The long-range transport of aerosols from northern China to Hong Kong – a multi-technique study

M. Fang[a],*, M. Zheng[b], F. Wang[a], K.S. Chim[c], S.C. Kot[c]

[a] *Institute for Environment and Sustainable Development, the Hong Kong University of Science & Technology, Clear Water Bay, Hong Kong, China*
[b] *Graduate School of Oceanography, University of Rhode Island, Narragansset, RI 02882, USA*
[c] *Department of Mechanical Engineering, the Hong Kong University of Science & Technology, Clear Water Bay, Hong Kong, China*

Abstract

The results of the inorganic and organic analyses of aerosol samples collected on the east and west sides of Hong Kong during a dust episode (9–10 May 1996) are reported. The origin of the dust was traced to Northern China. The dust reached Hong Kong by way of the East China Sea. The characteristics of the inorganic elements and organic compounds were quite different from the non-episodic samples collected on 1–2 April 1996, EPD (Environmental Protection Department, Special Administrative Region, Hong Kong, China) results for April–May 1994, and our early studies (Zheng et al., 1997. Atmospheric Environment 31(2), 227–237.). Results from X-ray spectrometry showed pronounced increase in the relative abundance of Al, Fe, Ca, S and Cl in the dust samples compared to the non-episodic samples. The high abundance of Cl in the dust samples suggested the aerosols experienced long-range transport by way of the sea. ICP-MS analysis revealed higher concentrations of Fe, Ca, S and Pb in the episodic samples relative to the values measured during April–May 1994 by EPD. The high Ca content in the soil samples is a characteristic of northern Chinese crustal material (Liu et al., 1985). Hong Kong aerosols are characterized by high octadecenoic acid concentration due to heavy urbanization and Chinese-style stir-fry cooking. A much lower $C_{18:1}/C_{18:0}$ ratio was found in the episodic samples, however, suggesting the aerosols were transported from a long distance. The high ratio of $\geqslant C_{20}/ < C_{20}$ in n-alkanoic acids, the high input of n-alkanes and n-alkanols from plant waxes, and the unusually similar distribution of the organic compounds in the east and west samples suggested the existence of non-local sources on 9–10 May 1996. The compositions and distributions of lipids in the aerosol samples collected during the episode corresponded well with those of the eolian dust samples over the Atlantic and Pacific Oceans. Back trajectories and low altitude (< 3 km) mesoscale flow modeling were performed, suggesting the existence of a mesoscale atmospheric structure off the east China coast, which could be responsible for the dust episode. Surface charts indicated the presence of suspended dust near Shanghai on 8 May 1996. Taiwan also experienced a similar episode on 8–9 May 1996. This integrated, multi-technique approach provides clear evidence that the 9–10 May episode was attributed to a consequence of Asian Dust. This is the first scientific report of Asian Dust in Hong Kong. ©1999 Published by Elsevier Science Ltd. All rights reserved.

Keywords: Hong Kong; Aerosol; Asian Dust; Inorganic elements; Solvent-extractable organic compounds; Long-range transport; Mesoscale modeling

* Corresponding author. Tel.: +852 2358 6916; fax: +852 2358 1334; e-mail: rcmfang@usthk.ust.hk

Introduction

The air quality in Asia is deteriorating on the whole in recent years due to the rapid, across-the-board economic and industrial development. The local sources of air pollution in urban areas are traffic and manufacturing plants while a great deal of biomass burning is still practiced in rural areas (Fang and Huang, 1998). Furthermore, due to the complicated air circulation patterns in Asia, long-distance transport of the pollutants occurs frequently.

The Atmospheric Research Team at the Hong Kong University of Science and Technology (HKUST) has been using an integrated, multi-technique approach to study the cross-boundary transport and dispersion of airborne pollutants. The technique includes ground-level monitoring, airborne measurements, mesoscale meteorological modeling, and satellite remote sensing. This technique is used because of the complicated terrain in and surrounding Hong Kong. In addition to the hills and mountains, Hong Kong is bordered on the east and south sides by the South China Sea and by the Pearl River Estuary on the west; severe land–sea breeze effects have thus been observed. All these factors contribute to the complexity of understanding the transport of atmospheric pollutants and suspended particulates, and sometimes a single method alone may not be sufficient to reveal the complete story. The 9–10 May 1996 Asian Dust episode will be used to demonstrate this approach in this paper. The dust was eventually traced to be originated from the northern part of China.

The atmosphere has been known to be a significant pathway for the transport of mineral dust and air pollutants from the continents to the coastal areas and oceans (Parrington et al., 1983; Bergametti et al., 1989). The mid-latitudes (25–40°N) are most strongly affected by Asian Dust derived from the arid regions of central and eastern Asia – the Takla Makan, Gobi and Ordos deserts and the regions where loess is deposited south of the Gobi and Ordos deserts (Zhang, 1984; Uematsu et al., 1983; Duce et al., 1980; Rahn et al., 1977). Asian Dust frequently occurs during the dry, east Asian spring, typically in April–May. Duce reported the long-range atmospheric transport of soil dust from Asia to the tropical North Pacific (Duce et al., 1980). Long-range transport of Asian desert dust may even contribute to the formation of haze layers in the North American Arctic (Rahn et al., 1977). A study of the mean surface wind fields in March–May 1979 showed that the dust rising over China could easily be transported at 700 mbar to the east (Darzi and Winchester, 1982; Duce et al., 1980). Air mass trajectory analysis indicated that the Asian Dust was transported in the free troposphere and that westerly winds carried the Asian Dust to the open ocean (Merrill et al., 1985, 1989).

There have been studies of selected crustal and pollutant elements in the Asian dust aerosols in several cities such as Xi'an, Beijing, Qingdao and Xiamen in China, Mallipo in Korea, and Okinawa in Japan, and of the deposition flux to coastal areas and oceans along the pathway of transport. The yearly atmospheric flux of mineral aerosols to the Yellow Sea was comparable to the northwestern Mediterranean Sea; the latter was strongly affected by the mineral dust from the Sahara desert (Prospero and Carlson, 1972; Gao et al., 1992). Lidar investigation has been applied to study the transport of mineral particles known as *kosa* in Japan and the results showed dust layers at 2–6 km (Iwasaka et al., 1988).

Asia is one of the major sources not only for mineral dust but also for nitrate and sulfate aerosols (Gao, 1994). The meteorological conditions favoring the long-range transport of Asian Dust do not discriminate the transport of anthropogenic materials (Arimoto et al., 1996). Studies carried out in Fujian and Guangdong Provinces in the southern part of China suggested that the acid rain problem in this region was primarily caused by the mesoscale long-range transport of pollutants from the northern provinces, where large and strong emission sources of SO_2 and NO_x existed (Wang et al., 1992; Yu et al., 1992).

Organic material in dust samples collected off the coast of northwest and southwest Africa and over the Pacific Ocean has been analyzed (Simoneit, 1977; Lepple and Brine, 1976; Peltzer and Gagosian, 1989). Cox concluded that eolian transport could be used as an input mechanism to explain the existence of terrestrial lipid detritus in some pelagic sediment at great distances from land in the North Atlantic (Cox et al., 1982).

In this paper, the 9–10 May 1996 dust episode in Hong Kong was studied using a combination of techniques described earlier and the results show that such a multi-technique is very useful in understanding complicated environmental issues.

2. The dust episode of 9–10 May 1996

The HKUST Atmospheric Research Group has been routinely collecting aerosol samples for solvent-extractable organic compound analysis using high-volume samplers. The sampling sites were located at the HKUST campus on the east side of Hong Kong, and on the west side at Shalowan near the new airport on Lantau Island (Fig. 1). This is a part of an on-going aerosol program to understand the transport of air pollutants (city plume) from the east end of the city to the west, which is still sparsely populated. The prevailing wind in Hong Kong is from the east, but, the presence of the Zhujiang (Pearl River) Delta on the west side causes changes in the micro-meteorology (land-sea breeze effect) and has a large impact on air dispersion (Kok et al., 1997).

