Aerial Observation of Atmospheric Nanoparticles on Fukue Island, Japan

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ABSTRACT

New particle formation (NPF) in the East Asian region is strongly influenced by photochemical processes during the long-range transport of air pollutants. Our previous measurements (Seto *et al.*, 2013; Chandra *et al.*, 2016) relating to the incomplete and weak NPF (onset diameter > 10 nm) on Fukue Island in Japan (downstream from the Asian continent) suggested nucleation in the upstream region. The vertical structure of atmospheric nanoparticle concentrations (particle size > 6 nm and altitude < 1.2 km) during NPF was observed using a Kite-Plane at Fukue Island. Three different event types were identified through aerial observations (different vertical profiles of nanoparticles), ground-based measurements (the initial detected diameter of nanoparticles, NPF starting time, and dominant chemical component in PM₁), and air mass backward trajectory analysis. A stronger NPF event (Event I: > 35,000 particles cm⁻³) than in our previous measurements (from 2012 to 2016, ~20,000 particles cm⁻³) with a particle size as small as 5 nm, affected by the long-range transport of air pollutants under a high-pressure system, was detected. A sudden increase in particle number and SO₂ concentrations with weak NPF caused by a change in the air mass origin (Event II: < 10,000 particles cm⁻³) as well as no NPF with aged sulfate particles during conditions with low particle number and SO₂ concentrations (< 0.15 ppb) (Event III: < 2,000 particles cm⁻³) were also observed.

Keywords: New particle formation; Vertical structure; Trans-boundary transport; East Asia.

INTRODUCTION

Secondary aerosol (SA) in East Asia exhibits complex features of condensation, evaporation, and chemical reactions due to the existence of large condensation and coagulation sinks (Kulmala *et al.*, 2001; Dal Maso *et al.*, 2002; Peng *et al.*, 2017), high SO₂ concentration (Zhang *et al.*, 2009; Lu *et al.*, 2010), large emission sources (Mönkkönen *et al.*, 2005; Zhang *et al.*, 2006) and long-range and trans-boundary transport (affecting other country located near the country of origin, e.g., air pollution) on westerly winds (Koike *et al.*, 2003; Weber *et al.*, 2003; Matsui *et al.*, 2011). SA has different impacts on local air quality, human health and global radiation balance depending on chemical composition and particle size distribution (Penner *et al.*, 2004; Davidson

et al., 2005; Yu *et al.*, 2010). New particle formation (NPF) is a key process that triggers SA formation via gas-to-particle conversion. NPF is associated with significant increase in the number concentration of nanoparticles (i.e., nucleation burst), and has been identified under various atmospheric conditions over East Asia. For example, Peng *et al.* (2017) and Kim *et al.* (2013, 2016) reported strong nucleation events in suburbs of Beijing and Xian in China and regional background sites in Korea, respectively. Several research groups also indicated the possibility of nucleation bursts during long-range transport in East Asia (Matsui *et al.*, 2013; Seto *et al.*, 2013; Chandra *et al.*, 2016). Weber *et al.* (2003) noted the importance of nanoparticle measurement at high altitudes using aircraft observation to clarify NPF process under long-range transport of air pollutants in East Asia.

As other studies have noted, aircraft measurement represent a direct, in-situ method of examining the vertical and horizontal distribution of atmospheric parameters. Several aircraft measurement campaigns have been conducted to investigate the spatial distributions and

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physical and chemical properties of gases and particulate matter (PM) over East Asia (Hoell *et al.*, 1997; Anderson *et al.*, 2003; Bahreini *et al.*, 2003; Hubert *et al.*, 2003; Matsuki *et al.*, 2003; Weber *et al.*, 2003; Hatakeyama *et al.*, 2004; Takegawa *et al.*, 2004; Hatakeyama *et al.*, 2011; Ren *et al.*, 2012; Fujiwara *et al.*, 2014; Hatakeyama *et al.*, 2014; Lee *et al.*, 2016; Hao *et al.*, 2017). Weber *et al.* (2003) observed atmospheric nanoparticles (down to 3–4 nm), with results indicating highly concentrated nanoparticles in upstream or high-altitude regions. This could be related to our recent observation of incomplete and “weak” NPF (onset diameter above 10 nm) in a downstream region, namely, Fukue Island in Japan (Chandra *et al.*, 2016). However, large-scale aircraft measurement is expensive and requires high air speeds and altitudes (> 1 km). Alternatively, lightweight unmanned aerial vehicles (UAVs) for conventional observation have been developed. In contrast to high-altitude manned-aircraft measurement studies, some studies have used UAVs to measure vertical and horizontal distributions of air pollutants at lower altitudes (Ramanathan *et al.*, 2007; Ramana *et al.*, 2010; Peng *et al.*, 2015; Pistone *et al.*, 2016; Schrod *et al.*, 2017; Mamali *et al.*, 2018). Wehner *et al.* (2010) studied NPF in Cabauw, the Netherlands, using a UAV and observed NPF in a turbulent layer before arriving at ground level. Altstädter *et al.* (2015) and Platis *et al.* (2016) investigated the vertical and horizontal distribution of ultrafine particles within the atmospheric boundary layer using UAVs with two condensation particle counters (CPCs) and studied the properties of the atmospheric boundary layer.

In this study, one type of the UAV, Kite-Plane (Sky Remote Co. Ltd.), was deployed to measure the vertical distribution (< 1.2 km) of atmospheric nanoparticles (> 6 nm) in the downstream region of the East Asian plume. The observation was combined with ground-based size distribution data (3–600 nm) and meteorological parameters at the Fukue observation supersite, Japan.

