen et al., 2022). This feature does not cause a major problem for  
300 investigating regional NPF, as long as the probability distribution of the particle formation  
301 rate is derived at large enough sizes, preferably at 5 nm and in minimum at 3 nm.  
302 Concerning sub-regional NPF, we need to investigate whether and how particle survival  
303 probabilities influence the connection between ion concentrations close to 2 nm and  
304 particles formation rates at larger sizes, as this connection is expected to depend on the  
305 environment and prevailing conditions.

306

307 It is well known that both the occurrence and intensity of NPF vary seasonally at most of the  
308 sites (Dall'Osto et al., 2018; Kerminen et al., 2018; Nieminen et al., 2018; Chu et al., 2019,  
309 Brean et al., 2023). The potentially large seasonal variability of the mixture and  
310 concentrations of vapors contributing particle formation and growth needs to be kept in  
311 mind when calculating particle formation and growth rates, and when determining  
312 representative distributions for these quantities.

313

314 Finally, although the tools introduced here provide a first-order estimate on particle growth  
315 rates in different environments, we are far from a full understanding on which vapors and  
316 processes determine GR in different environmental conditions. As a result, much future work  
317 is needed to define, characterize, and quantify the chemical regimes and processes that  
318 eventually determine GR and its variability, and how this variability feeds back into  
319 estimating particle formation rates during low-intensity NPF.

320

#### 321 **4. Conclusions**

322

323 In this opinion, we have proposed a new method/approach and elucidated a paradigm shift  
324 in investigating atmospheric NPF using field observations. Contrary to the traditional event-  
325 based classification of individual days, the new approach looks at atmospheric NPF in a more  
326 statistical sense, aiming to create a probability distribution of particle formation and growth  
327 rates for all the days from continuous measurements at individual sites. While generally  
328 applicable to regional NPF, we also present ideas on how this same framework could be  
329 extended to sub-regional, or local, NPF.

330



331 The new approach provides a method to quantitatively estimate the contribution of  
332 atmospheric NPF to particle number concentration budgets in a regional atmosphere. If  
333 supported by additional measurements in areas with distinct sources for NPF precursors, the  
334 relative contributions of such source areas to the regional NPF can, in principle, be  
335 estimated. The results from the new approach can be extended to continental scales,  
336 provided that continuous measurement data from different representative regions are  
337 available.

338

339 The approach proposed here should be thought as complementary to the traditional NPF  
340 event analysis and large-scale model simulations. The traditional NPF event analysis has  
341 been widely used in the past, so its application to an entirely new data set offers a simple  
342 way to get idea on how important NPF is in that particular environment, and how it  
343 compares to other environments investigated earlier. The traditional NPF event analysis  
344 remains to be a powerful tool to select cases (days) for some special investigation purposes,  
345 such as investigating atmospheric NPF pathways and associated precursor chemistry  
346 associated with atmospheric NPF. The large-scale view on atmospheric NPF, including its  
347 climatic and health effects, as well as the associated feedback processes, has relied almost  
348 entirely on model simulations in the past. The proposed approach brings atmospheric  
349 measurements on NPF closer to results from large-scale model simulations and, at the very  
350 least, the new paradigm offers an improved way to utilize measurement data to constrain  
351 and evaluate models simulating atmospheric NPF.

352

#### 353 **Author contribution**

354 Markku Kulmala and Veli-Matti Kerminen had the original idea for opinion paper. YC, AD  
355 and DRW contributed to developing the idea further. VMK, MK, KL and PP wrote the first  
356 version of the paper. DA, ST, RC, CY, FB, TP, VMK and MK developed the material and  
357 results behind the opinion paper. All coauthors contributed the final version of the paper.

358

#### 359 **Competing interests**

360

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376 **References**

377

378 Ahlm, L., Liu, S., Day, D. A., Russell, L. M., Weber, R., Gentner, D. R., Goldstedin, A. H.,  
379 DiGangi, J. P., Henry, S. B., Keutsch, F. N., VandenBoer, T. C., Markovic, M. Z., Murphy, J. G.,  
380 Ren, X., and Scheller, S.: *J. Geophys. Res.*, **117**, D00V08, doi:10.1029/2011JD017144, 2012.

381

382 Aliaga, D., Tuovinen, S., Zhang, T., Lampilahti, J., Li, X., Ahonen, L., Kokkonen, T., Nieminen, T.,  
383 Hakala, S., Paasonen, P., Bianchi, F. Worsnop, D. R., Kerminen, V.-M., and Kulmala, M.: Nano  
384 Ranking Analysis: determining NPF event occurrence and intensity based on the  
385 concentration spectrum of formed (sub-5 nm) particles. *Aerosol Research* (accepted for  
386 publication), 2023.

