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On secondary new particle formation in China

Kulmala, Markku

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111 **Abstract**

112

113 Formation of new atmospheric aerosol particles is a global phenomenon that has been
114 observed to take place in even heavily-polluted environments. However, in all
115 environments there appears to be a threshold value of the condensation sink (due to
116 pre-existing aerosol particles) after which the formation rate of 3 nm particles is no
117 longer detected. In China, new particle production has been observed at very high
118 pollution levels (condensation sink about 0.1 s^{-1}) in several megacities, including
119 Beijing, Shanghai and Nanjing as well as in Pearl River Delta (PRD). Here we
120 summarize the recent findings obtained from these studies and discuss the various
121 implications these findings will have on future research and policy.

122

123 1. Background

124

125 Atmospheric aerosol particles affect our life and its quality in multiple ways. First of
126 all, the interaction between aerosols and climate system is the dominant uncertainty in
127 predicting the radiative forcing and future climate [1]. Secondly, aerosol particles
128 deteriorate both human health and visibility, especially in urban areas [2, 3]. Thirdly,
129 aerosol particles modify the intensity and distribution of radiation that reaches the
130 Earth's surface, having direct influences on photosynthesis and terrestrial carbon sink
131 [4]. Better understanding of the various effects in the atmosphere requires detailed
132 information on how different sources (including those related to the biosphere) and
133 atmospheric transformation processes modify the properties of aerosol particle
134 populations.

135

136 One of the most important phenomena associated with the atmospheric aerosol
137 number concentrations is the secondary formation of new aerosol particles. This
138 includes the production of molecular clusters from gaseous precursor vapors, the
139 activation and growth of some of these clusters to detectable sizes, and the further
140 growth up to the sizes at which the particles may act as cloud condensation nuclei
141 [e.g. 5, 6]. Although atmospheric new particle formation has been observed to take
142 place almost everywhere at favorable conditions in the boundary layer [7], our
143 knowledge about this phenomenon is still far from perfect [5, 8]. The current
144 knowledge gaps in this regard range from the basic process-level understanding of
145 secondary atmospheric aerosol formation to its connection with anthropogenic
146 activities, biogenic emissions, atmospheric chemistry, and ultimately with climate
147 change and human health.

148

149 Secondary formation of new atmospheric aerosol particles is typically initiated by
150 photochemical reactions in the gas phase, so that especially the production of
151 extremely low volatility vapors like sulfuric acid [9, 10, 11] and highly-oxidized
152 organic compounds [e.g. 12, 13, 14] is crucial. Pre-existing aerosol particles act as a
153 sink for the low-volatile vapors, as well as for small clusters and growing
154 nanoparticles, thereby hindering or even suppressing atmospheric new particle
155 formation [e.g. 15, 16, 17]. The atmospheric new particle formation is affected by
156 several meteorological quantities and phenomena, particularly in the planetary
157 boundary layer, including the intensity of solar radiation and atmospheric mixing

158 processes. The recent findings indicate that critical clusters may be surprisingly small
159 in size, if existing at all, under atmospheric conditions [e.g. 18], and thus treatable by
160 advanced quantum chemistry methods [19]. It is very probable that the atmospheric
161 new particle formation is a two-step process, i.e. initial clustering and then
162 condensational growth after activation of clusters, as suggested by Kulmala et al. [21]
163 and verified by Kulmala et al. [18]. A summary of the current understanding of gas-
164 to-particle conversion is presented by Kulmala et al. [5].

165 New aerosol particles formed in the atmosphere become climatically important when
166 they reach sizes larger than about 50–100 nm in diameter [6]. Particles of this size
167 and larger are able to act as cloud condensation nuclei and scatter visible light,
168 thereby affecting cloud microphysical properties [e.g. 22], reducing the fraction of
169 solar radiation reaching the Earth's surface and contributing to visibility degradation
170 [e.g. 23]. Furthermore, health effects of airborne particles are related not only to the
171 amount and toxicity of the particulate material, but also to the particle size because
172 this property has a large effect on whether or not a particle is able to penetrate into the
173 lungs [e.g. 20] and even further into the blood circulation [e.g. 24].