MEASUREMENT AND METHOD

Monitoring Site

Atmospheric nanoparticle observation was conducted at a monitoring site located in Fukue Island (32.8°N, 128.7°E), Japan. As shown in Fig. 1, Fukue Island is located at the southwestern boundary of Japan, and represent a suitable site for monitoring NPF and long-range transported pollutions from the Asian continent with less influence of local anthropogenic emission sources. The size distribution, chemical composition of submicron particles, SO₂, O₃, and meteorological parameters (temperature, relative humidity (RH), wind speed and direction, and solar radiation) were measured at a ground-based monitoring site (G in Fig. 1). Previous researches (Seto *et al.*, 2013; Takami *et al.*, 2005) have described the details of the ground-based monitoring system. The aerial observation site (Location A in Fig. 1) is located at northwestern edge of Fukue Island, approximately 3.7 km from the ground-based monitoring site. Several aerial observation campaigns have been conducted at Fukue Island to measure NO_x, SO₂, O₃ (Hatakeyama *et al.*, 2004, 2011; Fujiwara *et al.*, 2014; Hatakeyama *et al.*, 2014), CO

Fig. 1. The location of observation sites at Fukue Island (A: Aerial observation site and G: Ground-based monitoring site).

(Hatakeyama *et al.*, 2011; Fujiwara *et al.*, 2014; Hatakeyama *et al.*, 2014), black carbon (BC) (Hatakeyama *et al.*, 2011; Fujiwara *et al.*, 2014) and the chemical composition of PM (Hatakeyama *et al.*, 2004, 2011; Fujiwara *et al.*, 2014; Hatakeyama *et al.*, 2014) in East Asia. However, most of these measurements have focused on the chemical characteristics of outflow from the Asian continent and only limited information on the vertical distribution of nanoparticles in East Asia is available. In this study, a CPC was deployed for the first time in aerial observation to measure the vertical structure of nanoparticles at Fukue Island. The aerial observation site was selected to enable investigation of inflow air masses to Fukue Island.

Aerial Observation (April 13–16, 2017)

Intensive aerial observations at Fukue Island were carried out using the Kite-Plane. The Kite-Plane's maximum flight altitude is 3500 m above sea level with the maximum speed and payload of 35 km hour⁻¹ and up to 5 kg, respectively (Yamashita *et al.*, 2005; Hayashi *et al.*, 2010). The detailed information regarding aerial observation is included in Table 2. A CPC (Model 3781, TSI Inc., USA)

with a time resolution of 2 seconds was installed in the Kite-Plane to measure the vertical concentration of nanoparticles (> 6 nm). A tablet PC (M80T, ASUS, Taiwan) was used to control the CPC and save the measured data during aerial observation. The highest permitted flight altitude was 1200 m above sea level and three flights were allowed each day.

The performance of CPC for aerial observation was evaluated and compared with another CPC (Model 3775, TSI Inc., USA) in the laboratory using Ag particles (6–20 nm) (not shown here). In addition, the effect of Kite-Plane exhaust was examined. After a preliminary test before the flights, an aerosol inlet for the CPC was installed at the front of the Kite-Plane to minimize the emission effect on particle concentration.

Ground-Based Observation (April 13–16, 2017)

Mobility size distribution and number concentration were measured using a long-scanning mobility particle sizer (long-SMPS) composed of a Long Differential Mobility Analyzer (DMA) (Model 3081, TSI Inc., USA) and a CPC (Model 3775, TSI Inc., USA) with a size range of 14–660 nm. For smaller nanoparticles (2.5–60 nm and 1.4–30 nm), a nano-SMPS (combination of Nano DMA (Model 3085, TSI Inc., USA) and Ultrafine CPC (UCPC; Model 3776, TSI Inc., USA)) and a 1nm SMPS (Model 3938E77, TSI Inc., USA), respectively, were used at the ground-based monitoring site. Meteorological parameters, including temperature, RH, wind direction and speed, and solar radiation were measured by Chiba University, Japan. SO_2 and O_3 concentration were measured using an SO_2 analyzer (Model 43i, Thermo Scientific, USA) and an O_3 analyzer (Model 49i, Thermo Scientific, USA), respectively. In addition, an Aerosol Chemical Speciation Monitor (ACSM, Aerodyne Research Inc., USA) was used to measure the chemical composition (sulfate, nitrate, ammonium, organic, and chloride) of submicron ($< 1 \mu\text{m}$) particles. Detailed information on measurement parameters is described in Table 1.

For the investigation of the origin, vertical distribution, and pathway of air masses, air mass backward trajectory analysis was conducted using the Hybrid Single-Particle Lagrangian Integrated Trajectory model, version 4 (HYSPLIT4; Draxler and Rolph, 2003); 48-hour air mass backward trajectory analysis was performed at elevations of 80, 200, 400, 600, 800, 1000, and 1200 m arrival height above the ground level at both the ground-based and aerial observation sites). In addition, data on aerosol optical depth (AOD) over Northeast Asia were retrieved to investigate aerosol spatial distribution during the observation periods (<http://worldview.earthdata.nasa.gov>).