387

388

389 Asmi, E., Kivekäs, N., Kerminen, V.-M., Komppula, M., Hyvärinen, A.-P., Hatakka, J., Viisanen,  
390 Y., and Lihavainen, H.: Secondary new particle formation in Northern Finland Pallas site  
391 between the years 2000 and 2010, *Atmos. Chem. Phys.*, **11**, 12959-12972, 2011.

392

393 Brean, J., Beddows, D. C. S., Shi, Z., Temime-Roussel, B., Marchand, N., Querol, X., Alastuey,  
394 A., Minguillon, M. C., and Harrison, R. M.: Molecular insights into new particle formation in  
395 Barcelona, Spain, *Atmos. Chem. Phys.*, **20**, 10029-10045, 2020.

396

397 Brean, J., Beddows, D. C. S., Harrison, R. M., Song, C., Tunved, P., Ström, J., Krejci, R., Freud,  
398 E., Massling, A., Skov, H., Asmi, E., Lupi, A., and Dall'Osto, M.: Collective geographical  
399 ecoregions and precursor sources driving Arctic new particle formation, *Atmos. Chem. Phys.*,  
400 **23**, 2183-2198, 2023.

401

402 Buenrostro Mazon, S., Riipinen, I., Schultz, D. M., Valtanen, M., Dal Maso, M., Sogacheva, L.,  
403 Junninen, H., Nieminen, T., Kerminen, V.-M., and Kulmala, M.: Classifying previously  
404 undefined days from eleven years of aerosol-particle-size distribution data from the SMEAR  
405 II station, Hyytiälä, Finland, *Atmos. Chem. Phys.*, **9**, 667-676, <https://doi.org/10.5194/acp-9-667-2009>, 2009.

406

407  
408 Cai, R., Deng, C., Stolzenburg, D., Li, C., Guo, J., Kerminen, V.-M., Jiang, J., Kulmala, M., and  
409 Kangasluoma, J.: Survival probability of new atmospheric particles: closure between theory  
410 and measurements from 1.4 to 100 nm, *Atmos. Chem. Phys.*, **22**, 14571-14587, 2022.

411

412 Chen, L., Qi, X., Niu, G., Li, Y., Liu, C., Lai, S., Liu, Y., Nie, W., Yan, C., Wang, J., Chi, X.,  
413 Paasonen, P., Hussein, T., Lehtipalo, K., Kerminen, V.-M., Petäjä, T., Kulmala, M., Ding, A.:  
414 High concentration of atmospheric sub-3 nm particles in polluted environment of eastern  
415 China: New particle formation and traffic emission. *J. Geophys. Res. Atmos.*, **128**,  
416 e2023JD039669, 2023.

417

418 Chu, B., Kerminen, V.-M., Bianchi, F., Yan, C., Petäjä, T., and Kulmala, M.: Atmospheric new  
419 particle formation in China, *Atmos. Chem. Phys.*, **19**, 115-138, 2019.

420

421 Dada, L., Paasonen, P., Nieminen, T., Buenrostro-Mazon, S., Kontkanen, J., Peräkylä, O.,  
422 Lehtipalo, K., Hussein, T., Petäjä, T., Kerminen, V.-M., Bäck, J., and Kulmala M.: Long-term