On 9 May 1996, Hong Kong experienced unusually high API (air pollution index) suggesting the existence of
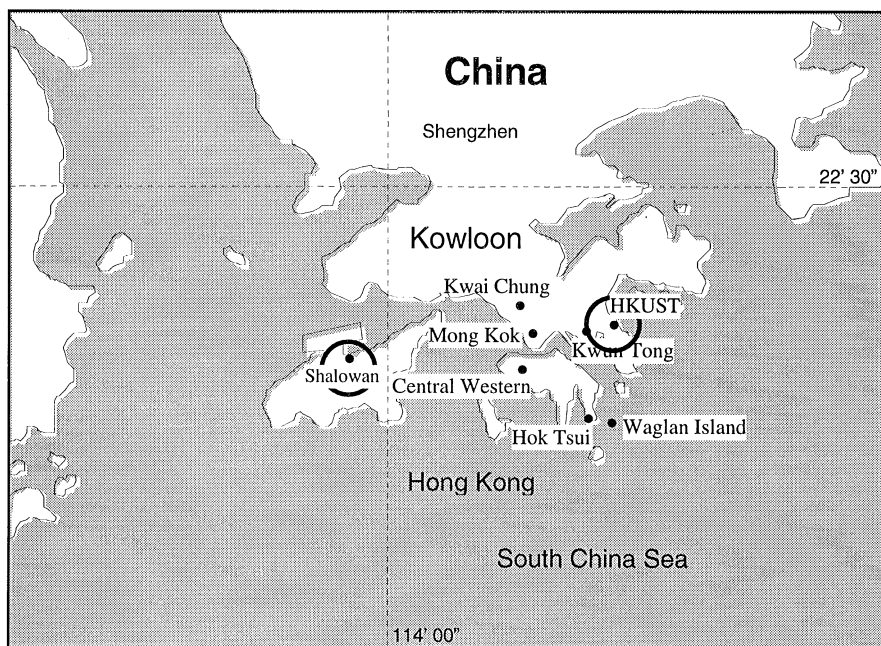
Fig. 1. Location of sampling stations.

a large concentration of pollutants or materials in the atmosphere that day. A strong easterly wind prevailed on the morning of 10 May. The total suspended particulates (TSP) samples collected were subjected to chemical analyses, and through the use of a multi-technique approach, the source of the airborne aerosols was traced back to Northern China.

Private communications with researchers at the National Taiwan University revealed that the presence of "yellow dust" at a station 40 km north of Taipei has been observed. The yellow dust reached Taiwan on 9 May, and the concentration was still high on 10 May. Asian Dust storms seldom occur in Hong Kong but they are a yearly event in Taiwan.

3. Experimental

Simultaneous aerosol samples were collected at HKUST and Lantau Island on 1–2 April and 9–10 May 1996 using commercial high-volume air samplers (Graseby GMWT 2200). The flow rate was 1.1–1.5 m³ min⁻¹ and the sampling time was around 24 h. Prior to sampling, the glass fiber filters (Graseby GMW, 20.3 × 25.4 cm) were annealed for 4 h at 550°C to remove any absorbed organic materials and the collected sample together with the filter was stored in a precleaned 500 ml glass jar with 3–5 ml of methylene chloride added to

prohibit microbial growth. The sample jars were stored in a refrigerator ($\sim 0°C$) (Simoneit and Mazurek, 1982).

Fig. 1 is a map indicating the sampling locations and the general geography of the area of interest. Samples previously collected at HKUST and Lantau Island on 1–2 April (designated as UST1 and LT1, respectively) were also analyzed to provide references and comparison. The weather condition for 1–2 April was a normal spring day in Hong Kong.

The dust samples were subjected to inorganic and organic analyses to identify, characterize and quantify their contents so that the origin and abundance could be determined. The details of the analytical procedure of the solvent-extractable organic compounds were presented in a previous publication (Zheng et al., 1997). Briefly, the filter was extracted with three 200 ml aliquots of dichloromethane, each for 15 min at room temperature. The total extract was combined and concentrated to a volume of 3–5 ml by a rotary evaporator (Buchi RE 111). The extract was then filtered and reduced further by gentle evaporation with a stream of high purity N_2 to a volume of 500 μl. The total extract was reacted with 14% BF_3 in methanol to esterify organic acids, then separated into aliphatics, PAHs, esters and alkanols using silica gel packed in a column. Each fraction was subjected to GC-FID (Hewlett Packard Model 5890A gas chromatograph) and GC-MS (a Hewlett-Packard 5971A Mass-selective Detector interfaced to a Hewlett Packard

Model 5890A gas chromatograph) analyses except the alkanol fraction, which was converted to trimethylsilyl ethers prior to the instrumental analysis. Organic compounds were quantified using the internal standards. Procedural and solvent blanks were determined and relative response factors for 63 standard compounds including alkanes, PAHs, methylated fatty acids, phthalates, and alkanols were calculated.

The inorganic analysis was performed by the Asia Pacific Competence Centre of Philips Electron Optics B.V. A Philips Sequential X-ray Spectrometer System (Model PW2400) was used to determine the elemental composition of the samples. Relative abundance of the detectable elements was obtained. The results presented in this paper were the averages of at least two scans of the samples. Seven scans were made to determine the blank sample composition.

The elemental composition of the dust samples was quantified by a Finnigan Element inductively coupled plasma mass spectrometer at the Graduate School of Oceanography, University of Rhode Island. Sample preparation was performed in a clean laboratory providing a Class 100 working environment. All plastic ware was acid-cleaned prior to use. Filters were digested in a 5 ml mixture of HCl, HNO_3 and HF on a hot plate at $110°C$ for 4 h. Using this total digestion method, the silicate lattices were completely dissolved and all the associated metals were released. After digestion, the samples were evaporated to dryness, redissolved in 2% nitric acid and filtered before ICP-MS analysis. Samples were introduced with a pneumatic nebulizer system. Operating conditions used were typical for an Ar-ICP-MS. The plasma power was maintained at 1350 W and the argon flow (in $l\,min^{-1}$) included cool gas – 13.95, auxiliary gas – 0.93, sample gas – 0.89, and additional gas – 0.41. Quantitative analysis was carried by the external calibration technique using a set of external calibration standards at concentration levels close to that of the samples. A drift correction was applied by running the set of external calibration standards before and after the ICP-MS analysis of the samples. Procedural and field blanks were also determined. Recoveries derived from the analysis of SRM 1646a (Standard Reference Material 1646a, estuarine sediments) were in the range of 80% for Zn to 123% for Cr.

4. Mesoscale meteorological modeling

For the long-range transport of air pollutants, it is possible to trace the movement of the pollutants by using the low-altitude flow fields computed from a mesoscale model. Wind field patterns computed from these models at low altitudes take ground effect into consideration, thus exhibiting behavior different from the general meteorological maps. These lower altitude winds are responsible for the dispersion of air pollutants. Backward and forward trajectories can be calculated to track the transport path.

The wind field calculations were made using the Penn State/NCAR (National Center for Atmospheric Research) Mesoscale Model (MM5) (Grell et al., 1994) on a Silicon Graphics Onyx 8-cpu mini-super computer. The computational domain was 108 grids in both horizontal directions with a grid size of 54 km. The area studied was $89.2–140°E$ and $3.45–48.04°N$ with the center at $28°N$ and $115°E$. A 10-min terrain resolution was used to represent the topography. Above the terrain, twenty-seven sigma levels were used for the vertical resolution. The initial field and boundary conditions were extracted from National Meteorological Center (NMC) synoptic model in 12-h intervals. An 84-h period starting at $00:00$ on 7 May to $12:00$ on 10 May 1996 was computed with a time step of 60 s.