RESULTS AND DISCUSSION

Data Overview and Meteorological Conditions

Fig. 2 shows the temporal variation in particle number size distribution, chemical composition, SO_2 concentration, and meteorological data. As shown in Fig. 2, the particle number concentration during the 4-day intensive

measurement period (from 2.5 nm to 600 nm) ranged from 197 to 32,110 particles cm^{-3} with an average of $2,965 \pm 1,948$ particles cm^{-3} . In calculating the average particle number concentration, 1-nm SMPS data were excluded because the 1-nm SMPS was only deployed during aerial observation. Our earlier studies (Seto *et al.*, 2013; Chandra *et al.*, 2016) reported that the average atmospheric nanoparticle concentration at Fukue Island was approximately 2,000 particles cm^{-3} , without any inflow from outside Fukue Island. The average concentration of PM_{10} (the sum of ammonium, chloride, organic, nitrate, and sulfate measured by ACSM) was $8.23 \pm 3.72 \mu\text{g cm}^{-3}$ during intensive measurement, with the daily average fraction of submicron particles differing from day to day. SO_2 concentration varied between 0.01 and 3.13 ppb, with an average of 0.75 ± 0.65 ppb. Based on the onset diameter (the starting diameter that could be measured by our system, i.e., SMPS) of NPF and growth (< 10 nm or > 10 nm), SO_2 concentration, the dominant chemical composition of submicron particles ($< 1 \mu\text{m}$), and air mass patterns, three different events were identified. During 4-day intensive measurements, long-range transport of air pollutants from northeast China with an NPF event was observed on April 13, 2017. On April 14, a high concentration of nanoparticles was detected due to a change of air mass origin starting at approximately 12:00. During the last two days (April 15–16), most of the air masses originated from the East China Sea and the particle number concentration decreased to 1,500 particles cm^{-3} . Regarding meteorological parameters, the maximum solar radiation during the intensive measurement period was 842.2 (April 13), 915.6 (April 14), 798.1 (April 15), and 761.6 (April 16) W m^{-2} . Wind speed ranged from 0.2 to 6.6 m s^{-1} , with an average of 1.6 m s^{-1} , and the wind direction was northwesterly and/or southwesterly. According to the weather map (Fig. 3), high pressure was dominant on April 13 and 14 near Fukue Island (Figs. 3(a) and 3(b)), whereas the island was situated between high and low fronts on April 15 and 16 (Figs. 3(c) and 3(d)). The average temperature and relative humidity were 18.0°C and 73.0%, respectively, during the measurement period.

During aerial observation, a total of 10 flights were conducted including one test flight without nanoparticle measurement. The flight area over Fukue Island was approximately $750 \times 1300 \text{ m}^2$ (as shown in Fig. 1) for investigation of long-range transport from other regions as well as NPF events. The particle number concentration ($< 1.2 \text{ km}$) during aerial observation varied between 0.3 and 35,750 particles cm^{-3} . The vertical distribution of atmospheric nanoparticles varied with the air mass origin and meteorological conditions. The details of each event are discussed in the following sections.

Event I (Strong NPF with Long-range Transport of Air Pollutants under a High-pressure System)

On April 13, a sudden increase of particle number concentration in the size range from the diameter of 5 nm started between 09:00 and 10:00 in the morning. Prior to this, the SO_2 concentration increased from early in the morning (before sunrise), representing a sufficient precursor

Table 1. Measurement parameter information at Fukue Island.

Parameters		Instrument	Time resolution	Typical value (range)
Aerial observation	Location	GPS, Kite Plane (Sky Remote Co. Ltd.)	1 second	Latitude: 32.76–32.77°N, Longitude: 128.64–128.65°E Height: 15–1200 m N: 0–38150 (# cm ⁻³) Temp: 7.9–27.2 (°C), RH: 8.0–79.2 (%), P: 875.3–1013.0 (hPa), WS: 0–26.5 (m s ⁻¹), WD: Northwest and Southwest N: 200–36970 (# cm ⁻³) N: 20–26470 (# cm ⁻³) N: 60–24880 (# cm ⁻³) Temp: 11.1–24.6 (°C), RH: 30.1–97.8 (%), WS: 0.2–6.6 (m s ⁻¹), WD: Northwest and Southwest, Solar radiation: 0–915.6 (W m ⁻²) 0.01–3.13 (ppb) 1.8–22.5 (µg m ⁻³)
	Number concentration Temp., RH, and P	CPC (> 6 nm, TSI Inc., model 3781) Meteorological sonde	2 seconds 1 second	
Ground-based observation	Number size distribution	Long-SMPS (14–600 nm, TSI Inc., model 3081 (DMA) + model 3775 (CPC))	3 minutes	
	Number size distribution	1 nm SMPS (1–30 nm, TSI Inc., model 3938E77)	3 minutes	
	Number size distribution	Nano-SMPS (2.5–60 nm, TSI Inc., model 3085 (Nano-DMA) + model 3776 (UCPC))	3 minutes	
	Temp, RH, WS, WD, and solar radiation	Met. Devices	10 seconds	
SO ₂ Chemical composition		Gas monitor (Thermo Scientific Inc., model 43i) Aerosol chemical speciation monitor (ACSM, Aerodyne Research Inc., USA)	1 minutes 15 minutes

Fig. 2. Temporal variation of (a) contour plot of particle number size distribution, (b) particle number concentration by 1nm SMPS, Nano SMPS, and long SMPS, (c) chemical composition (measured by ACSM), (d) SO_2 , (e) solar radiation, and (f) wind direction and wind speed observed at ground-based site in Fukue Island during the period from April 13 to 16, 2017.