- 423 analysis of clear-sky new particle formation events and nonevents in Hyytiälä, *Atmos. Chem.*  
424 *Phys.*, **17**, 6227-6241, 2017
- 425
- 426 Dada, L., Chellapermal, R., Buenrostro Mazon, S., Paasonen, P., Lampilahti, J., Manninen, H.  
427 E., Junninen, H., Petäjä, T., Kerminen V.-M., and Kulmala, M.: Refined classification and  
428 characterization of atmospheric new-particle formation events using air ions, *Atmos. Chem.*  
429 *Phys.*, **18**, 17883-17893, 2018.
- 430
- 431 Dall'Osto, M., Beddows, D. C. S., Asmi, A., Poulain, L., Hao, L., Freney, E., Allan, J. D.,  
432 Canagaratna, M., Crippa, M., Bianchi, F., de Leeuw, G., Eriksson, A., Swietlicki, E., Hansson, H.  
433 C., Heinzing, J. S., Cranier, C., Zemannova, K., Laj, P., Onasch, T., Prevot, A., Putaud, J. P.,  
434 Sellegri, K., Vidal, M., Virtanen, A., Simo, R., Worsnop, D., O'Dowd, C., Kulmala, M., and  
435 Harrison, R. M.: Novel insights on new particle formation from a pan-european observing  
436 system, *Sci. Rep.*, **8**, 1482, doi:10.1038/s41598-017-17333-9, 2018.
- 437
- 438 Doyle, G. J.: Self-nucleation in the sulfuric acid-water system, *J. Chem. Phys.*, **35**, 795-799,  
439 1961.
- 440
- 441 Gaman, A., Napari, I., Winkler, P. M., Vehkamäki, H., Wagner, P. E., Strey, R., Viisanen, Y., and  
442 Kulmala, M.: Homogeneous nucleation of n-nonane and n-propano mixtures: A comparison of  
443 classical nucleation theory and experiments, *J. Chem. Phys.*, **123**, 244502, 2005.
- 444
- 445 Gordon, H., Kirkby, J., Baltensperger, U., Bianchi, F., Breitenlechner, M., Curtius, J., Dias, A.,  
446 Dommen, J., Donahue, N. M., Dunne, E. M., Duplissy, J., Ehrhart, S., Flagan, R. C., Frege, C.,  
447 Fuchs, C., Hansel, A., Hoyle, C. R., Kulmala, M., Kürten, A., Lehtipalo, K., Makhmutov, V.,  
448 Molteni, U., Rissanen, M. P., Stozhkov, Y., Tröstl, J., Tsakogeorgas, G., Wagner, R., Williamson,  
449 C., Wimmer, D., Winkler, P. M., Yan, C., and Carslaw, K. S.: Causes and importance of new  
450 particle formation in the present-day and preindustrial atmospheres. *J. Geophys. Res.*  
451 *Atmos.*, **122**, 8739-8760, 2017.
- 452
- 453 Guo, S., Hu, M., Zamora, M. L., Peng, J., Shang, D., Zheng, J., Du, Z., Wu, Z., Shao, M., Zeng,  
454 L., Molina, M. J., and Zhang, R.: Elucidating severe urban haze formation in China, *Proc. Natl.*  
455 *Acad. Sci. U.S.A.*, **111**, 17373, 2014.
- 456
- 457 He, X.-C., Tham, Y. J., Dada, L., Wang, M., Finkenzeller, H., Stolzenburg, D., Iyer, S. Simon, M.,  
458 Kürten, A., Shen, J., Rörup, B., Rissanen, M., Schobesberger, S., Baalbaki, Wang, D. S., Koenig,  
459 T. K., Jokinen, T., Sarnela, N. Beck, L., Almeida, J., Amanatidis, S., Amorim<sup>1</sup>, A., Ataei, F.,  
460 Baccarini, A., Bertozzi, B., Bianchi, F., Brilke, S., Caudillo, L., Chen, D., Chiu, R., Chu, B., Dias,  
461 A., Ding, A., Dommen, J., Duplissy, J., El Haddad, I., Carracedo, L. G., Granzin, M., Hansell, A.,  
462 Heinritzi, M., Hofbauer, V., Junninen, H., Kangasluoma, J., Kempainen, D., Kim, C., Kong, W.,  
463 Krechmer, J. E., Kvashin, A., Laitinen, T., Lamkaddam, H., Lee, C. P., Lehtipalo, K., Leiminger,  
464 M., Li, Z., Makhmutov, V., Manninen, H. E., Marie, G., Marten, R., Mathot, S., Mauldin, R. L.,  
465 Mentler, B., Möhler, O., Müller, T., Nie, W., Onnela, A., Petäjä, T., Pfeifer, J., Philippov, M.,  
466 Ranjithkumar, A., Saiz-Lopez, A., Salma, I., Scholz, W., Schuchmann, S., Schulze, B., Steiner,  
467 G., Stozhkov, Y., Tauber, C., Tomé, A., Thakur, R. C., Väisänen, O., Vazquez-Pufleau, M.,  
468 Wagner, A. C., Wang, Y., Weber, S. K., Winkler, P. M., Wu, Y., Xiao, M., Yan, C., Ye, Q., Ylisirniö,  
469 A., Zauner-Wieczorek, M., Zha, Q., Zhou, P., Flagan, R. C., Curtius, J., Baltensperger, U.,



