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New particle formation under the influence of the long-range transport of air pollutants in East Asia

Indra Chandra [a], Seyoung Kim [a], Takafumi Seto [a, *], Yoshio Otani [a], Akinori Takami [b], Ayako Yoshino [b], Satoshi Irei [b], Kihong Park [c], Tamio Takamura [d], Naoki Kaneyasu [e], Shiro Hatakeyama [f]

[a] Department of Chemical and Material Engineering, Kanazawa University, Kanazawa, 920-1192, Japan
[b] Center for Regional Environmental Research, National Institute for Environmental Studies, 16-2 Onogawa, Tsukuba, 305-8506, Japan
[c] School of Environmental Science and Engineering, Gwangju Institute of Science and Technology (GIST), 261 Cheomdan-Gwagiro, Buk-gu, Gwangju, 500-712, Republic of Korea
[d] Center for Environmental Remote Sensing (CEReS), Chiba University, Chiba, 263-8522, Japan
[e] Global Environment Study Group, National Institute of Advanced Industrial Science and Technology, 16-1 Onogawa, Tsukuba, 305-8568, Japan
[f] Institute of Agriculture, Tokyo University of Agriculture and Technology, 3-5-8 Saiwaicho, Fuchu, Tokyo, 183-8509, Japan

HIGHLIGHTS

- New particle formation (NPF) events were observed at a rural island in East Asia.
- Two types of NPF were identified with different onset diameters of the growth.
- Correlation between the types of NPF and other air pollutants were investigated.
- NPF in East Asia is influenced by the long-range transported air pollutants.

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ABSTRACT

Field observations to investigate the correlation between New Particle Formation (NPF) and the long-range transport of air pollutants in the East Asia region were carried out on a rural Island of Japan in the East-China Sea (Fukue Island, 32.8°N, 128.7°E) over three periods (February 23 to March 7, 2013; November 7 to 20, 2013; and November 2 to 24, 2014). Frequent NPF events were identified (16 events in 50 days), typically in association with sudden increases in particle number concentrations and the successive growth of particles to mobility diameters of several tens of nanometers. The NPF events were classified into two types (A and B) according to the initially detected particle sizes (onset diameters). Type-A consisted of strong NPF events with onset diameters as small as 5 nm. Type-B consisted of NPF events whose onset (<10 nm) was not clearly identifiable. The correlations of SO_2 concentrations, solar radiation, $PM_{2.5}$ concentrations, and chemical composition were analyzed based on the types of NPF events.

1. Introduction

The trans-boundary transport of air pollutants in the East Asian region has become a serious concern in recent years. Air pollutants emitted from urban and industrial areas in China, Korea, and Japan travel over long distances are thought to bring negative impacts on plants and humans in downstream regions. Regional aerosol size distributions are influenced directly by primary particles transported over long distances, such as yellow sand and black carbon. In addition, gaseous pollutants transported over long distances indirectly affect the aerosol size distribution through a process of secondary particle formation. Gaseous pollutants such as SO_2 and volatile organic compounds, for instance, are photo-oxidized by solar radiation while they travel, and are converted to semi-volatile condensable vapors in downstream regions. When sufficient amounts of condensation sink are present, these vapors may condense into pre-existing particles. When aerosol concentrations in the background are low, these vapors freshly form

* Corresponding author.

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nanometer-sized particles through homogeneous nucleation, that is, the process known as new particle formation (NPF). For this reason, the particle size distribution in the boundary region of the East-Asian outflow exhibits a complex behavior that depends on both meteorological conditions and the level of air pollution.

There have been a large number of reports on NPF from around the world in the last decade (Kulmala et al., 2004). Few of them, however, provide data on the nanoparticle size distribution in the East Asia region. Weber et al. (2003) observed NPF events in East-Asian anthropogenic plumes in a series of TRACE-P aircraft experiments over East Asia. They measured high concentrations of 3–4 nm particles associated with pollution plumes using an ultrafine condensation particle counter equipped with a pulse height analysis function (PHA-UCPC). Lee et al. (2008) reported time-varying data on the mobility size distribution (10–487 nm) in the coastal region of Korea using a scanning mobility particle sizer (SMPS). They identified four classes of NPF and growth events and demonstrated that the nucleation events might have mainly originated from the air masses of the Asian continent. In an investigation of the cloud condensation nuclei (CCN) activity of submicron aerosol of the Gosan site (Jeju Island, Korea), Yum et al. (2007) reported that the CCN mostly behaved like ammonium sulfate. They also observed regional-scale NPF and growth events in the coastal region of Korea associated with air masses from northern China, Mongolia, or Russia. Shen et al. (2011) reported a long-term (1.5 years) observation of particle number concentrations at a rural site in the North China Plain using a TDMPS and an aerodynamic particle sizer (APS). They reported that clean air masses from inner Asia enhanced NPF while air masses from urban and industrial regions in China increased background aerosol concentrations. Recent studies report simultaneous observations in rural and urban sites (Jung et al., 2013; Wang et al., 2013; Yue et al., 2013), but few studies have observed events on a regional scale. As such, two important aspects of NPF and growth events in the East Asia region are still incompletely understood: (i) the distribution and chemical species of the initial nucleating species in the sub-10 nm range and (ii) the correlation between NPF and the long-range transport of air pollutants.

