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ATMOSPHERIC PARTICLES IN HOCHIMINH
SIZE DISTRIBUTION OF WATER-SOLUBLE INORGANIC IONS

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Abstract

Size distributions of water-soluble inorganic ions in atmospheric particles were studied in HoChiMinh City. Fine particles (< 2.1 μm) contributed 40% - 50% of the TSP mass for roadside air in any season, 40% for urban background air in the dry season and 20% for urban background and ambient air in the rainy season. The fraction at $0.65 - 1.1$ μm or $0.43 - 0.65$ μm was the most acidic. Sulphate was predominant water-soluble inorganic ion, followed by calcium. Sulphate was bimodal at $0.43 - 1.1$ μm and $3.3 - 4.7$ μm. In the dry season the fine mode was much higher than the coarse mode, but the two modes were comparable in the rainy season. Nitrate was mainly in the coarse particles. Ammonium ions were found mainly in the fine particles of the dry season. They were very few or not found in particles of the rainy season. The correlation between ions showed that sulphate in fine particles of the dry season was associated mainly with ammonium in the form acidic ammonium salts, while in the coarse particles it was mainly bound in $CaSO_4$.

Keywords: Andersen Impactor, atmospheric particles, size distribution, trajectory analyses, water-soluble inorganic ions.

1. Introduction

Evidence from human exposure to particles strongly suggests that fine ambient particles are closely correlated with observed adverse health effects (Thurston et al., 1994; R. Peled et al., 2005). Increase of fine particles increased mortality and morbidity (Ozkaynak et al., 1987; L.T. Gonçalves et al., 2005). Toxic mechanisms of particles associated with their composition. The substances of most concern are acidic sulphates H_2SO_4 and NH_4HSO_4. Not only a total mass, but also a size distribution and composition of particles are important for evaluating toxic effects of particles. For this reason much interest has been paid to size distribution. Size distribution and composition of particles depend on many factors, such as sources, meteorological factors, and topography of site (Kleeman et al., 2000; Funasaka et al., 2003; Hazi et al., 2003; Lestari et al., 2003; Cabada et al., 2004; Xiu et al., 2004; Yu et al., 2004; Sillanpää et al., 2005; Rosenbohm et al., 2005).

HoChiMinh City is the biggest commercial and industrial center in Vietnam, with more than 31000 enterprises, 2.3 million motorbikes, 0.2 million automobiles and a population of about 8 million persons. Particles are the most serious air pollutant in the city. In this research a distribution of water-soluble inorganic ions (SO_4^{2-}, NO_3^-, Cl^-, NO_2^-, NH_4^+, Na^+, K^+, Ca^{2+}, and Mg^{2+}) in particles was measured as a function of size and season. The obtained set of data was then examined by air mass trajectory analyses to identify source regions. .

2. Experimental

2.1. Sampling sites

Three sampling sites were a) University of Natural Sciences (VNU - about 20 meters above the ground level and 35 meters from the street), b) Department of Science and Technology of HoChiMinh

(DOSTE - about 1.5 meters above the ground level and 5 meters from pavement), c) residential area in My Tho (My Tho - about 4 meters above the ground level and 10 meters from small road). The first two sampling sites located in the center of HoChiMinh City. The first one represents urban background air and the second one represents roadside air. My Tho is small agricultural town in Mekong delta, about 60 km from the center of HoChiMinh City. My Tho is a clean area and was chosen for a comparison. The climatic characteristics of the sampling periods are given in Table 1.

Table 1. Climatic characteristics at the sampling days.

	Sample site	Season	Sampling period	Temperature	Humidity	Rain condition	Wind direction	Wind speed
1	VNU	dry	Feb 5-6/2005	27	69	No rain	NE	13
2	DOSTE	dry	Feb 5-6/2005	27	69	No rain	NE	13
3	VNU	rainy	July 24-25/2006	27	79	No rain	SW	13
4	DOSTE	rainy	July 30-31/2006	26	86	Short small rain	SW	18
5	MY THO	rainy	July 31-August 1/2006	25	99	Long small rain	SW	19