- 470 Kulmala, M., Kerminen, V.-M., Kurtén, T., Donahue, N. M., Volkamer, R., Kirkby, J., Worsnop,  
471 D. R., and Sipilä, M.: Role of iodine oxoacids in atmospheric aerosol nucleation. *Science*, 371,  
472 589-595, 2021.
- 473
- 474 Jaecker-Voirol, A. and Mirabel, P., Heteromolecular nucleation in the sulfuric acid-water  
475 system, *Atmos. Environ.*, 23, 2053-2057, 1989.
- 476
- 477 Kanawade, V. J., Sebastian, M., Hooda, R. K., and Hyvärinen, A.-P.: Atmospheric new particle  
478 formation in India: current understanding and knowledge gaps, *Atmos. Environ.*, 270,  
479 118894, 2022.
- 480
- 481 Kalivitis, N., Kerminen, V.-M., Kouvarikis, G., Stavroulas, I., Tzitzikalaki, E., Kalkavouras, P.,  
482 Daskalakis, N., Myriokefalitakis, S., Bougiatioti, A., Manninen, H. E., Roldin, P., Petäjä, T., Boy,  
483 M., Kulmala, M., Kanakidou, M., and Mihalopoulos, N.: Formation and growth of  
484 atmospheric nanoparticles in the eastern Mediterranean: results from long-term  
485 measurements and process simulation, *Atmos. Chem. Phys.*, 19, 2671-2686, 2019.
- 486
- 487 Kerminen, V.-M., Lehtinen, K. E. J., Anttila, T., and Kulmala, M.: Dynamics of atmospheric  
488 nucleation mode particles: a time scale analysis, *Tellus*, 56B, 135-146, 2004.
- 489
- 490 Kerminen, V.-M., Paramonov, M., Anttila, T., Riipinen, I., Fountoukis, C., Korhonen, H., Asmi,  
491 E., Laakso, L., Lihavainen, H., Swietlicki, E., Svenningsson, B., Asmi, A., Pandis, S. N., Kulmala,  
492 M., and Petäjä, T.: Cloud condensation nuclei production associated with atmospheric  
493 nucleation: a synthesis based on existing literature and new results, *Atmos. Chem. Phys.*, 12,  
494 12037-12059, 2012..
- 495
- 496 Kerminen, V.-M., Chen, X., Vakkari, V., Petäjä, T., Kulmala, M., and Bianchi, F.: Atmospheric  
497 new particle formation and growth: review of field observations, *Environ. Res. Lett.*, 13,  
498 103003, 2018.
- 499
- 500 Kirkby, J., Amorim, A., Baltensperger, U., Carslaw, K. S., Christoudias, T., Curtius, J., Donahue,  
501 N. M., El Haddad, I., Flagan, R. C., Gordon, H., Hansel, A., Harder, H., Junninen, H., Kulmala,  
502 M., Kurten, A., Laaksonen, A., Lehtipalo, K., Lelieveld, J., Mohler, O., Riipinen, I., Stratmann,  
503 F., Tome, A., Virtanen, A., Volkamer, R., Winkler, P. M., and Worsnop, D. R.: Atmospheric new  
504 particle formation from the CERN CLOUD experiment, *Nature Geosci.*, 16, 948-957, 2023.
- 505
- 506 Kuang, C., McMurry, P. H., McCormick, A. V., and Eisele, F., L.: Dependence of nucleation  
507 rates on sulfuric acid vapor concentration in diverse atmospheric locations, *J. Geophys. Res.*,  
508 113, D10209, doi:10.1029/2007JD009253, 2008.
- 509
- 510 Kulmala, M., Laaksonen, A., and Jokiniemi, J.: Numerical simulations of binary nucleation of  
511 hydrogen iodide and water vapors, *J. Aerosol Sci.*, 22, 149-157, 1991.
- 512
- 513 Kulmala, M., Vehkamäki, H., Petäjä, T., Dal Maso, M., Lauri, A., Kerminen, V.-M., Birmili, W.,  
514 and McMurry, P. H.: Formation and growth rates of ultrafine atmospheric particles: A review  
515 of observations, *J. Aerosol Sci.*, 35, 143-176, 2004.
- 516



