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Chemical characteristics of aerosols at coastal station in Hong Kong. I. Seasonal variation of major ions, halogens and mineral dusts between 1995 and 1996

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Abstract

Aerosols samples (total suspended particulate "TSP" and PM_{10} particulate) were collected at coastal monitoring station in Hong Kong between 1995 and 1996. They were analyzed to investigate the seasonal cycle among major ions (Na^+, NH_4^+–N, K^+, Mg^{2+}, Ca^{2+}, Cl^-, NO_3^-, and SO_4^{2-}), halogen elements (Br, I) and mineral dusts. The concentration of major ions showed a summer minimum and a winter maximum in a coastal region of Hong Kong. Halogen (Br, I) and dust concentrations exhibited a very similar variation. From the molar equivalence of Na^+ and Cl^-, it was found that the Cl^- in TSP or PM_{10} was deviated from sea-salt (NaCl) component. Bromine (Br) in aerosols evidently originated from marine source but iodine (I) in aerosols may have been generated from natural and anthropogenic sources including possible biomass emission. The ratios of nonsea-salt sulfate (nss-SO_4^{2-}) to nitrate (NO_3^-) and trace elements (As, Sb, Se, Pb, V and Zn) were used to explain the different pollution emissions. © 2000 Elsevier Science Ltd. All rights reserved.

Keywords: TSP; PM_{10}; Nonsea-salt sulfate; Trace elements; Dust

1. Introduction

In the East Pacific rim region, including Hong Kong, huge amount of SO_2, NO_x and anthropogenic species is observed and high emissions of SO_2 are also observed in the mainland area China. (Akimoto and Narita, 1994). Galloway (1989) anticipated that this region will become the largest source of SO_2 and NO_x in the world. Some fraction of the emitted SO_2 and NO_x, through the heterogeneous or homogeneous reaction, are oxidized to sulfate and nitrate aerosols before dry or wet deposition. These acidic aerosols have been recognized as one of the most important consequences of atmospheric pollution. They have harmful impact on human health (Vincent, 1989), natural environment and ecological system. The anthropogenic nonsea-salt sulfate (nss-SO_4^{2-}) aerosols can interact with solar and terrestrial radiation directly affecting climate (Charlson et al., 1992) and they alter the size distribution of cloud particles through nucleation processes to cause indirect climate effects. Acidic aerosols can react with gaseous ammonia (NH_3) to form completely or partially neutralized ammonium salts (Koutrakis et al., 1988). In southern parts of China, the lower availability of ammonia permits much of the sulfates to be deposited as acidic aerosols (Zhao et al., 1985). It is therefore necessary to characterize the chemical composition of aerosol at coastal area in Hong Kong.

During 1993–1994, the first TSP and PM_{10} aerosol particles were collected with the aim of understanding the aerosol pollution levels at the coastal area in Hong Kong (Lam et al., 1997). In this paper we present the results of one year (April 1995–April 1996) of daily aerosol collection at the same station. The major objective of the present study is to further understand the seasonal

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variation of aerosol composition and the impact of regional sources. The collected samples were analyzed for a range of major ions including sea salts, halogens (Br and I), mineral dusts and anthropogenic substances (NO_3^-, nss-SO_4^{2-}, nss-K^+ and trace elements). The results of these analyses are discussed along with the atmospheric processes controlling the aerosol concentrations.

1.1. Site location

The station established at Cape D'Aguilar is located at end of a long peninsula on the SE Hong Kong island. The station (22°13′N, 114°15′E, 60 m above sea level. Fig. 1) is sited on a cliff top facing the South China Sea and the West Pacific Ocean, where the average prevailing wind direction measured at the station is from the east. The station has more than 180° of sea view stretching from the northeast to the southwest. For over 60% of the time the wind comes off the sea. The population density on the peninsula is relatively low and the closest industrial town, Chai Wan, is located 10 km away.

The prevailing winds for Hong Kong are easterly and southeasterly in summer. During the winter season the prevailing winds are from north and northeast bring in the air pollutants from the Chinese mainland. Occasionally urban plumes affect the levels of atmospheric species.

2. Experimental

Atmospheric aerosol samples were collected between April 1995 and April 1996 (samples for major ions were extended to May 1996). The sampler was installed at a height of 60 m above sea level and high-volume air sampling techniques were used to collect the aerosol for analysis. The General Motor Works GMW (model SAUV-11H) high-volume air sampler was used for collecting PM_{10} and the GMWL-2000 high-volume air sampling system for collecting TSP. The samplers were manufactured by General Metal Works Inc. and Andersen Samplers Inc. Aerosols were collected by drawing

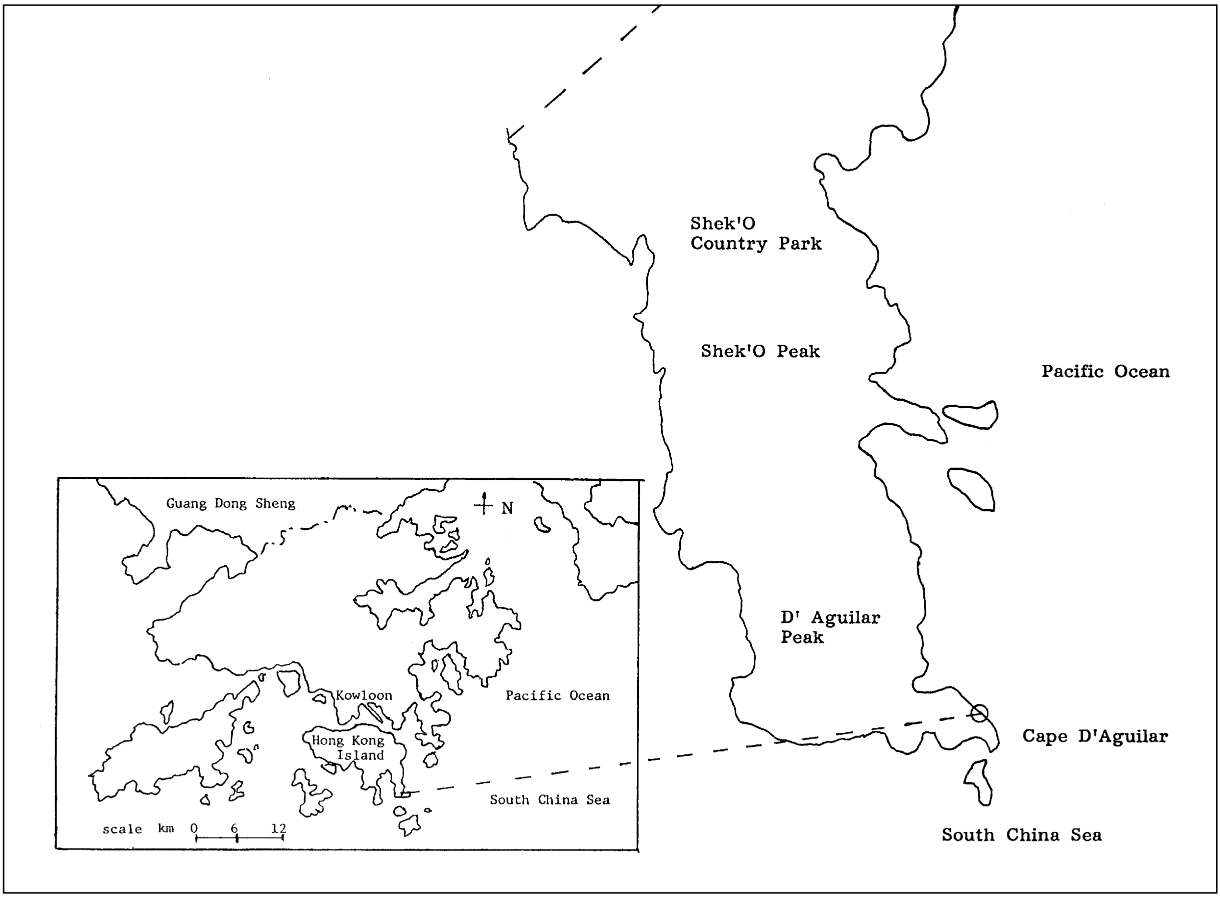

Fig. 1. Location of the site, Capte D'Aguilar of Hong Kong.

