Study of relationship between water-soluble Ca^{2+} and lidar depolarization ratio for spring aerosol in the boundary layer

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Received 30 May 2006; received in revised form 5 October 2006; accepted 8 October 2006

Abstract

The backscattering ratio, depolarization ratio and water-soluble ions of aerosols inside the boundary layer were measured by a depolarization lidar and an in situ ion chromatography (IC) in the spring of 2004 and 2005. To study the relationship between depolarization and aerosol compositions, depolarization ratios were compared with mixing ratio of soluble ions like calcium, potassium, and sodium from surface ground measurements. About 70% of the daily maximum depolarization ratios were distributed between 1.5% and 3.5% with a mean value of $2.14 \pm 0.83\%$. High correlation coefficients ($R > 0.8$) were found between depolarization and ion calcium for all of the depolarization episodes ($\text{DP} \approx 5\text{--}12\%$), which suggest the existence of mineral dust. As the tracer of biomass burning and sea salt, the mixing ratio of K^+ and Na^+ usually are greater than Ca^{2+} but did not show dependences with depolarization implying that biomass burning and sea salt aerosols may transport along with Asian dust but not exhibit significant non-spherical properties. © 2006 Elsevier Ltd. All rights reserved.

Keywords: Lidar; Depolarization; In situ IC; Water soluble ion; Dust; Atmospheric stability

1. Introduction

The Asian continent is an important source region of atmospheric aerosols with different origins including biomass burning, dusts, industrial and residential emissions. Based on the measurements of routinely operated depolarization lidar at Taipei Basin (25.01°N , 121.54°E), the depolarization ratio for aerosols within the boundary layer were found may increase from background value of $\sim 2\%$ to

greater than 10% when Taipei was suffering under Asian winter monsoon. Lidar depolarization observation indicates the existence of non-spherical particles. We notice most of depolarization episodes are accompanied with long-range transported anthropogenic pollutants (Chen et al., 2004; Lin et al., 2004). Aerosols inside boundary layer dominate the air quality and aerosol optical properties, therefore, understanding of aerosol would help for estimating the direct and indirect effects of the tropospheric aerosols on the radiation budget.

The particle depolarization is expected to relate with particle shape (Sassen, 1991; Chen et al., 2002; Noel et al., 2002). Murayama et al. (1999) had

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demonstrated the application of depolarization lidar observations combined with measurements of aerosol composition and found aerosol depolarization would be caused by dust or crystallized sea salt. Lidar measurements reveal particles such as Asian dust, biomass burning and sea-salt would exhibit clear depolarization properties owing to irregular shape. For spherical particles, depolarization is close to zero. For dry or crystallized sulfate, depolarization ratio is about 2% (Copper et al., 1974; Sassen et al., 1989). For Asian Dust, biomass burning, and sea salt, the depolarization ratio are about 5–30% (Liu et al., 2002; Sakai et al., 2002; Iwasaka et al., 2003; Kim et al., 2004; Shimizu et al., 2004), 6–11% (Wandinger et al., 2002; Murayama et al., 2004), and 8–22% (Murayama et al., 1999), respectively.

Dust, biomass burning and sea-salt are possible sources of non-spherical particle. In order to investigate what kind of aerosol causes high depolarization ratio, high temporal resolved (maximum 15 min) water-soluble ions Ca^{2+} (Mori et al., 1999; Kim and Park, 2001), K^+ (Andreae, 1983; Ma et al., 2003; Reid et al., 2005), and Na^+ are selected as the tracer of mineral dust, biomass burning, and sea-salt aerosol, respectively. Asian mineral dust had been identified to contain significant amounts of calcium (Nishikawa et al., 2000), thus Ca^{2+} is one of the species that most enhanced in dust storm events (Wang et al., 2004; Fung et al., 2005). Sea-salt particles may also contribute Ca^{2+} in coastal regions, however, during the sampling period, the measured soluble Ca^{2+} is clearly associated mainly with the dust events. Biomass burning in Asia is an important contributor to air pollution in the region. Gao et al. (2003) had shown biomass burning aerosols might contribute about $22 \pm 8\%$ to light scattering in outflow from East Asian. Therefore, biomass burning might exhibit significant depolarization if there is enough concentration.

Because of the observed value of depolarization reflects contributions from all kinds of aerosols, the higher value of depolarization ratio implies that the percentage of non-spherical particles is higher. Our results show Ca^{2+} is the only component that highly correlated with depolarization ratios for all depolarization episodes, which strongly suggest depolarization properties are contributed by mineral dust. The correlation between Na^+ and depolarization is scattered, which should be owing to low fraction of NaCl (0.4–2.5% of PM_{10}). Biomass burning aerosol may count remarkable mass frac-

tion, but our measurements show soluble K^+ did not well correlate with the depolarization, which implies the depolarization property of biomass burning aerosol were destroyed after long range transportation.