- 517 Kulmala, M., Petäjä, T., Nieminen, T., Sipilä, M., Manninen, H. E., Lehtipalo, K., Dal Maso, M.,  
518 Aalto, P. P., Junninen, H., Paasonen, P., Riipinen, I., Lehtinen, K. E. J., Laaksonen, A., and  
519 Kerminen, V.-M.: Measurement of the nucleation of atmospheric aerosol particles, *Nat.*  
520 *Protoc.*, 7, 1651-1667, doi:10.1038/nprot.2012091, 2012.
- 521  
522 Kulmala, M., Petäjä, T., Ehn, M., Thornton, J., Sipilä, M., Worsnop, D. R., and Kerminen V.-M.:  
523 Chemistry of atmospheric nucleation: On the recent advances on precursor characterization  
524 and atmospheric cluster composition in connection with atmospheric new particle  
525 formation, *Annu. Rev. Phys. Chem.*, 65, 21-37, 2014.
- 526  
527 Kulmala, M., Kerminen, V.-M., Petäjä, T., Ding, A. J., and Wang, L.: Atmospheric gas-to-  
528 particle conversion: why NPF events are observed in megacities?, *Faraday Discuss.*, 200, 271-  
529 288, doi:10.1039/c6fd00257a, 2017.
- 530  
531 Kulmala, M., Cai, R., Stolzenburg, D., Zhou, Y., Dada, L., Guo, Y., Yan, C., Petäjä, T., Jiang, J.,  
532 and Kerminen, V.-M.: The contribution of new particle formation and subsequent growth to  
533 haze formation, *Environ. Sci.: Atmos.*, 2, 352-361, 2022a.
- 534  
535 Kulmala, M., Junninen, H., Dada, L., Salma, I., Weidinger, T., Thén, W., Vörösmarty, M.,  
536 Komsaare, K., Stolzenburg, D., Cai, R., Yan, C., Li, X., Deng, C., Jiang, J., Petäjä, T., Nieminen,  
537 T., and Kerminen, V.-M.: Quiet new particle formation in the atmosphere. *Front. Environ. Sci.*,  
538 10, 912385, doi: 10.3389/fenvs.2022.912385, 2022b.
- 539  
540 Kulmala, M., Cai, R., Ezhova, E., Deng, C., Stolzenburg, D., Dada, L., Guo, Y., Yan, C., Peräkylä,  
541 O., Lintunen, A., Nieminen, T., Kokkonen, T. V., Sarnela, N., Petäjä, T., and Kerminen, V.-M.:  
542 Direct link between the characteristics of atmospheric new particle formation and  
543 Continental Biosphere-Atmosphere-Cloud-Climature (COBACC) feedback loop, *Boreal. Env.*  
544 *Res.*, 28, 1-13, 2023a.
- 545  
546 Kulmala, M., Ke, P., Lintunen, A., Peräkylä, O., Lohtander, A., Tuovinen, S., Lampilahti, J.,  
547 Kolari, P., Schiesti-Aalto, P., Kokkonen, T., Nieminen, T., Dada, L., Ylivinkka, I., Petäjä, T., Bäck,  
548 J., Lohila, A., Heimsch, L., Ezhova, E., and Kerminen, V.-M.: A novel concept for assessing the  
549 potential of different boreal ecosystems to mitigate climate change (CarbonSink+ Potential),  
550 *Boreal Env. Res.*, 28 (in press), 2023b.
- 551  
552 Kyrö, E.-M., Väänänen, R., Kerminen, V.-M., Virkkula, A., Petäjä, T., Dal Maso, M., Nieminen,  
553 T., Juhola, S., Shcherbinin, A., Riipinen, I., Lehtipalo, K., Keronen, P., Aalto, P. P., Hari, P., and  
554 Kulmala M.: Trends in new particle formation in eastern Lapland, Finland: effect of  
555 decreasing sulfur emissions from Kola Peninsula, *Atmos. Chem. Phys.*, 14, 4383-4396, 2014.
- 556  
557 Lee, S.-H., Gordon, H., Yu, H., Lehtipalo, K., Haley, R., Li, X., and Zhang, R.: New particle  
558 formation in the atmosphere: From molecular clusters to global climate, *J. Geophys. Res.*  
559 *Atmos.*, 124, 7098-7146, 2019.
- 560  
561 Lehtipalo, K., Yan, C., Dada, L., Bianchi, F., Xiao, M., Wagner, R., Stolzenburg, D., Ahonen, L.  
562 R., Amorim, A., Baccarini, A., Bauer, P. S., Baumgartner, B., Bergen, A., Bernhammer, A.-K.,  
563 Breitenlechner, M., Brilke, S., Buckholz, A., Buenrostro Mazon, S., Chen, D., Chen, X., Dias, A.,



