Atmos. Chem. Phys. Discuss., 13, 21801–21835, 2013
www.atmos-chem-phys-discuss.net/13/21801/2013/
doi:10.5194/acpd-13-21801-2013

Atmospheric Chemistry and Physics Discussions

Wet and dry deposition of mineral dust particles in Japan: factors related to temporal variation and spatial distribution

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Received: 21 June 2013 – Accepted: 12 August 2013 – Published: 23 August 2013

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Published by Copernicus Publications on behalf of the European Geosciences Union.

ACPD
13, 21801–21835, 2013

Abstract

Data of temporal variations and spatial distributions of mineral dust deposition fluxes are very limited in terms of duration, location, and processes of deposition. To ascertain temporal variations and spatial distributions of mineral dust deposition by wet and dry processes, weekly deposition samples were obtained at Sapporo, Toyama, Nagoya, Tottori, Fukuoka, and Cape Hedo (Okinawa) in Japan during October 2008–December 2010 using automatic wet and dry separating samplers. Mineral dust weights in water-insoluble residue were estimated from Fe contents measured using an X-ray fluorescence analyzer. For wet deposition, highest and lowest annual dust fluxes were found at Toyama (9.6 g m^{-2} yr^{-1}) and at Cape Hedo (1.7 g m^{-2} yr^{-1}) as average values in 2009 and 2010. Higher wet deposition fluxes were observed at Toyama and Tottori, where frequent precipitation (>60 % days per month) was observed during dusty seasons. For dry deposition among Toyama, Tottori, Fukuoka, and Cape Hedo, the highest and lowest annual dust fluxes were found respectively at Fukuoka (5.2 g m^{-2} yr^{-1}) and at Cape Hedo (2.0 g m^{-2} yr^{-1}) as average values in 2009 and 2010.

Although the seasonal tendency of the monthly dry deposition amount roughly resembled that of monthly days of *Kosa* dust events, the monthly amount of dry deposition was not proportional to monthly days of the events. Comparison of dry deposition fluxes with vertical distribution of dust particles deduced from Lidar data and coarse particle concentrations suggested that the maximum dust layer height or thickness is an important factor for controlling the dry deposition amount after long-range transport of dust particles. Size distributions of refractory dust particles were obtained using four-stage filtration: > 20, > 10, > 5, and > 1 µm diameter. Weight fractions of the sum of >20 µm and 10–20 µm (giant fraction) were higher than 50 % for most of the event samples. Irrespective of the deposition type, the giant dust fractions were decreasing generally with increasing distance from the source area, suggesting the selective depletion of larger giant particles during atmospheric transport. Because giant dust particles are an important mass fraction of dust accumulation, especially in the north

ACPD
13, 21801–21835, 2013

Pacific where is known as a high-nutrient, low-chlorophyll (HNLC) region, the transport height of giant dust particles is an important factor for studying dust budgets in the atmosphere and their role in biogeochemical cycles.

1 Introduction

Mineral dust particles in the atmosphere play an important role in meteorological effects and bio-geochemical cycles (Mahowald et al., 2005; McTanish and Strong, 2007; Shao et al., 2011). The meteorological and climatic importance of mineral dust particles includes the absorption and scattering of radiation in the atmosphere and the modifications of the optical properties of clouds and snow/ice surfaces (Tegen, 2003; Aoki et al., 2006). Atmospheric mineral dust also contributes to transport nutrients such as phosphorus (P), iron (Fe), and nitrate (NO_3^-) etc to marine and terrestrial ecosystems (Nishikawa et al., 1991; Okin, et al., 2004; Jickells et al., 2005). To evaluate these effects over regional and global scales, temporal variation and spatial distributions of deposition fluxes of mineral dusts must be ascertained. In addition to direct measurements at the ground, observations by ground-based and space-born Lidars etc provide vertical and spatial distributions of mineral dust particles in the atmosphere (for examples, Shimizu et al., 2004; Su and Toon, 2011; Mona et al., 2012). However, data related to temporal variations and spatial distributions of mineral dust flux to the ground are very limited (Washington and Wiggs, 2011; Schulz et al., 2012). Using appropriate deposition velocities and scavenging ratios with measured data on aerosols and precipitation, Duce et al. (1991) estimated the global dust deposition flux, providing insight into a global map of dust deposition over the ocean. However, details of the regional flux gradient and temporal variations remain unknown.

Asian mineral dust particles derived from Chinese and Mongolian deserts are transported by westerly winds to the northwestern Pacific. In Japan, located at the western edge of the north Pacific, visibility-reducing aeolian dust events, called *Kosa* in Japanese, have been observed mainly during spring (Koizumi, 1932; Arao et al., 2003).

ACPD
13, 21801–21835, 2013

Kosa events in Japan have also been studied in relation with perspectives not only of atmospheric sciences but also public health (e.g. Onishi et al., 2012). Routine forecasts of aeolian dust concentrations over Japan were begun in 2004 (Kinoshita and Maki, 2009) based on a numerical simulation model of the Japan Meteorological Agency: MASINGAR (Tanaka and Chiba, 2005). To improve the performance of dust concentration forecasts and the estimation of climatic effects of dusts in the atmosphere, validation data on dust simulation are necessary not only for atmospheric concentration but also for dust deposition flux.

Dust particles in the atmosphere are deposited onto the earth's surface accompanying precipitation (wet process) or without precipitation (dry process). Continuous multi-year data of deposition flux have been reported for dusts transported from desert areas such as central and eastern Asia (Inoue and Naruse, 1987; Yabuki et al., 2005; Hsu et al., 2009; Sun et al., 2003; Ta et al., 2004; Zhang et al., 1993). However, most such data are unsatisfactory for the validation of modern numerical model simulation of dust transport and deposition because of one or more reasons related to (1) no separation between wet and dry processes, (2) poor time resolution (monthly) of the sampling period, (3) suffering urban local dust contamination, (4) use of scavenging ratios to estimate wet deposition (namely, indirect data), and (5) single point of measurements. These issues will be addressed briefly.