air at a rate of $1270\,l\,m^{-1}$ ($\pm 12\%$) for PM_{10} and $1370\,l\,m^{-1}$ ($\pm 7\%$) for TSP through a Whatman-41 filter (20×25 cm). Under high-volume sampling conditions Whatman-41 filters are relatively efficient (Prospero, 1989). In the marine boundary layer the filters have efficiencies of greater than 95% for SO_4^{2-}, Na^+, K^+, Mg^{2+} and 90% for NH_4^+-N, NO_3^-. Compared with some alternative filters the nitrate and sulfate artifacts are relatively small (Savoie and Prospero, 1982). Thus Whatman-41 filter was used for the collection of aerosol during the entire program. The field blank was measured, i.e. the filters were exposed in the sampler without drawing air, while the other operating conditions were similar to those during the collection of aerosol samples.

After sampling, the filters were sealed in clean plastic bags, transported to the laboratory and then stored in refrigerator. The temperature was maintained at $-18°C$. Before analyzing the filters were cut into several portions for individual analysis. The water-soluble constituents were determined by suppressed ion chromatography (IC). The filters were extracted ultrasonically by water which was deionized to resistivity of $18\ M\Omega\,cm^{-1}$ and then distilled in a quartz vessel. The weak base eluent ($1.8\ mM\ Na_2CO_3 + 1.70\ mM\ NaHCO_3$) was used for anion ion detection, while the weak acid eluent (20 mM HCl) for cation ion detection. The instrument employed for ion measurement was a Dionex D-100 ion chromatography. The quality assurance and control were routinely carried out by using Standard Reference Materials produced by National Research Center for Certified Reference Materials, China and checked with NIST standards SRM 3182 chloride and SRM 3185 Nitrate. Uncertainties in the concentration of soluble ions are $\pm 3\%$ (Na^+), $\pm 5\%$ ($NH4^+$-N, K^+, Mg^{2+}, Ca^{2+}) and $\pm 3\%$ (Cl^-, NO_3^-, SO_4^{2-}). Al, Br, I, Fe, As, Sb, Se, Sc, V, Zn and total Mg, Ca, Na, Cl elements were measured by instrumental neutron activation analysis (INAA) operated by Institute of High Energy Physics, Chinese Academy of Sciences. The filters and reference material were placed in a nuclear reactor exposing them to neutrons (neutron flux $= 4.17 \times 10^{11}$ neutrons $cm^{-2}\,s^{-1}$ for "short" irradiation, 5 min; 6×10^{13} neutrons $cm^{-2}\,s^{-1}$ for "long" irradiation 24 h). SRM 1633a coal fly ash was used as quality assurance during routine INAA analysis. Due to the uncertainties in the flow measurement system and field blank correction, the uncertainties in the concentration of most elements are estimated to be about ± 12–15%. Pb was measured by ICP-MS techniques. The samples were digested with a mixture of nitric, perchloric and hydrofluoric acid at 150°C in platinum (or polytetrafluoroethylene) crucibles. SRM1648 urban particulate was used as quality control. Analysis was carried out on a Plasma Quard II ICP-MS Instrument from VG Elements, England. The analytical procedure will be described in next paper. The uncertainties were estimated to be about $\pm 15\%$.

An improved method was used for measuring the pH of aqueous extracts of TSP and PM_{10} (Koutrakis et al., 1988). These measurements represent total extractable acidity H^+_{extr}. The apparent hydrogen ion concentration, H^+_{extr}, was calculated as the following equation:

$$H^+_{extr} = 10^{-pH} - 10^{-pHb},$$

where pH and pHb are, respectively, the pH of the test solution and the blank solvent (diluted acidic water and field blank filter). The pH values were measured with reference standard buffer pH solutions (4.003 and 6.848, uncertainties = 0.005pH at 25°C) which were produced by the National Research Center for Certified Reference Materials, China. The particle acidity H^+_{estm} was estimated from ions [$neq\ m^{-3}$] balance model (Saxena et al., 1993):

$$\begin{aligned} H^+_{estm}\ (ng\ m^{-3}) = {} & ([SO_4^{2-}] + [NO_3^-] + [Cl^-]) \\ & - ([Na^+] + [NH_4^+\text{-}N] \\ & + [K^+] + [Mg^{2+}] + [Ca^{2+}]). \end{aligned}$$

3. Results and discussion

3.1. Water soluble ions

The monthly and annual average soluble ion concentration of aerosols were given in Figs. 2a, b and Table 1a, b. For TSP, the sea salt (Na^+, Cl^-) accounts for 60% of the total soluble aerosol. The next species are SO_4^{2-} and NO_3^- at 23% and 9%, followed by NH_4^+-N, K^+, Mg^{2+} and Ca^{2+} together accounting for 8.8%, while for PM_{10}, the SO_4^{2-} and NO_3^- account for 34% and 11%. The Na^+, Cl^- ions, at 19% and 25%, the other ions such as NH_4^+-N, Mg^{2+}, K^+ and Ca^{2+} together accounting for 12%. Table 1b shows that total concentrations of Na, Cl (measured by INAA) were close to concentrations of soluble ion Na^+, Cl^- (measured by IC) for TSP and PM_{10}. The maxium difference between concentrations of total Na and Na^+ for TSP was about 20%. The average concentrations of Mg_{total} were about twice as high as that of Mg^{2+}. The total extractable acidity H^+_{extr} for TSP or PM_{10}, make up $\sim 0.03\%$ of total soluble aerosol. The particle acidity H^+_{estm} for PM_{10} was close to H^+_{extr}, whereas the H^+_{estm} of TSP was about double the H^+_{extr}. The reason for this may be the loss of ammonia from the sample media. In Table 1a, it is demonstrated that the ammonium concentrations of PM_{10} are higher than those of TSP. This was caused by evaporation of volatile ammonium ion to the gas phase (Kim, 1997).The results show that a seasonal variation of summer low and winter high is clearly displayed in the concentration of cation ions (Na^+, NH_4^+-N, K^+, Mg^{2+} and Ca^{2+}) and concentration of anion ions (Cl^-, NO_3^- and SO_4^{2-}). Both clean marine air mass and scavenging effects due to wet

