



Atmospheric Environment 35 (2001) 5007-5018



www.elsevier.com/locate/atmosenv

Aerosol characters from the desert region of Northwest China and the Yellow Sea in spring and summer: observations at Minqin, Qingdao, and Qianliyan in 1995–1996

J. Zhang^{a,b,*}, Y. Wu^b, C.L. Liu^c, Z.B. Shen^d, Z.G. Yu^a, Y. Zhang^e

^a Department of Marine Chemistry, Ocean University of Qingdao, 5 Yushan Road, Qingdao 266003, People's Republic of China ^b State Key Laboratory of Estuarine and Coastal Research, East China Normal University, 3663 Zhongshan Road North,

Shanghai 200062, People's Republic of China

^c Institute of Marine Geology, Ministry of Geology and Mineral Resources, 40 Fuzhou Road, Qingdao 266071, People's Republic of China ^d Lanzhou Institute of Plateau Atmospheric Physics, Chinese Academy of Sciences, Donggangxilu 196, Lanzhou 730000, People's Republic of China

^eIMS, Canadian Centre for Remote Sensing, 588 Booth Street, Ottawa, Canada K1A OY7

Received 1 September 2000; received in revised form 10 April 2001; accepted 11 May 2001

Abstract

Aerosol samples were collected from Northwest China desert region (Minqin), coastal suburb (Qingdao) and interior of the Yellow Sea (Qianliyan) in spring and summer of 1995 and 1996. Samples were analysed for major components, carbon and sulphur. The results show that concentrations of aerosols change considerably in time and space. The crustal materials carried by cold front system increase notably the aerosol concentration (mass/unit vol.) over the Yellow Sea but reduce the percentage contribution of pollutants and sea-salt. The sea-salt and regional aerosols become dominant fractions in coastal atmosphere in summer when the dust storms are expired in source region and the Southeast monsoon starts in the Pacific Ocean. © 2001 Elsevier Science Ltd. All rights reserved.

Keywords: Aerosols; Chemical composition; NW China; Yellow Sea

1. Introduction

The impact of soil dust transported from East Asia over the North Pacific Ocean has well been recognised and documented in literature over last several decades. This includes a change in radiation forcing caused by aerosols, contribution to deep-sea sedimentation at midlatitude, and linkage between aeolian trace species and biological productivity at oligotrophic waters (Duce et al., 1980; Uematsu et al., 1983; Blank et al., 1985; Martin and Gordon, 1988; Chung, 1992; Zhang and Huang, 1992; Leinen et al., 1994). However, the information from coastal oceans (e.g. China Sea), where continental aerosols start to react substantially with the marine salts in atmosphere, is still limited (Uematsu et al., 1985; Zhang et al., 1992; Parungo et al., 1994; Gao et al., 1997; Hong et al., 1998).

The desert/Gobi region in Northwest of China is located at high elevation and covers an area of $1.3 \times 10^6 \text{ km}^2$, which is three-fold larger than the Loess Plateau ($0.4 \times 10^6 \text{ km}^2$) immediately at down-wind side (Fig. 1). The hypothesis is that desert/Gobi represents an important source of mineral aerosols over the East Asia and North Pacific Ocean, which was under-estimated in previous studies (cf. Zhang et al., 1993b; Chen et al., 1997).

An extensive sampling and analysis of aerosols from both source region and down-wind marine recipient is

^{*}Corresponding author. Present address: Department of Marine Chemistry, Ocean University of Qingdao, 5 Yushan Road, Qingdao 266003, People's Republic of China. Tel.: +86-532-2032005; fax: +86-532-2963005.

E-mail address: jzhang@sklec.ecnu.edu.cn (J. Zhang).



Fig. 1. Map of study area, it shows the distribution of desert region and loess covered area in Northwest China. The field observation sites of this study include Minqin, Qingdao, and Qianliyan. Stations where the dust storms were recorded in springs of 1995 and 1996 are also shown.

required to understand the atmospheric composition and to estimate deposition flux in North Pacific rim (cf. Uematsu et al., 1985; Zhang et al., 1993a; Xu et al., 1996; Gao et al., 1997; Hong et al., 1998; Zhang and Iwasaka, 1999). So far, very limited observations were carried out simultaneously at desert and coastal recipient in NW Pacific Ocean, that represent a reasonable time scale to compare the chemical composition of aerosols.