Numerical simulations of atmospheric aerosol particles have been validated by comparison of various atmospheric observation data such as Lidar (Uno et al., 2006) and optical thickness (Takemura et al., 2000). According to results from inter-comparison of dust transport models over Asia (Uno et al., 2006), improving treatments on dust removal is an important issue to make progress on the models. However, measurements of dust deposition are not so simple because of the difficulty of sampling, especially for dry processes (Washington and Wiggs, 2011; Huneeus et al., 2011). Although the time periods of precipitation or non-precipitation can be separated easily using a precipitation sensor, reliable sampling of the amount and size distribution for dry deposition are difficult because of variable collection efficiency depending on the particle size, wind

ACPD
13, 21801–21835, 2013

speed, and the collector shape (Goossens, 2005, 2007). Evaluations and improvements of various dry deposition collectors have been reported, but no perfect method of dry deposition exists (Vallack and Chadwick, 1992; Sow et al., 2006; Goossens and Rajot, 2008).

To compare with model outputs, the time resolution of observations is also a critical issue. Time scales of dust storms as well as precipitation processes are from sub-hourly to daily. Therefore, the dust deposition collection period should have a similar time range that is much shorter than monthly. Another sampling issue is to avoid contributions of local dust contamination. Based on deposition measurements at multiple sites in the northeastern US, Tai et al. (1999) pointed out that urban sites exhibit more coarse and irregular particles (possibly being windblown and anthropogenic sources) than non-urban sites do. Similarly, from observations at Tsukuba, an inland urban location (about 42 km from the nearest coast of the Pacific Ocean) in Japan, Inomata et al. (2009) reported local contamination by dry dust deposition under high wind speed conditions.

Some reports describe studies using scavenging ratios to estimate wet dust deposition flux without collecting precipitation samples (Uematsu et al., 1985; Duce et al., 1991; Hsu et al., 2009). Estimating wet deposition flux from aerosol concentration and precipitation amount with a scavenging ratio is convenient for reducing the labor of precipitation sampling. However, the estimation method includes great uncertainty of assumptions applying a single scavenging ratio and uniform aerosol concentration for a precipitating air column.

Considering all of the points presented above, process separation and multi-year, multi-site deposition data of mineral dusts are expected to be useful to validate numerical simulation models and to improve our knowledge of deposition processes. For this study, we conducted continuous 2.2 yr, weekly wet and dry deposition sampling at six sites in Japan. We present temporal and geographical variations of wet and dry dust deposition and discuss their controlling factors.

ACPD
13, 21801–21835, 2013

2 Observation sites, experiments, and meteorological data

Observation sites were selected to meet various criteria as described above. To collect dust deposition transported by westerly winds to Japan, the best location is the western edge of the Japanese archipelago. Furthermore, to minimize local dust contribution, the sampling sites are better located at non-urban sites near the ocean. In practice, an important factor was the employment of capable assistants who conducted sampler maintenance as well as geographical balance of the sites. Wet and dry dust deposition samples were collected weekly at Sapporo (43.08° N, 141.34° E), Toyama (36.70° N, 137.19° E), Nagoya (35.16° N, 136.97° E), Tottori (35.54° N, 134.21° E), Fukuoka (33.55° N, 130.36° E), and Cape Hedo (26.86° N, 128.25° E, Okinawa, see Fig. 1), using samplers that automatically separate wet and dry deposition, each of which is equipped with a precipitation sensor (US-330H for cold environment with a heater on precipitation funnel and US-330 for warm environment without a heater; Ogasawara Keiki Seisakusho Co., Ltd.). Distances of the sites from the nearest coast were 15 km at Sapporo, 6 km at Toyama, 10 km at Nagoya, 0.7 km at Tottori, 5 km at Fukuoka, and 0.2 km at Cape Hedo. The wet and dry samplers were placed in open areas on the rooftops of buildings, not on the ground, to minimize local soil contamination. To melt solid precipitation during the cold season, a ring heater was used at Sapporo, Toyama, and Tottori. The precipitation sensor detects precipitation at about 0.5 mm h^{-1} and starts a cover to move from a precipitation funnel to a dry deposition collector.

The wet deposition sample water was collected using a plastic funnel (20 cm diameter) and stored in a pre-cleaned wide-mouth 3 L polyethylene bottle in a refrigerator in the sampler. At the time of bottle replacement, the funnel was washed with 50 mL of ultrapure water to ensure that all dust on the funnel surface was transferred. Benzalkonium chloride (Osvan) was added to sample water as a > 0.01 % solution for sterilization.

ACPD
13, 21801–21835, 2013

The dry deposition sample was collected using a pre-cleaned plastic bag (Unipack L-8; Seisan Nipponsha Ltd.) fitted inside a deposition collector (20 cm diameter, 40 cm deep) during the non-precipitating period. The plastic bag was pre-cleaned using 40 mL of ethanol and 300 mL of ultrapure water. Clean plastic gloves were used to fit the plastic bag to the collector. The deposition collector with a plastic bag was used as a "surrogate surface" of dry deposition. As criticized in some reports (e.g. Goossens, 2007), collection of dust particles using this type of collector is not perfect for various size ranges, which potentially distorts the size distribution and collects a lesser amount of dust than an ideal collector. However, changing the plastic bag from the collector is easily done, making them suitable for frequent replacement at many sites and for transport to Nagoya. We used the same collector for all sites, providing at least a uniform sampling condition to study the geographical distribution of dry deposition in Japan.

Wet and dry samples were normally replaced on every Wednesday. The standard collection period was 1 week. In addition to the normal operation, we added event samples collected for shorter (one to several days) intervals according to various forecasts on dust concentration in the atmosphere and precipitation. We used dust forecasts provided by MASINGAR (Tanaka and Chiba, 2005) of JMA (http://www.jma.go.jp/jp/kosafcst/index.html) and CFORS (Uno et al., 2003) of Research Institute for Applied Mechanics/Kyushu University and National Institute of Environmental Studies (http://www-cfors.nies.go.jp/~cfors/index-j.html), and other information at NRL/Monterey Aerosol Page (http://www.nrlmry.navy.mil/aerosol/) and near real-time visibility map overlaid on image from MODIS (Kurosaki at Tottori University, personal communication, 2013). Wet and dry deposition samples were stored at room temperature and were sent to the laboratory at Nagoya University once a month. Deposition sampling was started in October 2008, ending in early January 2011.