2.2. Sampling procedure

Eight-stage Andersen impactor (Dylec. Co. Ltd., AN-200) was used to collect particles. The sampling was carried out for 24 hours at a flow rate of 28.3 L/min. Particles were collected on silica filters with a diameter of 8 cm. The sampling system was protected from rain. The Andersen sampler has 8 size-fractionated stages with nominal cut-off size of 9 (inlet), 5.8, 4.7, 3.3, 2.1, 1.1, 0.65, 0.43 μm and a back-up filter. Particles with a size less than 2.1 μm was considered as fine particles, and those bigger than 2.1 μm was considered as coarse particles. The samples were numbered by joining the name of the sampling site and season. VNU-Rainy indicates the sample collected in the rainy season at VNU, DOSTE-Dry indicates sample collected in the dry season at DOSTE. Particles were collected also by high-volume sampler for analyzing by XRD to identify chemical composition.

Before sampling, the filters were preheated in an electric furnace at 850°C for 4h and then conditioned in desiccators for 24 hours before weighing. All filters were kept in clean plastic bags that were sealed in aluminum envelops before and after sampling, and during transportation. Collected samples were stored in refrigerator at 5°C until chemical analysis.

2.3. Chemical analyses

Half of each filter was used for analyses of ions. The filters were cut into pieces and extracted ultrasonically for 20 minutes by 10 ml of pure water in clean plastic vials. A pH meter B-212 (Horiba) and a conductivity meter B-173 (Horiba) were used to record pH and conductivity of the extracts. The pH and conductivity meters were calibrated before measurements. The extracts were analyzed by using an ion chromatography system Dionex DX 500 IC equipped with auto-suppressed conductivity detector. Dionex CS12A and Dionex AS12A columns were used for analyses. The detection limits were 3, 6, 10, 30 and 9 ppb for Cl^-, NO_2^-, NO_3^-, PO_4^{3-} and SO_4^{2-}, and 0.02, 2, 0.2, 0.3 and 0.5 ppb for Na^+, NH_4^+, K^+, Mg^{2+} and Ca^{2+}, respectively.

2.4. Air mass trajectory analyses

Air mass back trajectories for the sampling days were used to identify source regions of collected particles. Air mass back trajectories were computed using Transport and Dispersion Model "Hybrid Single-Particle Lagrangian Integrated Trajectory version 4" (HYSPLIT 4) developed by the US National Oceanic and Atmospheric Administration (NOAA) Air Resource Laboratory (http://www.arl.noaa.gov/ready/hysplit4.html). The trajectories were computed for each quarter of day from the beginning of sampling. Five 5-day backward trajectories were computed for each sampling day using three starting heights (10 m, 25 m, and 50 m above ground level). Each set of trajectories then was analyzed to determine source regions of the majority of the trajectories in each set.

3.   Result and discussion

3.1. Size distribution of mass

Mass concentrations of TSP are given in Table 2. TSP clearly decreased in the rainy season. The fine particles contributed more than 40 % of TSP collected in the dry season at DOSTE and VNU as seen from an accumulative percentage of PM mass given in Fig. 1. Although the masses of particles in the rainy season were comparable from site to site, there is a big difference in their size distributions. PM at DOSTE (road side air) contained 50% of fine particles, while the samples at VNU (urban background) and My Tho (ambient air) contained only 20% of fine particles.

Fig. 1. Accumulative percentage of particles

Table 2. Mass concentration of TSP ($\mu g/m^3$).

VNU		DOSTE		My Tho
Dry season	Rainy season	Dry season	Rainy season	Rainy season
143	64	236	49	46