- 564 Dommen, J., Draper, D. C., Duplissy, J., Ehn, M., Finkenzeller, H., Fischer, L., Frege, C., Fuchs,  
565 C., Garmash, O., Gordon, H., Hakala, J., He, X., Heikkinen, L., Heinrzi, M., Helm, J. C.,  
566 Hofbauer, V., Hoyle, C. R., Jokinen, T., Kangasluoma, J., Kerminen, V.-M., Kim, C., Kirkby, J.,  
567 Kontkanen, J., Kürten, A., Lawler, M. J., Mai, H., Mathot, S., Mauldin, III R. L., Molteni, U.,  
568 Nichman, L., Nie, W., Nieminen, T., Ojdanic, A., Onnela, A., Passananti, M., Petäjä, T., Piel, F.,  
569 Pospisilova, V., Quéléver, L. L. J., Rissanen, M. P., Rose, C., Sarnela, N., Schallhart, S.,  
570 Schuchmann, S., Sengupta, K., Simon, M., Sipilä, M., Tauber, K., Tomé, A., Tröstl, J., Väisänen,  
571 O., Vogel, A. L., Volkamer, A., Wagner, A. C., Wang, M., Weitz, L., Wimmer, D., Ye, P., Ylisirniö,  
572 A., Zha, Q., Carslaw, K. S., Curtius, J., Donahue, N. M., Flagan, R. C., Hansel, A., Riipinen, I.,  
573 Virtanen, A., Winkler, P. M., Baltensperger, U., Kulmala, M., and Worsnop, D. R.:  
574 Multicomponent new particle formation from sulfuric acid, ammonia, and biogenic  
575 vapors, *Sci. Adv.*, 4, eaau5363, 2018.
- 576  
577 Leino, K., Nieminen, T., Manninen, H. E., Petäjä, T., Kerminen, V.-M., and Kulmala, M.:  
578 Intermediate ions as a strong indicator of new particle formation bursts in boreal forest,  
579 *Boreal Env. Res.*, 21, 274-286, 2016.
- 580  
581 Mäkelä, J. M., Aalto, P., Jokinen, V., Pohja, T., Nissinen, A., Palmroth, S., Markkanen, T.,  
582 Seitsonen, K., Lihavainen, H., and Kulmala, M.: Observations of ultrafine aerosol particle  
583 formation and growth in boreal forest, *Geophys. Res. Lett.*, 24, 1219-1222.
- 584  
585 Nemeth, Z., Rosati, B., Zikova, N., Salma, I., Bozo, L., de Espana, C. D., Schwartz, J., Zdimal, V.,  
586 and Wonaschutz, A.: Comparison of atmospheric new particle formation events in three  
587 Central European cities, *Atmos. Environ.*, 178, 191-197, 2018.
- 588  
589 Okuljar, M., Kuuluvainen, H., Kontkanen, J., Garmash, O., Olin, M., Niemi, J. V., Timonen, H.,  
590 Kangasluoma, J., Tham, Y. J., Baalbaki, R., Sipilä, M., Salo, L., Lintusaari, H., Portin, H., Teinilä,  
591 K., Aurela, M., Dal Maso, M., Rönkkö, T., Petäjä, T., and Paasonen, P.: Measurement report:  
592 The influence of traffic and new particle formation on the size distribution of 1–800 nm  
593 particles in Helsinki – a street canyon and an urban background station comparison, *Atmos.*  
594 *Chem. Phys.*, 21, 9931–9953, 2021.
- 595  
596 Paasonen, P.; Nieminen, T., Asmi, E., Manninen, H. E., Petäjä, T., Plass-Dülmer, C., Flentje, H.,  
597 Birmili, W., Wiedensohler, A., Hörrak, U., Metzger, A., Hamed, A., Laaksonen, A., Facchini, M.  
598 C., Kerminen, V.-M., and Kulmala, M.: On the roles of sulphuric acid and low-volatility organic  
599 vapours in the initial steps of atmospheric new particle formation, *Atmos. Chem. Phys.*, 10,  
600 11223-11242, 2010.
- 601  
602 Pierce, J. R. and Adams, P. J.: Efficiency of cloud condensation nuclei formation from ultrafine  
603 particles, *Atmos. Chem. Phys.*, 7, 1367-1379, 2007.
- 604  
605 Pushpawela, B., Jayaratne, R., and Morawska, L.: Differentiating between particle formation  
606 and growth events in an urban environment, *Atmos. Chem. Phys.*, 18, 11171-11183, 2018.
- 607  
608 Ren, J., Chen, L., Fan, T., Liu, J., Jiang, S., and Zhang, F.: The NPF effect on CCN number  
609 concentrations: a review and re-evaluation of observations from 35 sites worldwide,  
610 *Geophys. Res. Lett.*, 48, e2021GL095190, 2021.



