Interpretation of atmospheric particles at an urban area

Sonoyo Mukai [a], Itaru Sano [a], Masayoshi Yasumoto [a] and Yoshifumi Shinbo [b]

[a] *Research Institute for Science and Technology, Kinki University*
[b] *Graduate school of Science and Technology, Kinki University*

Abstract

Simultaneous measurements of atmospheric aerosols and suspended particulate matter (SPM) have been undertaken at Kinki University campus in Higashi-Osaka in order to monitor the urban environment. The sun/sky photometry has been made as a NASA/AERONET station since 2002, and the SPM-613D (Kimoto Electric) has been taking measurements of the SPM concentrations such as TSP, PM_{10}, $PM_{2.5}$, and OBC at the same site since March 15, 2004. The relationship between aerosol properties obtained from radiometry with AERONET and the SPM measurements is examined. It is found that there is a linear correlation between SPM concentrations and aerosol properties, which indicates that aerosol characteristics can be estimated from SPM data, and vice versa.

Furthermore chemical analysis of the SPM sampling strongly suggests the really complicated features of the atmospheric particles at Higashi-Osaka. In other words, it is shown that the air quality of the Higashi-Osaka site is poor due to not only anthropogenic particles by local emissions, such as diesel vehicles and chemical industries, but also due to dust particles coming from continental desert areas by large scale climatic conditions.

Keywords: Urban Atmosphere, Aerosols, PM_{10}, $PM_{2.5}$, OBC

1. Introduction

It is known that optical properties of atmospheric aerosols, such as optical thickness, particle size, single scattering albedo, etc., are important for the Earth's radiation budget [Nakajima *et al.* 2001]. It is also known that Asia is the most complicated region for aerosol study, because soil dust, which is named Kosa or Yellow sand, comes from China especially in the spring, and the emission of aerosols, e.g. sulfuric, nitric, and carbonaceous, is increasing due to economic growth [Sano et al. 2003]. Therefore urban areas in Asia are interesting and important sites for a feasible study of aerosols.

A multi-spectral photometer was set up as an NASA/AERONET site at Kinki University campus in Higashi-Osaka in 2002 for measuring urban aerosols. Higashi-Osaka is an industrial city located between Osaka Bay and the Ikoma Mountains, and is famous for its heavy air pollution. Because anthropogenic aerosols produced by industries are mixed with oceanic aerosols transported from Osaka Bay and trapped around Higashi-Osaka. Thus the aerosol properties here are especially complicated due to this mixing of anthropogenic and natural components.

It is known that anthropogenic small aerosol particles dominate the air over urban cities due to local emissions such as from diesel vehicles and, chemical industries [Wang et al. 2003] and that

small atmospheric particles play an important role in human health and climate change [Balasubramanian et al., 2003]. It is difficult to relate suspended particles matter (SPM) data directly to aerosol properties, but SPM data approximately represent the mass concentration of atmospheric particles at the surface. It has been shown that the concentration of SPM correlates with the column aerosol optical thickness [Goloub et al., 2000]. Thus air pollutants could bear some relation to the emission and transportation of aerosols. In order to elucidate the correlation between aerosol properties and concentrations of in-situ atmospheric particles at the surface, an instrument SPM-613D (Kimoto Electric, Japan) began taking measurements of SPM

concentrations such as TSP (total suspended particulates), PM_{10} ($<\approx 10\mu m$), $PM_{2.5}$ ($<\approx 2.5\mu m$), and OBC (optical black carbon) on March 15, 2004 at the AERONET/Higashi-Osaka site.

Simultaneous monitoring of aerosols with a multi-spectral photometer as a NASA/AERONET station and SPM by the SPM-613D at the Kinki University campus in Higashi-Osaka makes it possible to determine the relationship between aerosol properties and the particulate mass. It is of interest to mention that the SPM sampler works continuously, even on cloudy/rainy days, while radiometry is only available during the day time on a clear day.