3.2. Acidity and ion balance of the extracts

Fig. 2 shows pH of the extracts. The coarse fractions of all samples were almost neutral, with pH between 6 -7. The fine fractions collected in the rainy season at My Tho and VNU (ambient air and background air) were almost neutral also, while the fine fractions of DOSTE (roadside air) were more or less acidic depending on size. However a common feature characterized for all sample sets is that the fractions at 0.65 -1.1µm was almost the most acidic.
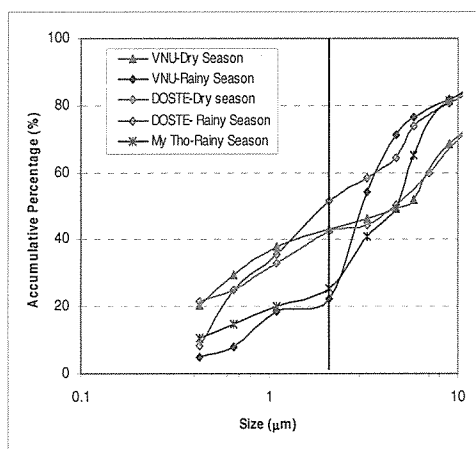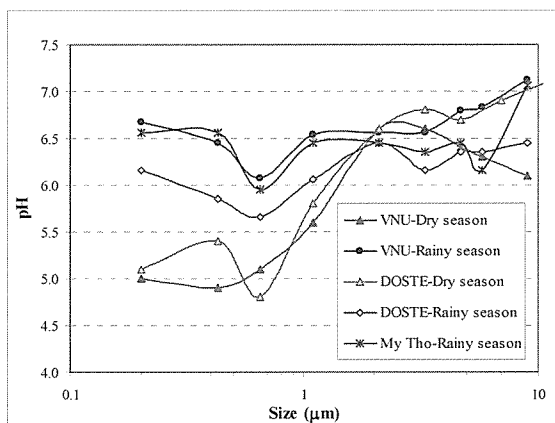

Fig. 2. pH of extracts of fractionated particles

Table 3 gives an ion balance Σcation / Σanion, where the amounts of ions are expressed in neq/m^3. The ion balance showed an anion deficiency for most of the coarse fractions. The same phenomenon was seen for most of the fine fractions collected in the rainy season, except for the most acidic fractions at 0.65 -1.1 µm at DOSTE (roadside air) where a cation deficiency was found instead. The cation deficiency was also found for all the fine fractions of the dry season. Similar results were obtained by Yao et al. (2003) and Rocha et al. (2005) for particulate pollutants in Beijing and Brazil.

Table 3. Ion balance Σcation / Σanion for the extracts of particles.

Fraction	My Tho Rainy season	VNU Dry season	VNU Rainy season	DOSTE Dry season	DOSTE Rainy season
>9	1.46	1.2	1.7	2.90	2.53
5.8 - 9	1.55	1.7	1.3	1.62	1.53
4.7 - 5.8	1.90	1.2	1.3	1.23	1.34
3.3 - 4.7	1.67	0.9	1.1	1.02	1.83
2.1 - 3.3	1.40	1.8	0.9	0.96	1.14
1.1 - 2.1	1.53	0.5	1.2	0.39	0.95
0.65 - 1.1	1.17	0.7	1.5	0.73	0.76
0.43 - 0.65	1.82	0.6	1.1	0.60	1.31
< 0.43	1.81	0.6	1.4	0.90	0.89

3.3. Size distribution of ions

Fig. 3 shows the concentrations (in µg/m^3) of common inorganic water-soluble ions in each fraction. The total concentrations of ions in the dry season were much higher than those in the rainy season, i.e. air in the rainy season was cleaner than in the dry season. The mass contribution of inorganic water-soluble ions in fine particle was 16-20% in the dry season and 24 – 50% in the rainy season.
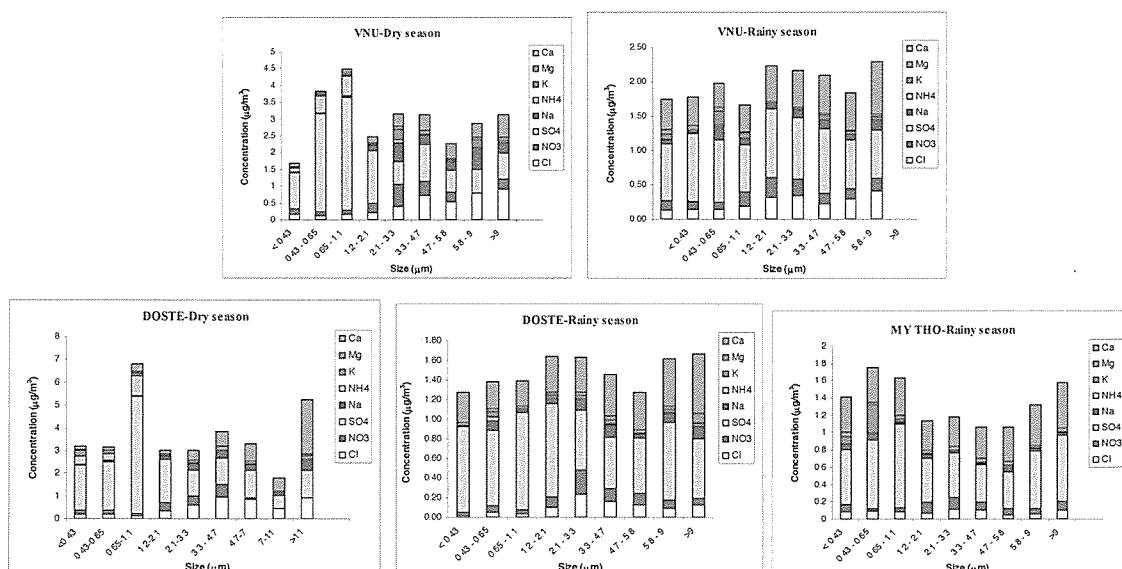

