C-061 Network observation of dust and sandstorms in Northeast Asia and its applications to real-time analysis of movement and to the evaluation of the effects on the environment (Abstract of the Final Report)

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1. Introduction

Mineral dust in the atmosphere has huge effects on the global environment. The mineral dust generated from arid areas in the interior of China and Mongolia is known as kosa aerosol (Asian mineral dust). The number of kosa events occurring in Northeast Asia has increased significantly, as have the associated environmental and social effects for Northeast Asian countries. There have been reports on SO_2 deposition onto kosa particles during their transport to Japan. Other anthropogenic gases might also deposit onto the particles, especially with the increasing industrialization and automobile use in Northeast Asian countries. As the social-environmental implications of kosa events increase, the need to develop and improve the forecasting of kosa phenomena based on real-time monitoring by a network system has also increased. The National Institute for Environmental Studies is conducting lidar network observations of kosa phenomena in cooperation with universities and other research institutes and also as part of the Ministry of the Environment's dust-monitoring project. Expansion of these lidar network programs is underway between Japan and Mongolia as well as between Japan and China. It is essential that real-time data from the network are shared among Northeast Asian countries to solve the problems associated with kosa. In this study, quality assurance and quality control (QA/QC) methods as well as methods for data processing and a system for real-time data processing are developed to monitor kosa events. The methods and system are applied to the existing network data to demonstrate the processing performance as well as to further understand the movement of kosa phenomena. A feasibility study of a four-dimensional variational (4DVAR) data assimilation system for regional dust modeling based on the lidar observation data has shown that 4DVAR can play an important role in improving dust modeling. The development of a model that incorporates 4DVAR should improve the forecasting of kosa phenomena in Northeast Asia.

2. Research Objectives

Our research objectives are to develop QA/QC methods and a real-time data processing system for the lidar dust-monitoring network. Another objective is to use the lidar network monitoring data to establish an useful forecasting system that incorporates the development of a 4DVAR assimilation system for regional dust modeling. It will allow us to study the three-dimensional movement of dust using the quality assured network data. We also studied the reaction mechanisms between kosa and air pollutants, e.g., for the evaluation of SO_2 deposition and oxidation on kosa particles during their transport.

3. Research methods

Our research project monitors kosa phenomena using precise real-time observation instruments (primarily lidar). We defined the data products of the lidar network for the real-time validation and assimilation of dust transport models (originally CFORS model) and developed data processing methods for automated real-time processing. We also analyzed monitoring data to study the movement of kosa phenomena and the reactions between air pollutants and kosa particles. To study such reaction mechanisms, we conducted laboratory experiments using a cylindrical flow reactor system that was developed for this study. The reactor is consisted of 10 or 11 glass cylinders arranged according to experimental condition. The inner walls of the cylinders were frosted and could be coated with kosa materials.

4. Results and discussion

1) Typical kosa phenomena observed by network monitoring

Ten distinct kosa events were observed from meteorological surveillance in Japan. Several typical kosa events in 2006-2007 except 2008 were examined in terms of the mixing of air-borne pollutants and kosa particles in the air mass. We used hourly operational monitoring data from the air quality surveillance network in Japan to monitor SO_2, NO_X, O_X, and SPM. During one kosa event (8-9, April 2006), extremely high SPM concentrations were observed after the kosa passed a cold front. A number of surveillance stations from the Chugoku district toward the Chubu district observed SPM concentrations of more than 0.2 mg/m^3. The hourly concentrations for the other pollutants were below daily average levels for non-kosa periods. This event was so-called "un-polluted kosa". During another kosa event (18-19, April 2006), however, moderate SPM concentrations were observed under anti-cyclonic weather conditions. The SO_2 and NO_X concentrations during this kosa event were higher than they were in the previous event. This event was so-called “polluted kosa”.

Among the nine extracted events in 2007, the two typical events in April 1-3and May 8-9 are monitored. In the former case, the maximum value of the SPM concentration in each measurement site was high near that of the large-scale dust event on 8-9 April, 2006. In the observation period of high concentration SPM, high concentration of SO_2, NO_x, and O_3 was not observed. This event can be considered “un-polluted kosa”, and the latter event must be “polluted kosa”. Because of comprehension for pollution mechanism of kosa particles, it will be important to verify adhesion phenomena of such pollutants to the kosa particle with laboratory experiments and with chemical analyses of field samples. As the raw material ratios on the fundamental energy for each city function are different, the major sources of pollution in China must be different from those in Japan. We compared the compositions of carbonaceous materials and the ratios of stable carbon isotopes ($\delta^{13}C$) of soot, and the ratio of organic carbon (OC) to elemental carbon (EC) in each urban aerosol in Beijing and Tokyo. There are clear differences between the $\delta^{13}C$ of the atmospheric aerosols in the winter and spring seasons in Beijing and Tokyo; we consider this to be a good index for examining long-range transport aerosols including “polluted dust”.