Fig. 3. Weather map (pressure map) during intensive measurement periods provided by Japan Meteorological Agency: (a) April 13, 09:00 JST, (b) April 14, 09:00 JST, (c) April 15, 09:00 JST, and (d) April 16, 09:00 JST (www.data.jma.go.jp).

of NPF. Moreover, strong solar radiation (max: 842.2 W m^{-2}) contributed to NPF, providing suitable conditions for photochemical reactions in the ambient atmosphere (Seto *et al.*, 2013; Chandra *et al.*, 2016). In addition, subsequent particle growth of particles to several tens of nanometers after NPF was observed. This event was identified by favorable conditions for NPF with a high SO_2 concentration (Fig. 2(d)) and strong solar radiation (Fig. 2(e)) under the effect of a high-pressure system (Fig. 3(a)). Many previous studies have shown that meteorological conditions such as a clear sky with few clouds, high solar radiation intensity, and no precipitation under a high-pressure system can affect NPF and particle growth in various regions (Covert *et al.*, 1996; Wehner *et al.*, 2004; Chandra *et al.*, 2016; Kim *et al.*, 2016). This high concentration of ultrafine particles ($< 100 \text{ nm}$) remained for 12 hours (until 22:00; Fig. 2(a)). Apart from the NPF event, high concentrations in other nanoparticle size ranges ($\sim 100 \text{ nm}$) were observed throughout the day; these may have been preexisting atmospheric particles. Regarding the chemical compositions in PM_{10} during the first day of intensive measurement, organics were predominant, followed by sulfate, nitrate, and ammonium. Based on air mass backward trajectory analysis, most of the air masses originated from polluted regions (northeast China, Fig. 4(b)). Two aerial observations (11:55–12:35 and 15:00–15:40) were conducted using the Kite-Plane with a

CPC. The vertical distribution of atmospheric nanoparticles, potential temperature, water vapor mixing ratio and the air masses recorded during the first aerial observation are shown in Figs. 4 and 5, respectively. During the first aerial observation, the particle number concentration ($2.5\text{--}600 \text{ nm}$) at ground level was between 15,000 and 20,000 particles cm^{-3} , while particle number concentration ($> 6 \text{ nm}$) increased to approximately 36,000 particles cm^{-3} at 400-m level (Fig. 4(a)). Above 400 m, the particle number concentration decreased to 2,000 particles cm^{-3} . The change in boundary-layer height (between 600 and 800 m) determined by the water vapor mixing ratio affected the vertical distribution of nanoparticles during aerial observation. The particle number concentration up to a height of 800 m was greater during the first flight than during the second, with the maximum difference in particle concentration ($\sim 24,000$ particles cm^{-3}) observed at approximately 400 m. This may have been caused by meteorological conditions such as vertical mixing, the boundary-layer height, or other factors (Table 2). However, from 900 to 1200 m, the particle number concentration was greater during the second flight, possibly because of a change of height in the air mass origin (Fig. 5(c)). Although the air mass origin at 80, 200, and 400 m was quite different during the first and second flights on April 13, most air masses moved near ground level. However, air masses above 600 m were less affected

Fig. 4. Vertical distribution of (a) atmospheric nanoparticles (left), potential temperature (middle), and water vapor mixing ratio (right) during aerial observation and (b) air mass backward trajectory on April 13, 2017 (JST).

by the surface concentration during transport (Figs. 5(a) and 5(b)). Based on the vertical distribution of air mass backward trajectory during aerial observation, most air masses below 400 m originated from below 1 km, and vertical movement within 300 m was observed (Fig. 5(a)). Therefore, we can speculate that various air pollutants, including organics and SO_2 , were transported from polluted regions that many nanoparticles could be formed during long-range transport.

During the second aerial observation in the afternoon, a higher particle number concentration ($> 8,000$ particles cm^{-3}) up to 500 m was observed (Fig. 4(a)). In addition, most air masses below 400 m moved near ground level. In light of other studies employing air mass backward trajectory analysis, this high concentration could have been due to long-range transport of anthropogenic sources from northeast China and/or Inner Mongolia (Zhang *et al.*, 2009; Chandra *et al.*, 2016). Moreover, an increased particle number

Fig. 5. Air mass backward trajectory during aerial observation on April 13, 2017: (a) 1st flight (11:55–12:35 JST), (b) 2nd flight (15:00–15:40 JST), and (c) air mass backward trajectory before and after during 2nd aerial observation at 800 m.

concentration was observed in the afternoon above 800 m, possibly caused by the inflow of different air masses. At 800 m, the air mass origin changed from over 3000 to approximately 1500 m between 15:00 and 16:00 (JST) (Fig. 5(c)). This height change may have caused vertical aerosol mixing in the source region, thereby influencing the increase in particle number concentration over 800 m (Fig. 5(c)).

Event II (Weak NPF with Change of Air Mass Origin on April 14)

Fig. 6 shows the vertical distribution of atmospheric nanoparticles, potential temperature, water vapor mixing ratio and air masses on April 14. In the morning, the average particle number concentration at the ground site

was less than $3,000 \text{ particles cm}^{-3}$. The particle number concentration also decreased with increasing flight altitude during the two aerial observations before noon. However, the particle number concentration in the size range from 20 to 100 nm began increasing at 14:00 and continued until 20:00 (Fig. 2; average number concentration between 14:00 and 20:00 JST was $5,273 \text{ particles cm}^{-3}$) at the ground site. In addition, the SO_2 concentration increased with the particle number concentration to 2.50 ppb between 16:00 and 17:00 JST. Based on SO_2 emissions data (700 t day^{-1} near Event II at Mt. Aso in Kyushu, Japan) and air mass backward trajectory analysis, the change of air mass origin (from the Yellow Sea to the Korean Peninsula and Kyushu) as well as SO_2 emitted from volcanic sources in Kyushu and anthropogenic local emissions over these areas could

Fig. 6. Vertical distribution of (a) atmospheric nanoparticles (left), potential temperature (middle), and water vapor mixing ratio (right) during aerial observation and (b) air mass backward trajectory on April 14, 2017 (JST).

have influenced the increase in particle number and SO_2 concentrations at the measurement site. Unfortunately, no aerial observation could be conducted due to strong winds in the afternoon of April 14. In contrast with the situation observed on April 13, the onset diameter of nanoparticles on this day started at approximately 10 nm, suggesting that these particles could have formed during transport or in the upstream region over the ground measurement site.