Fig. 3 Concentrations of ions as a function of size.

Sulphate was the most abundant among ions in any fraction. Table 4 gives concentrations of sulphate and its mass contribution in the fine and course particles. Fine particles in the dry season contained much more sulphate than coarse particles, while in the rainy season sulphate was comparable in the coarse and fine fractions. However the contribution of sulphate in particle mass for fine particles was always higher then for coarse particles.

In fine particles of the dry season ammonium was the second abundant after sulphate. However in fine particles of the rainy season and coarse particles of any season the second abundant ions were calcium. Nitrate contributed a much smaller portion of particle mass in comparison with sulphate. Sodium and chloride in particles of the dry season were much higher than those of the rainy season. Sodium was chosen as an indicator for sea-salt aerosols assuming that the composition of sea-salt aerosols is the same as of the sea water. The equivalent ratio of Cl^-/Na^+ in sea water is about 1.1. Among five set of samples three sets showed the ratio Cl^-/Na^+ of 1.3 to 1.6, the two others showed the values of 0.87 and 1.1. Therefore sea-salt aerosols are the main but not the only source of Cl^- in particles at HoChiMinh City. Equivalent ratios between concentrations of other ions and chloride concentration were much higher than those in the sea water, i.e. other ions had non-sea-salt origination.

Table 4. Concentrations of sulphate and their contributions in the mass of particles

	Fine particles (< 2.1 μm)		Coarse particles (> 2.1 μm)	
	Concentration ($\mu g/m^3$)	Contribution (%)	Concentration ($\mu g/m^3$)	Contribution (%)
VNU- Dry season	8.9	15	3.9	4.8
DOSTE- Dry season	11	11	5.3	3.9
VNU- Rainy season	3.4	24	4.2	8.5
DOSTE- Rainy season	3.6	14	3.1	13
My Tho - Rainy season	2.9	25	2.8	8.0