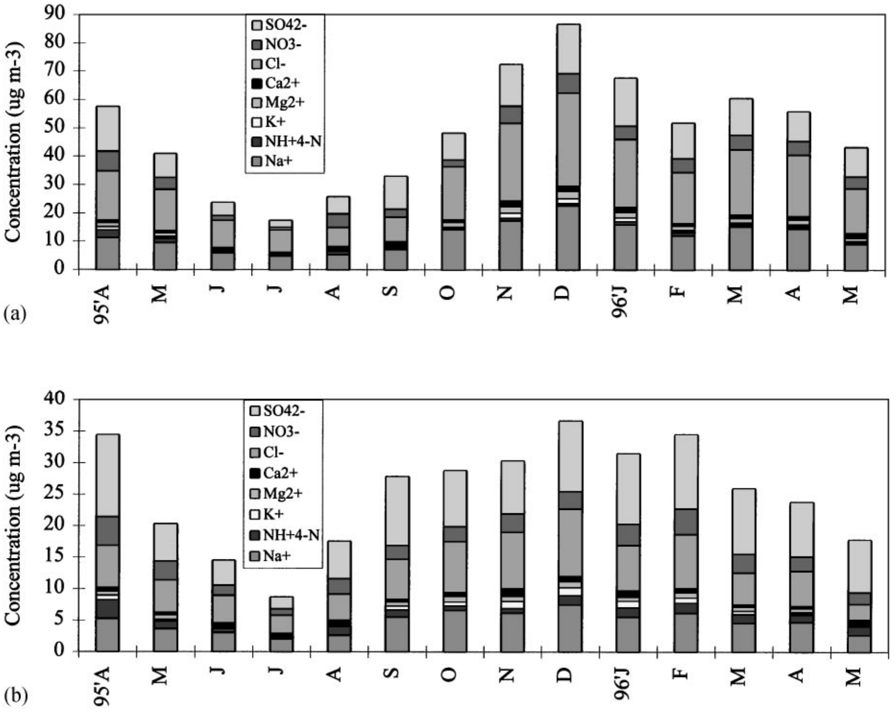

Fig. 2. (a) Average monthly composition of major ions (TSP); (b) Average montly composition of major ions (PM_{10}).

weather contribute to the summer low. In winter the continental outflow mainly from the mainland of China passes through vast source areas and this, combined with the dry and cold weather, contributes to high aerosol concentrations. The obtained results are similar to our previous aerosol studies (Lam et al., 1997).

3.2. Ion balance

The ion balance expressed by the ratio of cation sum to anion sum (μeq m^{-3}) was calculated from value of every ion species and the mean ratio of cation sum to anion sum ranges from 0.98 (stdev = 0.0903, std err* = 0.0082, $n = 120$, TSP) to 0.99 (stdev = 0.084, std err = 0.006, $n = 195$, PM_{10}). If the H^+_{extr} concentrations are ignored in the cation sum of TSP and PM_{10}, the ratio of cation sum to anion sum is 0.97 (stdev = 0.090, std err = 0.008, TSP) and 0.97 (stdev = 0.077, std err = 0.005, PM_{10}). Since aerosols were collected at a coastal monitoring site, it shows that Cl^- existing in the atmosphere of Cape D'Aguilar of Hong Kong can be expected at relative high concentrations.

To estimate the reference composition of a sea-water sample was collected on 13 June 1996 at the South China Sea. The sampling location was situated at a short distance (about 1–2 Km) east of station. Sampling depth was 0.5 m. The data sets incorporated with Wilson's (1975) are given in Table 2. The results demonstrated that ratios of sea salts (Cl^-/Na^+, K^+/Na^+, Mg^{2+}/Na^+, Ca^{2+}/Na^+ and SO_4^{2-}/Na^+) varied in terms of several percentages. The standard deviation estimated was ± 5%.

* std err stand for standard error of mean.

3.3. Chlorine

Table 2 shows that chlorine "loss" has occurred. This "loss" occurred during the 1993–1994 aerosol collection (Lam et al., 1997). The chlorine "loss" may be explained by the reaction of gases HNO_3/or H_2SO_4 with NaCl in sea-salt particles (Wall et al., 1988) generally formulated by

$$HNO_3 + NaCl\,(\text{sea salt}) \rightarrow NaNO_3(\text{sea salt}) + HCl$$

or by the absorption of gaseous SO_2 by sea-salt droplets that was oxidized to H_2SO_4 and the release of gaseous HCl (Hitchcock et al., 1980). The chlorine "loss" was also found in Sapporo site, Japan (Ohta and Okita, 1990; Kaneyasu et al., 1995). Based on the Ohta's calculation model (1990), the chlorine "loss" was estimated. In Cape D'Aguilar station the chlorine "loss" was 15–16% in summer (Jun, Jul and Aug) and in winter (Nov, Dec and Jan) was 15–20% for TSP and PM_{10}. These data show that the reaction between HNO_3/or H_2SO_4 and NaCl (sea salts) predominate all the year round.

Table 1

a. Annual concentrations of major ions at coastal station in Hong Kong between 1995 and 1996

	Na^+		NH_4^+–N		K^+		Mg^{2+}		Ca^{2+}		Cl^-		NO_3^-		SO_4^{2-}		H^+_{extr} ng m^{-3}		H^+_{estm} ng m^{-3}	
	TSP	PM_{10}	TSP	PM_{10}	TSP	PM_{10}	TSP	PM_{10}	TSP	PM_{10}	TSP	PM_{10}	TSP	PM_{10}	TSP	PM_{10}	TSP	PM_{10}	TSP	PM_{10}
Average	11.13	4.77	1.08	1.30	0.78	0.58	1.38	0.61	0.91	0.55	16.80	6.44	4.21	2.70	10.77	8.57	9.68	8.49	19.2	10.9
Median	10.36	4.41	0.66	1.03	0.64	0.42	1.24	0.56	0.69	0.38	14.59	5.26	3.17	2.29	10.15	8.20	5.73	6.48	14.1	8.64
Geomean	7.99	3.79	0.65	0.96	0.55	0.40	0.98	0.49	0.62	0.37	10.45	4.51	3.06	2.18	8.32	6.85	4.80	4.97	—	—
Stdev	7.98	2.98	1.10	1.01	0.60	0.48	1.02	0.38	0.91	0.58	13.19	4.86	3.22	1.79	6.94	5.18	11.45	9.16	84.7	34.2
Max.	32.67	18.19	4.96	5.42	2.78	2.21	4.45	2.23	7.42	4.70	52.39	29.82	15.70	8.82	34.67	27.03	62.50	60.70	323.4	152.1
Min.	0.98	0.24	0.073	0.097	0.075	0.024	0.13	0.035	0.09	0.032	0.19	0.098	0.20	0.31	1.40	0.95	0.022	0.011	− 354	− 111.8
n	120	195	119	195	120	195	120	195	120	195	118	195	120	195	120	195	117	172	120	195

b. Annual total concentrations of Na, Mg, Ca and Cl by INAA

	Na		Mg		Ca		Cl	
	TSP	PM_{10}	TSP	PM_{10}	TSP	PM_{10}	TSP	PM_{10}
Average	8.86	4.67	2.24	1.13	0.96	0.71	15.03	5.93
Median	8.20	4.35	1.93	0.96	0.73	0.47	11.00	4.88
Geomean	6.77	3.80	1.59	0.87	0.69	0.46	9.07	3.84
Stdev	5.90	2.76	1.84	1.01	0.83	0.89	13.91	6.36
Max.	24.80	13.80	11.60	11.20	4.71	8.99	89.60	67.60
Min.	1.10	0.26	0.24	0.074	0.14	0.061	0.39	0.03
n	114	189	114	189	114	189	114	189

Note: *n*, stdev stand for number of samples and standard deviation, concentration unit: μg m^{-3} except for H^+ (ng m^{-3}).