In this study, we collected aerosol samples from Minqin, Qingdao and Qianliyan in 1995 and 1996 (Table 1 and Fig. 1). The study is aimed to verify how and to what extent the coastal aerosol composition responds to dust storms of East Asia. Data are examined to reveal the link in composition between dust storms in Northwest China and episodic variation of aerosols over the Yellow Sea.

2. Sample collection and analysis

The geographic location of field observation sites and information of aerosol sample collection are shown in Table 1 and Fig. 1. Aerosol samples were collected by low volume pumps on pre-cleaned Nuclepore filter (pore-size: $0.4 \,\mu$ m) for inorganic species and on Whatman GF/F filter (pore-size: $0.7 \,\mu$ m) for carbon and sulphur. The sample holders were attached to the end of clean bamboo poles (ca. 4-5m), and the pumps were installed 10m down-wind side and/or isolated from the samplers. The sampling frequency was managed on daily base, except for the case of pump broken and checks. The flow meter was calibrated before use and checked again after sampling campaign. The blanks were prepared by exposing filters for 24h without pumping. Nuclepore filters were pre-cleaned with HCl (1:5 v/v), followed by thoroughly rinse with Milli-Q water, and dried at 50°C. Whatman GF/F filters were pre-cleaned by heating at 450°C over night. The sample change was undertaken in a clean plastic tent, with all protection and precaution (e.g. lab-coat and plastic gloves) necessary to avoid contamination.

In the laboratory, the Nuclepore filters were dried and weighed to estimate the concentration of aerosols (mass/ unit vol. of air). The samples were then digested by HNO₃–HF–HCIO₄, and solutions were analysed by inductively coupled plasma atomic emission spectrometry (ICP-AES) for Al, Ca, Na, Fe, Mn, Mg, K, V, Sr, and Zn, and by electro-thermal atomic absorption spectrophotometry (ETAAS) for Cd, Cr, Co, Cu, Ni and Pb. The samples collected on Whatman GF/F filters were washed by diluted HCl to remove carbonates and

Table 1

Station	Elevation (m)	Description of sample collection The sample collection was taken at 10 m above ground level in a plant garden of desert region. There is no habitation with a distance of 500–1000 m.			
Minqin	1340				
Qingdao	77	The collection site is down-wind the urban area with 2 million of population. The sampler was set on the top of a building at 500 m from coast.			
Qianliyan	75	The sample collection site is on an island located 50–60 km from the main land with 20–30 habitants. The sampler was in a meteorological observatory.			

Description of field observations and information of aerosol sample collection sites, Minqin, Qingdao, and Qianliyan, in 1995 and 1996

acid-soluble sulphur (e.g. sulphate), followed by rinsing with Milli-Q water to remove extra acids. The residual aerosol carbon (Rac: organic and elemental carbon) and sulphur (Ras: acid insoluble sulphur) were determined by LeCo-355 carbon and sulphur analyser (Zhang et al., 1997). The data quality of inorganic analysis was daily checked by the national (GSS-1, GSD-5, and GSMS-1) and Canadian MESS-1 standards (Liu, 1997). The precision of Rac and Ras determination was examined by the analysis of national standards (cf. Zhang et al., 1997).

Blanks of Nuclepore and Whatman GF/F filters were estimated by analysis of filters, which account generally for 5–10% of sample concentrations and are removed by subtraction from the data sets. The standardisation of method for carbonaceous aerosols is still not established and concentrations measured by different techniques need not always be comparable (cf. Countess, 1990; Hitzenberger et al., 1999).

3. Results

Totally about 180–200 sample sets were collected and analysed. The aerosol compositions from Minqin, Qingdao, and Qianliyan are summarised in Table 2 and Figs. 2–4. Two kinds of data are presented, i.e. concentration on air-volume-basis ($\mu g m^{-3}$) and in percentage of aerosols (%). In this study we present data for Al, Ca, Na, Rac, and Ras, data of other elements will be reported elsewhere.