174

175 The rapid, large-scale urbanization and industrialization of China are unique in
176 history. Consequently, China's air pollution situation has worsened dramatically
177 during the last 2–3 decades as emissions from industry, energy production and traffic
178 have increased. China is currently responsible for 30–35 % of the global SO_2 , NO_x ,
179 CO and Particulate mass (PM) emissions and 40% of global particle number (PN)
180 emission in the 20–1000 nm size range (see <http://gains.iiasa.ac.at/gains3/>).

181 Atmospheric concentrations of primary and secondary pollutants in China are 10 to
182 100 times (sometimes even 1000 times) higher than currently in Europe or Northern
183 America. However, highly non-linear processes, such as atmospheric chemistry and
184 aerosol dynamics, transform the urban pollution cocktail and generate secondary
185 pollution, such as ultrafine particles and ozone, during their residence in the
186 atmosphere [25, 26]. The fact that new particle formation does occur in polluted
187 Chinese megacities like Beijing [27] and Shanghai [28], or even during dust-storms
188 [29, 30], suggests that there are several major physical and chemical mechanisms in a
189 heavily-polluted atmosphere that have not been recognized before and may not even
190 be operating in clean or moderately-polluted environments. At present, atmospheric
191 air pollution in China threatens the health of hundreds of millions of people [e.g. 3,
192 31], and causes major problems to the environment and economy as a whole by
193 decreasing, e.g. severely the agricultural and industrial productivity of the nation as a
194 whole. This pollution also reduces visibility, thereby decreasing the attraction of these
195 mega-cities for tourists, and hinders the possibilities to use solar energy a source for a
196 clean energy on a local scale.

197
198 A holistic scientific understanding on the atmospheric phenomena associated with air
199 quality as a whole, as well as on the connection between air quality and climate, is
200 lacking at the moment [31-33]. Together with emission reductions, the key way to get
201 forward is to perform long-term, continuous and comprehensive observations on
202 aerosol particles (mass, number, chemical composition, optical properties), on
203 concentrations of trace gases (SO₂, NO_x, CO, VOCs, sulphuric acid, HONO, HNO₃,
204 NH₃ etc.), and on atmospheric oxidant levels (O₃, HO_x, RO_x, NO₃, Criegee

intermediates etc.), as well as on greenhouse gas concentrations [31]. With a network of such observation stations [34], we will be able to understand the interactions and feedbacks associated with the urban pollution mixture [e.g. 35-37], and ultimately, be ready to make targeted strategies for the pollution control. In the following we take recent advances in studying secondary new aerosol formation in China as an example to show how increased process-level understanding will help us to understand air quality-climate-weather interactions and how the feedbacks and interactions affect the air quality in highly-polluted environments such as those frequently encountered in Chinese megacities.

2. Results from recent studies on New Particle Formation in China

New particle formation events have been observed in many different locations in China, including coastal/marine, rural, regional and polluted urban environments [28, 30, 38-47].

The first long-term study on NPF events was performed in the urban of Beijing at PKU Urban Atmosphere Environment Monitoring Station (PKUERS), starting at 2004 [27, 48, 49]. On average, every fifth day (~21%) displayed a NPF event [50]. An evident seasonal variation profile for NPF events was observed, showing that a high frequency the NPF events (~ 40%) occurred during the spring and winter [27, 50], while fewer events were observed in summer [51, 52]. The observed formation rates

227 of 3-nm particles and their growth rates were in the ranges of $3.3\text{-}81.4\text{ cm}^{-3}\text{ s}^{-1}$ and
228 $0.1\text{-}11.2\text{ nm h}^{-1}$ [27, 50, 53], respectively.