2. Methods and instruments

In this paper, lidar observed vertical and temporal profiles are compared with surface ground aerosol water-soluble ions measured by an in situ ion chromatography (IC) system. Lidar and in situ IC are installed in a container placed at weather observatory (25.01°N, 121.54°E) of the National Taiwan University (NTU), which is located in the south-western part of the Taipei Basin. Fig. 1 shows the location of the sampling site, where National Taiwan University is marked as “N”. Aerosol samples were collected from the container placed at height about 3 m above ground level. Except for traffic pollution from nearby highway (about 2 km away), there is no other known pollution source in the vicinity. The sampling site is surrounded by grassland of 3000 m² and there is no obstruction in the direction of the prevailing easterly airflow. In the spring of 2004 and 2005, lidar was routinely operated on most of the clear day since 10th February 2004; in situ IC system was available at February 13 to April 8 of 2004 and March 4 to May 31 of 2005.

Fig. 1. Geophysical position of Lidar (NTU), EPA Wanhua station (W) and CWB Banchiao radiosonde station (B). The aerial distances from NTU to Wanhua and Banchiao are 4.6 and 9.9 km, respectively.

Radiosonde data provided by the Central Weather Bureau (CWB) of Taiwan and aerosol mass concentration PM_{10} and $PM_{2.5}$ provided by Environmental Protection Administration Executive Yuan (EPA) of Taiwan are used to compare with our measurements. CWB radiosonde site is located at Banchiao (25.01°N, 121.27°E), about approximately 9.9 km away from NTU. Radiosondes are launched twice daily at 08:00 LT (Local Time, 00:00 UTC) and 20:00 LT (12:00 UTC) to provide temperature, wind speed, wind direction, and humidity profiles between 0 and 30 km. At the spring of 2004, only PM_{10} measurements are available at NTU site, therefore, PM_{10} and $PM_{2.5}$ measured at EPA Wanhua station (25.03°N, 121.49°E) are selected as references of coarse and fine mode aerosols. The location of Banchiao and Wanhua are also shown in Fig. 1 and marked as “B” and “W”, respectively.

2.1. Depolarization ratio: lidar

RCEC/ASNTU Lidar is a dual-wavelength Raman and Depolarization Lidar system (manufactured by Zenon SA, Greece). The lidar system employs the second and third harmonics of Nd-YAG laser at 532 and 355 nm. More details of the system are provided in Table 1. This system is operated round the clock to probe the atmosphere in the height range between 0.3 and 8 km.

Aerosol backscattering ratio R is defined as

$$R_{\lambda}(z) = 1 + \frac{\beta_a(\lambda, z)}{\beta_m(\lambda, z)}. \quad (1)$$

For night-time measurement, the calculation of β_a (or R) is based on Raman inversion algorithm (Ansmann et al., 1992). For daytime measurement,

Klett's method (1981) is used to calculate the aerosol backscatter with lidar ratio obtained from night-time Raman measurements. The aerosol-free region (or match point) is set as $R = 1.06$ at 6 km. If Raman measurements were not available, a fixed lidar ratio 50 sr (Sugimoto et al., 2002) is applied. The return laser light was separated by a polarization beam splitting cube into perpendicular $P_{\perp}$ and parallel $P_{\parallel}$ components related to polarization of outgoing laser beam. The depolarization ratio is defined as the ratio of the return light in perpendicular to the parallel polarizations, as given by

$$DP = P_{\perp}/P_{\parallel}. \quad (2)$$

Total depolarization ratio includes information of air molecules, but owing to weather condition, the total depolarization ratio was not convert to particle depolarization (Cairo et al., 1999; Murayama et al., 1999). In this study, because the typical aerosol backscattering ratio for 532 nm is about 10 ± 5 , therefore, the total depolarization ratio should close enough to the particle depolarization.

2.2. Water-soluble ions: in situ IC

The sampled aerosol were analyzed by in situ IC system, which provides concentration of water-soluble ions Cl^{-} , NO_2^{-} , NO_3^{-} , SO_4^{2-} , Na^{+} , NH_4^{+} , K^{+} , and Ca^{2+} with maximum time resolution of 15 min. The in situ IC system consisted of the following: gas removal denuders, aerosol collecting device, and ion chromatograph (Model ICS-90, Dionex Corp., Sunnyvale, CA, YSA). The in situ IC system is attached to the bypass flow line of the TEOM monitor, to semi-continuously measure the water-soluble inorganic ions of PM_{10} (at spring of 2004) or $PM_{2.5}$ (at spring of 2005) at intervals of 15 min. Samples were collected from a container placed at height of 3 m above the ground level. Please refer to Chang et al. (2006) for more details about the in situ IC system.