3.1. Aerosol concentration

At Minqin, in 50% of sampling time the aerosol concentration is $< 100 \,\mu g \, m^{-3}$, and 75% of samples has concentration $< 200 \,\mu g \, m^{-3}$ (Fig. 2a, b). At Qingdao, 35% of samples have concentrations of $< 50 \,\mu g \, m^{-3}$,

and in 85% of sampling period aerosol concentrations are $<100 \,\mu g \, m^{-3}$ in spring 1995 (Fig. 3a). In 65% of sampling period of 1996, aerosol concentration is $<50 \,\mu g \, m^{-3}$, only 7% of samples has concentration $>100 \,\mu g \, m^{-3}$ (Fig. 3b).

In 1995, 90% of samples from Qianliyan have concentrations $<50 \,\mu g \, m^{-3}$ (Fig. 4a). Concentration of $\ge 50 \,\mu g \, m^{-3}$ was observed exclusively in April. In 1996, the aerosol concentrations are lower in average compared to 1995, presumably the sample collection started in the early May, and high aerosol events in spring (e.g. April) were missed (Fig. 4b).

3.2. Aluminium, Ca and Na

At Minqin, the elevated concentration of Al (e.g. $> 10 \,\mu g \,m^{-3}$) corresponds to dust storms in this region (Fig. 2c, d). The percentage concentrations (%) of Al in aerosols were 4.93–9.31%, similar to the composition (Al: 8%) of earth crust (Taylor and McLennan, 1995), and comparable to the composition of loess. The atmospheric Al concentration at coastal stations (Qing-dao and Qianliyan) is two orders of magnitudes lower, but highly variable as compared to the desert region (Minqin) (Figs. 3c, d and 4c, d).

The percentage concentrations of Ca in aerosols are found 4.55–8.40% in 1995 at Minqin, which are about twice higher than the average composition (i.e. 3.0%) for the upper continental crust (Taylor and McLennan, 1995). Suppose that Ca in aerosols comes from carbonates, the concentration corresponds to an equivalent CaCO₃ of 10–20% in aerosols (Table 2). Air volume based concentrations of Ca at Qingdao and Qianliyan can be higher than the level of upwind desert region (Figs. 3c, d and 4c, d). The corresponding percentage concentrations of 1.03-24.5% for Ca in coastal areas indicate the substantial incorporation of local and/or marine sources. Table 2

Composition of aerosols from Northwest China desert region and Yellow Sea. The data sets show (2a) air-volume-based concentration $(\mu g m^3)$ and (2b) percentage (%) of aerosols