2. RADIOMETRY OF AEROSOLS

A multi-spectral sun/sky photometer, the CE-318-2 (Cimel Electronique, France), was set up as an AERONET site at Higashi-Osaka in 2002 [Sano et al., 2003, Mukai et al., 2005]. This CE-318-2 instrument has four observing channels for photometry whose central wavelengths are 0.44, 0.67, 0.87 and 1.02 μm, and polarimetric capabilities at 0.87 μm. The radiometer is calibrated every year at NASA/GSFC by using a standard AERONET procedure [Holben et al., 1998]. Aerosol properties, such as the optical thickness, size distribution, complex refractive index, and single scattering albedo, are retrieved based on the AERONET standard inversion method [Dubovik et al., 2000, Dubovik and King, 2000].

The column aerosol optical thickness (AOT: τ_λ) at a wavelength λ is an important aerosol parameter retrieved from the transmittance measured by direct sun photometry. The resolution of the AOT is better than 0.01 in all observation channels, and cloud screening of the obtained data

was performed before aerosol retrieval [Smirnov et al., 2000]. The Ångström exponent (α) is calculated from the spectral tendency of AOT.

The values of α are closely related to the aerosol size. For example, small values of α indicate large particles and large values represent small particles such as artificial aerosols. In general, values of α from ~0 to 1 indicate particles such as sea salt aerosols and soil dust, whereas values of $1<\alpha<\sim2.5$ indicate anthropogenic particles such as sulfate and those associated with carbonaceous compounds.

The Ångström exponent (α) is the first derivative of ln τ_λ with respect to ln λ. Similarly the second derivative (α') of ln τ_λ with respect to ln λ is also defined. This value indicates the relative influence of fine mode particles upon the optical properties [Eck et al., 1999, O'Neill et al., 2003].

Furthermore the optical fraction of fine-mode particles ($\eta=\tau_f / \tau_\lambda$) is defined,

where τ_f represents a fine mode optical thickness of aerosol [O'Neill et al., 2001].

Figure 1 shows AOT (τ_λ, τ_f) at a wavelength of 0.87μm, Ångström exponent (α), its second derivative (α') and the optical fraction of fine-mode particles (η) derived for all days for which measurements are available from 15 March 2004 to 30 October 2005, at Higashi-Osaka. Note that the instrument has been calibrated at NASA/GSFC from January to March in 2005. The solid lines at 0.2 in the AOT (0.87μm) panel, at α=1.0, at α'=0.0 and at η=0.5 in another panels are referred to below. The value of AOT represents the aerosol loading, and small and large values of α indicate large and small particles, respectively, as mentioned above. The case of $\tau_{0.87} > \approx 0.2$ indicates lots of aerosol loading, what one calls aerosol events. In general, the Ångström exponent $\alpha > \approx 1$ indicates fine-mode particles, and $\alpha < 1$ indicates large particles such as soil dust. Variable η represents optical fraction of fine-mode particles, and hence $\eta > \approx 0.5$ means dominance of fine-mode particles.

The second derivative α' yields aerosol optical information such as carbonaceous compounds. At a glance of Figure 1, two types of aerosol events are found at Higashi-Osaka as: dust events in spring and anthropogenic particle-events in winter. Since $\alpha > \approx 1$ and the second derivative α' is negative for almost all cases, fine particles contaminated by anthropogenic emissions seem to always dominate at Higashi-Osaka.

The complicated features of aerosols at Higashi-Osaka are shown in Figure 1. Some aerosol events show characteristic feature of dust events as high AOT and low α, but rather high values of α are detected in other events. At any rate, long term radiometry at Higashi-Osaka indicates three typical types of aerosols [Mukai et al., 2005]:

(A) anthropogenic aerosol events with AOT>0.2, α>1.0, α'>0,

(B) background atmosphere with AOT<0.2, α>1.0, α'<0 and

(C) coarse dust events with AOT>0.2, α<1.0, α'<0.