229
230 Generally, NPF is an unexpected phenomenon in the polluted atmosphere of China
231 due to typically high loadings of pre-existing aerosol particles. For example, the mean
232 condensation sink (CS, [54]) values during the nucleation event days were 0.025 s^{-1}
233 ($0.003\text{-}0.086\text{ s}^{-1}$) and 0.026 s^{-1} ($0.004\text{-}0.082\text{ s}^{-1}$) at the rural (Kaiping) and urban
234 (Beijing) environments, respectively, which are approximately 5 to 10 times higher
235 than the values of CS observed in clean environments [55-57]. This high
236 concentration of pre-existing aerosol particles significantly inhibits the growth of
237 newly-formed particles. In fact, the observed NPF event is an end product of the
238 competition between the low-volatile vapor sources (such as SO_2 or sulfuric acid) and
239 sinks (such as pre-existing particles), as shown by Kulmala et al. [55]. The abundant
240 SO_2 emissions and high oxidation capacity in the polluted atmosphere of China
241 indicate that there is a sufficient source of sulfuric acid [40, 52]. Therefore, in the case
242 of both higher source and sink, their inter-competition is the most likely factor that
243 determines the occurrences of NPF events in polluted environments.

244
245 Two years (2011-2013) of continuous particle number size distribution measurements
246 were conducted at the Station for Observing Regional Processes of the Earth System
247 (SORPES [35, 36]) station about 20 km northeast of urban Nanjing. The location can
248 be considered as a regional background site of Yangtze River Delta in eastern China.
249 During this time period, 44% of the sampling days were NPF event days (see Figure 1

as an example). The formation rates of 6-nm particles varied from 0.24 to 10.9 cm⁻³ s⁻¹, the subsequent particle growth rates varied from 3.6 to 23 nm h⁻¹, and the values of CS during the event days varied from 0.007 to 0.068 s⁻¹ [47]. Most of the NPF events took place in spring, summer and autumn with the frequencies of 55, 54 and 49 %, respectively, whereas only 15 events (11.2%) were observed in winter.

Figure 1. A typical nucleation event measured using Air Ion Spectrometer (AIS) at the SORPES station, Nanjing, in China. The background cluster ions are seen in both negative and positive ion modes in the sub-2 nm size range. Negative ion clusters are smaller than positive ones. The new particle formation is seen in both polarities starting at around 8.30 am. Here J_6 is 1.8 cm⁻³ s⁻¹ and GR (6-30 nm) is 6.6 nm h⁻¹.

The typical NPF event in Nanjing is shown in Figure 1. In Nanjing, many of the NPF events occurred on the days associated with heavy pollution. As shown by Xie et al. [30], frequent NPF events were observed when the PM_{2.5} and PM₁₀ concentrations were in excess of 100 µg m⁻³ and 200 µg m⁻³, respectively. The reason for this is still an open question. One hypothesis is that nucleation can be promoted by heterogeneous reactions on the surface of the dust [29, 30]. This is supported by many observations from both SORPES station and another mountain top site, Mt. Heng in southern China. In the spring of 2009, relatively high new-particle formation rates (0.46 cm⁻³ s⁻¹) and growth rates (7.2 nm h⁻¹) were observed when the loading of pre-exist particles was higher than 600 µg m⁻³ at Mt. Heng. Combined with laboratory

investigations [58], dust-induced heterogeneous photochemical processes were supposed to provide additional gaseous oxidants to promote the NPF [29].

In urban Shanghai, particle size distributions were measured from November 2013 to January 2014 on the rooftop of a teaching building (31°18'N, 121°30'E) on the campus of Fudan University [28], which can be regarded as an urban site. During this 62-day campaign, 13 NPF events were identified with strong bursts of sub-3 nm particles and subsequent fast growth of these particles. The observed nucleation rate ($J_{1.34}$), formation rate of 3 nm particles (J_3), and CS were in the ranges of 112.4-271.0 cm⁻³ s⁻¹, 2.3-19.2 cm⁻³ s⁻¹ and 0.030-0.10 s⁻¹, respectively. The growth rates of the formed clusters and nanoparticle showed a clear size dependence, with average values of GR_{1.35~1.39}, GR_{1.39~1.46}, GR_{1.46~1.70}, GR_{1.70~2.39}, GR_{2.39~7} and GR_{7~20} being 1.6±1.0, 1.4±2.2, 7.2±7.1, 9.0±11.4, 10.9±9.8 and 11.4±9.7 nm h⁻¹, respectively. Nucleation of particles during this campaign might be explained by the activation theory, since the formation rate of the smallest particles was proportional to a 0.65±0.28 power of the sulfuric acid proxy. In addition, ammonia was very likely associated with NPF events, as the new particle formation rate was positively correlated with the concentration of gas-phase ammonia. The estimated sulfuric acid concentration was sufficient to explain the growth of 1.34–3 nm particles, but its contribution became smaller as the particle grew in size.

