Seasonal variation of polarization properties of tropospheric aerosols over western Japan based on polarization optical particle counter (POPC) measurement

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1. Abstract

Ground-based observation of the polarization properties of aerosol particles using a polarization optical particle counter (POPC) was carried out from October 2013 to January 2015 at a suburban site in the Kyushu area, Japan. Based on the long-term POPC and aerosol composition measurements, we successfully classified the size-dependent depolarization characteristics for three typical aerosol types (anthropogenic pollutants, dust, and sea salt). We found that a depolarization ratio value (the fraction of s-polarized signal in the total backward light scattering signal) of 0.1 could be used as a reliable threshold to classify the sphericity/non-sphericity for supermicron particles. Depolarization ratio of submicron particles during anthropogenic pollutants dominant periods varied between 0 and 0.2. We confirmed that air masses over the Northern China contained large amounts of non-spherical particles (i.e. dust) and anthropogenic pollutants, which could impact air quality in western Japan, especially in winter and spring.

2. Instrumentation

The light-polarization property of suspended aerosol particles (with particle diameter, Dp < 10 μm) was measured using a newly developed polarization optical particle counter (POPC; YGK Corp., Japan), located on a two-story building at Kyushu University (130.5°E, 33.5°N). We installed a 3 m-long vertical stainless steel tube through the roof of the building, and the ambient air was drawn into the room with a laminar flow rate of 13 L/min. The loss of coarse mode particles (Dp > 2.5 μm) due to gravity settling was negligible. The POPC uses a 780 nm linearly polarized laser source, and measures both forward scattering and backward scattering intensities at 60 degrees and 120 degrees, relative to the direction of incident light. The polarization direction of the incident laser was set parallel to the plane of the scattering angle, and the acceptance angle for the polarization detector was set to 45 degrees. This configuration was optimized to reduce uncertainties in optical measurements [*Kobayashi et al.*, 2014]. The size of particles was determined from the forward scattering intensity, and two polarized compounds (*s*-polarized/*p*-polarized, polarization direction perpendicular/parallel to the plane of the scattering angle) of backward scattering were measured simultaneously. In practice, the depolarization ratio (DR; the fraction of *s*-polarized to the total backward scattering, [$S/(S+P)$]) was a good indicator for particle sphericity because the direction of polarization of scattered light for spherical particles is identical to that of the incident light, while this is not the case for non-spherical particles (e.g., dust).

3. Results and discussions

The daily-averaged volume size distribution and size-resolved mode depolarization ratio (MDR; *Pan et al., 2015*) of aerosol particles measured at the observation site are shown in Figure 1. Volume concentrations of ambient particles had two pronounced peaks, in the submicron and/or coarse mode size ranges, particularly in winter and spring. The peak in the submicron range was related to anthropogenic pollution, while the coarse mode peak (6–8 μm in size) was attributed to mineral dust. The two peaks may occur concurrently or sequentially. In summer and autumn, the volume concentration of particles in both fine and coarse modes decreased significantly, firstly, because the air masses mostly originated from the west Pacific ocean, where anthropogenic emissions were limited; and secondly, because wet scavenging processes were significant due to frequent precipitation.

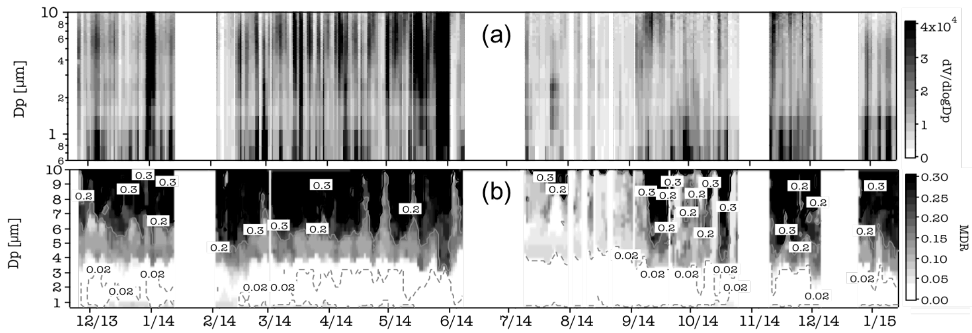

Figure 1 Time series of volume size distributions (a) and MDRs (b) of ambient particles at the observation site.

MDRs of aerosols showed obvious seasonal variation. In spring and winter, MDRs increased with increasing particle size. In general, particles with 1 < Dp < 2.5 μm normally had small MDR values, while particles with Dp > 5 μm had MDRs larger than 0.2. In summer, particles in the coarse mode also had relatively smaller MDRs (Figure 1d). This was mostly due to air masses that originated from clean marine regions, consisted of large amounts of sea salt aerosols, which typically have spherical morphology under high-humidity.

Monthly-averaged mass concentrations of $PM_{2.5}$ levels show a clear winter-spring high and summer low. In contrast, $PM_{2.5\text{-}10}$ mass had a pronounced peak in May, reflecting frequent dust events. Numbers of substandard days that daily-averaged mass concentrations of $PM_{2.5}$ exceeded the NAAQS of Japan (35 μg/m^3) were highest in spring. In particular, during May 2014, 12 days failed to meet the NAAQS; mineral dust was responsible for about 70% of these substandard days, followed by other mixed pollution types (20%). In March 2014, mixed pollution and dust dominated, accounting for 60% and 30% of substandard days, respectively. Mixed pollution types clearly had a maximum contribution to substandard air quality in the winter, and a minimum in autumn. This is because air masses pass through arid/semi-arid polluted regions in North China (see back trajectories in supporting materials) in winter, and during such LRT, dust particles and anthropogenic pollutants mix. In warmer seasons, only a few substandard days were related to anthropogenic pollution.