3. Results and discussions

During the sampling period (spring of 2004 and 2005, lidar was operated for 124 days. The typical backscattering ratio for aerosol below boundary layer is $R \approx 10 \pm 5$. The occurrence frequency of daily maximum depolarization ratio (hourly averaged) below the boundary layer is shown in Fig. 2. About 70% of the daily maximum total depolarization ratios are distributed between 1.5% and 3.5%,

Table 1
Lidar specification

Laser	Nd:YAG (Big-Sky CFR-400)
Wavelength	532/355 nm (linear polarization)
Pulse energy	65/60 mJ
Repetition rate	20 Hz
Transient record (Licel TR20-40)	12 bits A/D converter at 20 MHz and 250 MHz photon counting
Height resolution	7.5 m
Channels	532 nm (S and P)*, 355 nm, and 387 nm (nighttime only)
Telescope	Diameter 40 cm, focal length 160 cm
Field of view	0.5 mrad

* P : parallel; S : perpendicular (separated by polarizing beam splitter cube).

Fig. 2. Occurrence frequencies of total depolarization ratio (DP) observed at height below the boundary layer during the sampling period.

which is close to reported particle depolarization ratio of aerosols (Copper et al., 1974; Sassen et al., 1989).

During winter monsoon in Taiwan, the total depolarization ratio for aerosol below the boundary layer was found to usually increase from background value to 5–12%. The duration of each depolarization episode varies from a few hours to more than 3 days. The mean total depolarization ratio for background aerosols is found to be $2.14 \pm 0.83\%$ for high depolarization episodes with $6.22 \pm 1.48\%$.

Fig. 3 shows a typical depolarization episode occurred during February 14–17, 2004. A cold front system located between Taiwan and Japan during this episode. The ground surface wind measurements (Fig. 3(c)) show the wind direction and wind speed are east/northeast and $2\text{--}5.5\text{ ms}^{-1}$ at both NTU and Wanhau which indicates NTU and Wanhua are suffered by prevailing wind owing to winter monsoon. A clear depolarization layer with total depolarization ratio $DP \approx 2.5\text{--}6\%$ and backscattering ratio $R_{532} \approx 7\text{--}25$ could be found at the height mostly below 1.5 km between February 15 00:00LT (local time) and February 17 22:00LT, which implies the existence of non-spherical particles. The total depolarization ratio as shown in figure is not uniformly distributed (in vertical) within boundary layer during this episode.

Fig. 4(a) shows the vertical profiles of temperature and relative humidity measured by radiosonde launched on February, 15 at 08:00LT (00:00 UTC). A temperature inversion about 2°C can be seen between 1.25 and 1.6 km. At height above the temperature inversion layer, relative humidity is about $RH \approx 50\%$. Below the inversion layer, relative humidity decreases with height from $RH \approx 98\%$ at 1.2 km to $RH \approx 75\%$ at 0.5 km which might be caused by the temperature dependence of relative humidity. During 07:30LT–09:00LT, lidar signals are fully blocked by a dense cloud and, therefore, aerosol backscatter cannot be estimated.

Fig. 4(c)–(d) show the vertical profile of aerosol backscattering ratios and total depolarization ratios measured by lidar on February, 15 at 09:00LT, 09:15LT, and 10:15LT. As shown in figures, a cloud with backscattering ratio $R_{532} \gg 150$ could be found between 1.2 and 1.7 km in the morning of February, 15. Above the cloud top, aerosol backscattering ratio and total depolarization ratio are about $R_{532} \approx 1\text{--}2$ and $DP \approx 1\text{--}2\%$. Below the cloud base, total depolarization ratios tend to decrease monotonically with the backscattering ratios increase from $DP \approx 6\%$ at 0.5 km to $DP \approx 3\%$ at height around 1.2 km. If observed non-spherical particles are not easy to be deliquescent, the inhomogeneous vertical distribution of depolarization ratio may be caused by hygroscopic property of aerosol, which makes scattering cross section of spherical particle increase under higher relative humidity and hence there is a reduce in the depolarization ratio.

Inside the cloud, enormous cloud backscatter did not as expected to reduce depolarization ratio to zero that might be caused by multi-scattering effect. Laboratory simulations (Zaccanti et al., 1993; Gai et al., 1996) had shown that cloud depolarization owing to multi-scattering may up to 20–30% if cloud optical thickness exceeds 3, and they also indicate that the effect of multi-scattering is not significant on the depolarization if the cloud optical thickness is less than 1. As shown in Fig. 4, the maximum cloud depolarization ratio and backscattering ratio at 09:00 LT, 09:15LT, and 10:15LT are about $DP \approx 4.2\%$, 3.0% , 3.8% and $R_{532} \approx 150$, 33, 70, respectively, which are close to multi-scattering induced depolarization for evaporating acid droplets and pure water cloud contaminated by an aged aerosol from the evaporated acid cloud with optical thickness about 1 as shown by Sassen et al. (Sassen, 1974; Sassen and Petrilla, 1986; Sassen et al., 1989).

Fig. 3. Lidar observed (a) backscattering, (b) depolarization ratio, and (c) surface ground wind barbs measured at NTU and Wanhua during February, 14–17, 2004.