293 The observed new particle rates, condensation sink and particle growth rates in the
294 three megacities, i.e. Beijing, Nanjing and Shanghai, are of the same order of
295 magnitude. These similarities reflect the urban nature of the Beijing and Shanghai
296 sites, and hint that the Nanjing site, although considered as a regional background site
297 of Yangtze River Delta in eastern China, might be characterized with a similar
298 competition between the sources and sinks of low-volatility vapors. The seasonal
299 pattern of the NPF frequency is very different between the two sites having long-term
300 measurements, Beijing and Nanjing, in addition which the annual-averaged NPF
301 frequency is clearly higher in Nanjing. The fundamental reason for these differences
302 lies probably in a delicate balance between the factors that favor or suppress new
303 particle formation and growth. At both Beijing and Nanjing, for example, NPF is
304 favored by a low ambient relative humidity and low CS, whereas no consistent pattern
305 can be seen between the occurrence of NPF and either the ambient temperature or
306 sulfur dioxide concentration [27, 47]. The fact that high values of CS tend to suppress
307 NPF is fully in line with theoretical expectations [16, 17], and it might explain the low
308 NPF frequency observed in Shanghai during polluted winter conditions [28]. There is
309 strong, yet indirect evidence that NPF events in these three megacities are connected
310 to sulfuric acid [28, 29, 40, 52]. However, it is premature to conclude that the exact
311 nucleation mechanisms are identical in three megacities without direct measurements
312 of chemical composition of nucleating clusters and ions.

313

314 Besides the direct connection between pre-exist aerosols (e.g. mixed dust) and NPF, a
315 recent study found that biomass burning particles can enhance the conversion rate of

NO₂ to HONO which is one of the main sources of OH and can in turn promote the formation of secondary aerosol mass and number [59]. Furthermore, it was found that when biomass burning particles are mixed with anthropogenic pollution, the HONO production potential from the conversion of NO₂ to HONO tend to be enhanced even more. Given that biomass burning particles are easily mixed with anthropogenic pollution in eastern China, their influences on the HONO budget, radical pool, and thus the formation of secondary aerosols are expected to be important [59].

323

Heterogeneous, or multi-phase, processes influence the secondary aerosol formation. For example, most of aerosol sulfate has been believed to be formed from heterogeneous or aqueous-phase processes (cloud processes). Ozone and hydrogen peroxide are the major oxidants to drive these processes. Recent studies have shown that NO₂ can also be an important oxidant to convert SO₂ to sulfate when mineral dust and biomass burning plumes are present [30, 60]. Especially during the biomass burning-induced haze events [30], the oxidation processes by NO₂ became critical when the formation of other oxidants were suppressed. More interestingly, one of the “by-products” of the reaction of SO₂ and NO₂ is HONO, which can further enhance the atmospheric oxidation capacity. All these observations suggest that our current understanding on secondary aerosol formation processes need to be revised.

335

336 **3. On future NPF studies**

337

338 The importance of secondary aerosols has become apparent during the last decades,
339 so there is an increasing need for understanding their formation mechanisms and
340 atmospheric dynamics in detail. Although several field campaigns and a few long-
341 term (over several years) observations on NPF have already been conducted in China,
342 we need to perform additional long-term measurements, preferable continuous and
343 comprehensive observations utilizing the full capacity of the current state-of-the-art
344 instruments.

