Evolution of $PM_{2.5}$ Components in the Long-range Transport Plume accompanied with the Southward Asian Continental Outflow

Ming-Tung Chuang[1] , S-Yuan Joshua Fu[2], Ji-Cheng Carey Jang[3], Pei-Cheng Ni[4], Chang-Chuan Chan[5], Chung-Te. Lee[1]

[1]*Graduate Institute of Environmental Engineering, National Central University Jhongli, Taoyuan, Taiwan*

[2]*Department of Civil and Environmental Engineering, University of Tennessee, Knoxville, Tennessee, U.S.*

[3]*Office of Air Quality Planning and Standards United States Environmental Protection Agency*

[4]*CTCI Corporation, Taipei, Taiwan*

[5] *Institute of Occupational Medicine and Industrial Hygiene, National Taiwan University, Taipei, Taiwan*

1. Introduction

The main media of long-range transport originating from Asia continent is that Asia high pressure system moves eastward or southward. Dignon and Hameed (1998) has pointed out that Asia had become the area that emitted most sulfur globally. The main cause was the sustained increasing consumption of sulfur in China. In the last few years, there have been several publications that observed and estimated the contribution of acid deposition from China on a level from 20% to 50% (Kitada et al., 1992; Ichikawa and Fujita, 1995; Chung et al., 1996; Chang et al., 2000; Holloway et al., 2002).

In this study, the events occur in Taipei by long-range transport from Asian continent have been defined as HPP (Chuang et al., 2007). 53 events were observed in Taipei from March 2002 to February 2005. For HPP and non-HPP days observed air quality at TAS (in Fig. 1) is in Table 1. Especially, $PM_{2.5}$, $PM_{2.5}$ sulfate hourly average concentrations are 54.1 and 12.0 μ g m^{-3} respectively, which are much higher than 22.6 and 4.7 μ g m^{-3} that are observed on non-HPP days. It appears that the influence of pollutants originating from Asian continental outflow to downwind area is very significant.

Table 1. Basic statistics of aerosol characteristics and weather parameters for high pressure pushing (HPP) weather pattern and non-HPP time period from March 2002 to February 2005.

	Monitoring item	Effective hours	Avg.	s.d.	Max.	Min.
HPP	$PM_{2.5}$ (μg m^{-3})	937	54.1	23.8	148.3	6.5
	$PM_{2.5}$ sulfate (μg m^{-3})	819	12.0	7.2	55.1	1.3
	$PM_{2.5}$ nitrate (μg m^{-3})	806	2.6	2.0	14.9	0.4
	$PM_{2.5}$ OC (μg m^{-3})	803	4.5	1.0	13.9	1.0
	$PM_{2.5}$ EC (μg m^{-3})	524	1.7	0.8	8.6	1.0
non-HPP*	$PM_{2.5}$ (μg m^{-3})	8250	22.6	11.4	64.5	5.0
	$PM_{2.5}$ sulfate (μg m^{-3})	7662	4.7	3.5	28.5	0.4
	$PM_{2.5}$ nitrate (μg m^{-3})	6717	1.4	1.1	20.2	0.4
	$PM_{2.5}$ OC (μg m^{-3})	6747	3.5	1.7	18.0	1.0
	$PM_{2.5}$ EC (μg m^{-3})	2142	1.7	1.2	13.9	1.0

*Non-HPP time period is for days excluding June, July, August, and events time defined in Chuang et al. (2007).

2. Study area and monitoring data and model setup

From Figure 1, it is seen that Taiwan is located at the West Pacific, with 200 km wide Taiwan Strait, opposite to southeast of China mainland.

The greater Taipei area is located in the northern Taiwan which is the firstly influenced by long-range transport originating from the north.

In this study, the fifth-generation Pennsylvania State University-National Center for Atmospheric Research Mesoscale Model (MM5, Grell et al., 1994) is utilized to generate meteorological data as input for chemical model. For chemical model, the Models-3 Community Multiscale Air Quality (CMAQ, version 4.4, Byun and Ching, 1999; U.S.EPA, 1999) modeling system is utilized. In order to save computation time and reduce the size of output files, 25-layer output of meteorological model is transformed into 14 layers as inputs for CMAQ.