To investigate the reason that causes higher depolarization, water-soluble ions of aerosol were selected to study the correlations between depolarization and aerosol compositions. Soluble Ca^{2+} , K^+ and Na^+ are chosen as the indicator of mineral dust, biomass burning and sea salt aerosols. Fig. 5(a)–(c) show the temporal evolution of total depolarization ratio measured at 0.3 km and mass fraction of soluble ions (Ca^{2+} , K^+ , Na^+ , NO_2^- , NO_3^- , Cl^- , SO_4^{2-} , and NH_4^+) during February, 14 12:00LT to February, 17 24:00LT. Aerosol mass concentration measured at NTU (PM_{10}) and Wanhua (PM_{10} and $\text{PM}_{2.5}$) is displayed in Fig. 5(d). Fig. 6(a)–(c) are the scatter diagrams

show the relationship between DP and mixing ratio of water-soluble ions. As shown in figure, soluble ions Ca^{2+} , K^+ , and Na^+ count about 0.2–0.5%, 0.25–1%, and 0.2–0.5% of PM_{10} , respectively. In all of the soluble ions, only temporal variation tendency of $\text{Ca}^{2+}/\text{PM}_{10}$ is similar to that of total depolarization ratio. The correlation coefficients between depolarization ratio and mass fraction of ions are $R_{\text{Ca}} = 0.824$, $R_{\text{K}} = -0.247$, and $R_{\text{Na}} = 0.461$ indicating the more Ca^{2+} in aerosol the more depolarization can be observed. We also notice Na^+ is not correlated with K^+ or Ca^{2+} , which implies soluble Ca^{2+} and K^+ are not from sea-salt origin. Since soluble Ca^{2+} had been widely verified as tracer

Fig. 4. Profiles of (a) temperature and relative humidity measured by the radiosonde launched on February 15, 2004 at 08:00LT. Profiles of backscatter ratio and depolarization ratio measured by lidar on February 15, 2004 at (b) 09:00LT, (c) 09:15LT and (d) 10:15LT.

of mineral dust, above result suggests that the depolarization property is caused by mineral dust.

Low correlation between soluble Na^+ and total depolarization ratio might be possible to be caused by low concentration of NaCl or the hygroscopic property of sea-salt. In this episode, soluble Na^+ just counts about 0.2–0.5% of PM_{10} . As shown in Fig. 5(b), mass fraction of soluble Cl^- almost equals to that of Na^+ , in other words, equivalent concentration of Cl^- is about 1/3 lower than $[\text{Na}^+]$ which may be caused by depletion of chloride (Chang et al., 2006). If soluble Na^+ is totally contributed by sea-salt particle, then the corresponding mass fraction of NaCl is about 0.5–1.3% of PM_{10} . This could be one of the reason for the low correlation with depolarization, even if all NaCl are crystallized.

As the tracer of biomass burning aerosol, during February, 14 20:00LT to February, 15 04:00LT, soluble ion potassium is more concentrated as mass fraction about 0.75–1.0%, which is close to the mass

percentages (about 0.4–14%, typical about 2.5%) of potassium in the identified biomass burning events summarized by Reid et al. (2005). Therefore, considerable mass fraction of total biomass burning aerosols might not be ignored in this episode even though it is not reliable to estimate mass fraction of biomass burning aerosol by soluble ion potassium. Based on above assumption, which implies most of biomass burning aerosols are spherical-like shaped particle that may be caused by hygroscopicity or external mixing (Chou et al., 2005).

Dust particles usually are found in coarse mode. The scatter diagrams of water-soluble ions versus the aerosol mass concentration PM_{10} are shown in Fig. 6(d)–(f). The correlation coefficients are $R_{\text{Ca}} = 0.94$, $R_{\text{K}} = 0.41$, and $R_{\text{Na}} = 0.93$, which suggest soluble Ca^{2+} and Na^+ mainly concentrate in coarse mode particles. Because $\text{PM}_{2.5}$ measurement is not available at NTU in the year 2004, to investigate the relationship between water-soluble ions and particle size, PM_{10} and $\text{PM}_{2.5}$ measured at

Fig. 5. Time series of (a) depolarization ratio (at 300 m), mass fraction of ions Ca^{2+} , K^{+} , (b) Na^{+} , Cl^{-} , NO_2^{-} , (c) NO_3^{-} , SO_4^{2-} , NH_4^{+} , (d) aerosol mass concentration PM_{10} and $\text{PM}_{2.5}$ measured at NTU or Wanhua during February 14, 2004 12:00LT to February 17, 2004 24:00LT.