3. SAMPLING OF SPM

An instrument SPM-613D is set up on the roof of a university building (about 30 m above the ground and about 50 m above sea level). The SPM-613D makes it possible to determine the relationship between aerosol properties and the particulate mass, since it can separate the contributions of fine particles ($PM_{2.5}$), coarse particles (PM_c), and OBC. Namely the instrument can measure SPM concentrations such as TSP (total suspended particulates), PM_{10} ($<\approx 10$μm), $PM_{2.5}$ ($<\approx 2.5$μm) separately in size, and OBC (optical black carbon). The OBC value is measured from the optical density of $PM_{2.5}$. Tisch's standard inlet for TSP or PM_{10} is selected respectively in the case of

TSP measurements made during the dust season from March to May, or other usual time, i.e. from June to February, respectively. Note that the measurements of TSP on rainy days are manually removed from the data set in order to avoid contamination by rainy mist. Aspirate flow rate is constant at 16.7(l/min) throughout the year. The virtual impactor separates particles into fine- and coarse-modes. Those particles are sampled on a Teflon tape roll, and then their concentrations are measured by using a [147]Promethium beta ray.

Figure 2 shows measurements of PM_c (denoted by yellow color) and $PM_{2.5}$ (blue color) in the upper panel, and OBC in the

lower panel during the same period as Figure 1 from 15 March 2004 to 30 October 2005. Roughly speaking, the mass concentrations of SPM over Higashi-Osaka are divided into three seasons, spring, early winter and others. Each season has each character, for example in spring from March to May the coarse mode particles

PM_c, which are calculated from the difference betweenTSP/PM10 and PM2.5, are dominant. High concentrations of PM_c occurred simultaneously with the radiometric dust events in Figure 1. These data indicate that several dust events occurred at Higashi-Osaka during spring.

4. RELATIONSHIP BETWEEN AEROSOLS AND SPM

Some differences exist between aerosol radiometry and SPM sampling, because SPM sampler works continuously at the surface, while atmospheric radiometry is only available during daytime on a clear day. As shown in the previous sections, however, long term simultaneous monitoring of aerosols and SPM provides us with similar temporal variation, and indicates three types of aerosols according to the season, i.e. (type-A) anthropogenic aerosol events in early winter, (type-C) coarse dust events in spring and (type-B) others. For reference, Figure 3 presents the monthly averaged values of TSP/PM$_{10}$ (the solid curve), PM$_{2.5}$ (the dashed curve) and PM$_c$ (the dotted curve) in the upper panel, and OBC in the lower panel. From Figure 3 the following results are drawn.

(1) The concentration of SPM varies with month.

(2) Total amount (TSP/PM$_{10}$) of SPM gives the peaks in April, July and November in order.

(3) A peak of SPM in April is supplied with not only coarse particles (PM$_c$) but also fine particles (PM$_{2.5}$).

(4) Both SPM peaks in July and November are supported by fine particles (PM$_{2.5}$).

(5) The OBC shows the peaks simultaneous to PM$_{2.5}$ in November and December.

(6) Coarse particles (PM$_c$) present the maximum peak in April.

(7) Very low SPM amount is found in January.

These results coincide with the radiometry shown in Figure 1 and well explain the domestic facts. The SPM-613D data show the dominance of fine anthropogenic (PM$_{2.5}$) and coarse (PM$_c$) particles over Higashi-Osaka during dust events in March and April. Furthermore, PM$_{2.5}$ concentrations are larger than PM$_c$'s in several dust events. This feature has been also clearly shown by the aerosol size derived from the radiometry of AERONET 2002 [Sano et al., 2003, Mukai et al., 2005].