The physical options used in this simulation included simple radiative cooling, the high-resolution Blackadar Planetary Boundary Layer Scheme, Grell's cumulus parameterization scheme, the cloud effect included in the surface radiation, no snow covering effect, and normal evaporative cooling.

5. Results and discussion

5.1. General appearance of the samples

In the previous four years of aerosol collection in Hong Kong, all samples obtained had a black appearance with the exception of the background sample, which was light gray, that was obtained on Waglan Island. The black color strongly suggests the high content of carbonaceous materials (Fang et al., 1996). The 9–10 May 1996 samples were yellow–brownish.

5.2. Inorganic analysis

5.2.1. X-ray spectrometry analysis

The relative abundance of the elements in the 9–10 May samples and the non-episodic 1–2 April samples at HKUST and Lantau is presented in Fig. 2. Since the filter medium used was glass fiber, high background concentrations of some elements such as silicon, potassium, barium, and zinc were found while some elements such as Fe, S and Cl exhibited very low blanks.

There were distinct differences in the relative elemental abundance between the 1–2 April and the 9–10 May samples. A significant increase in the abundance of Al, Ca, Fe, S and Cl in the 9–10 May samples was found. For example, at HKUST, the abundance of Al, Fe, Ca, S and Cl was 8.76, 3.36, 11.22, 5.91, and 6.01%, respectively, in the dust sample, while it was only 4.49, 0.39, 5.62, 1.97,
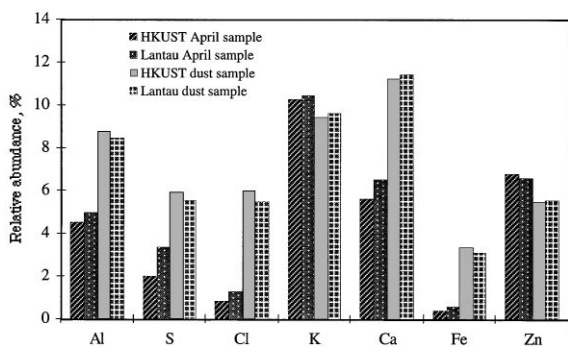
Fig. 2. Relative abundance of elements by XRF.

Fig. 3. ICP-MS results of dust samples and EPD 1994 average data.

and 0.82%, respectively, in the 1–2 April sample (Fig. 2). This increase was also observed in the Lantau sample and the magnitudes were very close to HKUST, suggesting that the pollutants were well mixed and the whole region was homogeneous.

The abrupt increase of Al, Fe and Ca in the aerosol samples indicated the input of crustal materials on 9–10 May which led to the unusually high API. Loess in Xi'an, China has been reported to have characteristically high Ca content (Liu et al., 1985) and Tadzhikistan loess has been found to be rich in Ca mainly as carbonate (Pye, 1987).

The high S concentration in the 9–10 May samples suggested a source from an area where there is burning of high S-content fuels (coal). This was most likely in the northern part of China, because Hong Kong banned the use of 2.5% S containing fuels in 1990 and, furthermore, most of the local manufacturing plants have been relocated to China and other southeastern Asian nations in recent years. Soil itself contains very small amounts of Cl (130 ppm) with a Fe/Cl ratio of 443 (Taylor, 1964) while the Fe/Cl ratio was 0.44–0.47 in the 1–2 April samples and 0.56–0.58 in the dust samples. The production of sea-salt aerosols by bubble bursting is an important natural source for elements such as Cl and Na in marine atmospheres (Arimoto et al., 1989). Considering the very low chlorine content in crustal materials, the increase in Cl content in the dust samples was probably due to the sea-salt particles, suggesting the transport of mineral aerosols via the East China Sea, and this supposition was supported by the mesoscale modeling and ground observations.

5.2.2. ICP-MS analysis

The dust samples were subjected to ICP-MS analysis for more detailed studies and Fig. 3 shows the elemental concentrations in the samples. Table 1 is a summary of all data. The concentrations measured at three stations (Sham Shui Po, Kwai Chung, and Kwung Tong) every six days during April–May in 1994 by EPD are also
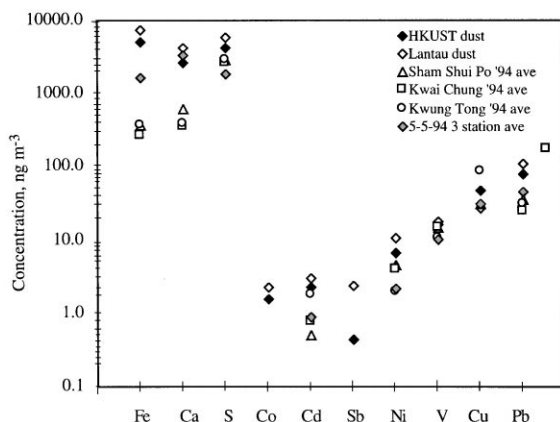
included in Table 1 for comparison (a total of 26 samples). The measurement of Al was hampered by the high background of the glass fiber filter and will not be discussed here.

The concentration of Fe in UST2 and LT2 was 5.183 and 7.585 μg m^{-3}, respectively, and was much higher than the average value of 497 ng m^{-3} for April–May 1994. The Fe concentration was higher than 1000 ng m^{-3} for all three stations only on 5 May (1400 ng m^{-3} at Kwung Tong, 1800 ng m^{-3} at Kwai Chung, and 1900 ng m^{-3} at Sham Shui Po). The concentration of Ca in UST2 and LT2 was 2.759 and 4.236 μg m^{-3}, respectively. The concentration of Ca reported by EPD from 5 April to 29 May 1994 was less than 710 ng m^{-3} on average except for the 5 May sample, which exhibited extremely high values with 2700 ng m^{-3} at Kwung Tong, 3100 ng m^{-3} at Kwai Chung, and 4500 ng m^{-3} at Sham Shui Po. These high values were very close to the dust samples. In fact, high concentrations of Al, Fe, and Ca were observed at 7 EPD stations on 5 May 1994. It is quite possible that a dust episode occurred on that day.

A high content of S (6.020 μg m^{-3} in LT2 and 4.292 μg m^{-3} in UST2) was found in the dust samples agreeing with the X-ray spectroscopy results. The EPD average concentration of S was 2743 ng m^{-3}. Sulfur has different sources, e.g. biogenic sulfur from the oxidation of dimethylsulfide (DMS) produced by marine biota and anthropogenic emissions from fossil fuel combustion (Andreae et al., 1985). In China, sulfate aerosol is a major problem; the sulfur dioxide emitted from coal combustion is estimated to be 20 Tg in 1987, accounting for $\sim$69% of the total SO_2 emissions in Asia (Kato and Akimoto, 1992). The acid rain problem in the southern part of China is caused by the mesoscale, long-range transport of pollutants from the northern provinces (Wang et al., 1992; Yu et al., 1992).