Fig. 4 shows a size distribution of ions in particles. SO_4^{2-} ions showed a bimodal distribution with two modes at 0.43 – 0.65 μm (or 0.65 – 1.1 μm), and 3.3 – 4.7 μm. In the dry season sulfate existed mainly in fine particles, while in the rainy season it was comparable in both fine and coarse particles. NO_3^- was polymodal. Cl^- was bimodal. Both NO_3^- and Cl^- were mainly in the coarse particles with the size of 2.1 – 5.8 μm for NO_3^- and 3.3 – 5.8 μm for Cl^-. For the fine fractions NO_3^- was very low in comparison with sulphate.
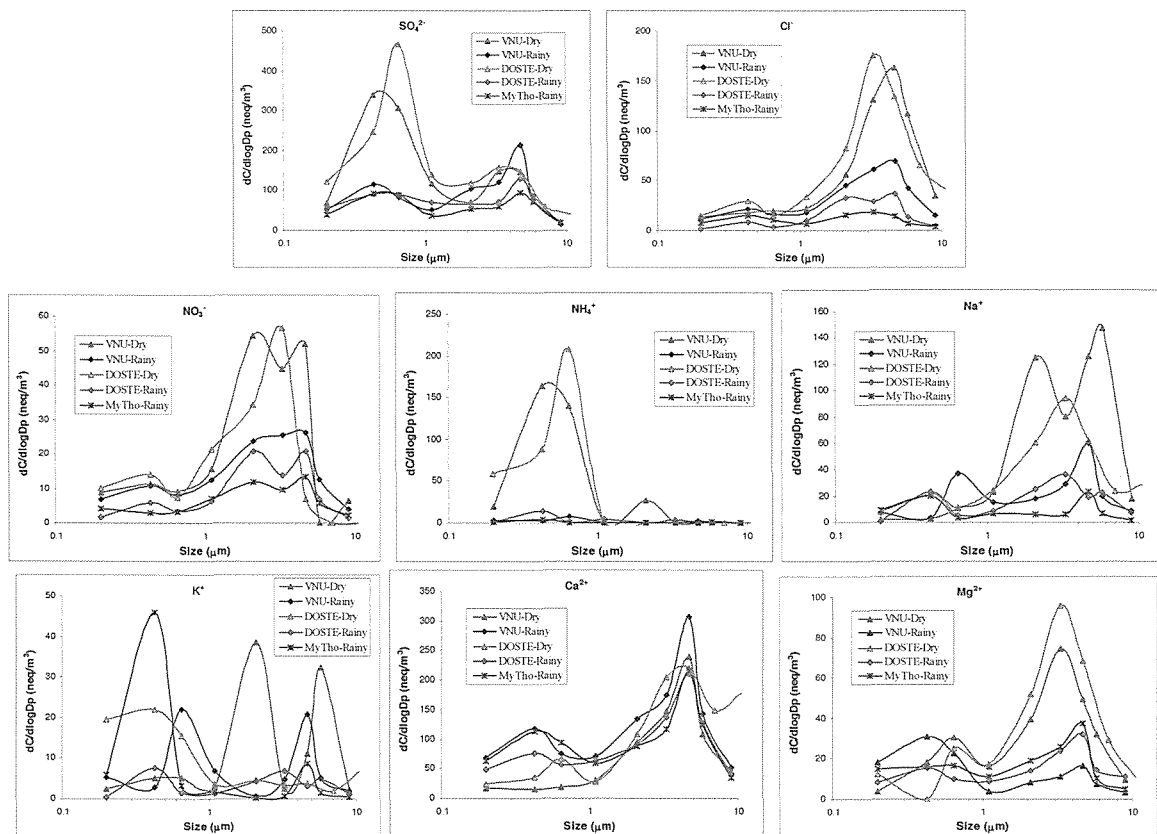
Fig. 4. Size distributions of ions

Ammonium is the most important basic atmospheric component that neutralizes acidic species in the air like SO_2, HNO_3, $HCl...NH_4^+$ was almost unimodal. Besides it was abundant in fine particles of the dry season, but was in small amounts or not detected in coarse particles of the dry season or in particles of any size in the rainy season. Na^+, Ca^{2+}, and Mg^{2+} were bimodal, but they were mainly in the coarse fractions.

The main source of Cl^- and Na^+ is sea-salt aerosols. Sea-salt aerosols form by evaporation of sea drops formed by wave shattering; hence they are in coarse fractions. The higher concentrations of Cl^- and Na^+ in the dry season due to monsoon coming from the sea to the land in the dry season, and washing effect of rains in the rainy season.