In the laboratory, wet deposition samples were filtered through preweighted, 25 mm diameter, 1.0 µm pore-size Nuclepore filters (Whatman Co. Ltd.). For dry deposition samples, the plastic bag was washed with 100 mL (50 mL each) of ultrapure water to ensure that all dust in the bag was transferred. The dry deposition sample water was

ACPD
13, 21801–21835, 2013

similarly filtered through Nuclepore filters (1.0 µm pore-size). After drying the filters at 40 °C for 2 h in an electric oven, we weighed the filters using an electric microbalance (ER182A and GH-252; A&D Co. Ltd.).

The water-insoluble residual weight on the filter includes some biogenic components such as pollens, especially in spring. Mineral dust amounts were estimated from the relation between Fe and mineral dust amount on the filters, assuming Fe constituting 3.7 % of mineral dust weight and that the relation does not vary among seasons or locations in Japan (Ura et al., 2011). Non-destructive X-ray fluorescence (MXF-1T; Kimoto Electric Co. Ltd.) analysis was used to measure Fe contents of the filter residue. Although elemental analysis using XRF provides Al and other elements, Fe was selected for our purposes because of (1) widest dynamic range of calibration response and (2) larger content than Ti that shows also wider dynamic range. A similar result of X-ray attenuation for light elements such as Al and Si was also reported for mineral dust filters by Formenti et al. (2010). Reference materials were used for calibration of Fe content of filter residue, which were provided from Geological Survey of Japan (sediment sample from Lake Biwa: JLK-1, Imai et al., 1996 and marine sediment sample from southern Pacific Ocean: JMS-2, Terashima et al., 2002).

The average Fe content of 3.7 % was calculated from linear correlation between Fe content and non-pollen weight of residue estimated from microscopic data (Ura et al., 2011). The average Fe content of the Earth's upper crust is 3.5 % (Taylor and McLennan, 1995). There might be some variations in the amount of Fe content depending on the size fraction (Cao et al., 2008; Wu et al., 2011) and on the source area (Formenti et al., 2011). Nonetheless, the average Fe content obtained by Ura et al. (2011) is consistent with the values reported for bulk Asian dust events in Korea (3.7 % at Seoul, Choi et al., 2001; 4.2–8.4 % at Seoul, Jeong, 2008) and in China (3.9–4.1 % at Beijing, Feng et al., 2008; 3.0–5.7 % at Beijing, Lue et al., 2010). In our sample treatment, both wet and dry deposition samples were washed with ultrapure water, so the water soluble fraction of Fe in the dust is lost during filtration. However, the fraction of water soluble

ACPD
13, 21801–21835, 2013

Fe is expected to be very small compared to the total Fe content (< 1 % of total Fe for Chinese loess, Schroth et al., 2009).

The dust size distribution was obtained using consecutive four-stage filtration: > 20 µm using a nylon net filter (25 mm diameter; Merck MilliPore), > 10 µm (25 mm diameter, Nuclepore Track-Etched Membranes; Whatman), > 5 µm (25 mm diameter, Nuclepore Track-Etched Membranes; Whatman), and > 1 µm (25 mm diameter, Nuclepore Track-Etched Membranes; Whatman). Before applying four-stage filtration, the original filter with residue was heated at 750 °C in an electric oven for 1 h to remove organic materials. It was gently loosened again by wetting and mixing with ethanol in an ultrasonic bath. Amounts of our filter residue samples were normally small (mostly below several milligrams). Therefore, multistage filtration was practically a best choice for measuring the size distribution. Heavier refractory samples (about > 1 mg) were selected for this filtration. The sample amount of filtration was restricted approximately 1–2 mg at the stage of > 10 µm to prevent clogging pores. By heating at 750 °C, some mineral dusts would be dehydrated and their mineral forms changed, so that the weight and size for dust particles with non-refractory minerals might be decreased after this treatment. Unfortunately, mineral compositions at the time of initial sampling were not known, and weight fractions of four-stage filtration were not corrected for heating effects.

Meteorological data such as precipitation were obtained from Japan Meteorological Agency (JMA) web sites (http://www.data.jma.go.jp/obd/stats/etrn/index.php). The numbers of Aeolian dust (visibility-reducing lithometeor) events at and near the sites were referred from tables at JMA web sites (http://www.data.kishou.go.jp/obs-env/kosahp/kosa_data_index.html). Precipitation data at Oku, the nearest (6 km distant) AMeDAS station, were used for Cape Hedo.

ACPD
13, 21801–21835, 2013

3 Results and discussion

3.1 Temporal variation and special distribution of dust deposition fluxes

Figure 2 shows the wet deposition flux of mineral dusts at six sites during October 2008–December 2010. Time intervals of flux shown in Fig. 2 are depicted with the original sampling duration. Therefore, the horizontal width is narrower for short-term sampling on a dust event predicted by *Kosa* event forecasts. Wet deposition flux was high in spring (February–May) and low in summer (July–September), irrespective of the site location and the year of observation. Temporal variations of wet dust deposition were not always synchronous among sites because of the migration of the precipitation region from west to east and the local effects of winter monsoon (drizzle precipitation) at Tottori, Toyama, and Sapporo. However, nearly simultaneous wet depositions at multiple sites were observed five times (late-January, late-February, mid-March, mid-April, and late-December) in 2009, and five times (mid-March, late-March to early-April, late-April, late-December) in 2010. These simultaneous depositions were roughly divided into two regional patterns as western to central parts and central to northern parts of Japan.

Wet dust deposition was often sporadic. Large wet fluxes were observed in Sapporo, Toyama, and Tottori, where drizzle precipitation was observed during the dusty season. In contrast, no such large sporadic flux was found at Nagoya and Cape Hedo, where drizzle precipitation was observed infrequently during the dusty season. Previously, Osada et al. (2011) reported that a combination of precipitation and *Kosa* events is necessary for the high wet-deposition flux of mineral dust based on an analysis of dust flux measurements, true color MODIS satellite images, and vertical distributions of dust concentration from Lidar observations. To portray this point better geographically, Fig. 3 presents monthly wet deposition flux (red line), monthly number of days of *Kosa* events near the sites (gray vertical bar), and the monthly number percentage of precipitation days (blue line). The winter monsoon from Eurasian continent brings snow or drizzling rain at sites along the Sea of Japan, which leads to high frequency of

ACPD
13, 21801–21835, 2013

precipitation in late fall to early spring (Barry and Chorley, 2003). The number of *Kosa* events was smallest at Sapporo, located in northern Japan. At Toyama, the tendency of monthly variation for wet deposition flux was similar to that of Kosa event days. Days of precipitation at Toyama were more than 60 % during November–April. In contrast, wet dust deposition flux was not so high in May 2010 when days of Kosa events were high (5 days), probably because of the low precipitation frequency (40 %). Therefore, the condition of frequent precipitation (> 60 %) is necessary to bring wet deposition of dusts. A similar tendency was also noted at Tottori. For Nagoya, Fukuoka, and Hedo, a tendency of monthly variation for wet deposition flux was not similar to that of *Kosa* events because of low precipitation frequency for many months, including the dusty season.