Table 2
Average of ionic concentration ratios (μeq m^{-3}/μeq m^{-3}) in aerosol samples and sea water

	Sea water ($n = 3$)		Ionic concentrations ratios			
Ratio	Present work	Wilson (1975)	TSP	PM_{10}	TSP (1993–1994)	PM_{10} (1993–1994)
Cl^-/Na^+	1.164(stdev, 0.012)	1.16	0.98	0.88	0.96	0.76
K^+/Na^+	0.0218(0.00006)	0.0218	0.037	0.073	0.032	0.041
Mg^{2+}/Na^+	0.223(0.0025)	0.227	0.24	0.24	0.20	0.14
Ca^{2+}/Na^+	0.0415(0.0014)	0.0439	0.094	0.13	0.14	0.17
SO_4^{2-}/Na^+	0.121(0.002)	0.121	0.46	0.86	0.75	1.23
Ca^{2+}/Mg^{2+}	0.186(0.005)	0.194	0.40	0.55		
Na^+/Mg^{2+}	4.47(0.05)	4.41	4.25	4.11		
K^+/Mg^{2+}	0.0976(0.001)	0.0961	0.18	0.30		
Cl^-/Mg^{2+}	5.21(0.104)	5.14	4.16	3.61		
SO_4^{2-}/Mg^{2+}	0.538(0.012)	0.532	1.97	3.53		

3.4. *Bromine and iodine*

The aerosol data were sorted by month, and each month's data were plotted in a "box and whisker". Figs. 3a, b and 4a, b show that seasonal variation in Br and I concentrations are presented at Cape D'Aguilar station. Concentrations of Br and I were the lowest in summer (Jun, Jul and Aug) but an increase in particulate occurred in winter (Nov to Dec). This is quite similar to the variation in the concentration of major ions. The marine enrichment factors (EF) were calculated by considering Na^+ in bulk sea water (Duce et al., 1983):

$$EF(x) = \{[X]_{air}/[Na]_{air}\}/\{[X]_{sea}/[Na]_{sea}\}.$$

The sea water enrichment factor for Br is around 1, whilst for I it ranged 120–450 for TSP and PM_{10}.

The annual concentration of nss-SO_4^{2-}, nss-K^+, NO_3^- and trace elements were given in Table 3. By examining correlation with other elements, the correlation coefficient (γ) shows that for TSP, Br correlated with the soluble ions Cl^-, Na^+, K^+, Mg^{2+}, I ($\gamma = 0.88, 0.87, 0.81, 0.81, 0.45$) and pollution elements Pb, Se ($\gamma = 0.55, 0.35$), whereas for PM_{10}, Br correlated also with Cl^-, Na^+, K^+, Mg^{2+}, I ($\gamma = 0.76, 0.80, 0.69, 0.81, 0.51$) and Pb, Se ($\gamma = 0.58, 0.56$). This suggests that sea-salt Br is the major Br component. There may also have been some conversion of pollutants (Cicerone et al., 1988; Yoshizumi, 1991) or natural (biogenic) (Sturges et al., 1992) gaseous Br into the particulate phase, but without data on the gaseous Br species and on the detailed size distribution of the particulate Br one can only speculate on these other Br sources. Although the oceans are a major source for atmosphere I (Cicerone, 1981), the correlation coefficient ($\gamma = 0.34$–0.25) shows that iodine slightly correlated with sea salt (Na^+, Cl^-) for TSP or PM_{10} and in contrast correlated with the pollution elements Sb, Se, nss-SO_4^{2-}, NO_3^-, K^+ and nonsea-salt soluble K^+ (nss-K^+). (0.46, 0.59, 0.68, 0.66 0.54 and 0.64 for TSP, 0.52, 0.69, 0.62, 0.61, 0.63 and 0.64 for PM_{10}). The nss-K^+ concentrations were calculated as nss-$K^+ = -[K^+] - [Na^+] \times 0.037$. (Hitchcock et al., 1980). Most of the Se is from fossil combustion and ocean (biogenic) emission. The correlation between Se and I ($\gamma = 0.59, 0.69$ for TSP and PM_{10}) suggests that the source of Se is also an important source of I. Combined EF values of I and correlation with pollution elements (Sb, Se, nss-SO_4^{2-}, NO_3^- and nss-K^+), indicate that iodine in TSP or PM_{10} is mainly generated from anthropogenic and marine source at Cape D'Aguilar station of Hong Kong. In terms of the aerosol constituents collected from a fire plume (Maenhaut et al., 1993) and correlated with Sb, Arimoto et al. (1995) suggested that one possible source for the iodine was biomass burning. Since the oceanic emissions of Sb are small compared with those from continental sources (Nriagu, 1989), Arimoto et al. (1995) suggested the use of soluble potassium (K^+) or black carbon as tracers for biomass burning. Echalar et al. (1995) observed that the potassium enrichment factors were very high (EF values > 10). These results show that K^+ may be regarded as a good tracer for the flaming phase of biomass burning. In Hong Kong in the coastal area iodine (I) in atmosphere is correlated with K^+ and nss-K^+ ($\gamma = 0.54, 0.64$ for TSP, 0.63, 0.64 for PM_{10}). The average sea water EF values of K^+ and nss-K^+ were 2,1(TSP) and 3,2 (PM_{10}). During the time of Autumn and Winter (Nov, Dec, Jan and Feb) the EF values of nss-K^+ were slightly enhanced for TSP and PM_{10}, but the maximum EF values of PM_{10} were 9–38. It seemed that some amount of biomass emission might have had an impact on the station. It was known that in China several portions of rice straws, wheat and maize stalks were burned in field during the Autumn and Winter. During the biomass burning a range of air pollutants are emitted and under certain meteorological conditions they are transported. The other evidence is