3.4. Mechanism responsible for modes of sulphate and nitrate

Sulphate and nitrate substances found by XRD analyses of sample collected by the high-volume sampler are $CaSO_4.2H_2O$, $Fe_2(SO_4)_3$, $K_3Fe(SO_4)_3$, $NH_4Fe(SO_4)_2$, NH_4HSO_4, $(NH_4)_2SO_4$, $(NH_4)_3H(SO_4)_2$, $(NH_4)_4(NO_3)SO_4$. In order to see better a correlation between ions the distribution curves of SO_4^{2-}, NO_3^-, NH_4^+, and Ca^{2+} ions are represented together in Fig. 5.
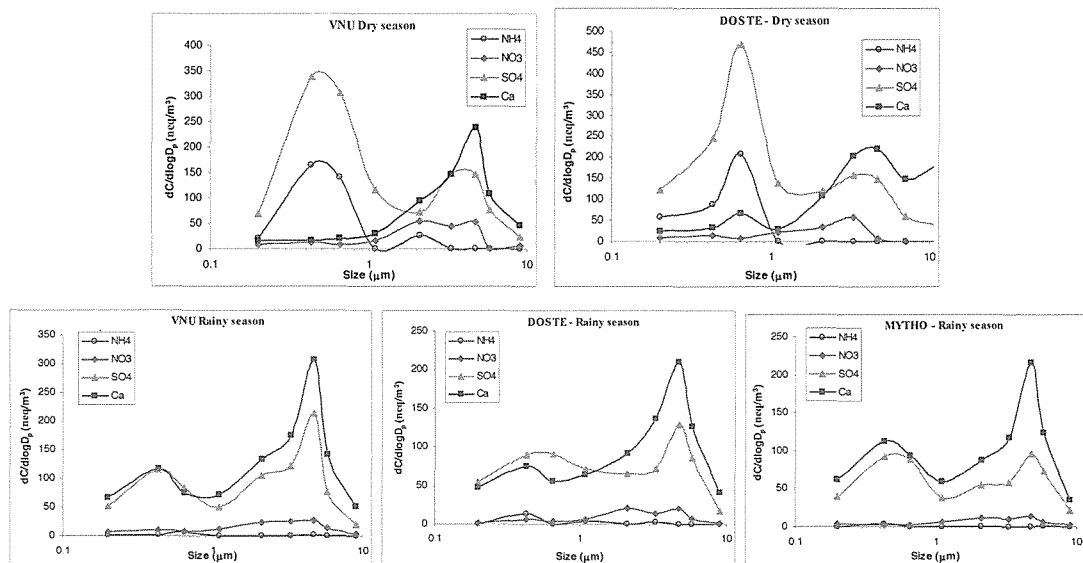
Fig. 5. Correlations between ions

Iron and calcium salts should be in coarse particles. A big part of course particles is a crustal dust that is blown up from the ground by winds and transportation means. The formation of calcium or iron sulphate is due to the reaction between solid crustal components, absorbed moisture and SO_2 (Lun, 2003; Ikuko, 2003; and LAN, 2004). This is confirmed by a good relation between size distribution of calcium and sulphate ions in coarse fractions. This implies that SO_4^{2-} was mainly in the form of gypsum ($CaSO_4.2H_2O$). SO_4^{2-} ions in the fine particles of the rainy season seemed to associate with calcium ions. Commonly calcium is mainly in the coarse fraction. Hence the mechanism related with the sulphate mode in the fine fraction of the rainy season is not understood

Sulphates should be in fine particles. They are formed by atmospheric reactions between SO_2, NH_3 and oxidizing agent in the air or by gaseous H_2SO_4 condensation. In larger fine particles sulphates are formed by coagulation or hygroscopic growth of smaller particles, or evaporation of sulphate-containing cloud water (Whitby, 1978; Hering and Friedlander, 1982; John et al., 1990; Ondov and Wexler, 1998; Meng and Seinfeld, 1994). It is clear that there is a good correlation between SO_4^{2-} and NH_4^+ ions only for the fine particles of the dry season. The equivalent ratio SO_4^{2-} / NH_4^+ was 2.1 – 2.2 (VNU) and 2.1 – 2.8 (DOSTE). Thus SO_4^{2-} should be mainly in the form of acidic sulphates. This was a reason of the high acidity and the cation deficiency of the extracts of the fine fractions as shown in section 3.2. Ammonium was found seldom or not at all in particles of the rainy season since ammonium sulphates were removed by wet deposition. The above suggestion is proved by the results obtained in the study on rain in South Vietnam (Lan, 2004). Lan found that sulphate and ammonium are predominant ions in rainwater collected at HoChiMinh.