Annual wet deposition fluxes are presented in Table 1. Maximum (minimum) annual flux was obtained at Toyama (Hedo). The ratio of maximum to minimum was about 6. As discussed above, places where precipitation was frequent during the dusty season received higher wet deposition of mineral dusts. The geographical distribution of annual wet flux in 2010 was almost the same as that in 2009, with smaller year-to-year variation at Nagoya and Tottori.

Figure 4 shows the dry deposition flux of mineral dusts at four sites during October 2008–December 2010. Dry deposition data at Sapporo and Nagoya were not used here because of the slight contribution of local urban dusts judging from microscopic analysis of filter residue samples, perhaps because of demolition and construction works and deflation of road side dusts near the sites. For other sites, the seasonal tendency of dry deposition fluxes was fundamentally the same as that for wet deposition, namely, high in spring and low in summer. Some sporadic dry deposition events at multiple sites were found three times in 2009 (21 February, 17 March, 19 May) and five times in 2010 (16 March, 21 March, 27 April, 4 May, 12 November; date as the widest distribution of *Kosa* reports based on a map of aeolian dusts by JMA web site). Small peaks around 13–20 May were noticeable at Toyama, Tottori and Fukuoka in May 2009 when no *Kosa* event was observed near the sites. Many other *Kosa* event days were

ACPD
13, 21801–21835, 2013

observed in Japan, as reported by JMA: 11 and 15 February, 25 April, 8 May, 19 October, and 26 December in 2009, and 13 March, 2 April, 21 and 25 May, 3, 11 and 23 December in 2010. However, dry deposition flux for these days was high only at a single site or not notable as a peak at any site, which implies that the air mass during *Kosa* events did not always carry dust particles that can deposit gravitationally on the ground.

The highest dry deposition flux among the sites was obtained at Fukuoka ($1.28\,g\,m^{-2}\,day^{-1}$) during 20–21 March 2010. Although sampling intervals were different, dry deposition fluxes for this event were also the highest at other sites; Toyama, $167\,mg\,m^{-2}\,day^{-1}$ for 20–22 March, Tottori, $169\,mg\,m^{-2}\,day^{-1}$ for 20–23 March, and Cape Hedo, $70\,mg\,m^{-2}\,day^{-1}$ for 17–24 March 2010. Many case studies were reported for this event because atmospheric dust concentrations were impressively high over wide areas in China and Korea including not only the East China Sea but also the South China Sea (Bian et al., 2011; Li et al., 2011; Wang et al., 2011; Lin et al., 2012; Tatarov et al., 2012).

Figure 5 shows monthly values of dry dust deposition flux (red line) and the monthly number of days for *Kosa* events observed near the sites (gray vertical). The annual number of *Kosa* events was higher at Tottori (13 days in 2009, 19 days in 2010) and Fukuoka (8 days in 2009, 19 days in 2010) than at the other sites. Dry deposition fluxes were also higher at Tottori and Fukuoka than at other sites (Table 1). Although the seasonal tendency of monthly dry deposition was roughly similar to that of monthly *Kosa* days, monthly dry deposition does not follow monthly Kosa days in some months, such as May 2010. As listed in Table 1, the ratio of maximum to minimum annual dry fluxes was about 3, which is about half of that for wet deposition. The geographical distribution of annual dry flux in 2010 was almost the same as that in 2009. The average wet/dry ratio was highest at Toyama (3.27) and lowest at Hedo (0.82), suggesting a larger contribution of dry deposition processes at the western and southern sites.

ACPD
13, 21801–21835, 2013

3.2 Dust in the atmosphere and dry deposition

Figure 6 shows the dry deposition flux at Tottori and daily mass concentrations of PM_{10} (red line) and $PM_{2.5}$ (green line; both from Network Center for EANET, 2009, 2011, 2012) at Oki Island locating at upstream (WNW) position, about 120 km from Tottori. The difference between PM_{10} and $PM_{2.5}$ corresponds to coarse particle concentration ($PM_{10} - PM_{2.5} = PM_c$). Mass median aerodynamic diameters of *Kosa* particles in Japan were typically reported at about 4 µm (Ishizaka and Ono, 1982; Mori et al., 2003). Therefore, a higher PM_c concentration implies a *Kosa* event condition. In fact, high dry deposition fluxes were observed mostly on days of high PM_c concentrations such as 17 March 2009 and 21 March 2010. In contrast, dry deposition fluxes were not so high for the periods on 26 December 2009 and 21–22 May 2010 (peaks with asterisks), although high PM_c concentrations were observed. We will discuss details of differences of these events.

Figure 7 shows details of the events in 2009 (upper part) and 2010 (lower part). In both parts, upper panels represent time–height sections of extinction coefficient of dust particles observed by lidar (http://www-lidar.nies.go.jp/index.html.en) at Matsue, middle panels show dry deposition fluxes at Tottori, and lower panels depict $PM_{2.5}$ and PM_{10} at Oki. The extinction coefficient of dust particles was estimated using the method described by Shimizu et al. (2004). Matsue is located about 110 km west from Tottori, as shown in the insert map (blue rectangle) of Fig. 1. As Fig. 7 shows, high extinction coefficients of dust particles were observed at the lower troposphere during the events of high PM_{10} concentrations near the ground level. Whereas the maximum heights of dust layers for high dust deposition fluxes with high PM_{10} events in March were both higher than 2 km, those for low dust deposition fluxes with high PM_{10} events on 26 December 2009 and 21 May 2010 were below 2 km. For the events of mid-March and end-December of 2009, a similar tendency of time–height dust data was widely observed at other windward lidar sites such as Beijing, Seoul, and Fukue (Sugimoto et al., 2013), suggesting that dust particles of these events were transported through

ACPD
13, 21801–21835, 2013

thick (> 2 km) and thin (< 2 km) layers, respectively, for a long distance. The correspondence of transport height and dry deposition flux imply that the maximum height or thickness of a dust layer is an important restriction factor for the long-range transport of dust particles.