EPA Wanhua station (about 4.5 km away from NTU) was applied. As shown in Fig. 5(d), PM_{10} measured at Wanhua is very close to that at NTU during this depolarization episode. Therefore, it is reasonable to assume aerosol properties including $\text{PM}_{2.5}$ are similar at both place during this depolarization episode. The correlations between water-soluble ions and mass concentration of coarse mode ($\text{PM}_{2.5-10}$) and fine mode ($\text{PM}_{2.5}$) aerosol are shown in Fig. 6(g)–(l). For coarse mode, the

correlation coefficients are $R_{\text{Ca}} = 0.93$, $R_{\text{K}} = 0.13$, and $R_{\text{Na}} = 0.84$. And for fine mode, the correlations are $R_{\text{Ca}} = 0.04$, $R_{\text{K}} = 0.55$, and $R_{\text{Na}} = 0.24$. The results indicate that the mineral dust and sea-salt particle are essentially distributed in coarse mode particles during this episode, but only mineral dust is the reason that causes the increasing of aerosol depolarization.

Fig. 7 shows another depolarization episode occurred during April 5–6, 2004. Between April 5,

Fig. 6. (a)–(c) Correlations between depolarization ratio (DP) and mass fraction of soluble Ca^{2+} , K^+ and Na^+ . (d)–(i) correlations between ion mass concentration and PM_{10} . (g)–(l) correlations between ions in the coarse mode ($\text{PM}_{2.5-10}$) or fine mode ($\text{PM}_{2.5}$).

18:00LT and April 6, 18:00LT, an aerosol layer with maximum depolarization ratio and backscattering ratio about $\text{DP} \approx 10\%$ and $R_{532} \approx 4$ could be found located mainly at the height above the boundary layer between 0.7 and 2.3 km. The height of nocturnal boundary layer at night of April, 5 could be easily figured out to descend from about 1.2 to 0.3 km. In this episode, wind speed is about $1\text{--}1.5\text{ ms}^{-1}$, and the wind direction is mainly south-eastward at NTU and northeastward at Wanhua

during April 5, 18:00LT to April 6, 07:00LT. At 07:00LT on April 6, wind starts to change direction from northwestward to eastward at NTU and Wanhua. Unlike previous case (during February 14–17, 2004), both wind speed and wind direction are quite different at NTU and Wanhua which implies the characteristics of aerosol may not be similar at these two places during this depolarization episode.

As shown in Fig. 8(b), this depolarization layer was separated by two thin low-depolarization layers

Fig. 7. Same as Fig. 3 but for the depolarization episode occurred during April 5, 2004 18:00LT to April 6, 2004 18:00LT.

with backscattering ratio $R_{532} \approx 7$ and depolarization ratio $DP \approx 1.5\%$ at height about 0.75 and 1.25 km during April 6, 04:00LT and April 6, 09:00LT. Higher backscattering ratio are associated with lower depolarization indicating that these two layer mainly consisted with spherical particles. Lower depolarization of these two layers may be caused by hygroscopic growth of aerosol because radiosonde data (Fig. 9(a)) shows two moist layers about $RH \approx 85\%$ located at same altitudes as the low-depolarization layers.

On April 6, between 9:00LT and 5:00LT, vertical distribution of depolarization ratio shows non-spherical particles extend to downward from depolarization layer to the surface, which might be

caused by elevating of internal boundary layer (mixing layer). Fig. 8(e) shows height of mixing layer (mixing height, MH), bottom height of depolarization layer, and lidar depolarization ratio measured at 0.3 km during April 6, 06:00LT to 18:00LT. In this study, the mixing height is determined by vertical backscattering ratio profile. For cases shown in Fig. 8(b)–(d), the mixing heights are 0.55, 0.67, and 1.43 km for April 6, 08:00 LT, 09:30LT, and 12:45LT, respectively. As shown in Fig. 8(e), the mixing height is about $MH \approx 0.5$ km before 10:00LT. After 10:00LT, mixing height rapidly ascended and reaches maximum height of 1.43 km at 12:45LT. The bottom of depolarization layer descended from 0.9 to 0.5 km and intersected

Fig. 8. Profiles of (a) temperature and relative humidity measured by the radiosonde launched on April 6, 2004 at 08:00LT. Vertical backscatter ratio and depolarization ratio profiles measured by lidar on April 6, 2004 at (b) 08:00LT, (c) 09:30LT and (d) 12:45LT. Temporal variation of (e) height of mixing layer and bottom of the depolarization layer determined by the lidar, and (f) the surface ground temperature measured at NTU and Wanhua.

with the MH at 10:00LT. Whereas, the depolarization ratio measured at 0.3 km is also found to increase rapidly from background value of 1–6% after intersecting the MH with the bottom of depolarization layer, which shows the non-spherical

particles found at 0.3 km are oriented from depolarization layer above the boundary layer.