Size-resolved depolarization characteristics of aerosols from different origins were evaluated in combination with backward trajectory analysis. The footprint regions for anthropogenic pollutants were determined based on ensemble simulations of 5 d backward trajectories of air parcels using HYSPLIT (v4.9; http://ready.arl.noaa.gov/HYSPLIT.php), offsetting the release point by a meteorological grid point in the horizontal, and 0.01 sigma units in the vertical. We divided the region of interest into four sub-regions, where emission flux of anthropogenic pollutants and their corresponding aerosol types were easily distinguished. When the air mass came from Region I (Figure 2a), which mostly covered the Sea of Japan and Japanese landmass, the DR vs. Dp plot showed one predominant peak at MDR = 0.05 and Dp = 1.8 μm (Figure 2e).

This suggested that spherical particles (e.g., sea salt) in the fine mode comprised most of the aerosol. Only 2% of this period was found to be substandard. When the air mass came from Region II (covering the polluted region of North China, Figure 2b), volume concentrations of particles in all size bins increased significantly, and volume size distributions of particles had three distinct modes at Dp = 0.7 μm (MDR = 0.1), Dp = 1.9 μm (MDR = 0.08) and Dp = 5 μm (MDR = 0.35), respectively (Figure 2f). The volume peaks in the submicron and coarse modes could be attributed to LRT of anthropogenic pollutants and mineral dust. The volume peak at Dp = 1.9 μm was associated with particles that were not spherical, in light of their large MDR values (from 0.08, up to 0.4), in contrast to particles from Region I. We surmised that these particles were a mixture of both anthropogenic and dust particles. This was also supported by our observations during a dust episode, which demonstrated that the DR values of supermicron dust particles are reduced when mixed with anthropogenic pollutants [*Pan et al.*, 2015]. We found that 32% of the days, dominated by northwesterly winds, were substandard, i.e., anthropogenic pollution, dust and mixed particle types contributed to 13%, 8% and 11% of particles, respectively. When air mass came from Region III (East China Sea, Figure 2c), 92% of these days met the air quality standard for Japan, and substandard days were mostly related to anthropogenic pollution (9%). The MDR value (0.03) for supermicron particles was very small (Figure 2g), indicating a high degree of sphericity. Air masses from Region VI (West Pacific ocean, Figure 2d) also contained a large volume fraction of supermicron particles, with small MDR values (~0.03). We found that the number of substandard days (16%), when air masses came from Region VI, were more frequent than for Region III, reflecting the mixing of maritime air with local anthropogenic emissions over western part of Japan (Figure 2h).

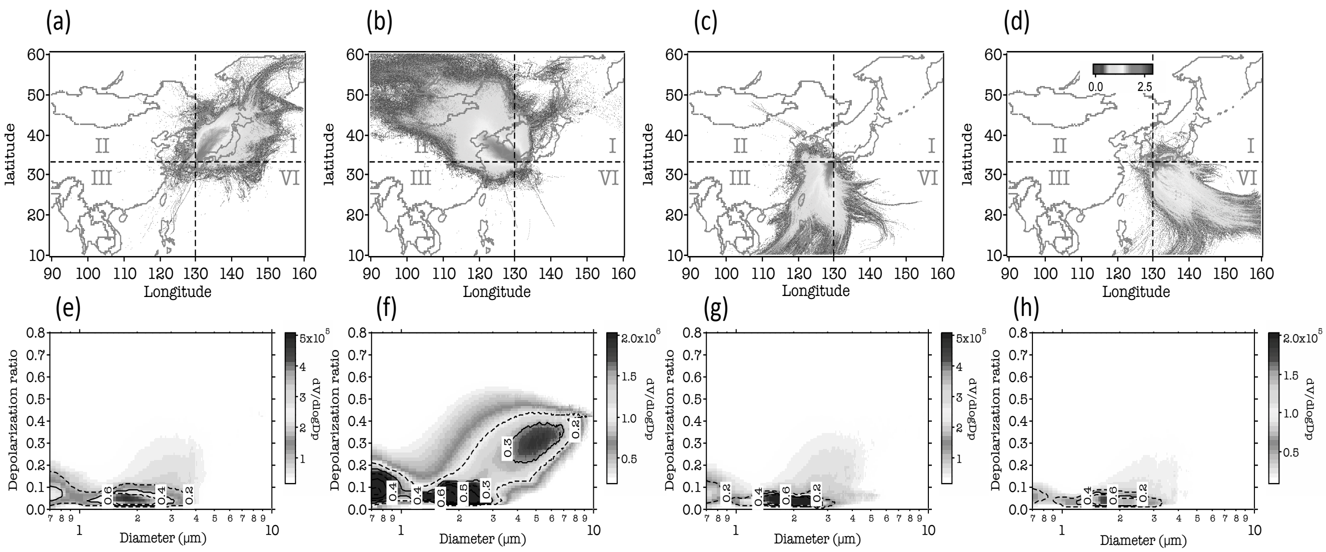

Figure 2 Air masses originating from different regions (a, b, c, d), and the corresponding size-resolved depolarization characteristics of their transported particles (e, f, g, h). The color scale (upper panel) represents the logarithm of the total number of trajectories that passed through the mixing layer of the target grid.

4. References

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