Event III (No NPF with Highly Aged Sulfate Particles on April 15 and 16)

The vertical distribution of atmospheric nanoparticles,

potential temperature and water vapor mixing ratio on April 15 and air mass backward trajectory on April 15 and 16 are shown in Figs. 7 and 8, respectively. During the last two days of intensive measurement, the average particle number concentration at the ground-based site was 1,812 and 1,207 particles cm^{-3} , respectively. Data measured using the Nano SMPS was not available from the afternoon of April 15 to the end of intensive measurement due to a technical problem; therefore, only long-SMPS data was used in the analysis of atmospheric nanoparticles at this time. Although the particle number concentration was less than 2,000 particles cm^{-3} and the SO_2 concentration mostly

Fig. 7. Vertical distribution of (a) atmospheric nanoparticles (left), potential temperature (middle), and water vapor mixing ratio (right) during aerial observation on April 15, 2017 (JST).

Fig. 8. Air mass backward trajectory on (a) April 15 and (b) April 16, 2017 (JST).

lower than the instrument's detection limit, the sulfate concentration in submicron size ($< 1 \mu\text{m}$) increased during these two days. The average sulfate concentration was $4.98 \mu\text{g m}^{-3}$, and with the non-sea-salt (nss) sulfate fraction over 0.95 during the last two days of measurement. Aerial observations were conducted five times during the last two days (three times on April 15 and twice on April 16). The particle number concentration was mainly $2,000 \text{ particles cm}^{-3}$ or less, and no increase of particle number concentration related to increasing sulfate could be observed during aerial observation. The average SO_2 concentration at the ground site was 0.15 ppb. According to air mass backward trajectory analysis, most air masses originated over the East China Sea and their movement was slower than on the previous two days (April 13–14). Satellite-retrieved AOD data was used to investigate the high sulfate concentration. Starting on April 15, high aerosol loading was observed over southeast China, including Shanghai. In addition, these high-concentration aerosols were observed moving over the East China Sea on April 15 and 16. Based on air mass backward trajectory (relatively slow air mass speed) and AOD data (over the southeast China region) (Fig. S1), the high aerosol loadings observed over southeast China could have affected the measurement site on April 15 and 16. To investigate particle aging, the ratio between sulfate (SO_4^{2-}) and the sum of sulfur dioxide (SO_2) and sulfate (SO_4^{2-}) was calculated, and found to be close to 1 on April 15 and 16, suggesting that the aerosols aged during the transport. Moreover, relatively low moving speed of air masses over the East China Sea could have ensured sufficient residence time for particle aging.

Comparison of the Vertical Distribution of Atmospheric Nanoparticles

Table 2 lists the characteristics of the three events during intensive measurement in April 2017. The vertical distribution of atmospheric nanoparticles, potential temperature, and water vapor mixing ratio are shown in Figs. 4, 6, and 7 for each event. The temperature lapse rate ranged from 0.6 to 0.8°C per 100 m. RH mainly increased with height from the ground to a level near the mixed layer (between 600 and 1000 m), then rapidly decreased within the mixed layer. The mixed-layer height varied according to different days and observation times. In most cases, the particle number concentration decreased with increasing height from the ground, except on April 13. During the ascent and descent of the first flight on April 13, the maximum particle number concentration was observed near 400 m, and was three times higher ($\sim 36,000 \text{ particles cm}^{-3}$) than that measured at the ground site. Although it is difficult to distinguish an NPF event based only on aerial observation data, higher particle number concentration ($36,000 \text{ particles cm}^{-3}$ near 400 m) than that measured on the ground was observed. In our earlier studies on five years of measurement, the maximum particle number concentration (3–670 nm) in ground-based measurement was less than $20,000 \text{ particles cm}^{-3}$ and such a high concentration ($36,000 \text{ particles cm}^{-3}$) was not observed. According to combined ground-based measurement and aerial observation

Table 2. Comparison of characteristics of three different events during the intensive measurement periods.

	Event I (April 13, 2017)	Event II (April 14, 2017)	Event III (April 15 and 16, 2017)
Type of event	Strong NPF	Weak NPF	No NPF
Ave. / peak number conc. ($\# \text{ cm}^{-3}$) ^a	4900 / 14900	3300 / 12200	1800 / 8000
Ave. / peak number conc. ($\# \text{ cm}^{-3}$) ^b	9900 / 35800 ($\sim 350 \text{ m}$)	3700 / 9300 (less than 50 m)	1700 / 4300 (less than 50 m)
Aerial observation time	11:55–12:35 / 15:00–15:40	08:35–09:35 / 11:10–11:45	08:40–09:30 / 11:05–11:50 / 14:00–15:00 (April 15) 08:55–09:35 / 11:00–12:00 (April 16)
Max. height of aerial observation (m)	1200 / 1200	1200 / 900	1200 / 1200 / 700 (April 15)
Mixing height (m)	400–700 / 400–600	600–800 / 400–700	700–1000 / 600–800 / N.A. (April 15)
Ave. / peak SO_2 conc. (ppb)	1.1 / 3.1	0.7 / 2.5	0.1 / 0.5
Ave. conc. ($\mu\text{g m}^{-3}$) of PM_{10} / dominant chemical species in PM_{10}	6.5 / Organic	6.1 / Organic and sulfate	10.2 / Sulfate
W.D. (degree) / W.S. (m s^{-1})	Northwest / 1.1	Northwest and Southwest / 2.4	Southwest / 1.4
Atmospheric pressure system	Under high pressure	Under high pressure	Between high and low pressure
Origin of air mass (48-hr)	Northeastern China	Yellow sea / Korea and Japan	East China Sea

^a particle number concentration for ground-based measurement (2.5–600 nm).