Composition	Concentration	Minqin		Qingdao		Qianliyan	
		1995	1996	1995	1996	1995	1996
(a) Air-volume	-based concentration	$(\mu g m^{-3})$ of major s	pecies				
Aerosol	Range	20-2060	14-378	16-180	6.5-327	4.1-57.0	3.3-35.5
	Geometric mean	90.8	98.6	60.2	33.0	18.4	12.1
	Average	187	131	68.8	49.2	23.0	15.0
Al	Range	1.39-153	0.72 - 28.7	0.82-9.32	0.16-23.1	0.11 - 2.28	0.03-1.87
	Geometric mean	6.35	6.25	2.42	1.27	0.57	0.25
	Average	13.6	8.79	2.93	2.53	0.83	0.45
Ca	Range	1.08-113	0.74-27.8	0.92-9.72	0.23-15.2	0.05-1.79	0.07-1.76
	Geometric mean	5.70	6.21	3.04	1.35	0.55	0.32
	Average	11.4	8.41	3.59	2.25	0.74	0.43
Na	Range	0.28-41.8	0.14-6.39	0.25-4.76	0.05-5.58	0.17-5.41	0.04-1.99
	Geometric mean	1.37	1.24	1.29	0.67	0.89	0.39
	Average	3.22	1.80	1.71	0.92	1.29	0.55
Rac	Range	1.52-23.8	1.41-11.3	3.86-18.4	0.65-30.4	0.29-13.2	0.45-6.56
	Geometric mean	3.12	3.71	8.15	3.71	2.07	1.71
	Average	4.27	4.28	8.88	5.24	3.02	2.13
Ras	Range	0.01-1.52	0.02-1.13	0.12-0.84	0.02 - 1.08	0.01-0.79	0.02-0.30
	Geometric mean	0.12	0.14	0.24	0.12	0.06	0.10
	Average	0.18	0.24	0.27	0.19	0.11	0.12
(b) Percentage	concentration (%) a	of major species in a	erosols				
Al	Range	5.43-9.31	4.93-7.96	1.85-6.36	1.23-7.08	0.96-6.79	0.42-7.06
	Geometric mean	6.99	6.34	4.02	3.84	3.12	2.08
	Average	7.04	6.40	4.23	4.21	3.71	2.55
Ca	Range	4.74-8.39	4.52-8.40	2.33-10.7	2.21-24.5	1.33-15.7	1.03-7.08
	Geometric mean	6.28	6.30	5.06	4.09	3.02	2.61
	Average	6.34	6.36	5.42	4.74	3.47	2.89
Na	Range	1.00-2.39	0.95-1.82	0.53-8.49	0.46-9.62	1.23-15.6	0.38-29.4
	Geometric mean	1.51	1.26	2.14	2.04	4.87	3.24
	Average	1.55	1.28	2.71	2.66	6.03	6.12
Rac	Range	0.73-23.4	0.92-47.6	7.71-70.0	3.89-22.7	4.18-58.8	6.64-42.6
	Geometric mean	3.51	3.77	14.1	11.2	11.3	14.5
	Average	4.67	5.91	16.2	12.0	13.8	16.3
Ras	Range	0.02-0.45	0.01-4.76	0.15-3.50	0.06-5.92	0.08-3.53	0.23-2.22
	Geometric mean	0.12	0.14	0.41	0.37	0.34	0.82
	Average	0.18	0.41	0.56	0.64	0.54	0.97

Sodium is of 0.95-2.39% for aerosols at Minqin (Table 2). These values are lower in average than that (2.3-2.9%) for continental crust (Taylor and McLennan, 1995). In coastal areas (e.g. Qingdao and Qianliyan), the percentage concentration of Na increases up to 30%, with differences by a factor of 20 between high and low Na samples. This indicates the dominance of Na in aerosols (i.e. Qianliyan) by the contribution of marine source, though the air-volume-based Na level can be lower than that of Minqin (Figs. 2c, d and 4c, d).

3.3. Carbon and sulphur

Compared with Al, Ca, and Na, a different distribution was observed for Rac and Ras (Figs. 2e, f, 3e, f and 4e, f). At Minqin, 75% of samples have Rac<5%, a value similar to the composition of soils in China. Exceptional high levels can be occasionally found with Rac up to 47.6%, presumably from burning of biomass in spring. The Ras is also low at Minqin, only 5% of samples have Ras > 0.5% in aerosols (Fig. 2e–f).

The Rac and Ras increase toward the coastal areas in this study (Table 2). At Qingdao, the estimated Rac and Ras could be 3.89-70.0%, Ras 0.06-5.92% (Table 2). At Qianliyan, Rac was found 4.18-58.8% and Ras 0.08-3.53%. Though the percentage Rac and Ras level at Qianliyan are lower than Qingdao in spring, the summer Rac concentration in this region could occasionally be up to 50-60% (Figs. 3e, f and 4e, f).



Fig. 2. Concentration and chemical composition of aerosol samples at Minqin in 1995–1996, which show the air-volume-based concentration ($\mu g m^{-3}$) of aerosols (a–b), Al, Ca and Na (c–d), and Rac and Ras (e–f).

4. Discussion

4.1. Comparison of aerosols between inland desert and coastal areas

The three sample collection sites can be compared with respect to elemental percentage concentrations for aerosols. Aluminium and Ca are higher at Minqin than Qingdao and Qianliyan, they decrease from inland deserts to the interior of Yellow Sea, especially when the data are presented as percentage of element in aerosols (Table 2). This implies a reduced influence of crustal aerosols in marine atmosphere. Air-volume-based concentration for



Fig. 3. Concentration and chemical composition of aerosol samples at Qingdao in 1995–1996, which show the air-volume-based concentration ($\mu g m^{-3}$) of aerosols (a–b), Al, Ca and Na (c–d), and Rac and Ras (e–f).