Basic emission data of domain 1 and domain 2 is from Asian emission database estimated by Streets et al. (2003). In order to reflect rapid economic development in China in the last few years, we refer to Hao and Wang (2005) and Heo and Feng (2005)'s statistics to estimate the anthropogenic growth factor from 2001 to 2004. Yearly growth factors of 1.13 and 1.19 is used for 2001 to 2003 and 2003 to 2004 respectively. Biogenic source is assumed to remain constant. Emissions of domain 3 and domain 4 are from TEDS6.1 (Taiwan Emission Database System for 2003, Fu et al., 2007) which is then spatially and temporally distributed by SMOKE (Houyoux and Vukovich, 1999) to estimates grid/hourly emission rate. It is expected that emission was not fluctuant too much from 2003 to 2004. Besides, it is noted that TEDS is the only emission database available in Taiwan.

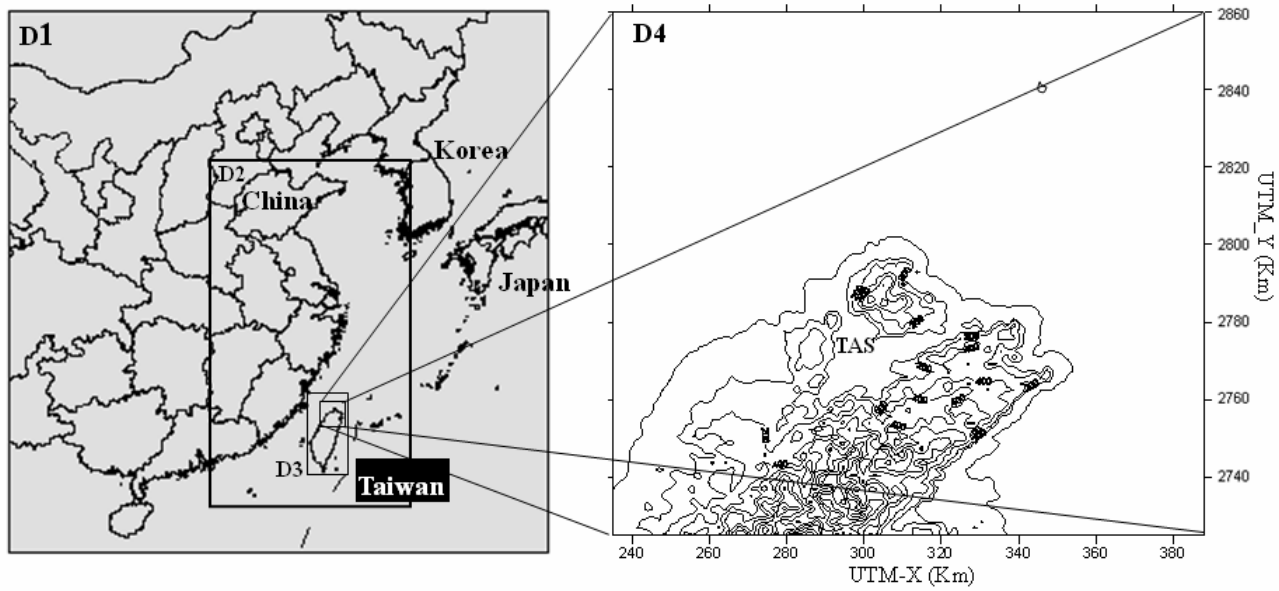

Fig. 1. Model simulation domains (D1 to D4) and geographical locations of the Taipei aerosol supersite (TAS).

3. Results and discussion

We select backward trajectory from 16:00 (at TAS, number 31 in Figure 2) on 20 December 2004. On that day, a high pressure system and $PM_{2.5}$ plume moved southward simultaneoutly as shown in Figure 3. Figure 4 characterizes the variation of percentile of $PM_{2.5}$ components along the path of moving plume shown in Figure 2.