Table 1
Elemental concentrations (ng m^{-3}) of the episodic samples UST2 and LT2 determined by ICP-MS, and 1994 HKEPD samples from three stations {Sham Shui Po (SSP), Kwai Chung (KC) and Kwung Tong (KT)}[a]

	Fe	Ca	S	Co	Cd	Sb	Ni	V	Cu	Pb	Se
UST2	5183	2759	4292	1.6	2.3	0.4	6.7	16.2	48.0	81.5	
LT2	7585	4236	6020	2.3	3.0	2.4	10.9	18.2	28.1	109.5	1.7
SSP ave[b]	373	629	2904		0.5		4.6	14.9	31.4	36.5	
KC ave[b]	273	363	2688		0.8		4.1	14.8	144.4	25.1	
KT ave[b]	380	399	2976		1.8		2.0	10.6	87.6	32.3	
SSP 5-4-94	270	480	3067		0.25		7.3	25	41	53	
SSP 11-4-94	360	640	5000		0.84		4.7	11	31	49	
SSP 17-4-94	340	710	3333		0.42		0.4	10.5	37	32	
SSP 23-4-94	340	680	2033		0.15		1.4	11	35	22	
SSP 29-4-94	390	590	2367		0.5		8.9	23	33	34	
SSP 5-5-94	1900	4500	2033		0.88		1.1	11.5	39	54	
SSP 11-5-94	420	630	2900		0.85		9	19	34	24	
SSP 17-5-94	420	660	1633		0.59		2.7	10	17	33	
SSP 23-5-94	440	640	2900		0.57		2.2	10	23	45	
KC 5-4-94	170	320	2700		1.4		7.4	18	240	46	
KC 11-4-94	330	260	4000		0.53		3.1	9.5	31	18	
KC 17-4-94	170	270	2767		0.42		0.36	9.5	260	17	
KC 23-4-94	240	410	2000		0.19		3.6	10	270	11	
KC 29-4-94	290	360	1967		0.47		8.6	29	290	22	
KC 5-5-94	1800	3100	1900		1.2		2.8	9.5	23	38	
KC 11-5-94	310	340	3333		0.96		5.5	21	18	25	
KC 17-5-94	310	470	1467		1.7		2.7	10	22	29	
KC 29-5-94	360	470	3267		0.63		1.3	11.5	24	33	
KT 11-4-94	490	370	5667		0.55		2.9	11	35	34	
KT 17-4-94	230	470	3333		0.51		0.4	11	360	35	
KT 23-4-94	270	230	1867		0.14		0.37	10.5	32	23	
KT 5-5-94	1400	2700	1833		0.65		2.6	10.5	34	43	
KT 11-5-94	320	260	2967		4.1		3.4	10.5	47	24	
KT 17-5-94	420	540	1833		6		3.3	10	50	35	
KT 23-5-94	380	400	2667		0.97		2.5	10.5	39	37	
KT 29-5-94	550	520	2500		0.6		1.15	10.5	50	38	

[a] Data Source: EPD (1997).
[b] ave = average of data listed for 1994.

Crustal material has a S/Fe ratio of 0.0046 while the S/Fe ratio was 0.83 in UST2 and 0.79 in LT2; the latter are much lower than the average value of 8.1 for the Hong Kong aerosols, suggesting the mixing of local aerosols with low S/Fe ratio aerosols. Compared to the normal samples, the higher absolute abundance of S in the dust samples was probably due to input from the high S emission regions along the transport path, and possibly from marine biogenic sulfur input from the East China Sea. Unfortunately, Cl derived primarily from sea-salt particles in coastal regions cannot be measured by ICP-MS due to the strong interference of chlorine such as $^{35}Cl^{16}O^{+}$ on $^{51}Ni^{+}$ and $^{40}Ar^{35}Cl^{+}$ on $^{75}As^{+}$. It is interesting to note that the lowest S/Fe and S/Ca ratios were found only in the 5 May samples of the EPD samples. These values were also very close to the dust samples. This provides added support that a dust episode may have occurred on 5 May 1994.

From Fig. 3, using EPD data as a comparison, the Pb content in the dust samples (81.51 ng m^{-3} in UST2 and 109.5 ng m^{-3} in LT2) was 2–3 fold higher than the average value of 33 ng m^{-3}. There are three major Pb sources: industrial Pb-rich carbonaceous fines, mainly from gasoline exhausts, and soil dusts containing both industrial and natural Pb (Maring et al., 1989). The high Pb and S contents in the dust samples could be due to the fuel usage and strong emissions north of the border.

To interpret the atmospheric trace element sources better, enrichment factors (EF) are calculated as follows:

$$EF_X = (X/Ref)_{air}/(X/Ref)_{crust}$$

where EF_X is the enrichment factor of X, and Ref is a reference element for crustal material (Fe is used because it is a good indicator of the presence of crustal material and is not affected by contamination). One can

Table 2
Enrichment factors (relative to Fe)

	Ca	Ni	Co	V	Cu	Pb	Cd	S
UST2	0.72	0.28	0.70	1.30	9.48	70.83	123.42	179.28
LT2	0.76	0.31	0.67	1.00	3.79	65.02	110.96	171.87
Ave 5-5-94 data	2.72	1.00		2.62	19.65	120.48	149.60	247.95
Ave EPD data	1.92	8.71		18.38	368.44	442.69	864.14	1945.13

infer that crust is the dominant source of element X if its EF value approaches unity. The enrichment factors thus calculated showed that the crust was the dominant source for Ca, Ni, Co, and V in dust samples (see Table 2 and Fig. 4). An enrichment factor close to 1 for V and Co has been reported for aerosols collected in April 1989 in Beijing, China and Mallipo, Korea, and the continental crust was regarded as the dominant source (Gao et al., 1992). The concentrations of Cu, Pb, Cd and S were about 4–180 times higher than those expected from mineral aerosols. Compared to the normal Hong Kong aerosols, the dust samples from HKUST and Lantau showed the lowest enrichment factors for all elements due to the well mixed air masses and the strong input of mineral aerosols from the outside. The enrichment factors of the 5 May 1994 samples were very close to those of the dust samples, while for the normal Hong Kong aerosols they were 4–19 times higher except for Ca.

5.3. Organic analysis

The concentration, composition and distribution of the solvent-extractable organic compounds in UST2 and LT2 were significantly different when compared to UST1 and LT1. The 1–2 April samples clearly exhibited local characteristics (Zheng et al., 1997) while UST2 and LT2 suggested outside sources.

The homologous distributions of n-alkanes, n-alkanoic acids and n-alkanols in UST2 and LT2 were correlated to the distribution patterns reported in dust samples collected over the Atlantic with the surface waxes of terrestrial vascular plants as the major source of lipids (Simoneit, 1977). Long-range transport of terrestrially derived lipids to the tropical North and South Pacific was reported in the Sea/Air Exchange Program (Gagosian et al., 1981, 1987).

Table 3 is a summary of the analytical data of the four fractions extracted from the samples: n-alkanes, PAHs, n-alkanoic acids, and n-alkanols. The CPI (carbon preference index), C_{max} (carbon number maximum), the ratio of unsaturated to saturated fatty acids, and the ratio of $\geqslant C_{20}/< C_{20}$ fatty acids provided supportive evidence for the sources (biogenic and anthropogenic) and the fate of the aerosols in the atmosphere (Mazurek and Simoneit, 1984).
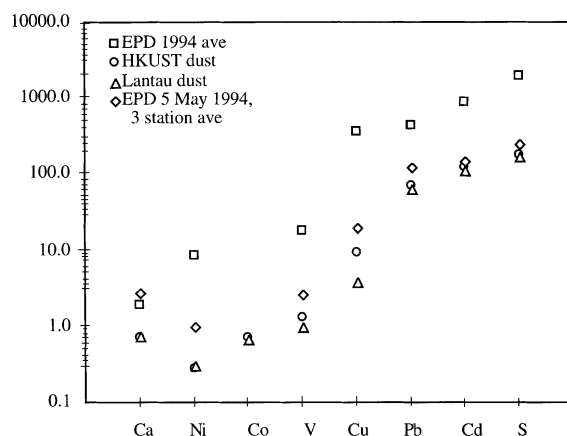

Fig. 4. Enrichment factors – relative to Fe.

The results from the 1–2 April samples collected from the same locations as UST2 and LT2 and from other sites in a previous study (Zheng et al., 1997) were used as references for comparison and analysis in this study.