Al, Ca and Na show a close correlation with aerosol levels at Minqin, with a slight increase in percentage concentrations during dust storms, indicating a character of mineral particles (Fig. 2c, d). Concentrations of Rac and Ras show a considerable reduce, e.g. by a factor of 5–10, in dust storms because of a different aerosol populations involved (Fig. 2e–f).



Fig. 4. Concentration and chemical composition of aerosol samples at Qianliyan in 1995–1996, which show the air-volume-based concentration ($\mu g m^{-3}$) of aerosols (a–b), Al, Ca and Na (c–d), and Rac and Ras (e–f).

Percentage concentrations of Rac and Ras are higher at Qingdao than both Minqin and Qianliyan, underlining the influence of coastal suburb by anthropogenic emission (Table 2). This is different from Al, Ca, and Na, for which higher levels are found at Minqin when the elements are crust dominated (e.g. Al), or at Qianliyan for the sea-salt species (e.g. Na). Using enrichment factors (EF), the non-sea-salt contribution of a given species (X)ns can be estimated by:

$$(X)ns = (X)air - (Na)air \times (X/Na)sea,$$
 (1)

$$(X)ns/(X)air = 1 - 1/EFs.$$
 (2)

Similarly, the non-crustal-material contribution for a given element (X)nc can be estimated by:

$$(X)nc = (X)air - (Al)air (X/Al)crust,$$
(3)

$$(X)nc/(X)air = 1 - 1/EFc,$$
(4)

where (X/Na) and (X/Al) are the concentration ratios between element X and end-member (i.e. Na and Al) index. In this study, we use the aerosol composition from Mingin as crustal, and seawater composition as marine references, respectively. The non-sea-salt contribution (X)ns for Ca are 0.5-1.0 at Qingdao and Qianliyan. The corresponding (X)ns is substantially increased in spring when the dust storms prevail. The decrease in (X)ns in summer follows the higher level of sea-salt (Na) in coastal region. The non-crustal contribution (X)nc for Ca illustrates lower values in spring (<0.1) at Qianliyan when the cold fronts pass through over the Yellow Sea. Higher values of (X)nc of Ca are found in later winter and summer (0.4-0.5), due to the expiry of dust storms, involvement of local/anthropogenic sources (winter) and marine materials (summer) in combination.

Data from Minqin indicate a Na/Al ratio of 0.22 ± 0.06 in 1995 and 0.20 ± 0.03 in 1996, which implies that observation of atmospheric Na at Yellow Sea includes the Na from dust particles in various proportions. Hence the estimate of (X)ns in Eqs. (1) and (2) turns to be:

$$(X)ns^* = (X)air - [(Na)air - (Al)air (Na/Al)crust] \times (X/Na)sea,$$
(5)

$$(X)ns^*/(X)air = 1 - 1/EFs^*,$$
 (6)

where (Na/Al)crust represent the value for crustal materials at Minqin. Again, the (X)ns and (X)ns^{*} for Ca can be compared with respect to the individual events at Qingdao and Qianliyan. While in non-dust storm conditions, the (X)ns^{*} matches (X)ns by 90–95%. In the heavy dust period of spring, the (X)ns^{*} to (X)ns ratio reduces to 0.7, indicating that a substantial amount of crustal Na is involved (data are not shown). The implication includes that an uncertainty of 30% is an optimistic estimate for non-sea-salt contribution over the Yellow Sea.