4.1 $PM_{2.5}$ nitrate

As $PM_{2.5}$ plume moves southward, $PM_{2.5}$ nitrate concentration decreases. The ratio of $PM_{2.5}$ nitrate to $PM_{2.5}$ decreases during long-range transport from 25% in Shanghai to 1% on the waters of north Taiwan. In the morning on 20 December, $PM_{2.5}$ nitrate is only about 1 μ g m^{-3}. Kin and Park (2001) found that NO_3^- was quite low in fine mode particles transported from Asia continent. This explains that the lifetime of NO_3^- in fine mode particles is very short. It is known that NO_2 can react with NO_3 radical and form N_2O_5 at night and then N_2O_5 may react with H_2O and form HNO_3 (Waston et al., 1994). However, N_2O_5 and NO_3 radical are only a few pptv at night on 19 December (unshown). When $PM_{2.5}$ nitrate decreases, HNO_3 apparently increases. It is concluded that HNO_3 is mainly evaporated from $PM_{2.5}$ nitrate. The high volatility of NH_4NO_3 makes itself dissociate into HNO_3 and NH_3 at higher temperature (Stelson and Seinfeld, 1982). NH_3 reacted with H_2SO_4 transformed from SO_2 would form NH_4HSO_4 or $(NH_4)_2SO_4$. It can be related to raise of $PM_{2.5}$ sulfate along the transport of $PM_{2.5}$ plume.

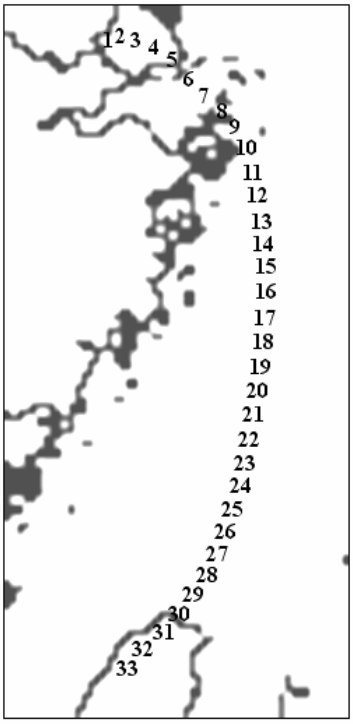

Fig.2. Hourly backward trajectory path of southward $PM_{2.5}$ plume

4.2 $PM_{2.5}$ sulfate

SO_2 can either react with HO_2 and form HSO_3- or oxidize to SO_3. HSO_3^- and SO_3 can react with H_2O_2 and H_2O (unshown, relative humidity was around 80% in the plume) respectively and form H_2SO_4, which reacts with NH_3 (evaporated from $PM_{2.5}$ nitrate) and forms $(NH_4)_2SO_4$ or NH_4HSO_4 through heterogeneous reaction. Thus $PM_{2.5}$ sulfate gradually increases in the early morning on 20 December. On the daytime on 20 December, less $PM_{2.5}$ sulfate is formed since $PM_{2.5}$ nitrate is low and less NH_3 is evaporated. Therefore, $PM_{2.5}$ sulfate slightly decreases due to dilution after 08:00 on 20 December. Relative to ammonium nitrate, ammonium sulphate is more stable. From Figure 4, it is seen that percentile of $PM_{2.5}$ sulfate increases from 10% in Shanghai at 10:00 19 December to 35% while the plume is near Taipei at 15:00 on 20 December.

4.3 $PM_{2.5}$ ammonium

Jordan et al. (2003) think that in the long-range transport plume there should be enough nss-SO_4^{2-} to react with NH_4^+. This implies that NH_3 evaporated from ammonium nitrate would transfer into sulfate ammonium. In Figure 4, the percentile of $PM_{2.5}$ ammonium remains around 13%. $PM_{2.5}$ ammonium may in the form of NH_4NO_3 or $(NH_4)_2SO_4$ or NH_4HSO_4 in source area. Since NH_3 can evaporate from NH_4NO_3 during long-range transport where SO_4^{2-} or HSO_4^- is present, it can be derived that $PM_{2.5}$ ammonium can be in the form of $(NH_4)_2SO_4$ or NH_4HSO_4 mostly when the plume is near Taiwan. Further, it is expected that the percentile of $PM_{2.5}$ ammonium remains nearly constant.