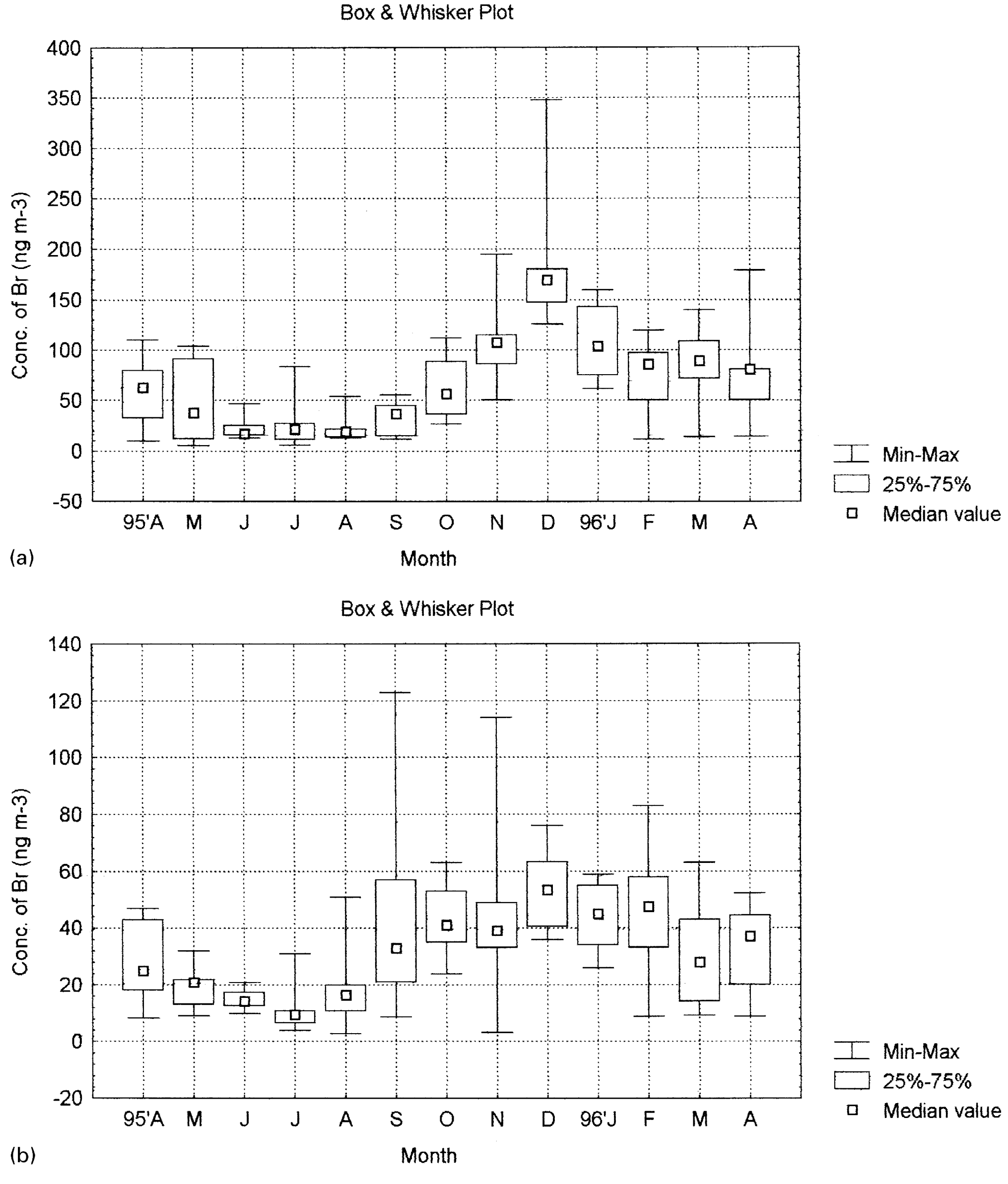

Fig. 3. (a) Monthly median concentrations of Br for TSP. (b) Monthly median concentrations of BR for PM_{10}.

that municipal incinerators also contain some form of biomass burning, and therefore incinerator emissions are also often rich in fine K. Based on the above-mentioned facts we suggest that the major part of iodine aerosols may be formed through the gas to particle reaction involving natural and anthropogenic species, including possible biomass emission.

3.5. *Nonsea-salt sulfates (nss-SO_4^{2-}) and nitrates*

The concentrations of nss-SO_4^{2-} are calculated as nss-SO_4^{2-} = [SO_4^{2-}] − [Na^+] × 0.2516 (Millero and Sohn, 1992). According to this calculation, the annual average of nss-SO_4^{2-} was 8.04 μg m^{-3} (stdev = 5.87, std.err = 0.54, $n = 119$) for TSP, 7.37 μg m^{-3} (stdev = 4.88, std.err = 0.35, $n = 195$) for PM_{10}. Comparison of nss-SO_4^{2-} concentrations measured at the same station during the PEM-WEST B (February–March, 1994) the mean concentration of nss-SO_4^{2-} measured by Prospero et al. (1995) were 10.55 μg m^{-3} (stdev = 5.25, $n = 34$), whilst in Feb–Mar, 1996 the mean value of nss-SO_4^{2-} measured by us was 11.78 μg m^{-3} (stdev = 5.61, $n = 17$) and 9.74 μg m^{-3} (stdev = 5.28, $n = 29$) for TSP and PM_{10}, respectively. Prospero et al. (1995) also observed

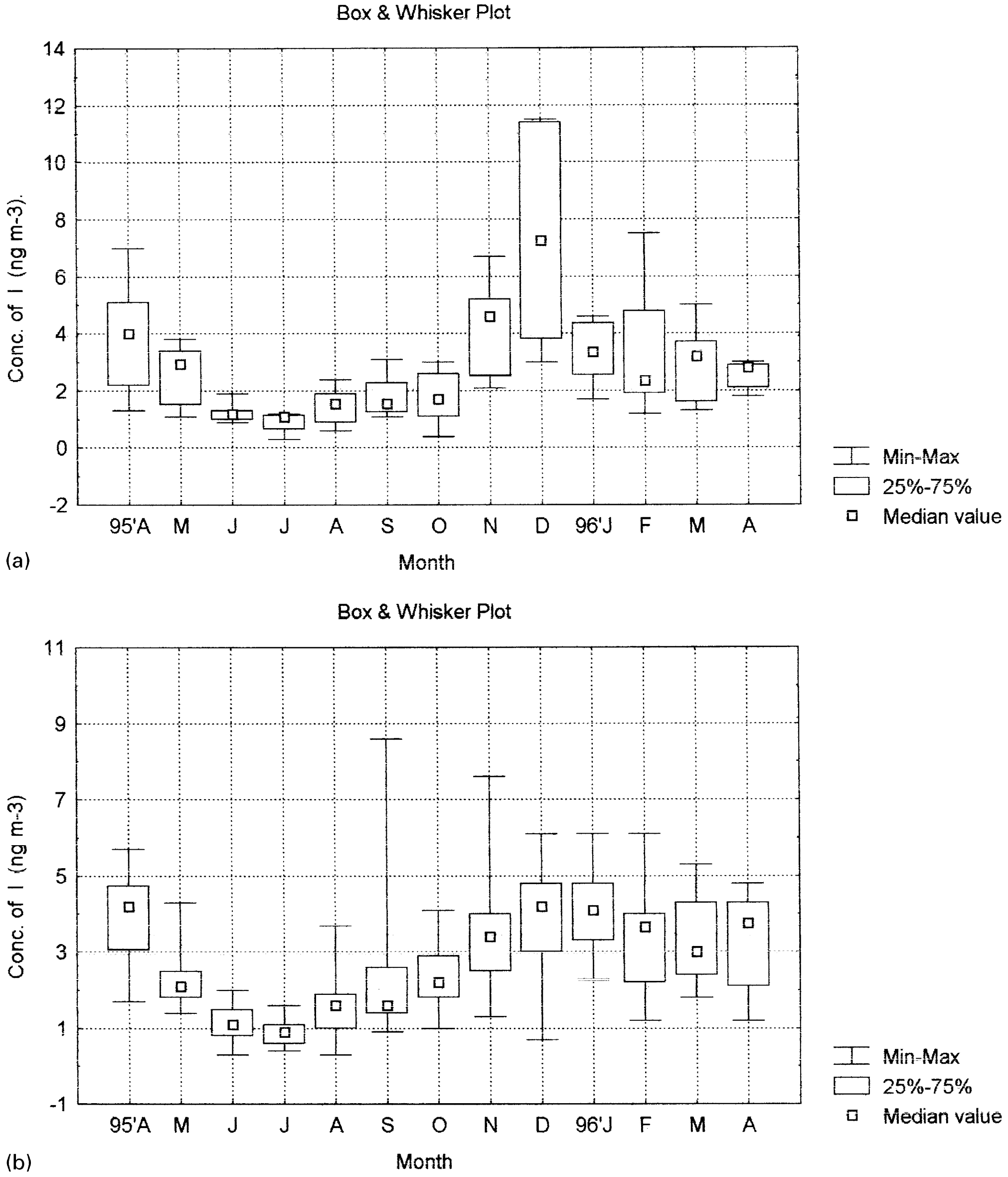

Fig. 4. (a) Monthly median concentrations of I for TSP. (b) Monthly median concentrations of I for PM_{10}.

