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Vertically-resolved profiles of mass concentrations and particle backscatter coefficients of Asian dust plumes derived from lidar observations of silicon dioxide

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HIGHLIGHTS

- A method for mass concentration profiles of mineral (desert) dust is developed.
- Dust backscatter coefficients are retrieved by the dust mass and the OPAC.
- The Asian dust plumes were separated into pure dust and pollution particles.

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ABSTRACT

This study presents a method to retrieve vertically-resolved profiles of dust mass concentrations by analyzing Raman lidar signals of silicon dioxide (quartz) at 546 nm. The observed particle plumes consisted of mixtures of East Asian dust with anthropogenic pollution. Our method for the first time allows for extracting the contribution of the aerosol component "pure dust" contained in the aerosol type "polluted dust". We also propose a method that uses OPAC (Optical Properties of Aerosols and Clouds) and the mass concentrations profiles of dust in order to derive profiles of backscatter coefficients of pure dust in mixed dust/pollution plumes. The mass concentration of silicon dioxide (quartz) in the atmosphere can be estimated from the backscatter coefficient of quartz. The mass concentration of dust is estimated by the weight percentage (38–77%) of mineral quartz in Asian dust. The retrieved dust mass concentrations are classified into water soluble, nucleation, accumulation, mineral-transported and coarse mode according to OPAC. The mass mixing ratio of 0.018, 0.033, 0.747, 0.130 and 0.072, respectively, is used. Dust extinction coefficients at 550 nm were calculated by using OPAC and prescribed number concentrations for each of the 5 components. Dust backscatter coefficients were calculated from the dust extinction coefficients on the basis of a lidar ratio of 45 ± 3 sr at 532 nm. We present results of quartz-Raman measurements carried out on the campus of the Gwangju Institute of Science and Technology (35.10°N, 126.53°E) on 15, 16, and 21 March 2010.

1. Introduction

Asian dust is a dominant aerosol component in spring in East Asia. Wind lifts large amounts of dust up into the air, causing serious air pollution in the local environment. Asian dust affects not only local air quality, but also local radiative forcing (Huang et al., 2008). Recently, special attention has been paid to understanding the effect of dust aerosols on regional and global climate (Huebert et al., 2003; Nakajima and Yoon, 2005; Mikami et al., 2006; Tesche et al., 2011). The impact of long-range transport of dust and air pollution from their continental sources over oceanic regions is one of the outstanding problems in understanding regional and global climate change (Ramanathan et al., 2001, 2007).

Recently, as the result of increasing industrialization, East Asia has experienced a significant increase of pollution levels from anthropogenic activities, such as emissions from industrial and agricultural sources (Ohara et al., 2007). Organic matter, soot, and dust emitted across the East Asian region make up for one-fourth to one-third of the total global emissions of aerosol pollution (Chin et al., 2002; Ginoux et al., 2004).

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Asian dust particles, which originate from desert areas in the Asian continent, can be mixed with polluted aerosols that contain black-carbon and/or smoke particles while they are transported over industrial regions (Ramanathan et al., 2001). This mixture of dust particles with anthropogenic particles causes changes in optical properties of dust layers (Kim et al., 2004a,b). Noh et al. (2012a,b) demonstrated that the increased radiative forcing exerted by East Asian dust plumes can be largely attributed to the presence of highly light-absorbing anthropogenic particles that are mixed in dust layers. However, the vertical structure and the degree of vertical mixing between dust and pollution layers during transport are poorly understood, primarily because of the lack of vertically resolved observations of aerosol pollution. There remains the need to understand the radiative effect of Asian dust and separate it from the radiative effect of anthropogenic particles within a dust layer.

Dust mass concentration is another important quantity if we want to estimate regional air quality. Studies like the ones carried out by, e.g., Wang et al. (2004), and Liu et al. (2013) show the importance of the impact of dust on air quality in China. However, these studies did not involve research on the vertical distribution of mass concentrations of dust because the measurements were performed by the in-situ instrument at the surface and/or 80 m tower. The vertical distribution of dust can provide evidence for long-range transport of dust across the Pacific Ocean (Edwards et al., 2004). The vertical distribution of dust however is also a critical factor that impacts the effect of dust on radiative forcing and climate (Won et al., 2004; Zhu et al., 2007; Forster et al., 2007; Noh et al., 2012a,b; 2014). Meloni et al. (2005) reported that the vertical distributions and optical properties of Asian aerosols are large sources of uncertainty when it comes to estimating aerosol radiative forcing.

Lidar is a powerful instrument for studying optical properties of dust aerosols. Lidar measurements have been utilized to obtain vertically resolved optical properties of Asian dust particles during the past 30 years (Iwasaka et al., 1988; Shimizu et al., 2004). Dust particles can be identified by lidar measurements of the linear particle depolarization ratio (Murayama et al., 1999; Sakai et al., 2000; Shimizu et al., 2004; Noh et al., 2012a; Noh, 2014). However, direct observations of the mass concentrations of dust using lidar have just recently started since it became possible to measure directly Raman signals of silicon dioxide (quartz) which acts as tracer for mineral dust (Müller et al., 2010; Tatarov et al., 2011).

