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Detection of internally mixed Asian dust with air pollution aerosols using a polarization optical particle counter and a polarization-sensitive two-wavelength lidar

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ABSTRACT

East Asia is a unique region where mineral dust (Asian dust) sources are located near urban and industrial areas. Asian dust is often mixed with air pollution aerosols during transportation. It is important to understand the mixing states of Asian dust and other aerosols, because the effects on the environment and human health differ depending on the mixing state. We studied the mixing states of Asian dust using a polarization particle counter (POPC) that measures the forward scattering and the two polarization components of backscattering for single particles and a polarization-sensitive (532 nm) two-wavelength (1064 nm and 532 nm) lidar. We conducted the simultaneous observations using the POPC and the lidar in Seoul from March to December 2013 and captured the characteristics of pure Asian dust and internally mixed polluted Asian dust. POPC measurements indicated that the density of large particles was lower in polluted Asian dust that transported slowly over the polluted areas than in pure Asian dust that transported quickly from the dust source region. Moreover, the backscattering depolarization ratio was smaller for all particle sizes in polluted dust. The optical characteristics measured using the lidar were consistent with the POPC measurements. The backscattering color ratio of polluted dust was comparable to that of pure dust, but the depolarization ratio was lower for polluted dust. In addition, coarse non-spherical particles (Asian dust) almost always existed in the background, and the depolarization ratio had seasonal variation with a lower depolarization ratio in the summer. These results suggest background Asian dust particles are internally mixed in the summer.

1. Introduction

East Asia has the highest concentration of anthropogenic aerosols in the world [1]. It is also a unique region where mineral dust (Asian dust) sources are located near urban and industrial areas. Asian dust is often mixed with air pollution aerosols during transportation. To study Asian dust and air pollution in East Asia, we conducted continuous observation of aerosols in East Asia using a network of ground-based lidars since 2001 [2–4]. The network is known as the Asian Dust and aerosol lidar observation Network (AD-Net), and currently two-wavelength (1064 nm and 532 nm) and polarization (532 nm) lidars are continuously operated at 20 locations. Some of the network stations are collocated with the SKYNET sky radiometers. The network also contributes to the World

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Meteorological Organization (WMO) Global Atmosphere Watch (GAW) program.

The data from the lidars in AD-Net are transferred to the data server at the National Institute for Environmental Studies in Tsukuba, Japan and processed in near real time to derive the attenuated backscattering coefficients at 1064 nm and 532 nm and the volume depolarization ratio at 532 nm. The extinction coefficient estimates for non-spherical particles (mostly Asian dust) and spherical particles (mostly air pollution aerosols) are also derived [2,4]. Although this method is based on the assumption of external mixture of the two types of aerosols (non-spherical and spherical aerosols), the features of the distributions can be clearly separated; and it is useful for various applications. The dust extinction coefficient was successfully used for data assimilation of Asian dust [5–9]. The dust and spherical aerosol extinction coefficient data were also used in climatological studies of aerosols [10,11].

The near-surface dust and spherical aerosol extinction coefficient data were used for epidemiological studies of the effects of Asian dust on human health [12–15]. Although the profile measurement feature of the lidars was not employed, the data were still very useful because the depolarization ratio measurement was sensitive to non-spherical dust, and the polarization lidar was the only instrument that could measure dust and other aerosols separately in real time. We studied the relationship between the dust and spherical aerosol extinction coefficients and mass concentration measurements, and we found a high correlation between the dust extinction coefficient and the dust mass concentration in PM2.5 [16–18]. The dependence of mass-scattering efficiency on relative humidity reported for pollution in Beijing [19] could be applied to the spherical extinction coefficient derived with the lidar [20]. In epidemiological studies, significant effects of Asian dust were reported (e.g., in asthma hospitalization in children) [12]. Some reports also indicated that the mixture of Asian dust and air pollution aerosols enhanced the effect, for example, on subjective symptoms of respiratory diseases [13]. These results suggest the importance of characterizing the mixture states of dust and air pollution aerosols for epidemiological studies. The effect could be different for external mixture and internal mixture.

Previous studies using in-situ measurements and sampling analyses reported that internal mixture of Asian dust was often observed e.g., [21,22]. Mixing states of Asian dust can also be analyzed using the polarization-sensitive two-wavelength lidars of AD-Net. By making a scatter plot between the particle depolarization ratio at 532 nm and the backscattering color ratio (backscattering coefficient at 1064 nm/backscattering coefficient at 532 nm), we can separate the clusters of spherical air-pollution aerosols and Asian dust as discussed in our previous paper [23]. External mixture of these two kinds of particles falls on the theoretical curve connecting these two clusters. We may attribute the data falling outside the curve having a smaller depolarization ratio and a larger color ratio to internally mixed Asian dust. However, the information contained in the volume scattering measurements with the polarization-sensitive two-wavelength lidar is not sufficient to conclude that such particles are an internal mixture. It is also possible to interpret such data as an external mixture of dust and large spherical particles (e.g., sea salt).

To identify the mixing state of Asian dust and air pollution particles, we introduced a polarization particle counter (POPC) and performed simultaneous observation with the AD-Net lidar in Seoul, Korea. The POPC is an in-situ instrument that measures forward scattering and the two polarization components of backscattering for each particle [24,25]. It consequently provides information on size and non-sphericity of the particle at the same time. Making a scatter diagram between particle size and the depolarization ratio (the ratio of intensities of two polarization components) for a large number of particles, the size distribution, non-sphericity, and mixing states of Asian dust and air pollution particles can be analyzed. The final goal of our study is to determine a reasonable optical model for internally mixed polluted dust with detailed analysis of the POPC data. In this paper, we report the initial results of observations indicating the optical characteristics of pure and polluted Asian dust particles.

2. Experiment

We performed continuous observation using a POPC at the AD-Net lidar site in Seoul (128.95E, 37.46N, 116 m above sea level) from March to December 2013. The POPC (YGK Corp., POPC-10) was installed in the lidar container, and the ambient air was sampled through a conductive tube from the inlet on the roof. The POPC measures forward scattering intensity (with a scattering angle of 60°) and intensities of the polarization components of the backscattering (with a scattering angle of 120°) for each particle using a linearly polarized diode laser with a wavelength of 780 nm. The three pulse height signals were recorded simultaneously for each particle. Details of the POPC instrument are presented in our previous papers [24,25]. Forward scattering intensity is related to particle size, and the depolarization ratio (the ratio of perpendicular component to parallel component) of backscattering is related to the non-sphericity of the particle. We analyzed the scatter diagram between the depolarization ratio and the forward-scattering intensity for every 1-hour period.

The AD-Net lidar is a two-wavelength (1064 nm and 532 nm) and polarization (532 nm) lidar. It also has a Raman scattering receiver for measuring the extinction coefficient at 532 nm; however the Raman signal was not used in this study. We used standard AD-Net data processing, where the extinction coefficient estimates for non-spherical particles and spherical particles are derived, as well as the attenuated backscattering coefficients at 1064 nm and 532 nm and the volume depolarization ratio at 532 nm [26] (http://www-lidar.nies.go.jp/AD-Net/). The backward Fernald method [27] is used to derive the extinction coefficient at 532 nm. We use a constant lidar ratio (extinction-to-backscatter ratio) of 50 sr [28,29]. Once the extinction coefficient is obtained, the particle depolarization is calculated. We then estimate the contribution of non-spherical and spherical aerosols in the