5.3.1. n-Alkanes

The distribution diagrams for n-alkanes are shown in Fig. 5. The n-alkane homologs were C_{14}–C_{36} and the total alkane concentrations ranged from 50.1 to 416.0 ng m^{-3}. The dust samples LT2 and UST2 exhibited similar distributions with maxima at n-C_{29}, n-C_{31} and n-C_{27} and CPI of 2.9 in LT2 and 2.8 in UST2. The non-episodic samples had quite a different distribution with C_{max} at n-C_{24}, n-C_{25}, CPI of 1.1 in LT1, and C_{max} at n-C_{29}, n-C_{31} and n-C_{25}, CPI of 1.4 in UST1. The CPI ranged from 1.09 in the urban samples to 1.61 in the rural samples in Hong Kong (Zheng et al., 1997).

It is generally recognized that the n-alkane fraction of petroleum has a CPI of 1 with maxima at n-C_{24} and n-C_{25} (Mazurek and Simoneit, 1984), it can therefore be concluded that in the LT1 sample with CPI of 1.1 (no odd to even predominance) and C_{max} at n-C_{24} and n-C_{25}, petroleum residues were dominant. Lantau Island is located on the far west side of Hong Kong and on the east bank of the Zhujiang (Pearl River) estuary. Aside

Table 3
Summary of the lipid fractions (concentration in ng m^{-3})

	UST1	LT1	UST2	LT2
Total, n-alkane	50.1	176.6	416.0	299.2
C_{max}, n-alkane	C_{29}	C_{24}	C_{29}	C_{29}
Total, PAHs	15.2	21.4	25.6	22.6
Total, fatty acids	160.4	302.2	148.1	224.0
C_{max}, fatty acid	C_{16}	C_{16}	C_{16}	C_{16}
Total, n-alkanols	15.9	17.6	75.6	52.9
C_{max}, n-alkanol	C_{18}	C_{18}	C_{28}	C_{28}
Total, lipids	241.6	517.8	641.0	598.7
CPI, n-alkane	1.4	1.1	2.8	2.9
Plant wax, %[a]	19	6	48	49
$C_{18:1}/C_{18:0}$, %	76.9	44.6	40.6	37.8
$\geqslant C_{20}/<C_{20}$ fatty acids	0.4	0.2	0.8	0.9
CPI (C_{12}–C_{31}), fatty acids	11.2	13.8	7.1	6.4
CPI, n-alkanols	5.0	5.4	11.5	9.7

[a] Plant wax, %: the percentage of total plant wax alkanes in total n-alkanes, Wax $C_n = [C_n] - [(C_{n+1} + C_{n-1})/2]$ (Simoneit et al., 1991).

from the construction of the new airport, the island is sparsely populated, has little commercial activity and vehicular emissions, and virtually no industry. However, Lantau Island is on the downwind side of the Hong Kong city plume since the prevailing wind is easterly. Furthermore, because of the Zhujiang estuary (approximately 25–30 km across), the land-sea breeze effect is severe and flow reversals or recirculations have been observed (Kok et al., 1997). When there is a northerly component, airborne pollutants from the Zhujiang Delta can also be easily detected at Lantau. Thus it is not unusual to find high petroleum residues in the aerosol samples collected at Lantau. The C_{max} of UST1 was n-C_{29}, n-C_{31} and n-C_{25}, and the CPI was 1.4, thus suggesting the contribution was a mixture of biogenic material and petroleum residues. The distribution of alkane homologs seen in LT1 and UST1 (collected on a non-episodic day), was representative of Hong Kong's aerosols, and agreed with our previous three-year across the territory study.

For the dust samples, the CPI was 2.9 and 2.8 for LT2 and UST2, respectively. This is the highest CPI measured in Hong Kong by far; even the background sample collected at Waglan Island with a CPI of 1.95 (Fang et al., 1996) was low by comparison. This strong odd to even predominance suggested a strong terrestrial vascular plant wax alkane input (Eglinton and Hamilton, 1963). Compared to the 1–2 April samples, the concentration of the n-alkanes in the dust samples increased due to the large increase of plant wax input. The plant wax component was 49 and 48% in the alkanes in LT2 and UST2, respectively, while it was only 6% in LT1 and 19% in UST1. Studies at the Enewetak Atoll in the North Pacific (northeast trade wind regime) where large amounts of Asian Dust have been observed during April–May,

showed a strong terrestrial plant surface wax input for the lipids analyzed, and the CPI range was 2.4–4.0; the major alkanes were n-C_{27}, n-C_{29} and n-C_{31}, and trace amount of aliphatic hydrocarbons $< C_{20}$ was also present (Gagosian et al., 1981). The alkanes in the eolian dust over the Atlantic were found to range from n-C_{23} to n-C_{35} with high CPI values; the maximum was n-C_{27} in North Atlantic, n-C_{29} in equatorial Atlantic, and n-C_{31} in the South Atlantic (Simoneit, 1977). The distribution of n-alkanes in LT2 and UST2 was similar to the eolian dust over the Atlantic and Pacific, suggesting that there was an eolian input to Hong Kong strong enough to overshadow the local aerosol characteristics.

5.3.2. PAHs

Polycyclic aromatic hydrocarbons were detected in all samples with unsubstituted PAH compounds as the major aromatic components. The main PAHs were phenanthrene, fluoranthene, pyrene, chrysene, benzo-(ghi)fluoranthene, benzo(e)pyrene (BeP), benzo(a)pyrene (BaP), indeno-(1,2,3-cd)pyrene, benzo(ghi)perylene, and coronene. PAHs are derived mainly from anthropogenic sources such as pyrolysis, incomplete combustion and carbonization processes. The distributions and concentrations of PAHs do not show much difference among the samples. PAHs were not reported in the eolian dust over the Atlantic Ocean by Simoneit (1977), nor in the Harmattan aerosols by Cox (1982), nor in the oceanic aerosols over the North and South Pacific by Gagosian (1981, 1987). The PAHs detected in LT2 and UST2 were primarily from local emissions.

5.3.3. Fatty acids

Compared with the other cities in the world, the concentration of unsaturated and saturated fatty acids is

very high in Hong Kong, where an annual average of 2212.4 ng m^{-3} was reported for Mong Kok. Mong Kok is one of the busiest districts in Hong Kong and the high fatty acids level is probably due to the high population density. It has also been suggested that kitchen emissions due to the characteristic Chinese-style stir-fry cooking from the overwhelming number of restaurants in the city may also be responsible. From our previous study, the concentration of $C_{18:1}$ fatty acid can be more abundant than $C_{18:0}$ at some stations (Fang et al., 1996; Zheng et al., 1997).

Alkanoic acids ranged from C_{12} to C_{32} with C_{max} at C_{16} and C_{18}, and strong even carbon number predominance (CPI range 6.4–13.8). The alkanoic acids in the dust samples (UST2 and LT2) exhibited a slight bimodal distribution with C_{max} at C_{16} and minor at C_{24}. The ratio of $\geq C_{20}/< C_{20}$ fatty acids in the dust samples was 0.9 in LT2 and 0.8 in UST2, which was higher than LT1 (0.2) and UST1 (0.4). Homologs $< n-C_{20}$ are ubiquitous in biota and derived in part from microbial sources

(Simoneit and Mazurek, 1982). Higher ratios of $\geq C_{20}/< C_{20}$ in the dust samples indicated a higher input of plant waxes since homologs $> n-C_{22}$ are derived primarily from vascular plant wax (Simoneit, 1977). The CPI was 6.4 in LT2 and 7.1 in UST2, while it was 13.8 in LT1 and 11.2 in UST1. The $> n-C_{20}$ fatty acids in the dust samples did not exhibit a high even carbon predominance, and the higher the $> n-C_{20}$ fatty acids the lower the CPI.

