On secondary new particle formation in China

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2016-10

Kulmala , M , Petaja , T , Kerminen , V-M , Kujansuu , J , Ruuskanen , T , Ding , A , Nie , W , Hu , M , Wang , Z , Wu , Z , Wang , L & Worsnop , D R 2016 , ' On secondary new particle formation in China ' , Frontiers of Environmental Science & Engineering in China , vol. 10 , no. 5 , 08 . https://doi.org/10.1007/s11783-016-0850-1

http://hdl.handle.net/10138/224159
https://doi.org/10.1007/s11783-016-0850-1

other
acceptedVersion

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Journal:	*Frontiers of Environmental Science and Engineering*
Manuscript ID	FESE-2016-0074.R1
Manuscript Type:	Special Issue: Progresses in understanding secondary air pollution
Date Submitted by the Author:	n/a
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Keywords:	aerosol particles, heavily-polluted environments, condensation sink, new particle production, megacities
Speciality:	Formation < POLLUTION, Particle-phase species < POLLUTION, Mass spectrometry < ANALYTICAL METHODS

The original question from guest editor: your view or comments or directions related to secondary aerosol formation in China

Journal: Front. Environ. Sci. Eng

Title:

On secondary new particle formation in China

Running title:

On secondary new particle formation in China

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Abstract

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

1. Background

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

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

Secondary formation of new atmospheric aerosol particles is typically initiated by photochemical reactions in the gas phase, so that especially the production of extremely low volatility vapors like sulfuric acid [9, 10, 11] and highly-oxidized organic compounds [e.g. 12, 13, 14] is crucial. Pre-existing aerosol particles act as a sink for the low-volatile vapors, as well as for small clusters and growing nanoparticles, thereby hindering or even suppressing atmospheric new particle formation [e.g. 15, 16, 17]. The atmospheric new particle formation is affected by several meteorological quantities and phenomena, particularly in the planetary boundary layer, including the intensity of solar radiation and atmospheric mixing processes. The recent findings indicate that critical clusters may be surprisingly small in size, if existing at all, under atmospheric conditions [e.g. 18], and thus treatable by advanced quantum chemistry methods [19]. It is very probable that the atmospheric new particle formation is a two-step process, i.e. initial clustering and then condensational growth after activation of clusters, as suggested by Kulmala et al. [21] and verified by Kulmala et al. [18]. A summary of the current understanding of gas-to-particle conversion is presented by Kulmala et al. [5].

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

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

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

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 [31-33]. Together with emission reductions, the key way to get forward is to perform long-term, continuous and comprehensive observations on aerosol particles (mass, number, chemical composition, optical properties), on concentrations of trace gases (SO_2, NO_x, CO, VOCs, sulphuric acid, HONO, HNO_3, NH_3 etc.), and on atmospheric oxidant levels (O_3, HO_x, RO_x, NO_3, 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 of 3-nm particles and their growth rates were in the ranges of 3.3-81.4 cm^{-3} s^{-1} and 0.1-11.2 nm h^{-1} [27, 50, 53], respectively.

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

Two years (2011-2013) of continuous particle number size distribution measurements were conducted at the Station for Observing Regional Processes of the Earth System (SORPES [35, 36]) station about 20 km northeast of urban Nanjing. The location can be considered as a regional background site of Yangtze River Delta in eastern China. 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 PM_{10} 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 rages of the formed clusters and nanoparticle showed a clear size dependence, with average values of $\mathrm{GR}_{1.35\sim1.39}$, $\mathrm{GR}_{1.39\sim1.46}$, $\mathrm{GR}_{1.46\sim1.70}$, $\mathrm{GR}_{1.70\sim2.39}$, $\mathrm{GR}_{2.39\sim7}$ and $\mathrm{GR}_{7\sim20}$ 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.

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

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

NO_2 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 NO_2 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].

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 NO_2 can also be an important oxidant to convert SO_2 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 NO_2 became critical when the formation of other oxidants were suppressed. More interestingly, one of the “by-products” of the reaction of SO_2 and NO_2 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.