Atmospheric boundary layer (ABL) and mixing layer is directly influenced by the presence of the earth's surface, and responds to surface forcing with

Fig. 9. Time series of (a) depolarization ratio (at 300 m), mixing ratio of ions Ca^{2+} , K^{+} , (b) Na^{+} , Cl^{-} , NO_2^{-} , (c) NO_3^{-} , SO_4^{2-} , NH_4^{+} , and (d) aerosol mass concentration (PM_{10} , $\text{PM}_{2.5}$) measured at NTU or Wanhua during April 5, 18:00LT to April 7, 24:00LT.

a timescale of about 1 h or less (Stull, 1988). The forces include heat transfer, friction drag, evaporation and transpiration, pollutant emission and terrain induced flow modification. Determination of the mixing height from radiosondings is the most common method since routine radiosondings of temperature, humidity and wind are available all over the world, but it is well known from field experiments that the mixing height strongly varies both in time and space (Stull, 1988). Some approaches (e.g. Khan and Simpson, 1997; Nath

and Patil, 2003) try to retrieve highly temporal resolved mixing height from ground-based meteorological parameters along with estimated atmospheric stability. The atmospheric stability is the tendency of the atmosphere to resist or enhance vertical motion and thus turbulence. An unstable atmosphere enhances turbulence, whereas a stable atmosphere inhibits mechanical turbulence.

Vertical mixing depends on the atmospheric stability and the existence and height of inversion layers. On April 6, 2004, Taipei is situated at outer

boundary of a high-pressure system centered at east of Japan, therefore, the growth of mixing height did not be suppressed by the sinking motion accompanied by anti-cyclone. Table 3 lists the estimated hourly atmospheric stability classes based on Pasquill's (1961) concept for daytime of April 6, 2004 according to surface atmospheric parameters measured at Central Weather Bureau, Taiwan (near downtown of Taipei). Atmosphere stability is found to change from extremely-to-moderately (class "A–B") to extremely (class "A") unstable during 9:00LT to 12:00LT and from extremely unstable to slightly (class "C") unstable during 12:00LT to 16:00LT. The temporal variation of atmospheric stability is agreed with the evolution of lidar observed mixing height and depolarization ratio (Fig. 8(e)). Khan and Simpson (1997) also shown the mixing height tends to be higher under unstable stability but they also shown there is no straightforward relationship between mixing height and stability. The mixing height is difficult to model and more studies are still needed because of complex interactions between the many physical processes that contribute to evolution of mixing layer. Since mixing layer has high temporal and

spatial variation, remote sensing instruments such as lidar can observe mixing layer variation almost continuously and provide good observational data of the dynamical process for the study of mixing layer.

Time series of total depolarization ratios at 0.3 km, mass fraction of aerosol water-soluble ions, and aerosol mass concentrations measured between April 5, 18:00LT and April 6, 18:00LT are shown in Fig. 9. The maximum and minimum values of PM_{10} are about 130 and $40 \mu\text{g m}^{-3}$. The scatter plots of mass fraction of ions as function of depolarization ratio and PM_{10} are shown in Fig. 10. Similar to previous case, only Ca^{2+}/PM_{10} exhibits similar variation tendency as depolarization. The correlation coefficients between DP and ions calcium, potassium, and sodium as shown in Fig. 10(a)–(c) are $R_{\text{Ca}} = 0.816$, $R_{\text{K}} = -0.014$, and $R_{\text{Na}} = -0.417$, respectively, which indicates that the non-spherical property is contributed by mineral dust. Since the non-spherical particles observed at surface ground is oriented from the depolarization layer above boundary, therefore, it can be concluded that the depolarization layer consists of mineral dust. In this episode, mass fraction of soluble ion potassium is

Fig. 10. Correlations between depolarization ratio (DP) and (a) Ca^{2+}/PM_{10} , (b) K^{+}/PM_{10} , (c) Na^{+}/PM_{10} . (d)–(f) correlations between aerosol mass concentration (PM_{10}) particles and Ca^{2+} , K^{+} and Na^{+} .

about $0.25 \pm 0.1\%$ implying biomass burning aerosol counts fewer parts of PM_{10} and results the low correlation with depolarization.

Both soluble Na^+ and Cl^- obviously exhibit quite different correlations with DP and PM_{10} at time before and after April 6, 06:00LT. To make the tendency more clear, data points are marked as dots and circles in Fig. 10 for data measured before and after April 6, 06:00LT. As shown in Fig. 9(a), mass fraction of Na^+ decreases monotonically from 1.2% to 0.25% during April 5, 18:00LT to April 6, 06:00LT and then maintaining about $0.25 \pm 0.05\%$ after April 6, 06:00LT. Fig. 7(c) also indicates wind at NTU changes direction from southeast to northwest. By comparing Na^+ with PM_{10} , Fig. 10(f) implies the source of Na^+ before April 6, 06:00LT is independent of local pollution. In this episode, equivalent concentration $[\text{Na}^+]$ is almost equal to $[\text{Cl}^-]$ (refer to Fig. 9(b)) representing the mass fraction of NaCl is about 0.6–3.1% of PM_{10} . Therefore, NaCl should not be expected to be well correlated with that of depolarization owing to low concentration.