2) Real-time data processing for the lidar monitoring network adapted to regional kosa modeling

If a kosa monitoring network using a lidar system covering Northeast Asia would be completed, the benefits would include improved accuracy of early warning and forecast modeling systems. We have begun to examine and develop what level of QA/QC and what lidar monitoring data assimilation protocol will be required to create a sufficient dust model. The sources of measurement errors with a two-wavelength (1064 nm, 532 nm) polarization (532 nm) lidar were studied. Methods for preventing daily maintenance problems were studied, as were possible methods for correcting problems in data processing. The insufficient removal of clouds from the lidar signals was also identified as a source of error. A cloud detection method using a 1064-nm signal was developed. A dust extinction coefficient for dust in an aerosol mixture was defined as the data product for model validation and assimilation. Data processing methods for both clear and cloudy conditions were developed. These methods were then combined to form an automated real-time data processing system. The real-time data processing was applied to kosa data from 2007. We analyzed Asian dust events in the spring of 2007 using the lidar data and CFORS. In the spring of 2007, several high oxidant events attributed to trans-boundary air pollution were reported. In the spherical aerosol extinction coefficients observed with lidar network, temporal variations and correlations between observation sites showing the characteristics of regional phenomena were observed. Comparing the dust extinction coefficient with CFORS without data assimilation, CFORS underestimates the dust emission in the event in the end of March to the beginning of April, and CFORS overestimates in the event in the end of May. For these three dust events as shown Figure1, the data assimilation experiments were carried out. The results demonstrated the data assimilation using the lidar network data is very useful for accurate estimation of dust emission areas and amounts.

We performed continuous observations with the lidar network, and also contributed to the dust information home page of Ministry of the Environment, Japan in real time. Based on the results, we improved the data analysis methods and the calibration method for the depolarization measurement.

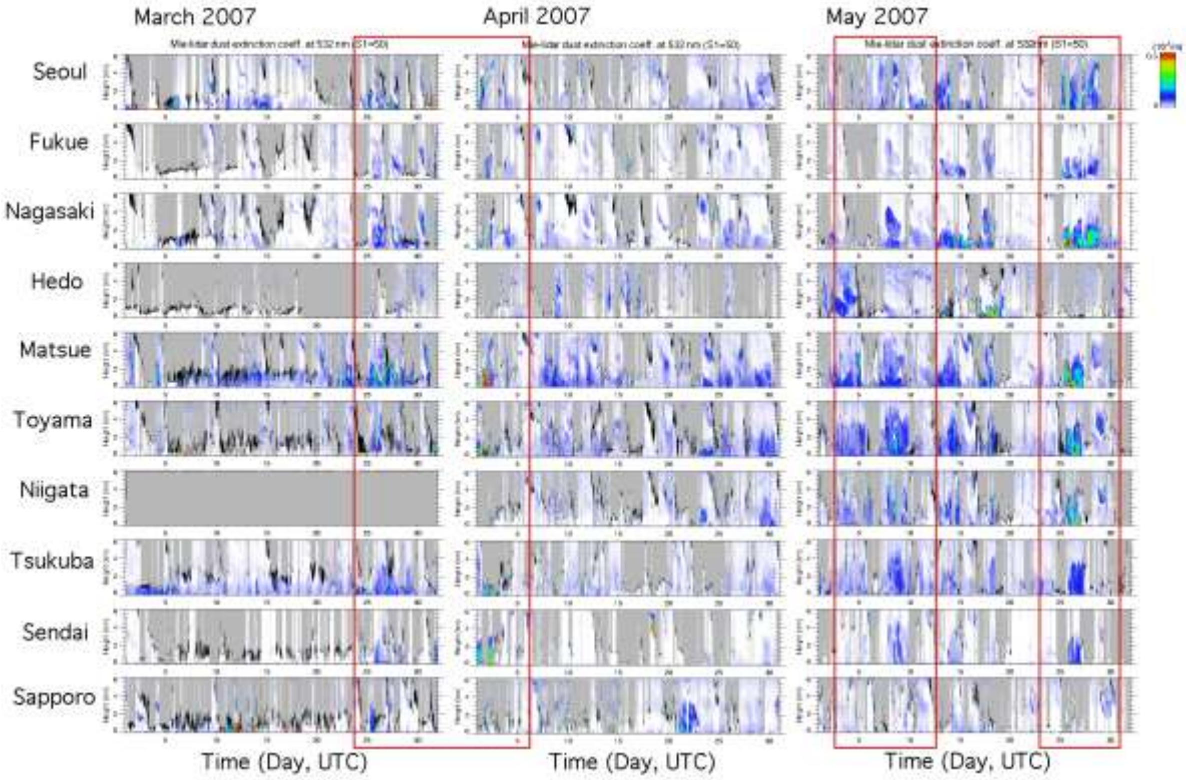

Figure 1 Lidar extinction coefficients in the DSS monitoring network by Mie-lidar system. Three square parts exhibit typical kosa events.