Whitby (1978) found the size distribution of sulfate was characterized by a trimodal model, consisting of the nucleation mode, the accumulation mode and the coarse mode. An accumulation mode of sulfate at 0.48 ± 0.1 μm was reported. Hering and Friedlander (1982) and John et al. (1990) later observed that the accumulation mode of sulfate consisted of two modes. A small accumulation mode of sulfate at around 0.27 ± 0.1 μm was attributed to gaseous H_2SO_4 condensation. A larger accumulation mode (the droplet mode) of sulfate at 0.77 ± 0.2 μm was attributed to aqueous phase reactions in fog and cloud droplets or to hygroscopic growth of aerosols from the smaller accumulation mode. More recently, Ondov and Wexler (1998) proposed that the accumulation mode of sulfate at 0.48 μm observed by

Whitby (1978) was from the oxidation of SO_2 with subsequent hygroscopic growth, but the droplet mode of sulfate at 0.7 μm was formed in clouds. Because of the large water content in clouds, SO_2 is oxidized rapidly within 1 hour by in-cloud processes (Meng and Seinfeld, 1994). The evaporation of cloud water results in droplet mode sulfate aerosols. We believe in the idea proposed by Ondov and Wexler, and Meng and Seinfeld; accordingly the droplet mode results from evaporation of cloud water and the formation of in-cloud ammonium sulphate is very fast. Therefore in-cloud ammonium sulphate should be higher than under-cloud ammonium sulphate. This can be seen clearly in Fig. 4 that in dry season sulphate in fractions 0.65 – 1.1 μm (droplet mode) was higher than that in fractions 0.43 – 0.65 μm (fine accumulation mode). In the rainy season in-cloud sulphate is deposited with rain; i.e. in-cloud sulphate particles are hard to form. Consequently the fine sulphate mode in the rainy season became much lower than that in the dry season and shifted to the smaller size (Fig. 4), and particles become less acidic in the rainy season.

Exposure to strongly acidic sulfate particles was found to cause an alteration in lung function and particle clearance rates (Amdur et al., 1978; Koenig et al., 1983; Bauer et al., 1988; Leikauf et al., 1984; Schlesinger et al., 1983; Lippmann et al., 1982, 1989). However, these effects were not observed following exposure to the weakly acidic ammonium sulfate particles (Schlesinger, 1989; Utell et al., 1982). Some studies have demonstrated a stronger association between SO_4^{2-} and mortality and morbidity than with any of the other measures of PM, such as total suspended particulate TSP, PM_{10} and $PM_{2.5}$ (Ozkaynak and Thurston, 1987; Thurston et al., 1994). In HoChiMinh sulphate in fine particles of the dry season is acidic. In addition its concentration in the dry season was high. Therefore the toxic effect of particles on the human respiratory tract in the dry season should be higher than that in the rainy season.

The distribution modes of nitrate are governed by the thermodynamic equilibrium of $HNO_3(g) + NH_3(g)$ $= NH_4NO_3(s, aq)$, which is affected by the relative humidity, temperature and concentrations of NH_4HSO_4 $(NH_4)_2SO_4$, $HNO_3(g)$ and $NH_3(g)$ (John et al., 1990). When the thermodynamic equilibrium favors the formation of $NH_4NO_3(s, aq)$, nitrate will dominate in the fine mode. On the other hand, if the formation of $NH_4NO_3(s, aq)$ is not favored; the reactions of HNO_3 with $CaCO_3$ or $NaCl$ in the coarse mode become important (Pakkanen et al., 1996; Zhuang et al., 1999). The fine particulate nitrate was shown to decrease with ambient temperature while the coarse particulate nitrate increased due to the shifting of the above equilibrium to the left (Gottlieb 1989, Lestari, 1996). At HoChiMinh City sulphate in the fine fractions is acidic. Also, the temperature in HoChiMinh City was high year-round, about $28 – 30^O C$, as a result the more preferable reaction is the neutralization of sulphate by ammonium, but not nitrate by ammonium. Therefore nitrate showed very low peak in the fine mode as it was showed in Fig. 4. In the coarse mode nitrate might associate with calcium. In other words, the peaks of sulphate and nitrate in the coarse fractions are present as surface coatings on these size particles, i.e. these peaks associate with heterogeneous chemical reactions on particle surface.