The correlation coefficients between water-soluble ions and PM_{10} are $R_{\text{Ca}} = 0.920$, $R_{\text{K}} = 0.825$, and $R_{\text{Na}} = -0.219$. Since temporal variations of wind and PM_{10} measurements show the meteorology and pollution environment at NTU and Wanhau are different (refer to Figs. 7(c) and 9(d)),

aerosol mass concentrations measured at Wanhau (PM_{10} and $\text{PM}_{2.5}$) are not referred as coarse or fine mode data for this case. By lacking fine mode aerosol information, above correlations could not support enough evidence to imply soluble Ca^{2+} and K^+ probably are mainly rich in the coarse mode.

The correlation coefficients between DP and aerosol water-soluble ions for the depolarization episodes observed in the spring of 2004 and 2005 are summarized in Table 2. The mean values of mixing ratio of ions for all depolarization ($\text{DP} > 3.5\%$) and low-depolarization episodes ($\text{DP} < 3.5\%$) are also listed. Please note that only PM_{10} particles were collected in the year 2004 and only $\text{PM}_{2.5}$ particles were collected in the year 2005. As shown in Table 2, even though PM_{10} is not available in 2005, $\text{Ca}^{2+}/\text{PM}_{10}$ and $\text{Ca}^{2+}/\text{PM}_{2.5}$ are highly positively correlated with depolarization ratio for all depolarization episodes. However, as the indicator of biomass burning and sea-salt particles, the mass fraction of soluble K^+ and Na^+ did not exhibit clear dependences with the depolarization. This suggests that all of the observed non-spherical particles in the spring of 2004 and 2005 were mineral dust aerosols. Moreover, Table 2 also imply fine mode soluble Ca^{2+} could also be applied as indicator of mineral dusts which is consistent with the results observed during ACE-Asia (Maxwell-Meier et al., 2004).

Table 2

The correlation coefficients between total depolarization ratio and mass fraction of water-soluble Ca^{2+} , K^+ and Na^+

Depolarization episodes	Maximum DP (at 0.3 km)	Correlation coefficient between DP and mass fraction of ions			Mean mass fraction of ions		
		R_{Ca}	R_{K}	R_{Na}	$\text{Ca}^{2+}/\text{PM}_{10}$	$\text{K}^+/\text{PM}_{10}$	$\text{Na}^+/\text{PM}_{10}$
Feb. 14–17, 2004	7%	0.824	−0.247	0.461	4.07×10^{-3}	4.35×10^{-3}	4.00×10^{-3}
Mar. 15–16, 2004	5%	0.783	−0.323	0.100	3.21×10^{-3}	1.54×10^{-3}	3.45×10^{-3}
Apr. 2–3, 2004	11%	0.808	0.285	0.074	13.70×10^{-3}	4.15×10^{-3}	4.84×10^{-3}
Apr. 6, 2004	6%	0.816	−0.014	−0.417	4.15×10^{-3}	2.82×10^{-3}	4.70×10^{-3}
	Cases above				6.28×10^{-3}	3.21×10^{-3}	4.25×10^{-3}
	Cases that DP < 3.5%				2.19×10^{-3}	4.66×10^{-3}	3.15×10^{-3}
		R_{Ca}	R_{K}	R_{Na}	$\text{Ca}^{2+}/\text{PM}_{2.5}$	$\text{K}^+/\text{PM}_{2.5}$	$\text{Na}^+/\text{PM}_{2.5}$
Mar. 18–19, 2005	8%	0.807	0.572	−0.641	8.14×10^{-3}	1.91×10^{-2}	2.67×10^{-2}
Mar. 25, 2005	5%	0.681	0.425	0.352	6.52×10^{-3}	1.19×10^{-2}	5.22×10^{-2}
Apr. 5, 2005	8%	0.752	0.348	−0.421	3.17×10^{-3}	9.61×10^{-3}	1.73×10^{-2}
Apr. 13–17, 2005	11%	0.777	0.564	−0.723	3.96×10^{-3}	1.94×10^{-2}	4.19×10^{-3}
Apr. 20–21, 2005	9%	0.724	0.275	0.612	2.95×10^{-3}	1.73×10^{-2}	6.30×10^{-3}
	Cases above				4.89×10^{-3}	15.46×10^{-3}	2.14×10^{-2}
	Cases that DP < 3.5%				1.69×10^{-3}	8.43×10^{-3}	1.03×10^{-2}