^b particle number concentration for aerial observation ($> 6 \text{ nm}$).

^c measured by aerosol chemical speciation monitor (ACSM).

N.A. (Not applicable).

data, NPF could have contributed to a large high-altitude particle concentration or occurred during transport and/or in the upstream region before reaching Fukue Island. Furthermore, the increased particle number concentration above 800 m that was observed during the second flight on April 13 was caused by the inflow of different trans-boundary air masses (Fig. 5). At a height of 800 m, the air mass origin changed from over 3000 m to approximately 1500 m between 15:00 and 16:00 (JST), which could have caused vertical aerosol mixing in the source region and thus influenced the particle number concentration at this altitude. The characteristics of the three different events during the intensive measurement periods are summarized in Table 2. The information on ground-based aerosol size distribution and vertical variations in particle number concentration may help reduce uncertainty when estimating radiative forcing caused by aerosols.

CONCLUSIONS

In this study, the vertical distributions and characteristics of atmospheric nanoparticles were investigated from April 13 to April 16, 2017, at the Fukue observation supersite in Japan. During 4-day intensive measurements, three different events were recorded via aerial observation and at a ground-based site. These events were analyzed using combined aerial and ground-based observation data. On April 13, NPF and an increased nanoparticle number concentration below 400 m were detected at the ground-based site and via aerial observation, respectively. In addition, an increased nanoparticle concentration between 800 and 1,200 m due to a change in the altitude of the air mass origin was identified via aerial observation. A sudden change in the origin of the air mass over the polluted area similarly caused increased nanoparticle numbers and SO₂ concentrations on the afternoon of April 14. A high concentration of sulfate in the PM₁ without increased SO₂ and nanoparticle concentrations was detected both at the ground-based site and via aerial observation on April 15 and 16. Starting on April 15, high aerosol loading over southeast China and aerosol transport over the East China Sea were observed. Most high-sulfate particles were affected by southeast China, including Shanghai, and these particles were highly aged during long-range transport over the East China Sea. These findings reveal the applicability of our UAV for detecting NPF processes in the downstream region of East Asia.

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SUPPLEMENTARY MATERIAL

Supplementary data associated with this article can be found in the online version at <http://www.aaqr.org>.

REFERENCES

- Altstädter, B., Platis, A., Wehner, B., Scholtz, A., Wildmann, N., Hermann, M., Käthner, R., Baars, H., Bange, J. and Lampert, A. (2015). ALADINA – An unmanned research aircraft for observing vertical and horizontal distributions of ultrafine particles within the atmospheric boundary layer. *Atmos. Meas. Tech.* 8: 1627–1639.
- Anderson, T.L., Masonis, S.J., Covert, D.S., Ahlquist, N.C., Howell, S.G., Clarke, A.D. and McNaughton, C.S. (2003). Variability of aerosol optical properties derived from in situ aircraft measurements during ACE-Asia. *J. Geophys. Res.* 108: 8647.
- Bahreini, R., Jimenez, J.L., Wang, J., Flagan, R.C., Seinfeld, J.H., Jayne, J.T. and Worsnop, D.R. (2003). Aircraft-based aerosol size and composition measurements during ACE-Asia using an Aerodyne aerosol mass spectrometer. *J. Geophys. Res.* 108: 8645.
- Chandra, I., Kim, S., Seto, T., Otani, Y., Takami, A., Yoshino, A., Iri, S., Park, K., Takamura, T. and Kaneyasu, N. (2016). New particle formation under the influence of the long-range transport of air pollutants in East Asia. *Atmos. Environ.* 141: 30–40.
- Covert, D., Kapustin, V., Bates, T. and Quinn, P. (1996). Physical properties of marine boundary layer aerosol particles of the mid-Pacific in relation to sources and meteorological transport. *J. Geophys. Res.* 101: 6919–6930.
- Dal Maso, M., Kulmala, M., Lehtinen, K.E., Mäkelä, J., Aalto, P. and O'Dowd, C. (2002). Condensation and coagulation sinks and formation of nucleation mode particles in coastal and boreal forest boundary layers. *J. Geophys. Res.* 107: PAR 2-1–PAR 2-10.
- Davidson, C.I., Phalen, R.F. and Solomon, P.A. (2005). Airborne particulate matter and human health: A review. *Aerosol Sci. Technol.* 39: 737–749.
- Draxler, R. and Rolph, G. (2003). Hysplit (Hybrid Single-Particle Lagrangian Integrated Trajectory) Model Access Via NOAA Arl Ready. NOAA Air Resources Laboratory, Silver Spring, Md. Dostupno na: <http://ready.arl.noaa.gov/HYSPLIT.php>, Last Access: 06 June 2010.
- Fujiwara, H., Sadanaga, Y., Urata, J., Masui, Y., Bandow, H., Ikeda, K., Hanaoka, S., Watanabe, I., Arakaki, T. and Kato, S. (2014). Aerial observation of nitrogen compounds over the East China Sea in 2009 and 2010. *Atmos. Environ.* 97: 462–470.
- Hao, J., Yin, Y., Kuang, X., Chen, J., Yuan, L., Xiao, H., Li, Z., Pu, M., Wang, J. and Zhou, X. (2017). Aircraft measurements of the aerosol spatial distribution and relation with clouds over Eastern China. *Aerosol Air Qual. Res.* 17: 3230–3243.
- Hatakeyama, S., Takami, A., Sakamaki, F., Mukai, H., Sugimoto, N., Shimizu, A. and Bandow, H. (2004).