3.3 Giant dust particles and geographical distribution of dry deposition

Figures 8 and 9 respectively show weight fractions of refractory dust particles for wet and dry deposition samples. Events shown in Figs. 8 and 9 were selected for samples of high refractory weight (about > 1 mg). Sampling sites in the figures were depicted from west (Fukuoka) to east (Tottori and Toyama), standing at shorter (left) to longer (right) distance from the source areas. Giant fractions (the sum of > 20 µm and 10–20 µm) were higher than 0.5 for the most of event samples. Weight fractions of 1–10 µm were somewhat larger at Toyama, especially for wet deposition samples. Although proportions of giant fractions for dry deposition were slightly larger than those for wet deposition, the proportional tendency of weight fraction on dry deposition samples was similar to that of wet deposition, implying that our dry deposition samplers might collect dust particles over a wide size range. Irrespective of deposition type, the giant fractions were generally decreasing with traveling to the east or increasing distance from the source area, suggesting depletion of giant particles during transport. Minor exception for the spatial tendency was the dry deposition event on 15–17 March 2010, which might be attributable to a shift of main dust stream toward Tottori according to lower visibility at Tottori rather than Fukuoka.

Shifting the size fraction of dust particles is attributable to differences in settling velocity with size. For a dust event in March 2001, Mori et al. (2003) reported aerodynamic mass size distributions sampled at Beijing, China, and at Yamaguchi, 1500 km downwind from Beijing, western Japan. Although the major peak of aerodynamic mass size distribution at Beijing was found at 4.7–7.0 µm, the peak in coarse mode at Yamaguchi was 3.3–4.7 µm. Assuming that the shift was attributed to solely dry processes, larger and heavier particles can be expected to be deposited earlier during their transport.

ACPD
13, 21801–21835, 2013

The modal diameter of the mass size distribution of deposited dust particles is expected to be much greater than the modal diameter of dust particles in the atmosphere (about ×4 as reported by Schneider et al., 1990).

The gravitational settling velocity of a spherical particle for physical diameter of 10 µm with density of 2.6 g cm^{-3} is estimated as about 685 m day^{-1} (Hinds, 1999). Because the distance from Chinese and Mongolian deserts to Japan is about 2500 km, the transport time of dust particles from the source areas is expected to be about 1–3 days, depending on meteorological conditions such as the injection height of dust particles and wind speed at the dust layers. Larger particles have higher settling velocity. Therefore, the abundance of such particles in the air would decrease quickly with transport time from the source area. Assuming the injection height of dust particles as 2 km a.s.l. and assuming that the wind speed at the dust layer as 10 m s^{-1}, giant particles of 10 µm with density of 2.6 g cm^{-3} would be mostly depleted by the time they reach Japan. In other words, a thicker or higher (> 2 km) dust layer is necessary to transport giant particles to distant areas. This inference is consistent with the observation of high deposition fluxes under thick dust layer conditions in March 2009 and 2010.

Based on monthly and seasonal sampling of atmospheric dust deposition at six sites on the Chinese Loess Plateau, Sun et al. (2003) reported that modal diameters of deposited dusts were 20–30 µm at all sites. Their sampling sites were close to major source deserts or were potentially located within the area of re-suspension of deposited dust. Therefore, modal diameters in their reports were much greater than in our results. Size measurements of dust particles in dusty snow collected at Mt. Tateyama, central Japan showed modal diameters of about 6–21 µm (Osada et al., 2004). Sampling conditions of spring snow at Mt. Tateyama are suitable to avoid local dust contamination. Therefore, their results are regarded as a good reference of the dust size distribution over Japan. Weight fractions in the present study are consistent with size data of snow at Mt. Tateyama. Giant mineral particles were found in the central Pacific near Oahu Island as a result of long-range transport of more than 10 000 km from Asian source regions (Betzer et al., 1988). Although their findings were striking in terms of distance

ACPD
13, 21801–21835, 2013

(> 4 fold compared to our cases) and size (> 75 µm) of dust particles transported, our data show the frequent transport of giant dust particles over Japan, suggesting that transport of giant dust particles is important for the total mass flux to remote ocean, especially in the North Pacific, which is known as an HNLC region. Large-scale transport models have difficulty reproducing the transport of large dust particles (Zendar et al., 2004). Therefore, data related to the frequent transport of giant dust particles toward this region are expected to be invaluable to ascertain actual dust effects on biogeochemical cycles as well as their climatic significance.

4 Summary and conclusions

Weekly wet and dry deposition samples at six sites in Japan were obtained during October 2008–December 2010 to study temporal variations and spatial distributions of mineral dust deposition fluxes. The sampling sites were chosen to cover the area from northeast to southwest: Sapporo, Toyama, Nagoya, Tottori, Fukuoka, and Cape Hedo (Okinawa) in Japan. We used automatic wet/dry separating samplers and Fe content of filter residues to estimate mineral dust amounts.

Wet and dry deposition fluxes of mineral dusts were both high in spring and low in summer, showing similar seasonal variations to frequency of visibility-reducing aeolian dust events (*Kosa*) in Japan. For wet deposition, higher annual deposition fluxes were observed at Toyama and Tottori, where frequent precipitation (> 60 % per month) was observed during dusty seasons. Annual wet deposition fluxes at Cape Hedo and Nagoya were much smaller because of infrequent precipitation in dusty season. For dry deposition among Toyama, Tottori, Fukuoka, and Cape Hedo, annual dry deposition fluxes were highest at Fukuoka and lowest at Cape Hedo. The average ratio of wet/dry deposition fluxes was the highest at Toyama (3.3) and the lowest at Hedo (0.82), indicating a larger contribution of the dry process at western sites, probably because of (1) distance from desert areas and relative location to main stream of dust

ACPD
13, 21801–21835, 2013

transport, and (2) effectiveness of the wet process such as frequent precipitation during the dusty season.

Temporal variations of dry deposition fluxes were compared with the frequency of Kosa events and atmospheric concentrations of coarse particles (PM_c). Although higher dry deposition fluxes were generally obtained during frequent *Kosa* events with higher PM_c, fluxes for some events were not so high during *Kosa* events. Examination of the vertical distribution of dust particles indicated by Lidar suggests that transport through the shallow (about < 2 km) dust layer engendered low dry deposition of dust, although high PM_c concentrations were observed.