The scattergrams of PM$_{2.5}$ against AOT (0.87µm) are shown in Figure 4, which presents total aerosol optical thickness (τ_λ) and fine mode aerosol optical thickness (τ_f) vs. PM$_{2.5}$ in the left and right panels, respectively. Each equation written within each panel represents the solid regression line. The value of γ represents the correlation coefficient. The correlation is good, and the value of γ (=0.93) against τ_f is better than that (=0.89) against τ_λ. The relationship between the aerosol properties derived from radiometry and the particulate mass simultaneously obtained with the SPM-613D instrument looks promising in that aerosol characteristics can be estimated from SPM data, and vice versa. For example, the linear correlation between SPM and AOT allows the aerosol properties to be estimated from SPM data in areas without an AERONET site, on

cloudy or rainy days, or at night. Alternatively, satellite-derived aerosol information is useful for indicating air quality on a global scale.
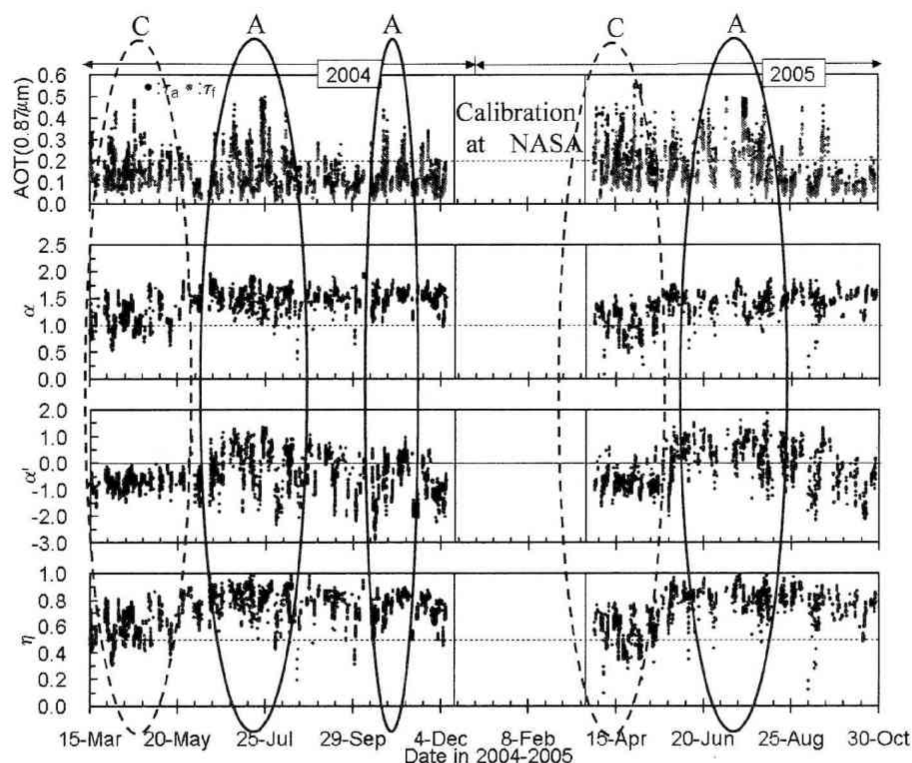

Fig.1 Radiometry of aerosols at Higashi-Osaka site from 15 March 2004 to 30 October 2005, where AOT (τ_λ, τ_f) at a wavelength of 0.87µm, Ångström exponent α, its second derivative α' and the optical fraction of fine-mode particles η are presented. Refer to the text in section 2 with respect to clusters (A) and (C).