Our group conducted a field study to investigate NPF and the influence of air pollution transported over large distances in the East Asia region as part of the ASEPH (Impacts of Aerosols in East Asia on Plants and Human Health) project. We selected Fukue Island (32.8°N, 128.7°E), a rural island located in the outflow region of the East-Asian plume, as a supersite for field observation. In a previous study we collected time-resolved data on the mobility size distribution and number concentration at this supersite from March 9 to 16, 2012 (Seto et al., 2013). We identified several NPF and growth events associated with the large scale (>65 μg m^{-3} of $PM_{2.5}$ concentration), long-range transport of polluted air masses from the East Asian region. Our instrumentation, however, lacked the sensitivity to detect the initial stages of the NPF (the instrumentation measured a mobility size range of 14–670 nm). In the present study we measured the time-resolved mobility size distribution using nano-mobility particle sizers ranging from 2.5 to 64 nm. We analyzed how the concentration of the possible precursor (SO_2 gas) and chemical composition of the pre-existing particles influenced the condition of the NPF, based on data taken over a 50-day observation period.

2. Field observation: overview and methods

2.1. Monitoring site

The observation was conducted at the Fukue Island Supersite (32.8°N, 128.7°E), Nagasaki, Japan, for a total of 50 days divided between three periods: February 23 to March 7, 2013; November 7 to 20, 2013; and November 2 to 24, 2014. Detailed descriptions of the Fukue site and the equipment for meteorological and atmospheric observation have been described earlier (Takami et al., 2005; Seto et al., 2013). The island is located in the outflow region of the East-Asian plume, particularly in the winter-to-spring season. The supersite is situated in a rural area of the island where anthropogenic emissions such as automobile and factory exhaust gas have no significant influence on the local environment. In our previous study (Seto et al., 2013) we observed frequent NPF events at this site but were unable to identify the initial stage of the particle formation (mobility diameter of <10 nm). This observation campaign was organized by the ASEPH project, and various meteorological data were available from the observation network for aerosol-cloud-radiation interaction (SKYNET) and measurements of the concentrations and chemical composition of atmospheric aerosols and gases were available from the observation system of National Institute for Environmental Studies (NIES).

2.2. Instruments

Mobility size distributions, meteorological parameters, $PM_{2.5}$ mass concentrations, aerosol chemical components, and SO_2 gas concentrations were measured concurrently. Mobility size distributions between 2.5 nm and 64 nm were measured using a Nano-Scanning Mobility Particle Sizer (nano-SMPS) system composed of a nano-differential mobility analyzer (nano-DMA) (model 3085, TSI, Inc.) and an ultrafine condensation particle counter (UCPC) (model 3776, TSI, Inc.). In addition, a long-SMPS system composed of a long-DMA (model 3081, TSI, Inc.) and condensation particle counter (CPC, model 3775, TSI, Inc.) was used to determine the particle size distributions from 14 to 670 nm in the observations in November 2013 and 2014.

Meteorological data on solar radiation (*I*), temperature (*T*), and relative humidity (*Rh*) were measured during the observation periods using a measuring system provided by Chiba University, Japan. The $PM_{2.5}$ mass concentration was measured by a Tapered Element Oscillating Microbalance (TEOM) for 60-min signal averaging. The chemical composition of particles, such as chloride, nitrate, sulfate, organic, and ammonium, were measured by an Aerodyne Aerosol Chemical Speciation Monitor (ACSM) with a time resolution of 15-min (Ng et al., 2011). Aerodynamic lens systems with transmission window in the rage of aerodynamic diameter from 40 nm to 1 μm was equipped to the inlet of the ACSM. The SO_2 concentration was measured by an SO_2 analyzer (model 43i, Thermo Scientific, Inc.) with a time resolution of 1-min.

A Nano Sampler (model 3180, Kanomax Japan, Inc.) was employed to analyze the size-dependent chemical composition (water-soluble ions) of the aerosol particles. The Nano Sampler was equipped with four impactor stages for PM_{10}, $PM_{2.5}$, PM_1, and $PM_{0.5}$. Quartz fiber filters (φ55 mm, 2500 QAT-UP, Pallflex Products Corp.) were placed on the each impactor stage to prevent rebound of the particles collected. An inertial filter (stainless steel fiber) was also attached after the $PM_{2.5}$ stage to collect the particles larger than 100 nm via an inertial filtration mechanism (Otani et al., 2007). The particles penetrating the inertial filter (<100 nm in aerodynamic diameter) were collected in a backup quartz filter. The aerosol samplings were typically conducted for 7–16 h at a flow rate of 40 L min^{-1}. The samples were prepared in both daytime (typically from 10:00 to 17:00) and nighttime (from 17:30 to 9:30), except on rainy days. Once the size-segregated aerosol samples on the quartz filter were prepared, the entire samples were carried to the laboratory and extracted using ultrapure water by ultra-sonication. The water-soluble cation (NH_4^+) and anion (SO_4^{2-}, NO_3^-, and Cl^-) were analyzed with an Ion Chromatograph (IC, model DX-120, Thermo