3) A feasibility study of a 4DVAR data assimilation system for regional dust modeling with real-time lidar network monitoring data

The lidar dust extinction coefficient is used directly to drive the 4DVAR evaluation of the cost function. We used the 4DVAR at four sites (Sendai, Sapporo, Toyama, and Tsukuba). We specifically examined the RAMS/CFORS-4DVAR capability in assessing the heavy dust episode that occurred on 30 April 2005 in northern Japan, especially at Sendai. The results revealed the following: (1) Estimates of dust concentrations were considerably improved using data assimilation (they had previously been underestimated), and the assimilated concentrations were in better agreement with lidar observations. (2) There was a 31% (3.2 Tg) increase in estimated dust emissions using data assimilation, especially over the Mongolian region. (3)A four-dimensional variational (4D-VAR) data assimilation system for a regional dust model has also been applied to a heavy dust event which occurred between 30 March to 4 April 2007 over eastern Asia. The vertical profiles of the dust extinction coefficients derived from 5 NIES LIDAR observation sites are directly assimilated. The assimilation resulted in increase of dust emissions over the Gobi Desert and Mongolia considerably; especially between 29 and 30 March. The heavy dust event is caused by the heavy dust uplift flux over the Gobi Desert and Mongolia during those days. We obtained the total optimized dust emissions of 57.9 Tg (57.8 % larger than before assimilation).

As a feasibility study of a 4DVAR data assimilation system for regional dust modeling with real-time network monitoring data, the following system has been developed. We present detailed 3D structure of Asian dust outflow from a dust source region to the northwestern Pacific ocean retrieved by NASA/CALIOP onboard CALIPSO and results simulated by a four-dimensional variational (4DVAR) data assimilation version of a dust transport model (RC4) based on the NIES Lidar network in Figure 2. The modeled and CALIOP dust extinction showed good agreement, both for horizontal scale (600-1200 km) and vertical depth (1600-3600 m) near the dust source regions. Cross-section analyses of CALIOP and RC4 assisted by forward trajectory revealed that dust from the Gobi Desert on 5 May traveled approximately 1000-1500 km/day to the east and passed over Japan on 8 May. The elevated dust subsequently passed to the Pacific Ocean while maintaining the major dust layer height of 2500-4000 m MSL trapped within the potential temperature of 302-306 K. Results of our analyses demonstrate the importance of integration of CALIOP measurement and the dust model for clarifying the overall structure of an Asian dust event.