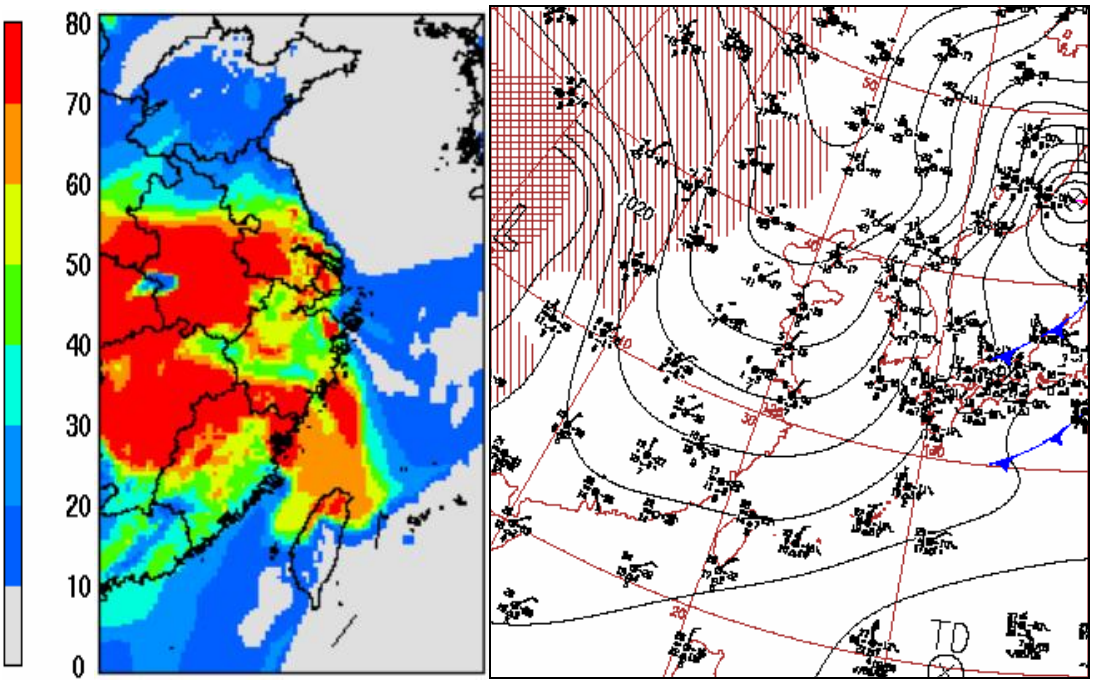

Fig. 3. Simulated near-surface $PM_{2.5}$ concentration of domain 2 and surface weather map (announced by JMA, Japan Meteorology Agency, http://www.jma.go.jp/jp/g3/) at 14:00 on 20 in December 2004.