Meteorology and HYSPLIT back-trajectory (not shown) analysis show the most possible mineral dust source is outflow of Asian dust oriented from North or Northwest China. As shown in Fig. 11, depolarization ratios observed in spring of 2004 exhibit similar dependence on $\text{Ca}^{2+}/\text{PM}_{10}$ indicating that the observed mineral dusts may have similar depolarization or microphysical properties. Fig. 11 also implies that if mass fraction of soluble Ca^{2+} (or dust) is large enough, we should observe depolarization as large as measured at mainland China and Japan (e.g. Iwasaka et al., 2003; Murayama et al., 2001). However, Ca-containing particles with spherical shape have been recently identified in dust particles. Matsuki et al. (2005) had found spherical Ca-rich particle at Beijing, China and concluded that might be transformed from calcite, which is one of the major components of Asian dust particle (Okada et al., 1990). The morphology of dust particle might be caused by external mixing process such as coating dust with aqueous solution (Sakai et al., 2002) or heterogeneous chemical process. Krueger et al. (2003a, b) have shown that the heterogeneous chemical process would transform CaCO_3 from solid irregular particles into spherical liquid droplets after reaction with gas-phase HNO_3 in the presence of water vapor. For such cases, DP should be expected to not exhibit good correlation with $\text{Ca}^{2+}/\text{PM}_{10}$.

As shown in Table 2, mean value of $\text{Ca}^{2+}/\text{PM}_{10}$ and $\text{Ca}^{2+}/\text{PM}_{2.5}$ in depolarization episodes are about 3 times than in the low-depolarization episodes which is consistent with previous studies about correlation between total depolarization

Table 3

Pasquill stability classified according to ground-based measurement for daytime of April 6, 2004

Local time (h)	Solar Insolation (W m^{-2})	Surface wind speed (ms^{-1})	Pasquill stability ^a
7:00	83	0.9	B
8:00	278	0.7	B
9:00	506	0.3	A-B
10:00	689	0.2	A-B
11:00	833	0.3	A
12:00	833	0.6	A
13:00	797	3.5	B
14:00	747	2.7	A-B
15:00	417	2.1	B
16:00	297	1.8	C
17:00	208	0.6	B
18:00	39	1.8	C

^aA: extremely unstable; B: moderately unstable; slightly unstable.

ratio and Ca^{2+} . In 2004, mean values of soluble $\text{K}^+/\text{PM}_{10}$ and $\text{Na}^+/\text{PM}_{10}$ in the depolarization episodes are close to those in the low-depolarization episodes. In 2005, mean values of soluble $\text{K}^+/\text{PM}_{2.5}$ and $\text{Na}^+/\text{PM}_{2.5}$ in the depolarization episodes are about twice of those in the low-depolarization episodes, which imply biomass burning and sea salt aerosols may transport along with Asian dust and mainly exist in the fine mode particles. Sea salt aerosol is a possible source of K^+ . We found K^+/Na^+ could be as high as ~ 2 . Since the value of K^+/Na^+ in sea water is about 0.04 (Carmichael et al., 1997). Therefore, soluble K^+ would be easily interfered by biomass aerosol, and should not applied as tracer of sea salt in this study.

4. Conclusion

By seeking the correlation between mass fraction of aerosol soluble ions and lidar-observed total depolarization, we found soluble Ca^{2+} is the component that highly correlated with total depolarization ratio which strongly suggests that the depolarization property is contributed by mineral dust. HYSPLIT back-trajectories indicate the mineral dusts oriented from north or northwest China, therefore, the most possible dust source is outflow of Asian Dust. The maximum observed dust depolarization ratio in each depolarization episode is about 6–10%, which is much lower than dust measured at China or Japan and that should be caused by the low dust mass fraction. With high

Fig. 11. Correlation between DP and $\text{Ca}^{2+}/\text{PM}_{10}$ for cases occurred in 2004.

vertical and temporal resolution of lidar and in situ IC system, we also found non-spherical particles can be used as tracer to study the dynamics of internal boundary.

In this study, soluble Na^+ may count up to 1% of PM_{10} (in average, 0.3–0.4%) but did not show regular dependences with aerosol mass concentration or depolarization. Low (or none) correlation between soluble Na^+ and depolarization could be caused by low mixing ratio of Na^+ (NaCl). Na^+ may be rich in coarse mode (as shown in first case) but more detail meteorological and aerosol analysis are needed to understand the behavior of Na^+ . Soluble K^+ is also not well correlated with depolarization. Though it is hard to estimate the concentration of biomass burning by soluble ions, our measurements of soluble K^+ reveal mass fraction of biomass burning may be remarkable in PM_{10} or $\text{PM}_{2.5}$. Therefore, the depolarization property of biomass burning aerosol may be reduced during long-range transport (if original biomass burning particle is indeed highly irregular in shape). Though K^+ and Na^+ did not well correlate with depolarization, mass fraction of fine mode K^+ and Na^+ (in 2005) in depolarization episodes is about twice than that in low-depolarization episodes implying aerosols containing K^+ may be transported from Mainland China to Taipei area along with Asian Dust.

Acknowledgments

This work was supported by the Nation Science Council of Taiwan, R.O.C. through Grants NSC-90-2119-M-002-013, NSC-94-EPA-Z-008-003, NSC-94-2111-M-001-004, and NSC-95-2111-M-001-004. Partial funding support for the lidar instrument from the College of Science, National Taiwan University is greatly appreciated.

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