4.2. Impact of dust storms in deserts on the aerosol composition over the Yellow Sea

In April 1995, two important cold front systems move south-eastwards, dominating over the North China and adjacent coastal ocean (Fig. 5). The first one passed by Mingin in 19-20 April, and resulted in a strong dust storm with aerosol concentration up to $215 \,\mu g \,m^{-3}$ (Fig. 2a). On 22 April 1995, another cold front came from Mongolia with strong northerly wind and reached the Yellow Sea in a few days (i.e. 23-27 April). The combination of these two cold fronts induced elevated aerosol concentrations over the Yellow Sea, i.e. 85- $90 \,\mu g \,m^{-3}$ at Qingdao and $50-55 \,\mu g \,m^{-3}$ at Qianliyan (Figs. 3a and 4a). The percentage of lithogenic elements (e.g. Al and Ca) increases, followed by reduce of Rac and Ras, compared to non-front conditions (Figs. 3c, f and 4c, f). For example, at Qianliyan, concentrations of Al and Ca reach 6.5-7.0% and 4.0-4.5%, respectively, with a significant decrease (ca. 50%) for Rac and Na in aerosols during the pass-through of these cold fronts over the Yellow Sea.

In the 3–10 of May 1996, there were frequent lower pressure systems, which moved eastward due to smooth circulation (Fig. 5). Several cold front systems passed over Minqin and induced a high concentration of aerosols of $300-350 \,\mu g \,m^{-3}$ (Fig. 2b). The first cold front (03 May) goes slowly eastward over East China and to the adjacent coastal ocean, joined later with other front systems (8–11 May). In these cold front events (i.e. 9–11 May), high aerosol level of $30-40 \,\mu g \,m^{-3}$ was found at Qianliyan (Fig. 4b). Concentration of Al reached up to 6.0–7.0%, with a subsequent reduce by two-fold of Na, Rac and Ras in cold fronts (Fig. 4c–f).

To examine further the influence of the dust storms in desert region on the aerosol composition in coastal area (i.e. Yellow Sea), we normalise the percentage Al concentration of aerosol samples from Qianliyan [Al (QLY)] to that of dust storms at Minqin [Al (MQ)] and compare with the data of Na (%) (Fig. 6). Clearly, the Al (QLY)/Al (MQ) ratio increases and approaches 1.0 when the cold fronts pass over through the Yellow Sea, which indicates the nature of crustal materials from desert region. The air-volumebased concentration of Al at Qianliyan is 1-2 orders of magnitudes lower than Minqin, however (Figs. 2c, d and 4c, d). The low Al (QLY)/Al (MQ) in winter (March) indicates a different local source involved, although the air-volume-based levels $(\mu m m^{-3})$ is high in down-wind and coastal areas, as reported previously (cf. Gao et al., 1997). In Fig. 6, the percentage Na level is reduced in spring when cold fronts are active over the Yellow Sea, whereas Na (%) increases due to atmospheric reloading following the expiration of dust storms (e.g. summer). Compared with previous studies in this region, data of percentage



Fig. 5. Cold fronts in Northwest China in spring, which shows two examples of cold front system in April 1995 (upper panel) and May 1996 (lower panel), respectively. Both indicate the influence of dust storms of desert region on the marine atmosphere of Yellow Sea.

concentration reveal fine structures of aerosol dynamics, as the fingerprint of Fig. 6 could not be deduced from bulk air-volume-based concentrations.

As shown in Fig. 6, that a regular change in aerosol composition was found in Yellow Sea, indicating the scavenging and atmospheric reloading of aerosols. For instance, the percentage Na concentration ranges from <1.0 to 20-25% within a typical period of 1 week to 10 days, corresponding to the passage of episodic dust storm events (Fig. 4c, d). The Al to end-member ratio [Al (QLY)/Al (MQ)] at Qianliyan falls to 0.1 when the

dust events expire. While atmospheric reloading of seasalt is sustained by the surface ocean, removal/scavenging mechanisms is not straightforward, either due to precipitation, incorporation of crustal materials and local emission, and reaction with vapour/gas species, or both (Yao et al., 1998).