3. On future NPF studies

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

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

The aerosol and ion instruments together with the high-resolution mass spectrometers, such as Atmosperic Pressure interface – Time of Flight mass spectrometer (APiTOF, [68] and Chemical Ionization APiTOF [69], will make it possible to connect the NPF to the concentrations of different vapors participating in this process. Such vapors include sulfuric acid [9, 70, 71], ammonia [72], amines [12, 73] and organic vapors [11, 13, 74]. Furthermore, the mass spectrometers can also be utilized in determining atmospheric radical concentrations [75, 76, 77] responsible for the oxidation of precursor vapors in the atmosphere.

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

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

4. Capacity building

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

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

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

Solution-oriented thinking, need for updating skills as well as knowledge of rapidly changing air quality situation, are crucial. Reliable, research-based education that has a holistic view on the whole big picture of causes and effects and their interactions and feedbacks affecting air quality will support long lasting solutions. Also basic understanding of the processes behind atmospheric phenomena is needed for building a foundation for evaluating new information. Learning lasts a lifetime, which actually is underlined by the fact that the university professors have pointed out that they deepen their knowledge when lecturing to students.

5. Future Outlook

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

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

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

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

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

Reducing the use of fossil fuels does not only reduce emissions of air pollutants, but also CO_2 and black carbon (BC), thereby decreasing radiative forcing in national and global scales. Also, agricultural production and ecosystem services will benefit from lowered pollutant levels. Healthier food will further improve peoples' health, and less pollution damage improves yields of vegetables and crops. Better insulation of buildings will lower the need for indoor heating, thus reducing emissions, but can also reduce outdoor-indoor penetration of air pollutants. New technology in industry, traffic and energy production will decrease emissions. The reduced pollution will increase solar radiation in ground level and increase potential for solar energy.

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

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Acknowledgements

The work in this manuscript is supported by Academy of Finland via Center of Excellence in Atmospheric Sciences (project no. 272041) and the Finnish Funding Agency for Technology and Innovation TEKES via Beautiful Beijing project (3667/31/2013) and European Research Council Advanced Grant (ATMNUCLE, 227463) and InGOS DEFROST and CRAICC (no 26060) and Nordforsk CRAICC-PEEX (amendment to contact 26060) funded by Nordforsk. The SORPES station was supported by Nanjing University and the Collaborative Innovation Center of Climate Change in Jiangsu Province, China. Part of Aijun Ding's work was supported by the excellent young scientist fund of National Natural Science Foundation of China (D0512/ 41422504).

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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.

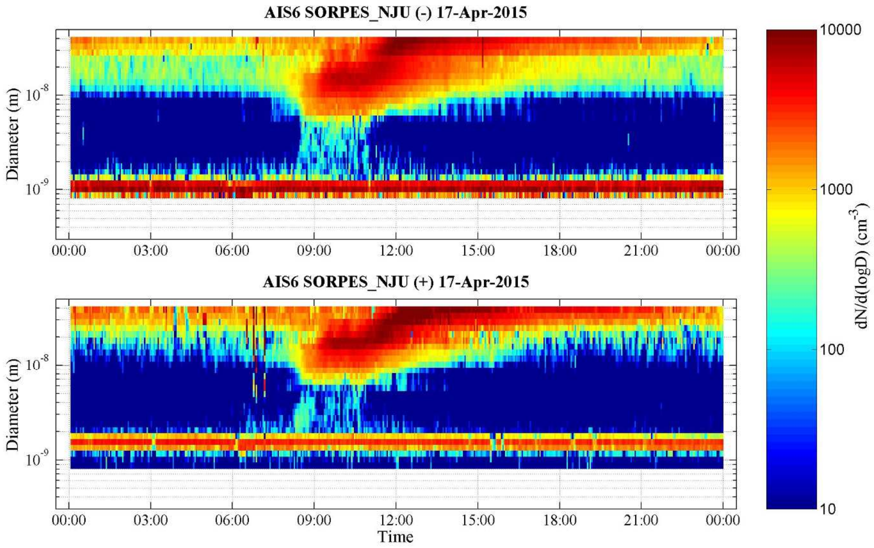

Figure 1. A typical nucleation event measured using Air Ion Spectrometer (AIS) at the SORPES station, Nanjing, in China. The background cluster ions are seen in both negative and positive ion modes in the sub-2 nm size range. Negative ion clusters are smaller than positive ones. The new particle formation is seen in both polarities starting at around 8.30 am. Here J6 is 1.8 cm-3s-1 and GR(6-30 nm) is 6.6nm/hr.

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