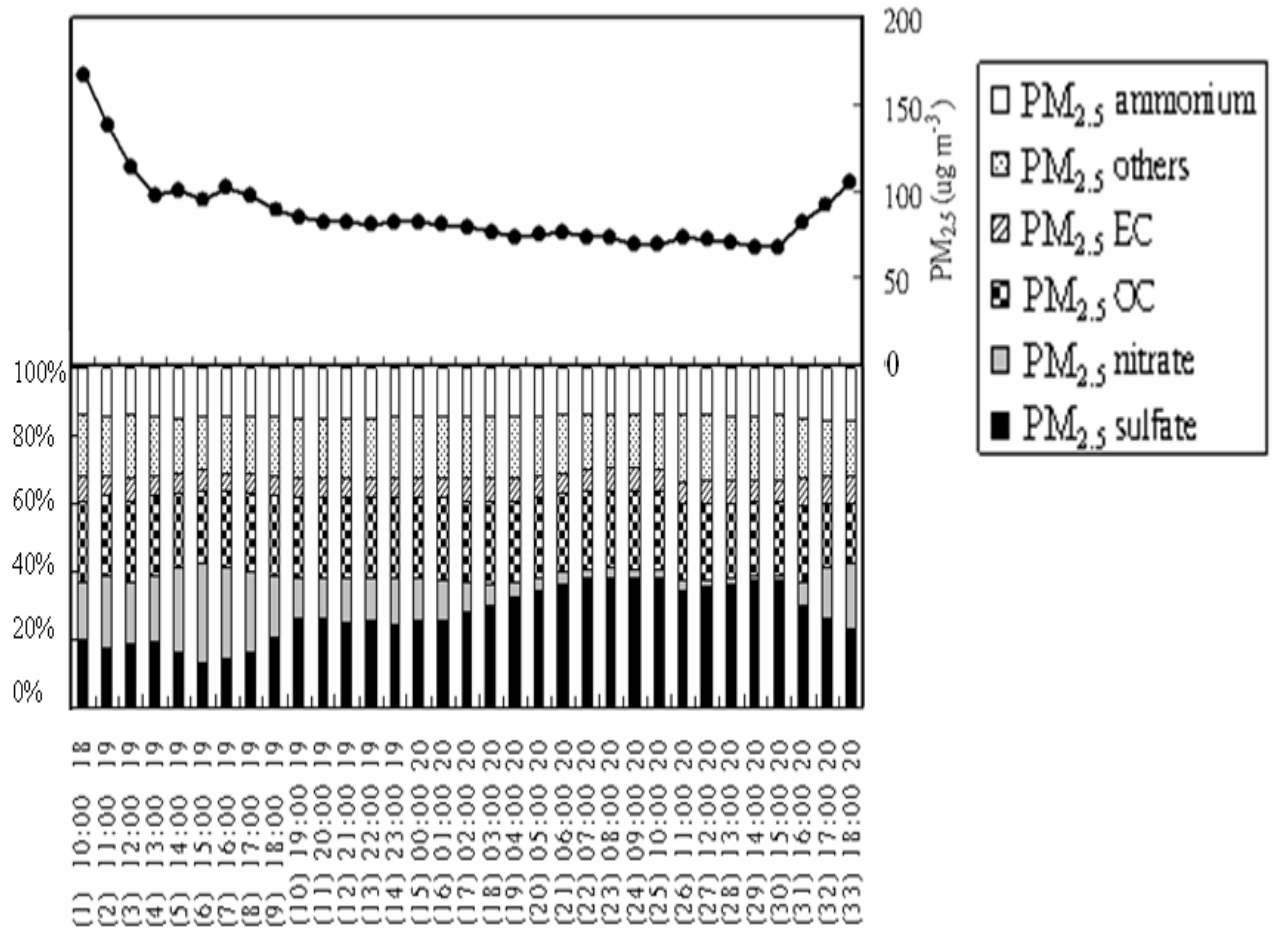

Fig. 4. The $PM_{2.5}$ mass concentration and the percentage of $PM_{2.5}$ chemical components in $PM_{2.5}$ during the path of Fig. 2

4.4 $PM_{2.5}$ carbonaceous content

$PM_{2.5}$ OC and $PM_{2.5}$ EC slowly decrease in the moving plume. Notably, $PM_{2.5}$ OC decreased faster than $PM_{2.5}$ EC. Therefore OC/EC ratio decreased from 4.1 near the outlet of Changjiang River to 2.7 in Taipei. From Shanghai to Taipei, the ratio of $PM_{2.5}$ OC to $PM_{2.5}$ just slightly decreases from 24% to 21%. In contrast, percentile of $PM_{2.5}$ EC remains nearly the same, 6%.

4. Summary

Pollutants from Asia continent would influence air quality in Taipei countries by means of southward long-range transport. Especially $PM_{2.5}$ sulfate is the major chemical components. A classical long-range transport case from Asia continent to Taiwan occurred on 20 December 2004 is simulated in this study. Simulation results show that the percentage of OC in $PM_{2.5}$ only slightly decreases from 24% to 21%. However, percentage of nitrate $PM_{2.5}$ decreases from 25% to a very lower value 1%. Relatively, the percentage of $PM_{2.5}$ sulfate increased from 10% to 35%.

5.Acknowledgements

We would like to express our deep gratitude to the Taiwan National Science Council, Taiwan Environmental Protection Administration (TWEPA) and U.S EPA for providing financial support to this study. We would also like to thank Dr. Streets for providing Asian emission database. In addition, we acknowledge NCEP (National Centers for Environmental Prediction) and DBAR (Data Bank of Atmospheric Research, managed by the Department of Atmospheric Sciences, National Taiwan University) for sharing the atmospheric monitoring data used in this study.