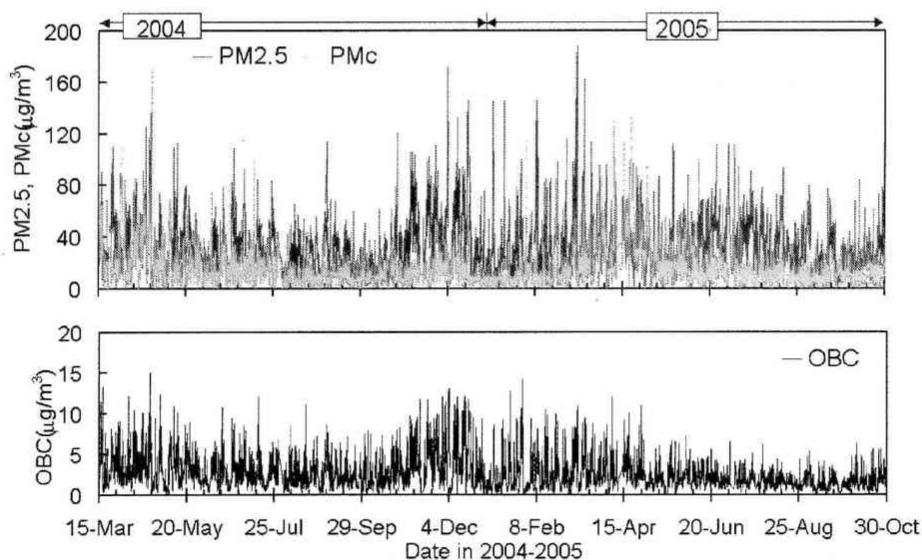

Fig.2 SPM measurements during the same period as Figure 1, where PM_c and $PM_{2.5}$ are denoted by yellow and blue color in the upper panel, and OBC in the lower panel.

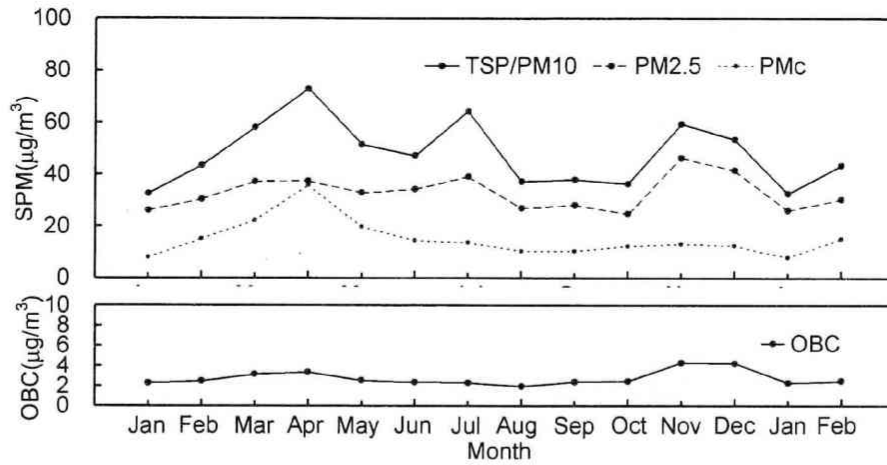Fig.3 Monthly averaged values of TSP/PM$_{10}$ (the solid curve), PM$_{2.5}$ (the dashed curve) and PM$_c$ (the dotted curve) in the upper panel, and OBC in the lower panel.
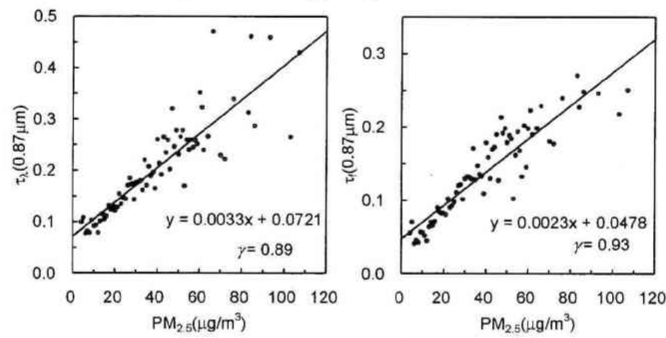

Fig.4 Scattergrams of PM$_{2.5}$ against AOT (0.87μm) as total aerosol optical thickness (τ_λ) (left) and fine mode-optical thickness (τ_f) (right).

5. CONCLUSION

Long term simultaneous monitoring of aerosols and SPM provides us with typical aerosol types over an industrial city Higashi-Osaka according to the season as: anthropogenic aerosols in early winter, coarse dust in spring, and others. It can be seen that fine anthropogenic particles dominate at Higashi-Osaka even during dust events. It is found from NOAA/HYSPLIT particle transport analysis that air masses come from the China continent passing through East Asia especially over desert area. It is of interest to mention that dust events at Higashi-Osaka seem to be caused by a mixture of non-absorbing coarse dust and other small haze particles. As a result, the air quality is not good at Higashi-Osaka.