In this study, aerosol data are examined in combination with weather map. The knowledge obtained is in agreement with results of individual particle analysis and backward isentropic air trajectories (cf. Zhang and Iwasaka, 1999). The backward air trajectory technique



Fig. 6. Ratio of percentage Al levels [Al (QLY)] at Qianliyan to average Al concentration (%) in dust storms from Minqin [Al (MQ)], and percentage Na (%) concentration at Qianliyan. Note that in late winter (March) the Al (QLY)/Al (MQ) ratio is relative low, averaging 0.5. The value of Al (QLY)/Al (MQ) approaches 1.0 in spring when episodic cold fronts pass through over the Yellow Sea. Sea-salt contribution (i.e. Na) to aerosols is considerably reduced by crustal materials in cold front events, and becomes the major component in non-storm periods of summer.

provides information on the passage of event-specific air-mass, the uncertainty of simulation could be a few hundreds of kilometres over a real time of ca. 50 h. The weather map allows to examine the movement of airmass and change in aerosol composition in a threedimensional meso-scale over a broad geographic regime. Both techniques present evidence that dust-storms in desert region of Northwest China and episodic high aerosol events over the Yellow Sea can be closely related/ matched in time and composition (i.e. percentage concentration). In this study, 15 dust-storm events were sampled at Minqin in springs 1995 and 1996, and ca. 10 corresponding high aerosol events observed at Yellow Sea, with a delay of 3–4 days in average. The recorded dust storms at desert region (e.g. Minqin) at upwind side and higher altitude are much more serious than downwind loess covered area in both frequency and aerosol concentration (data are not shown).

4.3. Comparison with previous studies from the Yellow Sea

There are very limited data available in literature from Northwest Pacific Rim that can be directly compared

with this study. Gao et al. (1992, 1997) reported Al and Na at two urban/suburb stations around the Yellow Sea, with 0.6–6.7 μ g m⁻³ for Al and 0.5–11 μ g m⁻³ for Na at Mallipo and $2.5 \pm 2.2 \,\mu g \,m^{-3}$ for Al at Qingdao. These values are comparable to our observations at Qingdao, but higher than Qianliyan. Carmichael et al. (1996) and Kim et al. (1998) reported water-soluble concentration for Na $(1-8 \mu g m^{-3})$ and Ca $(0.1-3.0 \mu g m^{-3})$ from aerosols at Cheju Island. These concentrations are higher in average compared to Qianliyan, but lower for Ca when compared to Qingdao. Part of the aerosol Ca and Na is not soluble and hence not covered by the previous studies (Carmichael et al., 1996; Kim et al., 1998). None of above studies were based on the simultaneous and extensive sample collection and analysis, however, that covers a wide geographic dimension to allow source and recipient comparison and/or a considerable time scale to conduct seasonal and inter-annual examination. Moreover, elemental and organic carbon (EC+OC) reported by Kim et al. (1998) are $0.27 \pm 0.05 \,\mu g \,m^{-3}$ and $3.74 \pm 0.39 \,\mu g \,m^{-3}$, respectively, which together are lower than those from Qingdao $(6.98 \pm 4.38 \,\mu\text{g m}^{-3})$ but similar to Qianliyan $(4.54 \pm 2.17 \,\mu g \,m^{-3})$ in summers of 1995 and 1996. The elevated Rac values from Qingdao is due to its downwind approximation to strong emissions in North China. Total Rac is similar between Cheju and Qianliyan, because a large amount of Rac is found to exist in the iso-kinetic size $> 2.5 \,\mu\text{m}$ (Parungo et al., 1994). Kim and co-workers (1999) reported the $PM_{2.5}$ and PM₁₀ of EC+OC of 17.5–19.5 μ g m⁻³ at Seoul and $4.0 \,\mu g \,m^{-3}$ for Cheju Island. These data in combination with our study reveals that chemical composition of aerosols is readily comparable between west (Qianliyan) and east (Cheju) part of Yellow Sea. The EC+OC from coastal urban and suburb region (Seoul and Qingdao) can be one order of magnitude higher than the remote region of Yellow Sea.

All previous measurements as mentioned above were unable to provide information relevant to the percentage concentration (%) of aerosols, hence the significance that crustal and marine sources contribute differently the aerosol composition in the Yellow Sea (cf. Fig. 6).