that about two-thirds of nss-SO_4^{2-} and NH_4^+ were in the size fraction below 1.2 μm diameters, while nitrate was predominantly in super-micron fraction. The mass median diameter (MMD) of nitrate in Hong Kong in the coastal area was greater than that measured at other sites. The arithmetic average ratios of nss-SO_4^{2-} to NO_3^- (2.08 ± 1.15, $n = 119$, 2.98 ± 1.92, $n = 195$ for TSP and PM_{10}) were approximately close to coastal site at Xiamen (24°N, 118°E) (1.8 ± 0.60, $n = 19$) (Gao et al., 1996) and PEM-WEST B (Feb–Mar, 1994) (2.50 ± 1.00, $n = 34$) (Prospero et al., 1995). The correlation coefficients between nss-SO_4^{2-} and NO_3^- were $\gamma = 0.68$, slope = 1.85 and $n = 195$ for PM_{10}, $\gamma = 0.74$, slope = 1.36 and $n = 119$ for TSP. The other East Asian sites (Cheju East, Cheju West, Kato, Kenting and Okinawa), the correlation coefficients and slope ranged from 0.89 to 0.51 and from 4.8 to 1.7. This similar relationship might be explained by some similarities in the sources and sinks of sulfur and nitrogen compounds. The similar trends in concentrations of nss-SO_4^{2-} and NO_3^- at Cape D'Aguilar site were also probably caused by the combined effects of sources, meteorology and transport.

Table 3
Annual average concentrations of nss-SO_4^{2-}, nss-K^+, NO^{3-} and trace elements

	nss-SO_4^{2-} ($\mu g\,m^{-3}$)		nss-K^+ ($\mu g\,m^{-3}$)		NO_3^- ($\mu g\,m^{-3}$)		As ($ng\,m^{-3}$)		Sb ($ng\,m^{-3}$)		Se ($ng\,m^{-3}$)		V ($ng\,m^{-3}$)		Zn ($ng\,m^{-3}$)		Pb ($ng\,m^{-3}$)	
	TSP	PM_{10}	TSP	PM_{10}	TSP	PM_{10}	TSP	PM_{10}	TSP	PM_{10}	TSP	PM_{10}	TSP	PM_{10}	TSP	PM_{10}	TSP	PM_{10}
Average	8.04	7.37	0.37	0.41	4.21	2.70	3.11	3.49	1.60	1.66	1.18	1.40	7.04	6.33	70.08	73.00	40.73	49.22
Median	6.70	6.92	0.22	0.24	3.17	2.29	1.90	2.40	0.91	1.30	0.82	1.20	4.95	3.90	46.50	58.00	25.45	35.33
Geomean	5.77	5.57	0.20	0.22	3.06	2.18	1.81	1.98	0.67	0.84	0.80	1.02	5.37	4.50	32.96	37.90	16.68	22.18
Stdev	5.87	4.88	0.38	0.42	3.22	1.79	3.05	3.44	2.18	1.69	1.00	1.01	6.47	10.36	74.91	65.64	43.70	49.08
Max	27.78	26.01	2.26	1.97	15.70	8.82	13.00	24.00	17.00	9.50	4.80	6.50	48.00	131.0	390.0	340.0	226.30	255.58
Min	0.67	0.48	0.011	0.003	0.20	0.31	0.15	0.10	0.032	0.023	0.012	0.0083	0.80	0.60	1.00	0.69	0.273	0.376
n	119	195	119	193	120	195	90	165	113	189	113	181	112	189	111	189	120	189

The differences in slopes suggested that there were regional differences in emissions of these compounds and possibly in their sink. (Arimoto, 1996). The correlation among the nss-SO_4^{2-} and Sb, Se, As, Pb, Zn were $\gamma = 0.50, 0.77, 0.68, 0.81$ and 0.69 for TSP and $\gamma = 0.62, 0.68, 0.66, 0.68$ and 0.62 for PM_{10}. In the case of Sb, Se both arithmetic average ratios of nss-SO_4^{2-} to Sb and Se (12470 ± 12724, $n = 113$, 10660 ± 29677, $n = 113$ for TSP, 9780 ± 10920, $n = 189$, 8488 ± 25759, $n = 181$ for PM_{10}) are higher than those at Xiamen (5200 ± 2900, $n = 19$, 2800 ± 1000, $n = 19$) and Kato (5380 ± 6100, $-$). It is not strange that the ratios of nss-SO_4^2 to Sb and to Se in the present site are higher than at Xiamen, Kato. After all, nss-SO_4^{2-} is a secondary aerosol component which is constantly formed from its precursor (SO_2) during the long-range transport, whereas Sb and most of the Se are primary aerosol constituents. What seems more strange is that the ratios of nss-SO_4^{2-} to V and to Zn (1645 ± 1383, $n = 112$, 283 ± 406, $n = 111$ for TSP, 1819 ± 1473, $n = 189$, 263 ± 510, $n = 189$ for PM_{10}) are close to those obtained at Xiamen (2700 ± 1400, 200 ± 110, $n = 19$). This could point to local (regional) sources of V and Zn at Hong Kong. Thus, the present station might be influenced by the combined effects of sources, meteorology and pollutants transported from other regions, i.e. multiple sources may have an impact on the Cape D'Aguilar station.

3.6. *Ammonium*

Ammonium (NH_4^+–N) may be formed through the gas-phase reactions involving vapor gas NH_3 from agricultural activities with NH_4^+–N as an end product. The sub-micron of NH_4^+–N and its shifting to larger sizes have been observed at Hong Kong station during PEM-WEST B measurement (Prospero et al., 1995). The $NH4^+$–N is correlated with nss-SO_4^{2-} ($\gamma = 0.70$ for TSP, 0.85 for PM_{10}). As shown in Table 1a the ammonium concentrations in PM_{10} were higher than those in TSP, this may be due to collecting particles in a coastal environment, sampling artifacts and evaporation of volatile ammonium ion to the gas phase. The ammonium concentrations in TSP might be comparable to or less than those in PM_{10} particles (Wollf et al., 1986; Kim, 1997). When the southwesterly flow came across to the station (Jun, Jul) the lowest NH_4^+–N concentrations of TSP and PM_{10} were 0.25 ± 0.11, $n = 12$, 0.32 ± 0.13, $n = 17$ $\mu g\,m^{-3}$. The ammonium concentrations in PM_{10} were still higher than those in TSP. Ammonium particles in the atmosphere also consist of ammonium nitrate which is a product of gas-phase reactions between fine ammonia and fine nitric acid below the cloud to form the 0.1–0.3 μm (aerodynamic diameter) particles and reaction of nitric acid with pre-existing fine particles to form the 0.5–0.7 μm ammonium nitrate (Wall et al., 1988). This formation process and its size distributions may be used to explain the correlations between ammonium and nitrate.