611  
612 Rose, C. et al.: Seasonality of the particle number concentration and size distribution: a  
613 global analysis retrieved from the network of Global Atmosphere Watch (GAW) near-surface  
614 observations, *Atmos. Chem. Phys.*, 21, 17185-17223, 2021.  
615  
616 Rönkkö, T., Kuuluvainen, H., Karjalainen, P., Keskinen, J., Hillamo, R., Niemi, J. V., Pirjola, L.,  
617 Timonen, H. J., Saarikoski, S., Saukko, E., Järvinen, A., Silvennoinen, H., Rostedt, A., Olin, M.,  
618 Yli-Ojanperä, J., Nousiainen, P., Kousa, A., and Dal Maso, M.: Traffic is a major source of  
619 atmospheric nanocluster aerosol, *Proc. Natl. Acad. Sci. U.S.A.*, 114, 7549-7554, 2017.  
620  
621 Salma, I. and Nemeth, Z.: Dynamic and timing properties of new aerosol particle formation  
622 and consecutive growth events, *Atmos. Chem. Phys.*, 19, 5835-5852, 2019.  
623  
624 Sihto, S.-L., Kulmala, M., Kerminen, V.-M., Dal Maso, M., Petäjä, T., Riipinen, I., Korhonen, H.,  
625 Arnold, F., Janson, R., Boy, M., Laaksonen, A., and Lehtinen, K. E. J.: Atmospheric sulphuric  
626 acid and aerosol formation: implications from atmospheric measurements for nucleation  
627 and early growth mechanisms, *Atmos. Chem. Phys.*, 6, 4079–4091, doi:10.5194/acp-6-4079-  
628 2006, 2006.  
629  
630 Spracklen, D. V., Carslaw, K. S., Kulmala, M., Kerminen, V.-M., Sihto, S.-L., Riipinen, I.,  
631 Merikanto, J., Mann, G. W., Chipperfield, M. P., Wiedensohler, A., Birmili, W., and Lihavainen,  
632 H.: Contribution of particle formation to global cloud condensation nuclei concentrations,  
633 *Geophys. Res. Lett.*, 35, L06808, doi:10.1029/2007GL033038, 2008.  
634  
635 Stolzenburg, D., Cai, R., Blichner, S., Kontkanen, J., Zhou, P., Makkonen, R., Kerminen, V.-M.,  
636 Kulmala, M., Riipinen, I., and Kangasluoma, J.: Atmospheric nanoparticle growth, *Rev. Mod.*  
637 *Phys.*, 95, 045002, 2023.  
638  
639 Tammet, H, Komsaare, K., and Horrak, U.: Intermediate ions in the atmosphere, *Atmos. Res.*,  
640 135-136, 263-273, 2014.  
641  
642 Tuovinen, S., Cai, S., Kerminen, V.-M., Jiang, J., Yan, C., Kulmala, M., and Kontkanen, J.:  
643 Survival properties of atmospheric particles: comparison based on theory, cluster population  
644 simulations, and observations in Beijing, *Atmos. Chem. Phys.*, 22, 15071-15091, 2022.  
645  
646 Tuovinen S., Lampilahti J., Kerminen V.-M. & Kulmala M. 2023. Ion clusters as indicator for  
647 local new particle formation. *Egusphere* preprint 2023-1108,  
648 <https://doi.org/10.5194/egusphere-2023-1108>.  
649  
650 Vehkamäli, H., Kulmala, M., Napari, I., Lehtinen, K. E. J., Timmreck, c., Noppel, M., and  
651 Laaksonen, A.: An improved parameterization for sulfuric acid-water nucleation rates for  
652 tropospheric and stratospheric conditions. *J. Geophys. Res.*, 107, D22, 4622,  
653 doi:10.1029/2002JD002184, 2002.  
654  
655 Viisanen, Y., Kulmala, M., and Laaksonen, A.: Experiments on gas-liquid nucleation of sulfuric  
656 acid and water. *J. Chem. Phys.*, 107, 920-926, 1997.  
657