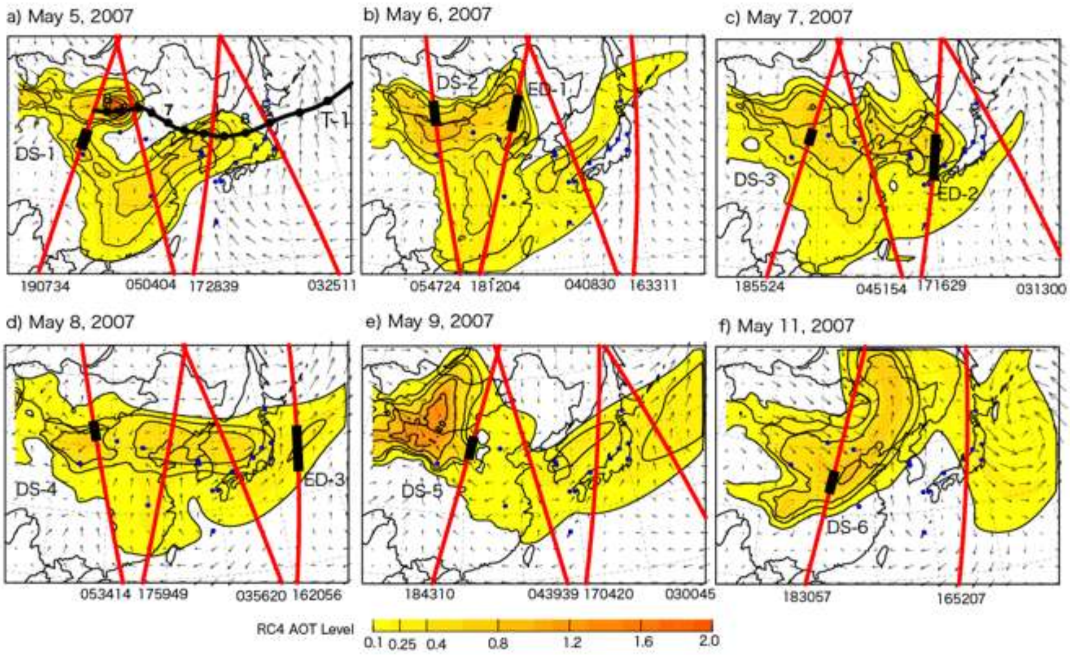

Figure 2 CALIPSO orbit paths (red) and RC4 dust AOT (color). Vectors indicate the wind field at 140 m AGL. Six-digit numbers at the bottom of each figure show the CALIPSO path time (HHMMSS). Dots show Lidar observation sites.

4) Experimental verification of the reaction of kosa particles with SO_2 and/or HF, $(COOH)_2$ gases

During transport, kosa particles can mix with the polluted air masses that contain primary pollutants such as SO_2 and HF emitted from coal combustion and secondary pollutants such as HNO_3, O_3, and oxalic acid generated during photochemical smog. Then, acidic SO_2 can be adsorbed and neutralized by kosa; in addition, deposition of SO_2 onto kosa particles can be followed by oxidation of sulfite to sulfate.

SO_2 reacted rapidly with deposited kosa (loess) particles, especially under humid conditions.

The calculated deposition velocity (V_d) changed dramatically at an R.H. value of ca. 60 %. By assuming that the deposition of SO_2 on the yellow sand particle was a pseudo-first-order reaction, we calculated the initial SO_2 uptake coefficient calculated with a geometric surface area. The coefficient, which depended on the SO_2 concentration at the reactor outlet, was estimated to be on the order of 10^{-4}, and, like V_d, it changed at R.H. = 60 %. These marked changes at R.H. = 60 % were probably caused by the formation of a bulk water film on the loess particles (Table 1); the formation of the water film substantially changed the properties of the mineral dust surface. An SO_2 uptake coefficient that takes R.H. into consideration should be used in atmospheric models to predict the dynamics of the interaction between kosa particles and SO_2.

Table 1 Calculated slope *k* and initial uptake coefficients (γ_0) among *R.H.* < 5 － 80 %

Conditions(*R.H.*)	5 %	10 %	20 %	30 %	40 %	50 %	60 %	70 %	80 %
Slope *k*	5.35	5.20	5.26	5.48	5.27	5.29	6.22	6.26	6.84
Uptake Coef.γ_0 ($\times 10^{-4}$)	1.16	1.13	1.14	1.19	1.14	1.15	1.35	1.36	1.48

Exposure experiments of deposited kosa(loess) particles to SO_2 and HF were performed by using the cylindrical flow reactor with 10 series glass cylinders. The deposition velocities of HF and SO_2 in the exposure to the mixed gases were estimated from the value of reaction probability. Comparing the deposition velocities in the exposure to single component gas with those in the exposure to mixed gas, it can be considered that the competitive depositions of SO_2 and HF would occur in the latter exposure. The SO_2 deposition was reduced in the presence of HF, possibly owing to blockage of reactive sites on the particle surfaces by the deposition of high concentration HF. However, because of high reactivity of HF, deposition of HF onto loess particles could occur even if SO_2 level were more significant.

Table 2 U_{dep} and *ORS* values for exposure experiments

	U_{dep} (10^9 μmol s mg^{-1} cm^{-2})		*ORS* (%)
	SO_2	$(COOH)_2$	
SO_2	0.0167±0.0018	-	48.9±6.2
$(COOH)_2$	-	0.0354±0.0019	-
SO_2 + $(COOH)_2$	0.0127±0.0006	0.0353±0.0028	46.0±5.4

The deposition coefficient of $(COOH)_2$ was higher than that of SO_2 because $(COOH)_2$ can be deposited at any of a number of sites in the cylinders, whereas SO_2 deposition part is limited. In the presence of $(COOH)_2$, the deposition coefficient of SO_2 was approximately 24 % lower than in the absence of $(COOH)_2$, but the oxidation ratio of sulfur remained almost the same as that observed upon exposure to SO_2 alone (Table 2). Therefore, our results indicate that gaseous $(COOH)_2$ in the atmosphere can suppress the deposition of SO_2 on kosa particles under dry conditions.

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