3.7. Mineral dusts

The dust particles originating from arid and semiarid regions of China prove to be a major component in the atmosphere over the mid-latitudes of the Northern Hemisphere. (Prospero et al., 1989). Aluminum (Al) is usually selected as a tracer material of soil. The concentration of Al in a surface soil sample collected from the central area of station was measured by INAA and its value was, 7.26%, close to average crustal materials 8% (Taylor, 1964; Mason, 1982). The mineral dust aerosols concentration was estimated to be [Al]/7.26%. The annual dust concentrations were 9.03 $\mu g\,m^{-3}$ (stdev = 12.27, std.err = 1.15, $n = 113$) and 7.38 $\mu g\,m^{-3}$ (stdev = 8.43, std.err = 0.61, $n = 189$) for TSP and PM_{10}. Figs. 5a and b show that the highest peak values in winter were 27.16 (TSP) and 15.70 $\mu g\,m^{-3}$ (PM_{10}), whilst the lowest peak values in summer for TSP and PM_{10} were, respectively, 1.12 and 0.86 $\mu g\,m^{-3}$. The seasonal cycle of dust aerosols is similar to that of major ions. It is further proved that the station is greatly

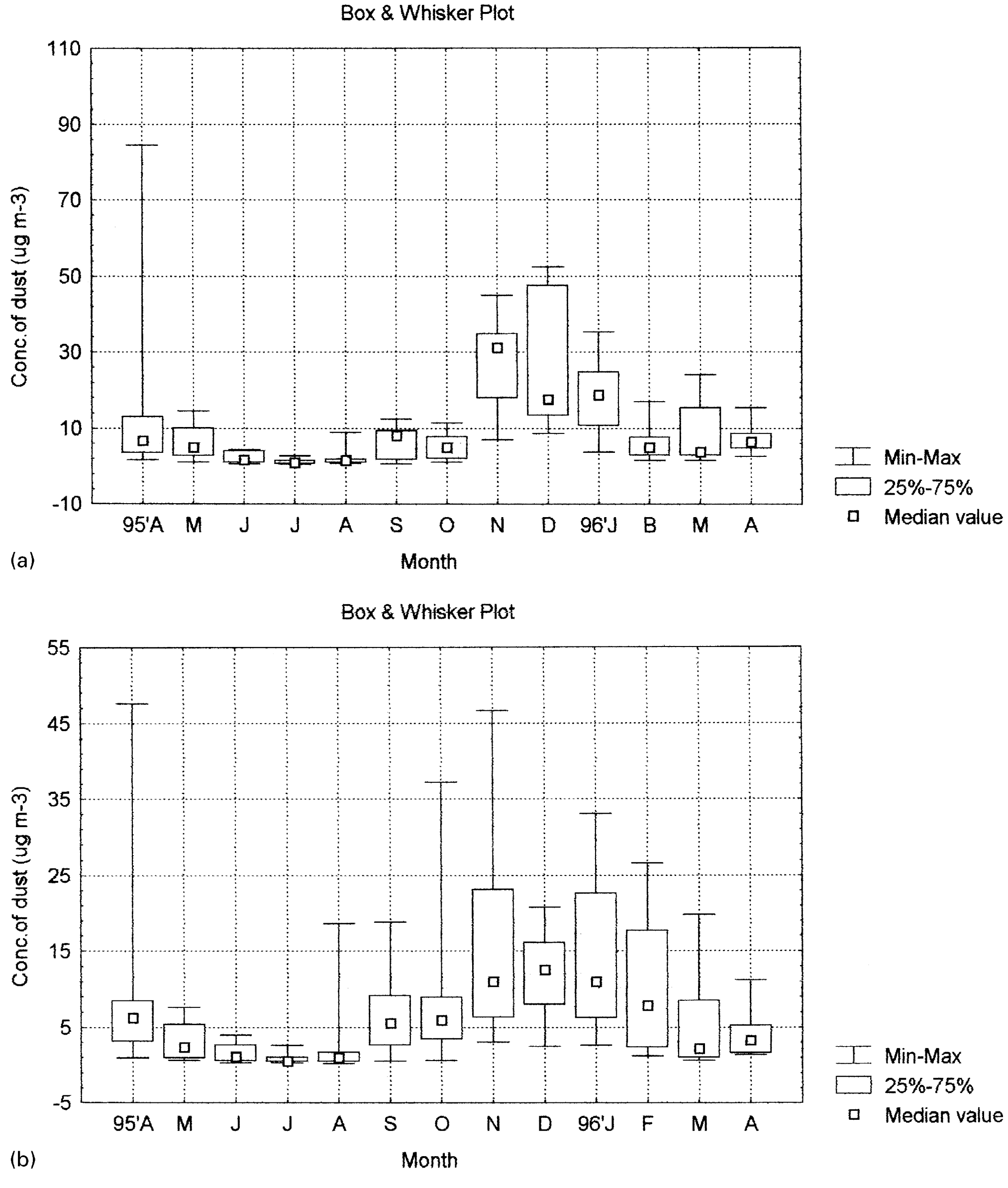

Fig. 5. (a) Monthly median concentrations of dust for TSP. (b) Monthly median concentrations of dust for PM_{10}.

influenced by the meteorological conditions and other pollution emissions. Recently, Zhang et al. (1997) suggested the use of ratios Fe/Al, Mg/Al and Sc/Al as an elemental tracer to evaluate the source distributions from Asian dust. The ratios values (geometric mean) which represented three types of dust regions, ranged from 0.83 to 0.44 (Fe/Al), 0.31 to 0.19 (Mg/Al) and 0.27 to 0.34×10^{-3} (Sc/Al). Our measurement was the geometric mean ratio values of Fe/Al, Sc/Al were 0.71, 0.18×10^{-3} for TSP ($n = 113$), 0.72, 0.22×10^{-3} for PM_{10} ($n = 189$) which was close to tracer values Fe/Al 0.83–0.44 and Sc/Al 0.27×10^{-3}–0.34×10^{-3}. However, the ratio values of Mg/Al were 2.20 ($n = 99$), 1.25 ($n = 186$) for TSP and PM_{10}. Mg concentration was calculated by total Mg – soluble Mg^{2+}, are greater than 0.31–0.19 of Zhang et al. (1997). There is no evidence, including tracer values of Fe/Al, Mg/Al and Sc/Al, to demonstrate the long-range transport of dust originated from N.W. China to the present station. Hong Kong is much farther from the dust source regions than the Xi' an sampling site located 40 Km northwest to Xi' an city, China. The average dust concentrations there, are lower roughly by a factor of 10. Therefore, it is more likely that the dust particles at Hong Kong were fly ash or locally generated mineral matters as opposed to dust transported long distances (Arimoto et al., 1997).

3.8. Factor analysis

In Hong Kong the possible sources were identified by Fung and Wong (1995). They briefly stated as

Sources	Elements
Incineration and automobile	Zn, Pb, Cd.
Coal-fired power plant	Se, As.
Soil-related sources	Ba, Fe, Mn, Ca, Sr.
Oil combustion	S, V.

Electricity generator is also a major source for SO_2 and NO_x emissions (Wu and Chan, 1997) and Gao et al. (1996) suggested that pyrometallurgical nonferrous metal production is the dominant source over the South China Sea.