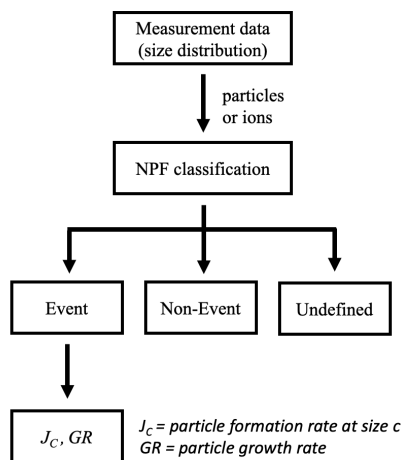
- 658 Wang, Z., Birmili, W., Hamed, A., Wehner, B., Spindler, G., Pei, X., Qu, Z., Cheng, Y., Su, H.,  
659 and Wiedensohler, A.: Contributions of volatile and nonvolatile compounds (at 300 °C) to  
660 condensational growth of atmospheric nanoparticles: An assessment based on 8.5 years of  
661 observations at the Central Europe background site Melpitz. *J. Geophys. Res. Atmos.*, **122**,  
662 485-497, 2017.
- 663
- 664 Weber, R. J., McMurry, P. H., Eisele, F. L. and Tanner, D. J.: Measurement of expected  
665 nucleation precursor species and 3 to 500 nm particles at Mauna Loa Observatory, Hawaii, J.  
666 *Atmos. Sci.*, **52**, 2242-2257, 1995.
- 667
- 668 Weber, R. J., Marti, J. J., McMurry, P. H., Eisele, F. L., Tanner, D. J, and Jefferson A.: Measured  
669 atmospheric new particle formation rates: Implications for nucleation mechanisms, *Chem.*  
670 *Eng. Commun.*, **151**, 53–64, 1996.
- 671
- 672 Wiedensohler, A., Cheng, Y., Nowak, A., Wehner, B., Achtert, P. Berghof, M., Birmili, W., Wu,  
673 Z. J., Hu, M., Zhu, T., Takegawa, N., Kita, K., Kondo, J., Lou, S. R., Hofzumahaus, A., Holland, F.,  
674 Wahner, A., Gunthe, S. S., Rose, D., Su, H., and Pöschl, U.: Rapid aerosol particle growth and  
675 increase of cloud condensation nucleus activity by secondary aerosol formation and  
676 condensation: a case study for regional air pollution in northeastern China, *J. Geophys. Res.*  
677 *Atmos.*, **114**, D00G08, doi:10.1029/2008JD010884, 2009.
- 678
- 679 Woo, K. S., Chen, D. R., Pui, D. Y. H., and McMurry, P. H.: Measurement of Atlanta size  
680 distributions: observations of ultrafine particle formation events, *Aerosol Sci. Technol.*, **34**,  
681 75-87, 2001.
- 682
- 683 Wyslouzil, B. E., Seinfeld, J. H., Flagan, R. C., and Okuyama, K.: Binary nucleation in acid-  
684 water and a comparison with methanesulfonic acid-water, *J. Chem. Phys.*, **94**, 6842-6850,  
685 1991.
- 686
- 687 Yan, C., Yin, R., Lu, Y., Dada, L., Yang, D., Fu, Y., Kontkanen, J., Deng, C., Garmash, O., Ruan, J.,  
688 Baalbaki, R., Schervish, M., Cai, R., Bloss, M., Chan, T., Chen, T., Chen, Q., Chen, X., Chen, Y.,  
689 Chu, B., Dällenbach, K., Foreback, B., He, X., Heikkinen, L., Jokinen, T., Junninen, H.,  
690 Kangasluoma, J., Kokkonen, T., Kurppa, M., Lehtipalo, K., Li, H., Li, H., Li, X., Liu, Y. Ma, Q.,  
691 Paasonen, P., Rantala, P., Pileci, R. E., Rusanen, A., Sarnela, N., Simonen, P., Wang, S., Wang,  
692 W., Wang, Y. Xue, M., Yang, G., Yao, L., Zhou, Y., Kujansuu, J., Petäjä, T., Nie, W., Ma, N., Ge,  
693 M., He, H., Donahue, N. M., Worsnop, D. R., Kerminen, V.-M., Wang, L., Liu, Y., Zheng, J.,  
694 Kulmala, M., Jiang, J., and Bianchi, F.: The synergistic role of sulfuric acid, bases, and oxidized  
695 organics governing new-particle formation in Beijing, *Geophys. Res. Lett.*, **48**,  
696 e2020GL091944, 2021.
- 697
- 698 Yao, L., Garmash, O., Bianchi, F., Zhen, J., Yan, C., Kontkanen, J., Junninen, H., Buenrostro  
699 Mazon, S., Ehn, M., Paasonen, P., Sipilä, M., Wang, M., Wang, X., Xiao, S., Chen, H., Lu, Y.,  
700 Zhang, B., Wang, D., Fu, Q., Geng, F., Li, L, Wang, H., Qiao, L., Yang, X, Chen, J., Kerminen, V-  
701 M., Petäjä, T., Worsnop, D. R., Kulmala, M., and Wang, L.: Atmospheric new particle  
702 formation from sulfuric acid and amines in a Chinese megacity, *Science*, **361**, 278-281, 2018.
- 703



- 704 Zhang, R., Khalizov, A., Wang, L., Hu, M., and Xu, W.: Nucleation and growth in the  
705 atmosphere, *Chem. Rev.*, 112, 1957-2011, 2012.  
706  
707 Zhou, Y., Dada, L., Liu, Y., Fu, Y., Kangasluoma, J., Chan, T., Yan, C., Chu, B., Daellenbach, K. R.,  
708 Bianchi, F., Kokkonen, T. V., Liu, Y., Kujansuu, J., Kerminen, V.-M., Petäjä, T., Wang, L., Jiang, J.,  
709 and Kulmala, M.: Variation of size-segregated particle number concentrations in wintertime  
710 Beijing, *Atmos. Chem. Phys.*, 20, 1201-1216, 2020.

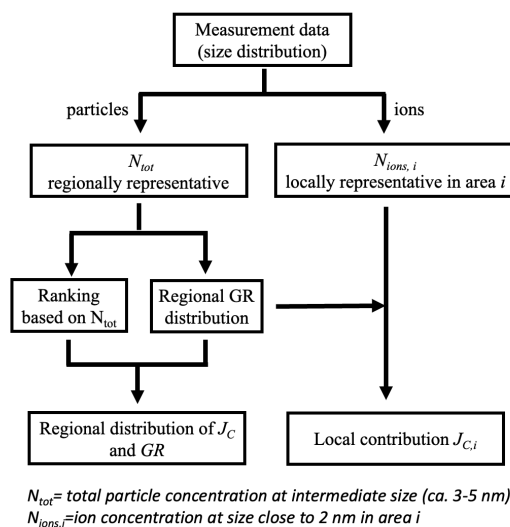


711 **Figures**



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Figure 1. Schematics of the traditional method used to characterize regional atmospheric NPF.



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Figure 2. Schematics of the new method proposed in this paper to characterize both regional and local NPF.