Unsaturated fatty acids such as $C_{16:1}$, $C_{18:2}$ and $C_{18:1}$ were present in all samples. Unsaturated fatty acids are very unstable and are rapidly oxidized and degraded in the environment (Simoneit and Mazurek, 1982; Kawamura and Gagosian, 1987). The aged dust aerosols had a low level of unsaturated compounds due to their oxidative and photochemical lability relative to their saturated counterparts (Gagosian et al., 1981). This low level of the $C_{18:1}/C_{18:0}$ ratio in LT2 and UST2 provided supportive evidence for the long-range transport of the aerosols.
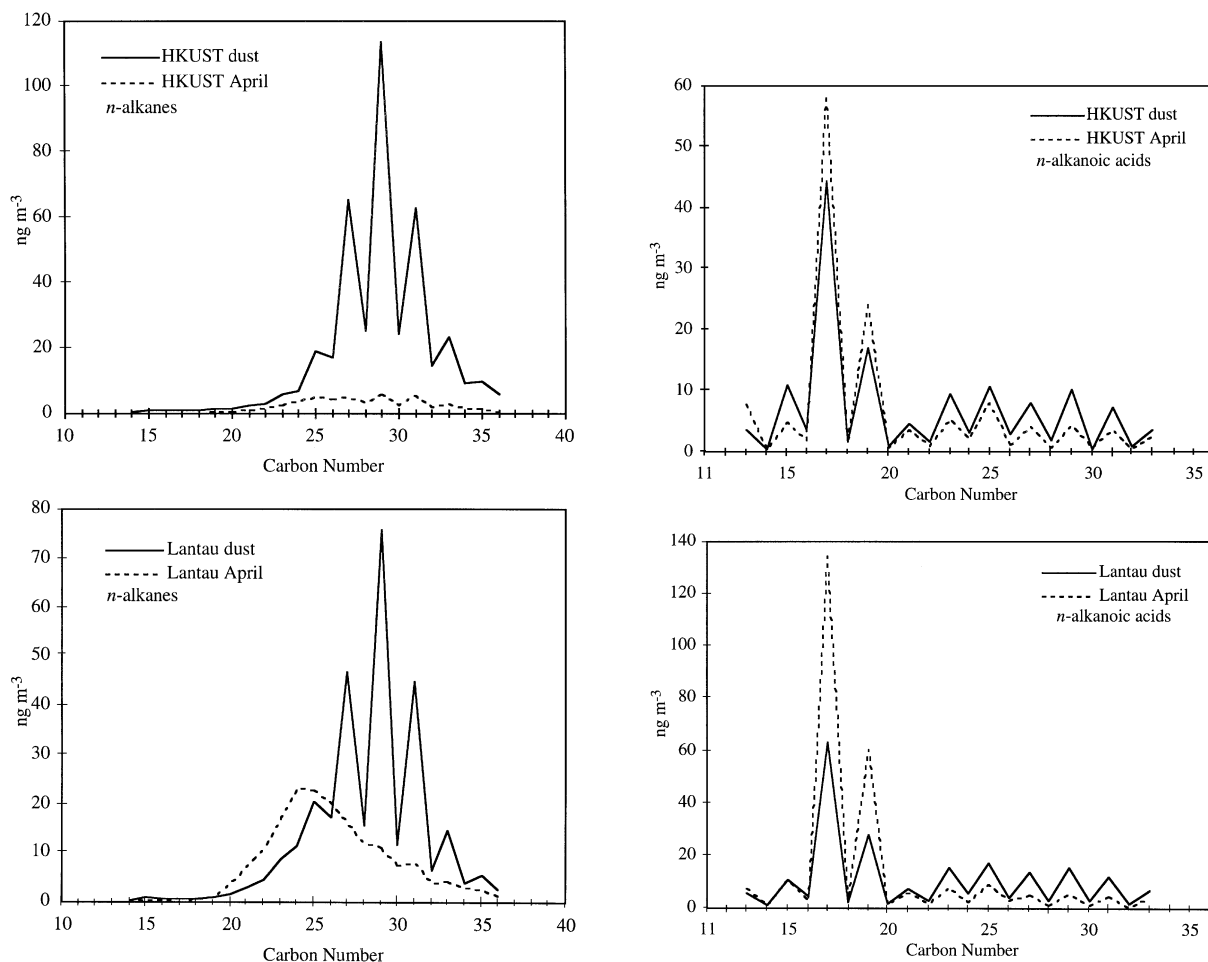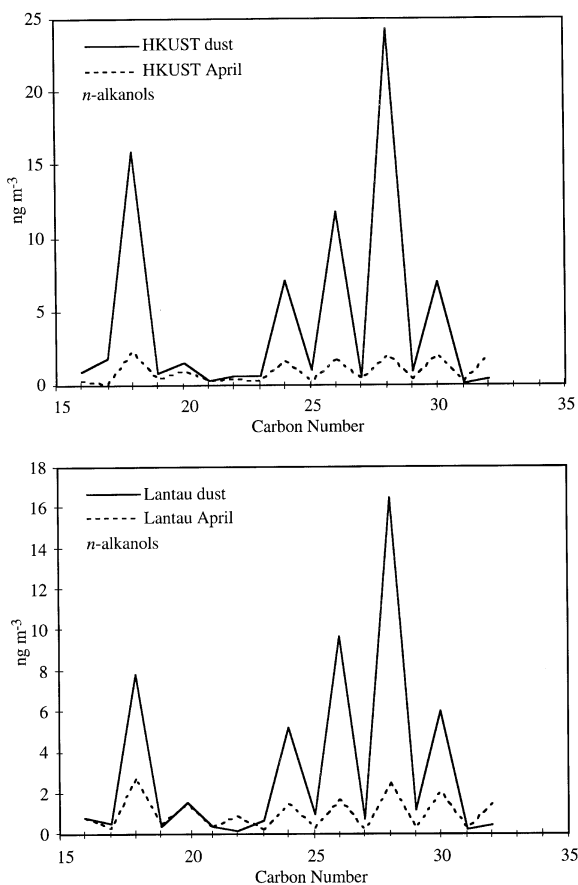

Fig. 5. (*Continued overleaf*).

Fig. 5. Distribution diagrams of solvent-extractable organic compounds including n-alkanes, n-alkanoic acids and n-alkanols in the non-episodic (1–2 April 1996) and episodic (9–10 May 1996) samples at HKUST and Lantau.

5.3.4. n-Alkanols

The alkanols ranged from C_{16} to C_{32} with very strong even to odd predominance (CPI range 5.0 – 11.5) and C_{max} at C_{28}, C_{18}. Higher CPI values were reported for the dust samples, e.g., 9.7 in LT2 and 11.5 in UST2. The concentration of n-alkanols in the dust samples at both stations was about three to five times that of the April samples, suggesting the increased input from higher plant waxes. From the long-range transport studies over the Pacific, it was found, of the terrestrially derived compounds, the n-alkanes and C_{21}–C_{36} fatty alcohols were generally the most abundant (Peltzer and Gagosian, 1989). In the epicuticular waxes of vascular plants, n-alkanes and $> C_{20}$ n-alkanols account for a major weight percentage of the waxes.

5.4. Mesoscale modeling

Due to the length of the present discussion, a paper on the simulation and the synoptic atmospheric phenomena is under preparation for presentation elsewhere, only some of the results pertinent to the provision of support of the dust episode are presented here.

The wind direction in the upper atmosphere was found to be mainly westerly. All times used are local time and China and Hong Kong are in the same time zone. On 9 May, 8 : 00 (24 h clock) a high surface pressure region existed at 990 mbar (27 °N, 114 °E) and the wind direction changed from west to east and south–east near that level. The model showed that the air mass from the north mixed with a clean air mass in the Bashi Channel, due to the vertical wind shear induced turbulence. From the meteorological point of view, the collapse of these two air masses divided the dust stream into two parts. One followed the clockwise motion induced by the surface high-pressure region and eventually reached Hong Kong. At 8 : 00 on 10 May, the high-pressure region (28°N, 128°E) caused the wind direction below 800 mbar to blow from the southeast. This could be the system which brought the dust to Hong Kong. Fig. 6a,b,c,d is a series of wind barb diagrams at the 990 mbar level over the area of interest at 1200, 7 May; 800, 8 May; 1200, 8 May; and 0000, 10 May 1996, respectively. It can be seen that the high-pressure region induced a clockwise rotational motion which was responsible for the transport of the dust to Hong Kong.