Refractory parts of deposited dust particles were size-segregated using multiple filters: > 20, > 10, > 5, and > 1 µm diameter. Weight fractions of giant dust (the sum of > 20 µm and 10–20 µm) were higher than 50 % for the most heavy event samples (> 1 mg of refractory weight). Irrespective of deposition type, the giant fractions were generally decreasing with increasing distance from the source area, suggesting the selective depletion of larger giant particles during transport. Combined with the relation of dry deposition flux, dust layer thickness in the atmosphere, and dust size information, transport through a thicker (> 2 km) dust layer would be able to carry more giant dust particles to Japan. Because giant dust particles are important mass fractions of dust accumulation, especially in the north Pacific, precise simulation of giant particles in the atmosphere is necessary for the study of dust budgets through the atmosphere, their climatic effects and their roles in the biogeochemical cycle.

Acknowledgements. We are grateful to Y. Takeda in Cape Hedo, H. Sasaki in Sapporo, T. Nakayama and Y. Kobayashi in Toyama, T. Maruyama and A. Naganuma in Fukuoka, A. Kato and A. Abulaiti in Tottori, K. Kawakita and S. Ueda in Nagoya for helping with sample collection of dust deposition. We also thank Y. Hara at Kyusyu University for helpful discussions. Data of PM_{10} and $PM_{2.5}$ at Oki were provided by Acid Deposition Monitoring Network in East Asia (EANET) (http://www.eanet.asia/product/index.html). This research was financially supported by Grants-in-Aid for Scientific Research (A) (20244078) and (B) (20310009), and a Grant-in-Aid for Scientific Research in Priority Areas, Grant No. 18067005 (W-PASS), provided by the Ministry of Education, Culture, Sports, Science and Technology, Japan. This research is

ACPD
13, 21801–21835, 2013

a contribution of IGBP/SOLAS activity. This research was also supported by the Environment Research and Technology Development Fund (5B-1202) of the Ministry of the Environment, Japan.

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13, 21801–21835, 2013

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ACPD
13, 21801–21835, 2013

Table 1. Annual deposition flux ($g\,m^{-2}\,yr^{-1}$) of mineral dust.

	Wet			Dry			Wet/dry ratio		
	2009	2010	AVG	2009	2010	AVG	2009	2010	AVG
Sapporo	4.12	5.36	4.74						
Toyama	8.66	10.62	9.64	2.77	3.12	2.95	3.12	3.40	3.27
Nagoya	2.44	2.06	2.25						
Tottori	5.11	5.93	5.52	3.22	4.86	4.04	1.58	1.22	1.36
Fukuoka	3.96	5.46	4.71	4.73	5.65	5.19	0.84	0.97	0.91
Cape Hedo	1.14	2.16	1.65	1.51	2.50	2.00	0.76	0.86	0.82

ACPD
13, 21801–21835, 2013

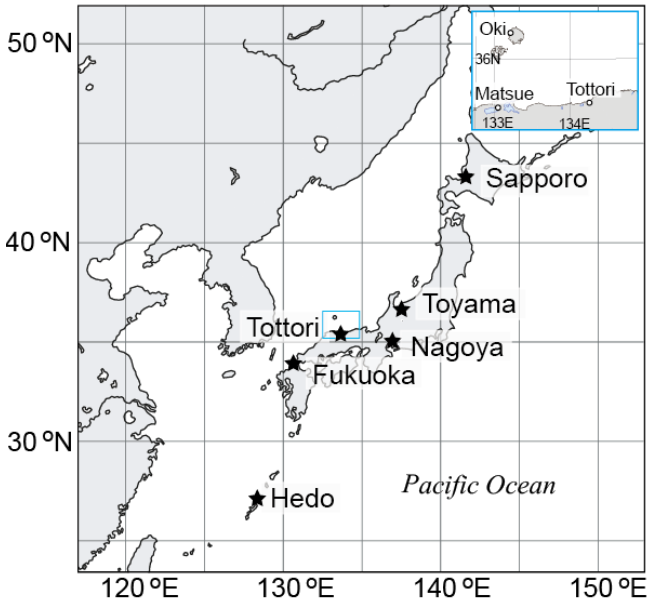

Fig. 1. Map of observation sites. The blue rectangle in the upper right corner is an enlargement of the map near Tottori, showing positions of Oki Island (PM_{10} and $PM_{2.5}$ measurements by EANET) and Matsue (Lidar observation by NIES).

ACPD
13, 21801–21835, 2013

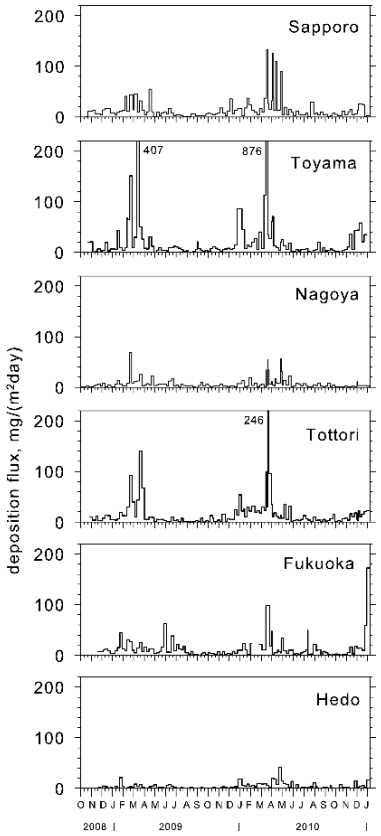

Fig. 2. Wet deposition flux (mg m^{-2} day^{-1}) of mineral dust during October 2008–December 2010.