3.4. Source region identification

Air parcels coming to HoChiMinh on February 5th and 6th 2005 (Fig. 6) began in Shanghai region, traveled over the Pacific Ocean through the polluted city of Metropolitan Manila in the Philippines, and passed directly from the sea to HoChiMinh City. There were small rains along with the trajectory at the area closed to Shanghai. The trajectories for the sampling days in the rainy season have similar courses. All of them began in the Indian Ocean, crossed tourist and agriculture area of Phuket, Surat Thani… in the South of Thailand, or clean Aceh area in Malaysia, then traveled over the Gulf of Thailand, and finally passed through the Mekong Delta to HoChiMinh. Besides, there were long and heavy rains along these trajectories. Thus Shanghai and Manila can be considered as source regions of air pollution in HoChiMinh in the case of the two samples collected in the dry season. From section 3.3, in the dry season sulphate in

fine particles at HoChiMinh exits mainly in the form of acidic sulphates.Rodhe and Grandell (1981) showed that fine acidic sulfates tend to persist in the atmosphere and may be transported long distances from their original sources. High acidic sulfates concentrations have been measured not only near SO_2 emission sources, but also at locations far away from the emission sources (Stevens et al., 1980; Huntzicker et al., 1984). Ambient ammonia concentrations decrease with altitude. Therefore, when acidic sulfate particles are emitted above the inversion layer, they can travel long distance without being neutralized by the ammonia rich ground level air (Tanner et al., 1984). This implies that besides local source the other important source of acidic sulphates is long-range transportation. Therefore, long-range transportation should be considered when examining sources of this pollutant. Thus a part of particle acidic sulphate of the two first samples might come from Shanghai and Manila.

NOAA HYSPLIT MODEL
Backward trajectories ending at 06 UTC 05 Feb 05
GDAS Meteorological Data

Fig. 6. Typical backward 5-day trajectory of incoming air parcel to HoChiMinh on February 5th 2005

Conversely, in the rainy season the air parcels coming to HoChiMinh and My Tho spent most of their advection over the Pacific Ocean and the lands which have no SO_2 sources. Therefore the local pollutants are the only source of air pollutants at HoChiMinh and My Tho for the samples of the rainy season.

4. Conclusion

The mass concentrations of particles as well as the concentrations of common soluble inorganic ions in particles at HoChiMinh were high in the dry season and low in the rainy season. The mass contribution of the fine particles smaller then 2.1 μm in TSP was 20 – 50% and it was higher for the dry season than for the rainy season, and higher for roadside air then for urban background air. Size distribution of sulphate is bimodal. Particles of the dry season contained a lot of acidic ammonium sulphate and mainly in fine particles, hence are considered more harmful than those of the rainy season. Particle sulphate in the rainy season was low and distributed evenly in fine and coarse particles. Nitrate was mainly in coarse particles. The amount of nitrate was very small in comparison with the amount of sulphate. Coarse sulphates are products of surface reactions between crustal components and gaseous precursors. NH_4^+ ions were found only in the fine fractions of particles collected in the dry season. Most of chloride ions originated from sea-salt aerosols and existed mainly in the coarse fractions. Sea-salt aerosols were higher in the dry season when prevailing winds came directly from the east sea to HoChiMinh City. Trajectory analyses showed that Manila and Shanghai were the source regions of air pollution in HoChiMinh. The particles collected in the rainy season were less than in the dry season owing to the two factors: 1) rains; 2) air masses coming to HoChiMinh in the rainy season spent most of their advection over the sea and clean areas.
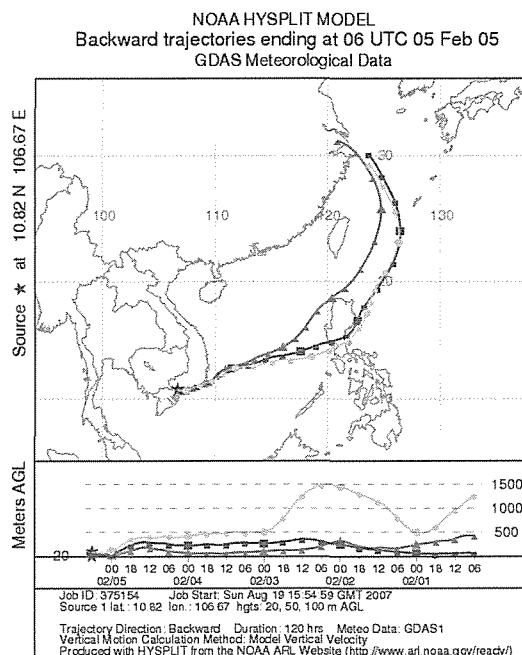
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