This study presents vertically-resolved dust mass concentrations that follow from measurements of quartz Raman signal at 546 nm. The measurements we done in mixed Asian dust plumes. We also separated the contribution of mineral dust from the contribution of anthropogenic particles in these mixed-dust layers. We retrieved profiles of backscatter coefficients of dust by using profiles of mass concentrations of dust and OPAC (Optical Properties of Aerosols and Clouds), see Hess et al. (1998).

The paper is organized as follows. Section 2 describes the GIST (Gwangju Institute of Science and Technology) multi-wavelength quartz-Raman lidar and the methodology used to retrieve the vertically-resolved mass concentrations and backscatter coefficients of dust. Section 3 presents measurement examples. In Section 4, the results are compared with depolarization ratios measurements of dust. Section 5 contains a summary and conclusions.

2. Methodology

2.1. Quartz-Raman signal acquisition

The quartz-Raman signals were acquired by our multi-wavelength quartz-Raman lidar system which is located on the campus of the Gwangju Institute of Science and Technology (GIST), 35.10°N, 126.53°E, in Gwangju, South Korea. The GIST multi-wavelength Raman lidar is operational since 2004 (Noh et al., 2007, 2008, 2009, 2011, 2013a,b, 2014) and since then has extensively been used for vertical profiling of optical properties of East Asian dust and pollution, as for example the linear volume depolarization ratio at 532 nm, extinction coefficients at 355 and 532 nm, backscatter coefficients at 355, 532, and 1064 nm, and the extinction-to-backscatter ratio (lidar ratio) at 355 and 532 nm.

Since 2009 the system has undergone significant hardware upgrades including the installation of *quartz-Raman channels* that detect quartz Raman signals at 361 and 546 nm (Müller et al., 2010; Tatarov et al., 2011). Quartz is one main component of dust particles. The measurement technology was first applied using a high spectral resolution lidar (Tatarov and Sugimoto, 2005). The quartz channel setup used in our multi-wavelength quartz-Raman lidar system, the used optical components, the analysis method of the quartz-Raman backscatter signals, and details of the operation mode are described by Tatarov et al. (2011).

2.2. Retrieval of vertical resolved dust mass concentration

The mass concentration of quartz in the atmosphere can be estimated from the Raman backscatter coefficient of quartz (Tatarov and Sugimoto, 2005; Tatarov et al., 2011). The Raman backscatter coefficient is connected to the Raman backscatter differential cross section ($d\sigma(\lambda_L, \lambda_R, \pi)/d\Omega$)) of quartz and the number concentration of quartz molecules (N_q). This relation is defined as:

$$\beta_R(r, \lambda_L, \lambda_R) = N_q(r)\frac{d\sigma(\lambda_L, \lambda_R, \pi)}{d\Omega} \tag{1}$$

Eq. (1) allows us to estimate the number concentration N_q, if the differential cross-section is known. The mass concentration of mineral quartz is then obtained by multiplying N_q with the molecular mass of quartz.

Quartz is one of the main components of dust. If the weight percentage of quartz in Asian dust is known, we can take the next step and estimate the dust concentration. The weight percentage of quartz, measured in Asian dust on the basis of elemental and mineral composition analyses at source regions and in the far field after long-range transport of dust varies between 38% and 77% (Ivanov et al., 1989; Ma et al., 2001; Feng et al., 2002; Ganzei and Razzhigaeva, 2006; He et al., 2013). We did not measure the weight percentage of quartz in dust in this contribution.. We used these minimum and maximum values for our computations of the dust mass concentration.

2.3. Retrieval of dust backscatter coefficient

Dust backscatter coefficients are retrieved from dust concentrations according to the method described in Section 2.2. The retrieval procedure includes the use of the software package OPAC (Hess et al., 1998). OPAC provides optical particle properties in the solar and terrestrial spectral range. OPAC separates dust in the components denoted as *water soluble*, *nucleation*, *accumulation*, and *coarse mode*, and the component *mineral transported*. The *mineral transported* component is used to describe desert dust that is transported over long distances, where by large particles are removed by gravitational settling (Hess et al., 1998). The mass mixing ratios of the components *water soluble*, *nucleation*, *accumulation*, and *coarse mode* are 0.018, 0.033, 0.747 and 0.201, respectively.

Although the entrainment of dust particles into the air is initiated by saltation of sand-sized dust particles, only particles with sizes less than 10 μm diameter reside in the atmosphere long enough to significant distances downwind (Zender et al., 2003). Maring et al. (2003) report that mineral dust particles with diameter above 7.3 μm were preferentially removed by gravitational

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