We have examined the relationship between the aerosol properties derived from radiometry and the particulate mass simultaneously obtained with the new SPM-613D instrument. Strong correlations were found between the PM2.5 concentrations and the fine mode aerosol optical thickness. Combining radiometric aerosol information and the surface-level particulate mass is useful when studying air quality and aerosol properties.

ACKNOWLEDGEMENTS

REFERENCES

Balasubramanian, R., W.-B.Qian, M.C. Facchini, and S.Fuzzi, Comprehensive characterization of PM2.5 aerosols in Singapore.,J. Geophys.Res., **108**, 4523,doi:10, 1029/2002JD002517, 2003.

Dubovik, O., A.Smirnov, B.N.Holben, M.D.King, Y.J.Kaufman, T.F.Ech and I.Slutsker, Accuracy assessments of aerosol optical properties retrieved from AERONET sun and sky-radiaometric measurements, *J. Geophys.Res.*, **105**, 9791-0806, 2000.

Dubovik, O. and M.D. King, A flexible inversion algorithm for retrieval of aerosols optical properties from sun and sky radiance measurements, *J. Geophys.Res.*, **105**, 20673-20696, 2000.

Eck, T.F., B.N. Holben, J.S. Reid, O. Dubovik, A. Smirnov, N.T. O'Neill, I. Slutsker, and S.Kinne, Wavelength dependence of the optical depth of biomass burning, urban, and desert dust aerosols. *J. Geophys. Res.*, **104**, pp. 31,333–31,349, 1999.

Goloub, P., J.L.Deuze, M. Herman, A.Marchand, D.Tanre, I.Chiapello, B.Roger, R.P.Singh, Aerosol Remote Sensing Over Land From The Spaceborne Polarimeter POLDER, *Proc.IRS2000*,115-116,2000.

Holben, B.N., T.F. Eck, I. Slutsker, D. Tanré, J.P. Buis, A. Setzer, E. Vermote, J.A. Reagan, Y. Kaufman, T. Nakajima, F. Lavenu, I. Jankowiak, and A. Smirnov, AERONET - A federated instrument network and data archive for aerosol characterization, *Rem. Sens. Environ.*, **66**, 1–16, 1998.

Mukai, S., I.Sano, and B.N.Holben, Aerosol properties over Japan by sun/sky photometry. *Water, Air and Soil Pollution* (in press).

Nakajima, T., Higurashi, A., Kawamoto, K., Penner, J.E., A possible correlation between satellite-derived cloud and aerosol microphysical parameters. *Geophys. Res. Lett.*, 28, pp.1171–1174, 2001

O'Neill, N.T., O.Dubovik and T.F.Eck, Modified Angstrom exponent for the characterization of submicrometer aerosols, *Appl. Optics*, **40**, pp.2368-2375, 2001.

O'Neill, N.T., T.F. Eck, A. Smirnov, B.N. Holben and S. Thulasiraman, Spectral discrimination of coarse and fine mode optical depth, *J. Geophys. Res.*, **108**, 4559, doi:10.1029/2002JD002975, 2003.

Sano, I., S. Mukai, Y.Okada, B.N.Holben, S.Ohta, and T.Takamura, Optical properties of aerosols during APEX and ACE-Asia experiments, *J.Geophys.Res.*, **108**, 8649, doi:10.1029/2002JD003263, 2003.

Smirnov, A., B.N.Holben, D.Savoie, J.M.Prospero, Y.J.Kaufmann, D.Tanre, T.F. Ech, and I.S.Slutsker, Relationship between column aerosol optical thickness and in situ ground based dust concentrations over Barbados, *Geophys. Res. Letter*, 27, 1643-1646., 2000.

Wang, J., and S.A.Christopher, Intercomparison between satellite-derived aerosol optical thickness and PM2.5 mass: Implications for air quality studies, *Geophys. Res. Letter*, **30**, 2095, doi:10, 1029/2003GL018174, 2003.