Wind trajectories were launched to trace the path the dust storm traveled in this episodic period using Vis5D (Hibbard, 1995). In Fig. 7, arrows are used to indicate the locations of the trajectories of the dust during the three days of travel over China and the China Seas before reaching Hong Kong.

5.5. Surface chart

Surface charts of 7–8 May 1996 of the Northern Hemisphere were examined. These charts contained ground meteorological observations reported from the various participating members. "Suspended dust" was reported at almost the same time as our simulation calculations near Shanghai and Hangzhou Bay on 8 May.

6. Conclusions

The results of the inorganic and organic analyses of the 9–10 May 1996 dust episode samples collected on the east and west sides of Hong Kong and the simulated mesoscale wind field study, demonstrated the transport of mineral aerosols from Northern China to Hong Kong (a distance of some 2500 km). Qualitative and quantitative results were obtained from X-ray spectroscopy and ICP-MS analysis, respectively. Compared to a normal spring day (1–2 April 1996), Fe, Al, Ca, S, and Cl were found to be enriched in the dust samples. Al and Fe are good indicators of crustal materials and high Ca content
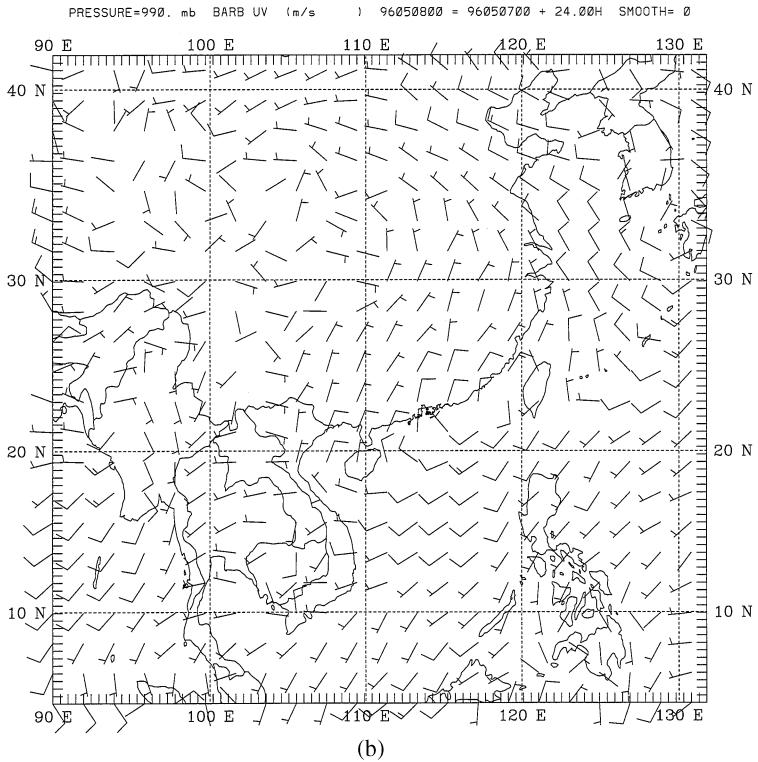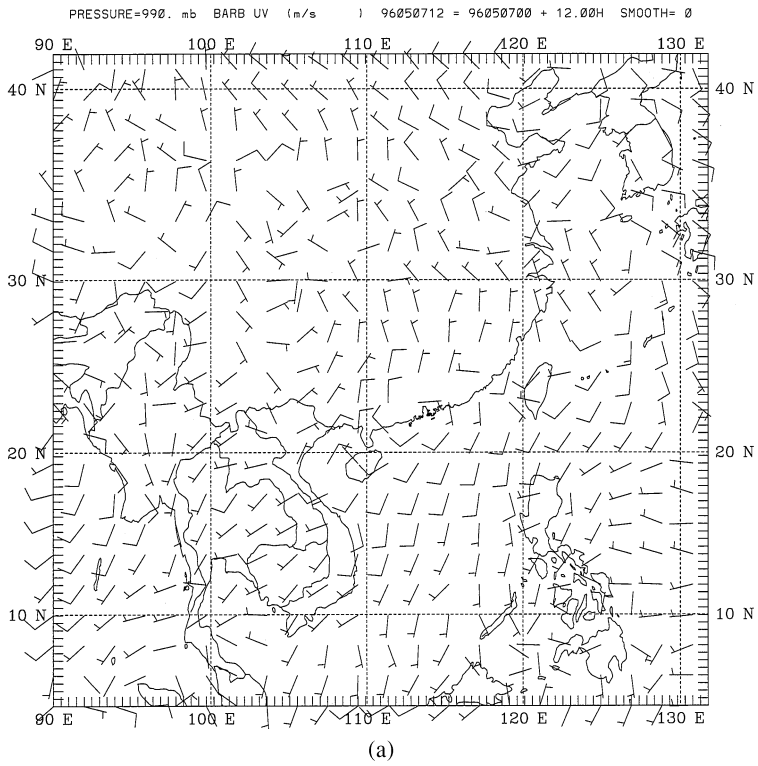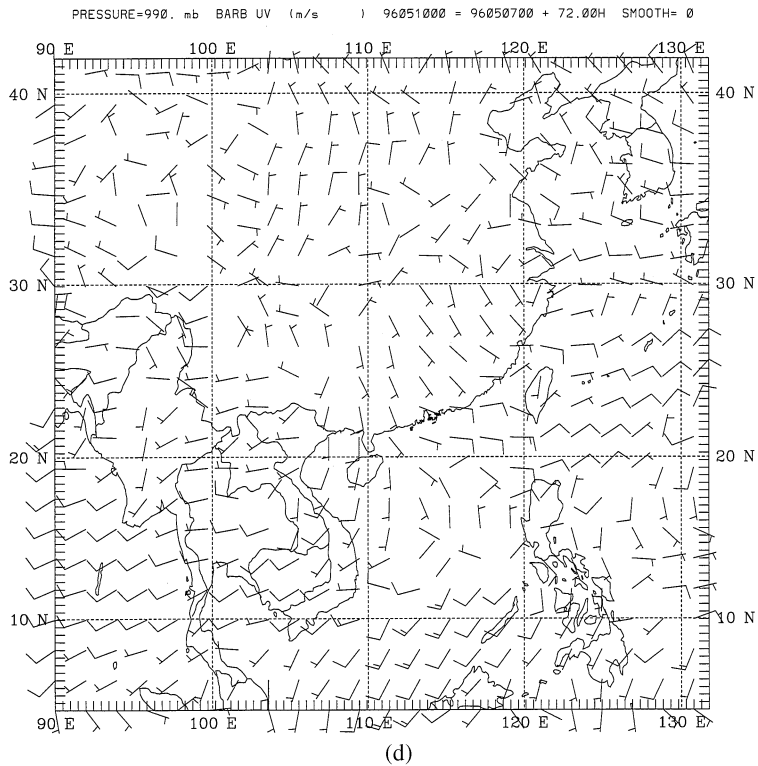
PRESSURE=990. mb BARB UV (m/s   ) 96050712 = 96050700 + 12.00H SMOOTH= 0

(a)

PRESSURE=990. mb BARB UV (m/s   ) 96050800 = 96050700 + 24.00H SMOOTH= 0

(b)

Fig. 6. (*Continued overleaf*).