ACPD
13, 21801–21835, 2013

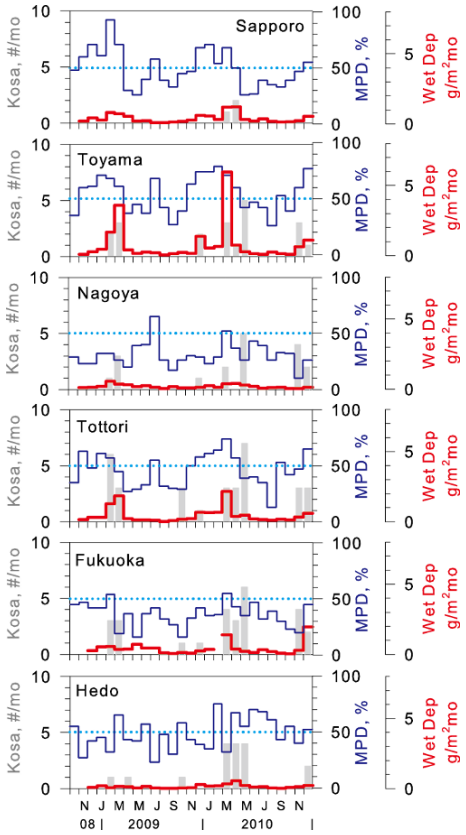

Fig. 3. Monthly numbers of aeolian dust event (*Kosa*) days near the sites (gray), monthly wet deposition fluxes (red), and monthly precipitation days in percent (blue). Horizontal dotted light blue line depicts 50 % of monthly precipitation days.

ACPD
13, 21801–21835, 2013

Wet and dry deposition of mineral dust particles in Japan

K. Osada et al.

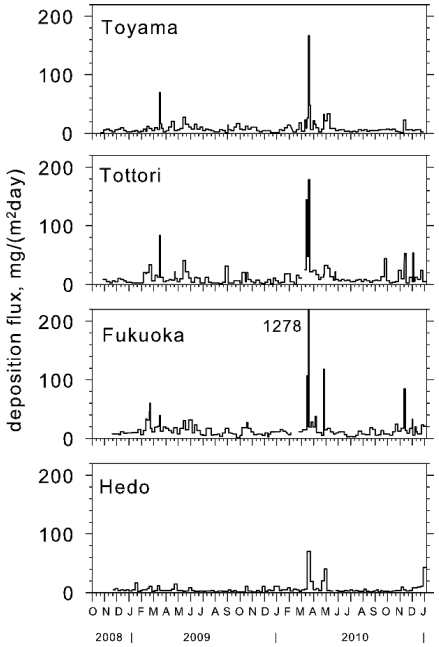

Fig. 4. Dry deposition flux ($mg\,m^{-2}\,day^{-1}$) of mineral dust during October 2008–December 2010.

ACPD
13, 21801–21835, 2013

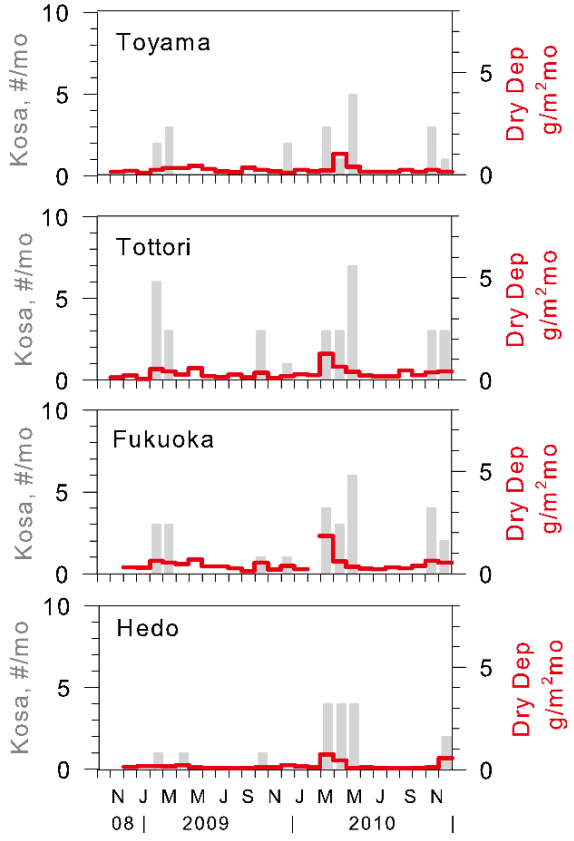

Fig. 5. Monthly numbers of *Kosa* days near the sites (gray) and monthly dry deposition fluxes (red).

ACPD
13, 21801–21835, 2013

Wet and dry deposition of mineral dust particles in Japan

K. Osada et al.

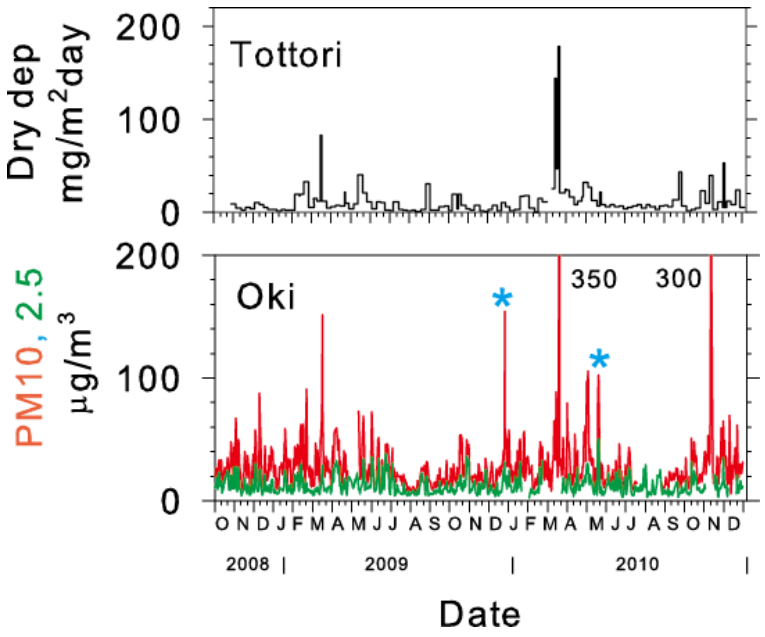

Fig. 6. Upper panel, dry deposition flux (mg m^{-2} day^{-1}) of mineral dust at Tottori; lower panel, daily average concentrations of PM_{10} and $PM_{2.5}$ at Oki Island (EANET). Blue asterisks denote days of high PM_{10} concentrations but with low dry deposition flux.