345

346 In the coming decade, we need to utilize the full capacity of new aerosol and ion
347 instruments, such as the Particle Size Magnifier (PSM, [61]), Neutral cluster and Air
348 Ion Spectrometer (NAIS, [62]) and Sigma [63]. With these instruments, we will be
349 able to detect and analyze the frequency of NPF events, as well as to determine cluster
350 concentrations, particle formation rates and size-dependent particle growth rates [e.g.
351 64, 65]. Furthermore, we will be able to quantify the contribution of ion and neutral
352 pathways to NPF [66, 67].

353

354 The aerosol and ion instruments together with the high-resolution mass spectrometers,
355 such as Atmospheric Pressure interface – Time of Flight mass spectrometer (APiTOF,
356 [68] and Chemical Ionization APiTOF [69], will make it possible to connect the NPF
357 to the concentrations of different vapors participating in this process. Such vapors
358 include sulfuric acid [9, 70, 71], ammonia [72], amines [12, 73] and organic vapors
359 [11, 13, 74]. Furthermore, the mass spectrometers can also be utilized in determining

360 atmospheric radical concentrations [75, 76, 77] responsible for the oxidation of
361 precursor vapors in the atmosphere.

362

363 To support the NPF analysis, aerosol number size distributions need to be measured
364 with harmonized instruments [78], enabling quantification of the condensation sink of
365 a pre-existing particle population. On-line chemical analysis is important as well,
366 since such information can be used to attributing the relative contributions of different
367 aerosol sources [e.g. 26, 79].

368

369 In order to have reliable data which can also be compared from one site to another,
370 instruments need to be calibrated often enough in the laboratory. This should be
371 conducted within specific calibration centers. In order to assure the data quality, open
372 data flows and joint data analysis are preferable, which will lead to joint publications
373 and provides novel avenues to exploit the data to improve both regional air quality
374 and global climate.

375

376 **4. Capacity building**

377

378 Capacity building related to scientists, engineers and technicians operating
379 instruments and stations are necessary pre-requisites for obtaining good data. For
380 example, a proper use of instruments will optimize the efforts, improve the data
381 quality and enhance data and publication flows.

382

383 The new insights gained on the secondary aerosol formation and atmospheric
384 phenomena associated with air quality as a whole need to be disseminated from the
385 academia to the public and to the private sector. The academic experts need to keep
386 their knowledge and skills up-to-date and widen their knowledge base with horizontal
387 learning of the adjunct fields in science and technology. Atmospheric research
388 involves several fields of science, such as chemistry, physics, meteorology and Earth
389 system sciences, so deepening and widening the expertise is required. The horizontal
390 learning principle has been shown to be a good example of collaborative problem
391 solving and participatory action research [80]. The shift from discipline-tied
392 fundamental education towards a multi-disciplinarity is imperative for a successful
393 career in climate and global change science [81].

394

395 In capacity building, we actually need to answer several questions: What are the target
396 groups? What knowledge needs to be transferred and what are the skills that each
397 target group needs? Concerning the comprehensive atmosphere earth system
398 measurements: Which kind of observation infrastructures is best to improve the air
399 quality in China? Concerning the effective knowledge transfer and innovative
400 thinking methods: What kind of knowledge transfer is needed for sustainable air
401 quality solutions?

402

403 Solution-oriented thinking, need for updating skills as well as knowledge of rapidly
404 changing air quality situation, are crucial. Reliable, research-based education that has

405 a holistic view on the whole big picture of causes and effects and their interactions
406 and feedbacks affecting air quality will support long lasting solutions. Also basic
407 understanding of the processes behind atmospheric phenomena is needed for building
408 a foundation for evaluating new information. Learning lasts a lifetime, which actually
409 is underlined by the fact that the university professors have pointed out that they
410 deepen their knowledge when lecturing to students.