Factor analysis (SPSS for window 6.0 release SPSS Inc, 1993) is used to identify the possible source for major ions, halogens, crustal elements and nonsea-salt sulfates. Four factors (after varimax rotation) are shown in Tables 4 and 5. Table 4 presents TSP factor loading. The first factor (Pc 1), eigenvalues greater than 1 (i.e. 12.04) explaining 60.2% of the variance, has high loadings for Cl^-, Na^+, Mg^{2+}, Br, K^+ and Ca^{2+} indicating a marine source. The second factor (Pc 2) includes As, Sb, Se, Pb and Zn and eigenvalues 3.20, accounting for 16.0% of the variance, mainly represent the fossil combustion, vehicle, municipal incineration and nonferrous metal production emissions. Because there are two large thermal power stations in Hong Kong, both consuming low sulfur coal (< 1% sulfur), they are located at the west of the station (~ 30 Km). Hong Kong had more than 462,000 licensed vehicles at end of 1994, petrol was used for these vehicles, about 75% of which was unleaded fuel (Environmental Protection Department of Hong Kong, 1995). A waste incinerator is also located at the northwest, the waste burning being a source of nss-K^+ and K^+. All of these emissions clearly have had an impact on the station. The third factor (Pc 3) and eigenvalues 1.37 explaining 5.7% of the variance appear to represent the by-products of oxidation of SO_2 and NO_x which are from fossil fuel combustion including possible biomass burning. The loadings of NH_4^+–N, NO_3^-, nss-SO_4^{2-}, SO_4^{2-}, I and Se are moderately high. As mentioned above, most of Se is from fossil combustion and ocean emission. The correlation between Se and I (0.59, 0.69 for TSP and PM_{10}) suggests that the source of Se is an important source of I. The fourth factor (Pc 4) and eigenvalues 0.80, accounting 4.0% of the variance, represent the crustal elements Fe, Al and Sc. These elements are obviously from crustal or solid component. Table 5 presents the results of factor analysis for PM_{10}. Because the particle sizes of PM_{10} are smaller than those of TSP, the enrichment factor of anthropogenic elements in PM_{10} such as

Table 4
Factor loadings of concentrations of the different elements of TSP aerosol (Varimax rotated factor pattern)

Factor isolated	Pc 1	Pc 2	Pc 3	Pc 4	Communality
Cl^-	**0.98**				0.97
Na^+	**0.96**	0.16		0.11	0.96
Mg^{2+}	**0.90**	0.17			0.85
Br	**0.85**	0.29		0.22	0.86
Ca^{2+}	**0.70**	0.37	0.22	0.43	0.87
K^+	**0.70**	0.56	0.33	0.24	0.97
Sb	0.15	**0.82**	0.15	0.13	0.74
Zn	0.24	**0.79**	0.34	0.28	0.88
As	0.27	**0.78**	0.26	0.36	0.88
Pb	0.30	**0.73**	0.51	0.27	0.95
nss-K^+	0.35	**0.73**	0.44	0.28	0.92
NH_4^+-N	− 0.10	0.11	**0.89**		0.82
NO_3^-		0.17	**0.79**	0.32	0.76
nss-SO_4^{2-}	0.19	0.46	**0.79**	0.12	0.88
SO_4^{2-}	0.44	0.45	**0.71**	0.16	0.93
I	0.11	0.28	**0.65**	0.46	0.72
Se	0.13	0.61	**0.63**		0.78
Al	0.20	0.29	0.27	**0.79**	0.81
Sc	0.47	0.52	0.22	**0.59**	0.89
Fe	0.39	0.53	0.21	**0.53**	0.75
Eigenvalue	12.04	3.19	1.14	0.80	
Pct of Var	60.2	16.0	5.7	4.0	

Note: Total matrix sampling adequacy 0.858.

Table 5
Factor loadings of concentrations of the different elements of PM_{10} aerosol (Varimax rotated factor pattern)

Factor isolated	Pc 1	Pc 2	Pc 3	Pc 4	Communality
Pb	**0.82**	0.27	0.32	0.29	0.93
As	**0.82**	0.19	0.28	0.31	0.88
nss-K^+	**0.81**	0.27	0.25	0.38	0.90
Zn	**0.76**	0.23	0.33	0.31	0.83
K^+	**0.74**	0.45	0.24	0.37	0.95
Se	**0.71**	0.25	0.34	0.28	0.76
Sb	**0.69**	0.14	0.36	0.14	0.65
Na^+	0.20	**0.95**		0.20	0.98
Cl^-	0.15	**0.94**		0.24	0.96
Mg^{2+}	0.21	**0.92**	0.12	0.28	0.98
Br	0.43	**0.76**		0.16	0.79
NH_4^+-N	0.35	− 0.18	**0.86**		0.89
NO_3^-	0.10	0.13	**0.84**	0.27	0.80
nss-SO_4^{2-}	0.51		**0.80**		0.90
SO_4^{2-}	0.51	0.22	**0.77**		0.91
I	0.49	0.15	**0.52**	0.27	0.61
Fe	0.27	0.32	0.25	**0.82**	0.90
Ca^{2+}	0.22	0.45	0.15	**0.81**	0.92
Al	0.38	0.33	0.18	**0.80**	0.94
Sc	0.29			**0.67**	0.54
Eigenvalue	11.57	3.13	1.37	0.97	
Pat of var	57.9	15.7	6.8	4.9	

Note: Total matrix sampling adequacy 0.853.

As, Sb, Se and Pb are higher than those of TSP. Thus the first factor (Pc 1) and eigenvalues 11.57 explaining 57.9% of the variance, have high loadings for Pb, As, nss-K^+, Zn, K^+Se, and Sb indicating to be likely from fossil combustion and anthropogenic emission. The second factor (Pc 2) and eigenvalues 3.13 accounting 15.7% of the variance, Cl^-, Na^+, Mg^{2+} and Br can be attributed to sea-salt component. The third factor (Pc 3) and eigenvalues 1.37 accounting 6.8% of the variance, included NH_4^+–N, NO_3^-, nss-SO_4^{2-}, SO_4^{2-} and I, which are indicative of fossil fuel combustion, vehicle and possible biomass emissions. At coastal station in Hong Kong the concentration of Al in PM_{10} is lower than that in TSP. Al is predominantly in the super-micron fraction and the mass median diameter (MMD) of Al is about 7 μm (Prospero et al., 1995; Arimoto et al., 1997). The fourth factor (Pc 4) and eigenvalues 0.97 accounting 4.9% of the variance, Fe, Ca^{2+}, Al and Sc represent mineral dust composition. The factor analyses show that at coastal station in Hong Kong, sea salt has the greatest loading for TSP while anthropogenic species are high loading for PM_{10}.

Due to the complication of emission patterns, it may not be easy to identify quantitatively some sources at in the coastal area of Hong Kong, including Xiamen, which is located in the coast of the South China Sea. Furthermore, the present station might be influenced by the combined effects of sources, meteorology and pollutant transport from other regions, especially under meteorological conditions that favor mixing of air masses (Gao et al., 1996). In addition, the incomplete regional inventories and lack of long-term aerosol data set also make a quantitative source apportionment for I and nss-SO_4^{2-} difficult at present.

4. Conclusion

The analysis of aerosol samples collected from a coastal station in Hong Kong during the period of April 1995 to April 1996 are presented. The aim of this study is to characterize the chemical composition of major ions, halogen elements and dusts for TSP, PM_{10}. This study also assesses the pollution emissions impact on ground level aerosol in a coastal area of Hong Kong. The Asian monsoon system brings clean air masses during summer time (Jun, Jul and Aug) and dust laden air masses in winter (Dec, Jan and Feb), so the seasonal variation in major ion concentrations obviously shows a summer minimum and a winter maximum. Chlorine "loss" occurs at the station. It is similar to the last collection period (1993–1994) and resembles other Asian stations. The halogen aerosols (Br, I) have a similar seasonal variation. The sources of Br are mainly marine while most of I may have originated from different sources such as natural and anthropogenic sources, including possible biomass emission. The nss-SO_4^{2-} concentrations and its various ratios to trace elements indicate that pollution emissions dominate the Asian region, including Hong Kong.

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