6. References

Byun, D.W., Ching J. K.S., Science Algorithm of the EPA Models-3 Community Multiscale Air Quality(CMAQ) Modeling System", EPA/600/R-99/030, USEPA/ORD, March 1999.doi:10.1029/2002JD003116.

Chang, K.H., Jeng, F.-T., Tsai, Y.L., Lin, P.L., 2000. Modeling of long-range transport on Taiwan' s acid deposition under different weather conditions. Atmospheric Environment 34, 3281–3295.

Chuang, M.T., Lee, C.T., Chiang, P.C., Wang, C.F., Chang, Y.Y., 2007. The effects of synoptical weather pattern and complex terrain on the formation of aerosol events in the greater Taipei area, submitted to Science of the Total Environment.

Chung, Y.S., Kim, T.K., Kim, K.H., 1996. Temporal variation and cause of acidic precipitation from a monitoring network in Korea, Atmospheric Environment 30, 2429-2435.

Dignon, J., Hameed, S., 1989. Global emissions of nitrogen and sulfur oxides from 1860 to 1980. Journal of Air Pollution Control Association 39, 180-186.

Fu, J.S., Yeh, F.L., Chien, H.C., Carey, R.J. C., Chuang, M.T., 2007. Environmental Technology Implementation: Modeling air quality impacts on Taiwan island. International Journal of Environmental Technology Management. (accepted)

Grell, G. A., J. Dudhia and D. R. Stauffer, 1994: A description of the fifth-generation Penn State/NCAR mesoscale model (MM5). NCAR Technical Note, NCAR/TN-398+STR, 117 pp.

Hao, J., Wang, L., 2005. Improving urban air quality in China: Beijing case study. J. Air & Waste Manage. Assoc. 55, 1298-1305.

Heo, E., Feng, Y., 2005. Recent development of energy use in China at the industrial sector level. http://www.cenet.org.cn/cn/CEAC/2005 in /zyhj011.doc

Holloway, T., Levy II, H., Carmichael, G., 2002. Transfer of reactive nitrogen in Asia: development and evaluation of a source – receptor model. Atmospheric Environment 36, 4251 – 4264.

Houyoux, M., Vukovich, J., 1999. Updates to the Sparse Matrix Operator Kernel Emissions (SMOKE) modeling system and integration with Models-3, Presented at The Emissions Inventory : Regional Strategies for the Future Conference, Air & Waste Management Association, Raleigh, NC.

Ichikawa, Y., and S. Fujita, 1995: An analysis of wet deposition of sulfate using a trajectory model for east Asia. Water, Air, Soil Pollut., 85, 1927–1932.

Jordan, C.E., Dibb, J.E., Anderson, B.E., Fuelberg, H.E., 2003. Uptake of nitrate and sulfate on dust aerosols during TRACE-P. Journal of Geophysical Research 108(D21), 8817, doi:10.1029/2022JD003101.

Kitada, T., and Tanaka, K., 1992. Simulated semi-global scale transport of SO_2 and $SO^{=}_4$ from East Asia to the Northern Pacific in spring season: The role of low and high pressure systems. Air Pollution Modeling and Its Application IX, H. van Dop and G. Kallos, Eds., Plenum Press, 445–454.

Kleinman, L.I., 1991. Seasonal dependence of boundary layer peroxide concentration: the low and high NOx regimes J. Geophys. Res. 96, 20721-20733.

Stelson, A.W., Seinfeld, J.H., 1982. Relative humidity and temperature dependence of the ammonium nitrate dissociation constant. Atmos. Environ. 16, 983-992.

Streets, D.G., Bond, T.C., Carmichael, G.R., Fernandes, S., Fu, Q., He, D., Klimont, Z., Nelson, S.M., Tsai, N.Y., Wang, M. Qm., Woo, J.-H., Yarber, K.F., 2003. An inventory of gaseous and primary aerosol emissions in Asia in the year 2000. Journal of Geophysical Research 108 (D21), 8809.

Waston, J.G., Chow, J.C., Lurmann, F.W., Musarra, S.P., 1994. Ammonium nitrate, nitric acid, and ammonia equilibrium in wintertime Phoenix, Arizona. J. Air & Waster Manage. Assoc. 44, 405-412.