412 **5. Future Outlook**

414 Atmospheric new particle formation contributes significantly to local, regional and
415 global aerosol number and CCN loads [e.g. 6]. Therefore, understanding of this
416 phenomenon is central to solving the secondary air pollution problem as a whole. The
417 following steps are needed in this process:

- 418 1) to perform long-term continuous, comprehensive observations on aerosol
419 precursors, oxidants, clusters, ions and aerosol particles together with proper
420 metadata and meteorological data. If needed, new Station for Measuring
421 Ecosystem – Atmosphere Relations II (SMEAR II, [82]) -type flagship stations
422 should be established, since they will help understanding the connections between
423 NPF and land surface – atmosphere interactions and feedbacks,
- 424 2) to establish calibration centers for mass spectrometers, PSMs and ion
425 spectrometers,
- 426 3) to organize joint data workshops for analyzing atmospheric data in proper,
427 comprehensive manner,

- 428 4) to ensure open data and metadata fluxes to other users, and
429 5) to organize joint paper writing workshops and publish the joint papers in peer-
430 reviewed journals.

431 It would be a big step forward to establish tight connections between different
432 Chinese research groups and support further deep collaborations in the future. The
433 second challenge is to establish open data policy and knowledge transfer at all levels.
434 The access to data is crucial to be able to answer research questions and to solve air
435 pollution problem(s). As a good sign, during the last years we have already seen
436 improvements regarding these issues. The third point is the capacity building,
437 including new infrastructures, data flows, databases etc. Furthermore, a new
438 generation of scientists needs to be educated to improve the knowledge base and
439 optimal use of infrastructures and data [86].

440

441 Understanding the formation of secondary pollutants is extremely important, since it
442 enables deep understanding of air pollutant dynamics crucial to air quality.
443 Improving air quality in China has several co-benefits, as it will lead to reduced
444 greenhouse-gas and black carbon emissions and concentrations, together with
445 improved fresh water quality and food supply. The cleaner air will decrease adverse
446 health effects caused by pollutants significantly [83, 84]. Efforts to prevent adverse
447 health effects must be well planned and should occur on multiple levels and places
448 simultaneously. Successful efforts will lead to significant gains in population health,
449 personal well-being and environmental quality as well as improving economy in
450 personal, local and national levels together with other significant co-benefits [85].

451 Reducing the use of fossil fuels does not only reduce emissions of air pollutants, but
452 also CO₂ and black carbon (BC), thereby decreasing radiative forcing in national and
453 global scales. Also, agricultural production and ecosystem services will benefit from
454 lowered pollutant levels. Healthier food will further improve peoples' health, and less
455 pollution damage improves yields of vegetables and crops. Better insulation of
456 buildings will lower the need for indoor heating, thus reducing emissions, but can also
457 reduce outdoor-indoor penetration of air pollutants. New technology in industry,
458 traffic and energy production will decrease emissions. The reduced pollution will
459 increase solar radiation in ground level and increase potential for solar energy.

461 Thus, tackling the air quality rapidly can lead to significant improvement on the
462 quality of life of the population as a whole and can lead to a positive feedback cycle,
463 which will encourage further progress towards cleaner environment.

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Research highlights

- 1) Formation of new atmospheric aerosol particles is a global phenomenon that has been observed to take place in even heavily-polluted environments. A holistic scientific understanding on the atmospheric phenomena associated with air quality as a whole, as well as on the connection between air quality and climate, is lacking at the moment.
- 2) In China, new particle production has been observed at very high pollution levels (condensation sink about 0.1 s^{-1}) in several megacities. With a network of observation stations, we will be able to understand the interactions and feedbacks associated with the urban pollution mixture, and ultimately, be ready to make targeted strategies for the pollution control.
- 3) This paper summaries the recent advances in studying secondary new aerosol formation in China to show how increased process-level understanding will help us to understand air quality-climate-weather interactions and how the feedbacks and interactions affect the air quality in highly-polluted environments such as those frequently encountered in Chinese megacities.

Figure 1. A typical nucleation event measured using Air Ion Spectrometer (AIS) at the SORPES station, Nanjing, in China. The background cluster ions are seen in both negative and positive ion modes in the sub-2 nm size range. Negative ion clusters are smaller than positive ones. The new particle formation is seen in both polarities starting at around 8.30 am. Here J_6 is $1.8 \text{ cm}^{-3}\text{s}^{-1}$ and $\text{GR}(6\text{-}30 \text{ nm})$ is 6.6 nm/hr .
871x523mm (72 x 72 DPI)