- Aerial measurement of air pollutants and aerosols during 20–22 March 2001 over the East China Sea. *J. Geophys. Res.* 109: D13304.
- Hatakeyama, S., Hanaoka, S., Ikeda, K., Watanabe, I., Arakaki, T., Sadanaga, Y., Bandow, H., Kato, S., Kajii, Y. and Sato, K. (2011). Aerial observation of aerosols transported from East Asia—Chemical composition of aerosols and layered structure of an air mass over the East China Sea. *Aerosol Air Qual. Res.* 11: 497–507.
- Hatakeyama, S., Ikeda, K., Hanaoka, S., Watanabe, I., Arakaki, T., Bandow, H., Sadanaga, Y., Kato, S., Kajii, Y. and Zhang, D. (2014). Aerial observations of air masses transported from East Asia to the Western Pacific: Vertical structure of polluted air masses. *Atmos. Environ.* 97: 456–461.
- Hayashi, M., Kamiya, Y., Ozuka, K., Yamashita, K., Higashi, H., Ashida, M., Okabe, K. and Shigeta, T. (2010). Mineral Particles internally mixed with sea salt over Karatsu Bay, Southwest of Japan, observed with a small unmanned aerial vehicle. *J. Fukuoka Univ. Sci. Learn.* 40: 245–253.
- Hoell, J., Davis, D., Liu, S., Newell, R., Akimoto, H., McNeal, R. and Bendura, R. (1997). The pacific exploratory mission-west phase B: February–March 1994. *J. Geophys. Res.* 102: 28223–28239.
- Huebert, B.J., Bates, T., Russell, P.B., Shi, G., Kim, Y.J., Kawamura, K., Carmichael, G. and Nakajima, T. (2003). An overview of ACE-Asia: Strategies for quantifying the relationships between Asian aerosols and their climatic impacts. *J. Geophys. Res.* 108: 8633.
- Kim, Y., Yoon, S.C., Kim, S.W., Kim, K.Y., Lim, H.C. and Ryu, J. (2013). Observation of new particle formation and growth events in Asian continental outflow. *Atmos. Environ.* 64: 160–168.
- Kim, Y., Kim, S.W., Yoon, S.C., Park, J.S., Lim, J.H., Hong, J., Lim, H.C., Ryu, J., Lee, C.K. and Heo, B.H. (2016). Characteristics of formation and growth of atmospheric nanoparticles observed at four regional background sites in Korea. *Atmos. Res.* 168: 80–91.
- Koike, M., Kondo, Y., Kita, K., Takegawa, N., Masui, Y., Miyazaki, Y., Ko, M., Weinheimer, A., Flocke, F. and Weber, R. (2003). Export of anthropogenic reactive nitrogen and sulfur compounds from the East Asia region in spring. *J. Geophys. Res.* 108: 8789.
- Kulmala, M., Maso, M.D., Mäkelä, J., Pirjola, L., Väkevä, M., Aalto, P., Mikkiläinen, P., Hämeri, K. and O'dowd, C. (2001). On the formation, growth and composition of nucleation mode particles. *Tellus B* 53: 479–490.
- Lee, G., Oh, H.R., Ho, C.H., Kim, J., Song, C.K., Chang, L.S., Lee, J.B. and Lee, S. (2016). Airborne measurements of high pollutant concentration events in the free troposphere over the West Coast of South Korea between 1997 and 2011. *Aerosol Air Qual. Res.* 16: 1118–1130.
- Lu, Z., Streets, D.G., Zhang, Q., Wang, S., Carmichael, G.R., Cheng, Y.F., Wei, C., Chin, M., Diehl, T. and Tan, Q. (2010). Sulfur dioxide emissions in China and sulfur trends in East Asia since 2000. *Atmos. Chem. Phys.* 10: 6311–6331.
- Mamali, D., Marinou, E., Sciare, J., Pikridas, M., Kokkalis, P., Kottas, M., Biniotoglou, I., Tsekeri, A., Keleshis, C. and Engelmann, R. (2018). Vertical profiles of aerosol mass concentration derived by unmanned airborne in situ and remote sensing instruments during dust events. *Atmos. Meas. Tech.* 11: 2897–2910.
- Matsui, H., Kondo, Y., Moteki, N., Takegawa, N., Sahu, L., Koike, M., Zhao, Y., Fuelberg, H., Sessions, W. and Diskin, G. (2011). Accumulation-mode aerosol number concentrations in the Arctic during the ARCTAS aircraft campaign: Long-range transport of polluted and clean air from the Asian continent. *J. Geophys. Res.* 116: D20217.
- Matsui, H., Koike, M., Takegawa, N., Kondo, Y., Takami, A., Takamura, T., Yoon, S.J., Kim, S.W., Lim, H.C. and Fast, J.D. (2013). Spatial and temporal variations of new particle formation in East Asia using an NPF-explicit WRF-chem model: North-south contrast in new particle formation frequency. *J. Geophys. Res.* 118: 11647–11663.
- Matsuki, A., Iwasaka, Y., Osada, K., Matsunaga, K., Kido, M., Inomata, Y., Trochkin, D., Nishita, C., Nezuka, T. and Sakai, T. (2003). Seasonal dependence of the long-range transport and vertical distribution of free tropospheric aerosols over east Asia: On the basis of aircraft and lidar measurements and isentropic trajectory analysis. *J. Geophys. Res.* 108: 8663.
- Mönkkönen, P., Koponen, I., Lehtinen, K., Hämeri, K., Uma, R. and Kulmala, M. (2005). Measurements in a highly polluted Asian mega city: observations of aerosol number size distribution, modal parameters and nucleation events. *Atmos. Chem. Phys.* 5: 57–66.
- Peng, Y., Dong, Y., Li, X., Liu, X., Dai, J., Chen, C., Dong, Z., Du, C. and Wang, Z. (2017). Different characteristics of new particle formation events at two suburban sites in northern China. *Atmosphere* 8: 258.
- Peng, Z.R., Wang, D., Wang, Z., Gao, Y. and Lu, S. (2015). A study of vertical distribution patterns of PM_{2.5} concentrations based on ambient monitoring with unmanned aerial vehicles: A case in Hangzhou, China. *Atmos. Environ.* 123: 357–369.
- Penner, J.E., Dong, X. and Chen, Y. (2004). Observational evidence of a change in radiative forcing due to the indirect aerosol effect. *Nature* 427: 231.
- Pistone, K., Praveen, P.S., Thomas, R.M., Ramanathan, V., Wilcox, E.M. and Bender, F.A.M. (2016). Observed correlations between aerosol and cloud properties in an Indian Ocean trade cumulus regime. *Atmos. Chem. Phys.* 16: 5203–5227.
- Platis, A., Altstädter, B., Wehner, B., Wildmann, N., Lampert, A., Hermann, M., Birmili, W. and Bange, J. (2016). An observational case study on the influence of atmospheric boundary-layer dynamics on new particle formation. *Boundary Layer Meteorol.* 158: 67–92.
- Ramana, M., Ramanathan, V., Feng, Y., Yoon, S., Kim, S., Carmichael, G. and Schauer, J. (2010). Warming influenced by the ratio of black carbon to sulphate and the black-carbon source. *Nat. Geosci.* 3: 542.
- Ramanathan, V., Ramana, M.V., Roberts, G., Kim, D., Corrigan, C., Chung, C. and Winker, D. (2007). Warming trends in Asia amplified by brown cloud solar absorption. *Nature* 448: 575.