PRESSURE=990. mb BARB UV (m/s   ) 96050812 = 96050700 + 36.00H SMOOTH= 0
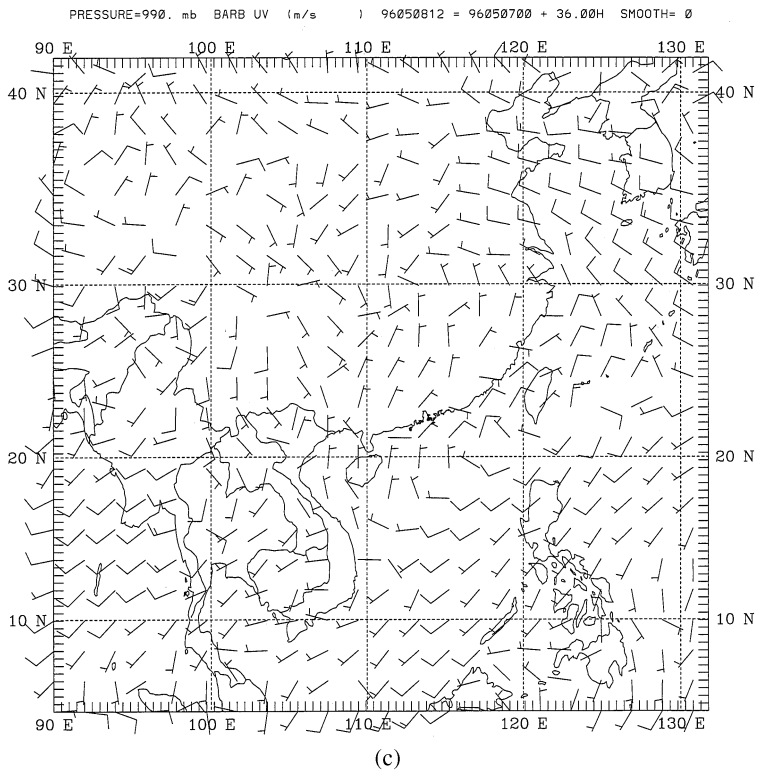
(c)

PRESSURE=990. mb BARB UV (m/s   ) 96051000 = 96050700 + 72.00H SMOOTH= 0

(d)

Fig. 6(a)–(d). Wind barb diagrams at 900 mbar – 96050712Z, 96050800, 96050812, and 96051000.
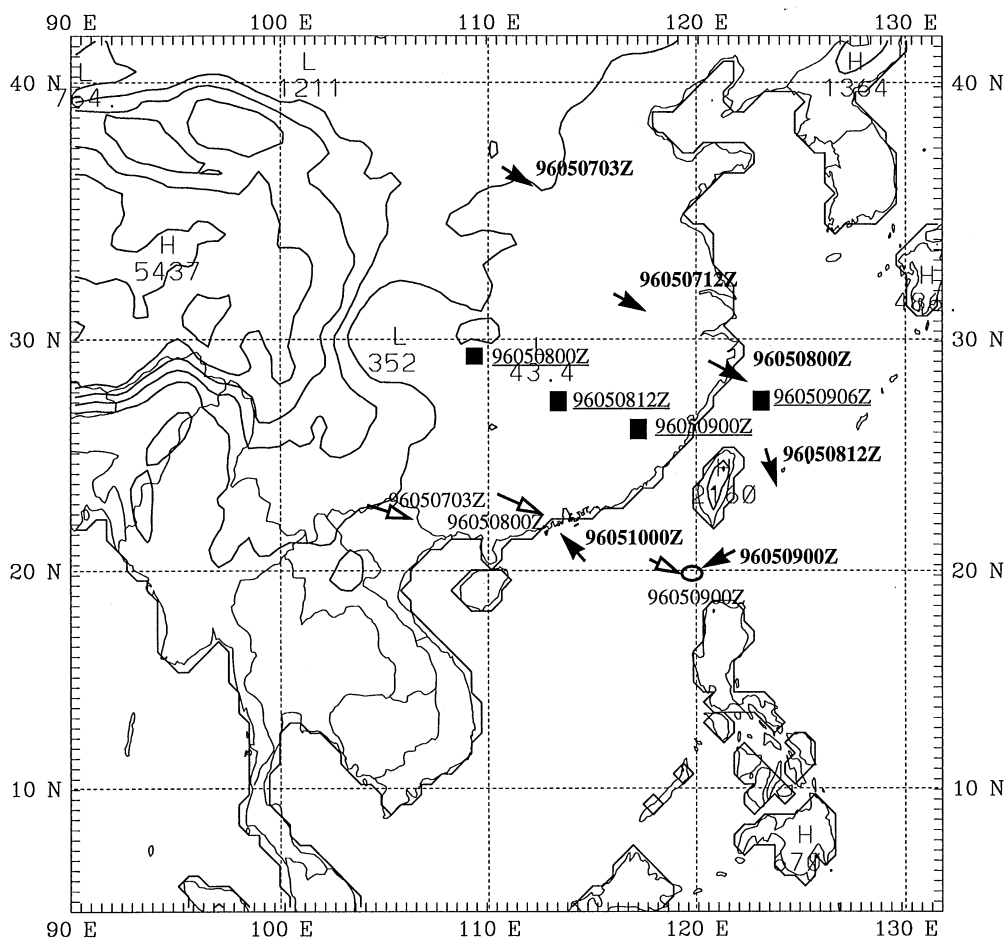
Fig. 7. Back trajectories. Black arrows are the dust trajectories; white arrows are air mass from Vietnam; ellipse is the location where the two air masses mixes. Bold dates are for the dust while plain dates are for the Vietnam air masses. Black boxes with underlined dates are centers of high surface pressure.

is a typical characteristic of soil in the northern part of China. The higher abundance of Cl in the dust samples suggested the aerosols transported from a long distance by way of the East China Sea, while the increase in S and Pb content in the aerosols was probably a result of the transport of pollutants along with the dusts. Coal is widely used in China, particularly, for residential heating in winter. A comparison with EPD data collected at three stations from April to May in 1994 showed an extremely high abundance of Al, Fe, Ca in the 5 May 1994 samples, suggesting that a similar dust event might have occurred. Compared to EPD data of the normal samples, all elements in the 9–10 May 1996 samples showed the lowest enrichment factors.

The organic characteristics of the 9–10 May samples were significantly different from the non-episodic 1–2 April samples and the results obtained from our previous

three-year territory-wide study. The latter showed typical local contributions, especially at Lantau, which tends to accumulate pollutants from the city plume due to micro-meteorology and the terrain effect. The 9–10 May samples at both sites, however, showed an unusual distribution and composition of lipids. Indicating parameters such as CPI, C_{max}, ratio of $\geqslant C_{20}/<C_{20}$ fatty acids, the distribution and concentration of n-alkanes and n-alkanols all provided supportive evidence that the sources were not all local. As a matter of fact, a strong eolian input with high plant wax content overshadowed local emissions. This eolian input exhibited the same characteristics compared to the long-range transport dust samples collected over the Pacific. A low ratio of $C_{18:1}/C_{18:0}$ fatty acids in the dust samples indicated that the aerosols were aged due to long-range transport.

The simultaneous increase in the concentration of crustal elements, e.g. Fe, and some pollution elements such as Pb and S, and the similarities in the organic characteristics of the dust samples collected at the east and west sides of Hong Kong suggested that the episode was region-wide.

Mesoscale modeling showed the path of dust during the three days of travel over China and the China Seas before reaching Hong Kong. Not only winds in the upper troposphere transport the Asian Dust to a large distance, the winds near the surface also play a significant role in long-range transport. Ground observations of suspended dust in Shanghai on 8 May 1996 provided additional support to the chemical analyses and modeling. With the multi-technique approach, it is possible to conclude that the dust in the aerosol samples had its origin in Northern China.

Acknowledgements

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