ACPD
13, 21801–21835, 2013

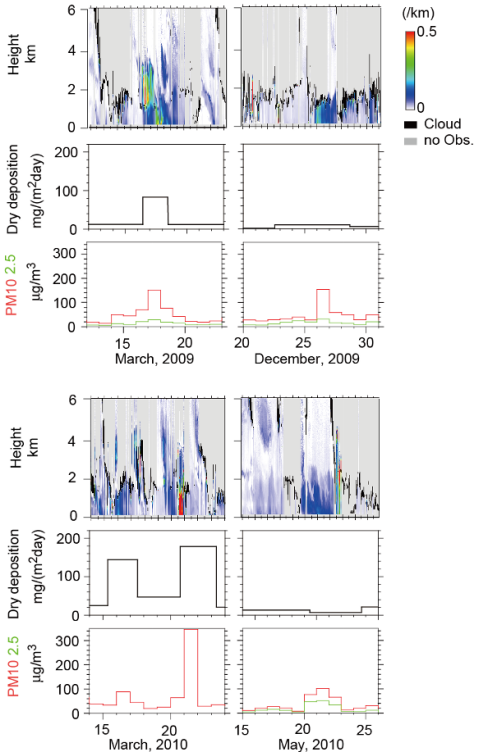

Fig. 7. Comparison of high PM_{10} events with high (left) and low (right) dry deposition events in 2009 (upper part) and 2010 (lower part). Upper panels of the year show time–height sections of extinction coefficient of dust particles based on lidar measurements at Matsue (NIES web site: http://www-lidar.nies.go.jp/index.html.en). Black and gray shading respectively represent clouds and height ranges of no data. Horizontal black lines in middle panels represent dry deposition flux of dusts at Tottori. Red and green horizontal lines in lower panels show PM_{10} and $PM_{2.5}$ concentrations at Oki Island (EANET).

ACPD
13, 21801–21835, 2013

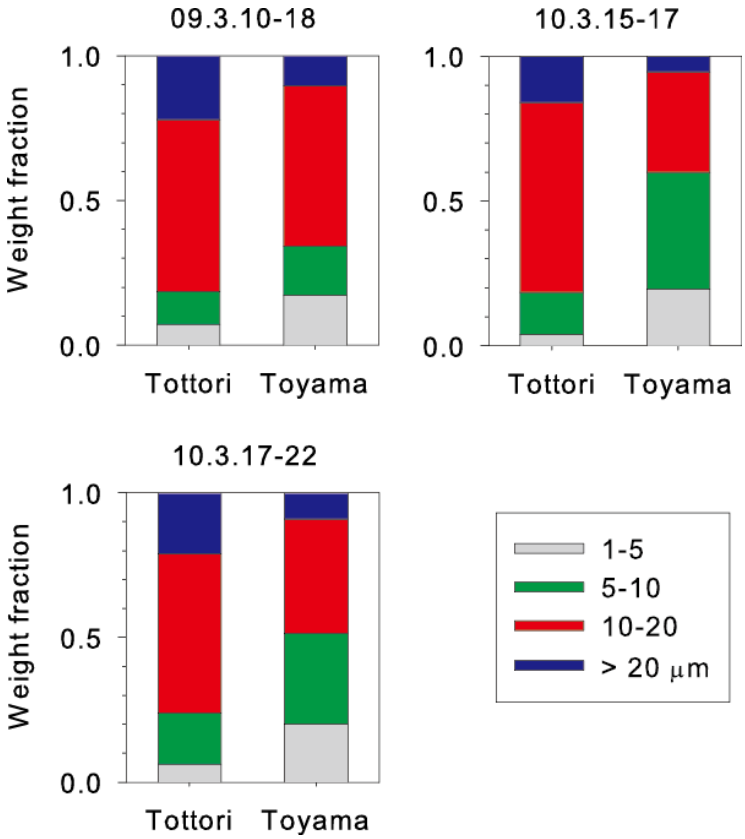

Fig. 8. Weight fraction of size-segregated refractory dust deposited by wet processes. Sampling periods are shown at the top of each panel as yy.mm.dd-dd.

ACPD
13, 21801–21835, 2013

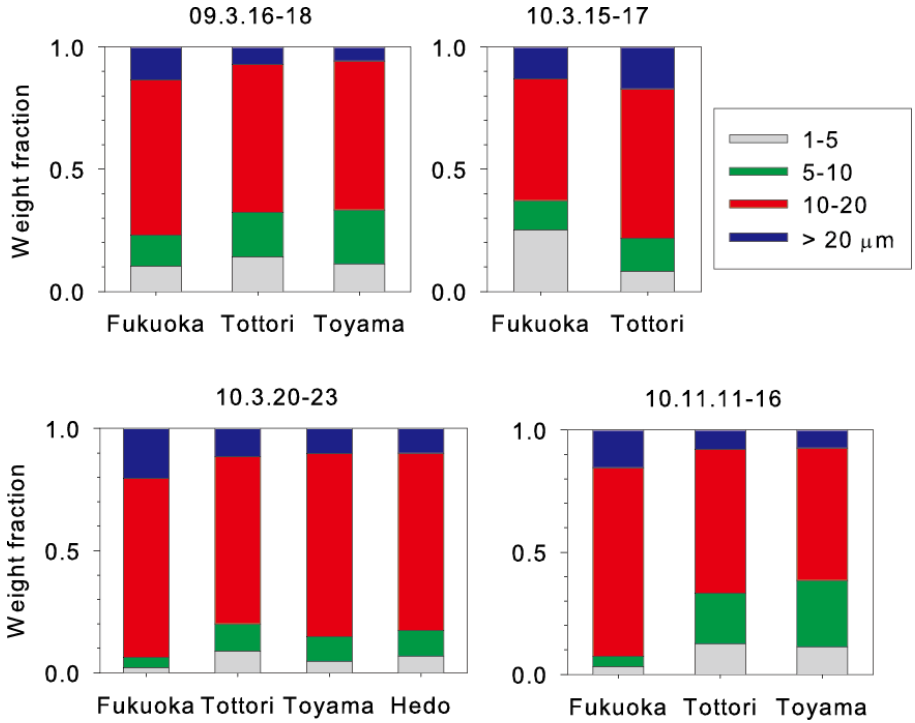

Fig. 9. Weight fraction of size-segregated refractory dust deposited by dry processes Sampling periods were indicated at the top of each panel as yy.mm.dd-dd.

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13, 21801–21835, 2013