- Ren, L., Zhang, R., Bai, Z., Chen, J., Liu, H., Zhang, M., Yang, X. and Zhang, L. (2012). Aircraft measurements of ionic and elemental components in PM_{2.5} over Eastern Coastal area of China. *Aerosol Air Qual. Res.* 12: 1237–1246.
- Schrod, J., Weber, D., Drücke, J., Keleshis, C., Pikridas, M., Ebert, M., Cvetković, B., Nickovic, S., Marinou, E. and Baars, H. (2017). Ice nucleating particles over the Eastern Mediterranean measured by unmanned aircraft systems. *Atmos. Chem. Phys.* 17: 4817–4835.
- Seto, T., Kim, S., Otani, Y., Takami, A., Kaneyasu, N., Fujimoto, T., Okuyama, K., Takamura, T. and Hatakeyama, S. (2013). New particle formation and growth associated with East-Asian long range transportation observed at Fukue Island, Japan in March 2012. *Atmos. Environ.* 74: 29–36.
- Takami, A., Miyoshi, T., Shimono, A. and Hatakeyama, S. (2005). Chemical composition of fine aerosol measured by AMS at Fukue Island, Japan during APEX period. *Atmos. Environ.* 39: 4913–4924.
- Takegawa, N., Kondo, Y., Koike, M., Chen, G., Machida, T., Watai, T., Blake, D., Streets, D., Woo, J.H. and Carmichael, G. (2004). Removal of NO_x and NO_y in Asian outflow plumes: Aircraft measurements over the western Pacific in January 2002. *J. Geophys. Res.* 109: D23S04.
- Weber, R., Lee, S., Chen, G., Wang, B., Kapustin, V., Moore, K., Clarke, A., Mauldin, L., Kosciuch, E. and Cantrell, C. (2003). New particle formation in anthropogenic plumes advecting from Asia observed during TRACE-P. *J. Geophys. Res.* 108: 8814.
- Wehner, B., Wiedensohler, A., Tuch, T., Wu, Z., Hu, M., Slanina, J. and Kiang, C. (2004). Variability of the aerosol number size distribution in Beijing, China: New particle formation, dust storms, and high continental background. *Geophys. Res. Lett.* 31: L22108.
- Wehner, B., Siebert, H., Ansmann, A., Ditas, F., Seifert, P., Stratmann, F., Wiedensohler, A., Apituley, A., Shaw, R. and Manninen, H. (2010). Observations of turbulence-induced new particle formation in the residual layer. *Atmos. Chem. Phys.* 10: 4319–4330.
- Yamashita, K., Hayashi, M., Irie, M., Yamamoto, K., Saga, K., Ashida, M., Shiraishi, K. and Okabe, K. (2005). Amount and state of mineral particles in the upper mixed layer and the lower free troposphere over Mt. Raizan, Southwestern Japan: Unmanned airplane measurements in the spring of 2003. *J. Meteorolog. Soc. Jpn.* 83: 121–136.
- Yu, F., Luo, G., Bates, T.S., Anderson, B., Clarke, A., Kapustin, V., Yantosca, R.M., Wang, Y. and Wu, S. (2010). Spatial distributions of particle number concentrations in the global troposphere: Simulations, observations, and implications for nucleation mechanisms. *J. Geophys. Res.* 115: D17205.
- Zhang, Q., Zhao, C., Tie, X., Wei, Q., Huang, M., Li, G., Ying, Z. and Li, C. (2006). Characterizations of aerosols over the Beijing region: A case study of aircraft measurements. *Atmos. Environ.* 40: 4513–4527.
- Zhang, Q., Streets, D.G., Carmichael, G.R., He, K., Huo, H., Kannari, A., Klimont, Z., Park, I., Reddy, S. and Fu, J. (2009). Asian emissions in 2006 for the NASA INTEX-B mission. *Atmos. Chem. Phys.* 9: 5131–5153.

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