5. Summary and problems remained

The data sets of this study indicate that percentage concentration of major inorganic components (e.g. Al, Ca, and Na) is considerably stable at desert/Gobi region, though the aerosol levels vary by five- to tenfold between dust storms and non-dust conditions. Aerosol samples from the Yellow Sea show much lower level of aerosols compared to the upwind desert area, presumably due to the remote location and an increased contribution from sea-salt and/or regional emission. It is found that coastal ocean (e.g. Yellow Sea) atmosphere responds to the spring episodic dust-storms in Northwest China by a dramatic increase in aerosol levels and percentage concentration of crust-dominated elements. Such crustal aerosols may considerably reduce the proportion of sea-salt and regional source. The regional material and sea-salt aerosols become the dominant components in summer, when the dust storms in deserts/ Gobi are expired and southeast monsoon prevails in the NW Pacific Ocean.

Acknowledgements

This study was funded by the Natural Science Foundation of China (No. 49525609). The analysis of Rac and Ras was undertaken at the State Key Laboratory of Gas Geochemistry (Lanzhou), Chinese Academy of Sciences under the contract of No. 92-04. We are very grateful to Prof. P. Brimblecombe, Dr. J. Austin and two anonymous reviewers for their comments and suggestions, which greatly improved the original manuscript.

References

- Blank, M., Leinen, M., Prospero, J.M., 1985. Major Asian aeolian inputs indicated by the mineralogy of aerosols and sediments in the western North Pacific. Nature 314, 84–86.
- Carmichael, G., Zhang, Y., Chen, L.L., Hong, M.S., Ueda, H., 1996. Seasonal variation of aerosol composition at Cheju Island, Korea. Atmospheric Environment 30, 2407–2416.
- Countess, R.J., 1990. Interlaboratory analyses of carbonaceous aerosol samples. Aerosol Science and Technology 12, 114–121.
- Chen, L.L., Carmichael, G.R., Hong, M.S., Ueda, H., Shim, S., Song, C.H., Kim, Y.P., Arimoto, R., Prospero, J., Savoie, D., Murano, K., Park, J.K., Lee, H.G., Kang, C., 1997. Influence of continental outflow events on the aerosol composition at Cheju Island, South Korea. Journal of Geophysical Research 102, 28551–28574.
- Chung, Y.S., 1992. On the observations of yellow sand (dust storms) in Korea. Atmospheric Environment 26A, 2743– 2749.
- Duce, R.A., Unni, C.K., Ray, B.J., Prospero, J.M., Merrill, J.T., 1980. Long-range atmospheric transport of soil dust from Asia to the tropical North Pacific: temporal variability. Science 209, 1522–1524.
- Gao, Y., Arimoto, R., Duce, R.A., Lee, D.S., Zhou, M.Y., 1992. Input of atmospheric trace elements and mineral matter to the Yellow Sea during the spring of a low-dust year. Journal of Geophysical Research 97, 3722–3767.
- Gao, Y., Arimoto, R., Duce, R.A., Zhang, X.Y., Zhang, G.Y., An, Z.S., Chen, L.Q., Zhou, M.Y., Gu, D.Y., 1997. Temporal and spatial distributions of dust and its deposition to the China Sea. Tellus 49B, 172–189.

- Hitzenberger, R., Jennings, S.G., Larson, S.M., Dillner, A., Cachier, H., Galambos, Z., Rouc, A., Spain, T.G., 1999. Intercomparison of measurement methods for black carbon aerosols. Atmospheric Environment 33, 2823–2833.
- Hong, G.H., Kim, S.H., Yang, D.B., Lim, G.H., 1998. Atmospheric input of trace metals over the Yellow Sea: shipboard results, April 1995. In: Hong, G.H., Zhang, J., Park, B.-K. (Eds.), Health of the Yellow Sea. The Earth Love Publication Association, Seoul, pp. 211–235.
- Kim, Y.P., Lee, J.H., Baik, N.J., Kim, J.Y., Shim, S.G., Kang, C.H., 1998. Summertime characteristics of aerosol composition at Cheju Island, Korea. Atmospheric Environment 32, 3905–3915.
- Kim, Y.P., Moon, K.C., Lee, J.H., Baik, N.J., 1999. Concentrations of carbonaceous species in particles at Seoul and Cheju in Korea. Atmospheric Environment 33, 2751– 2758.
- Leinen, M., Prospero, J.M., Arnold, E., Blank, M., 1994. Mineralogy of aeolian dust reaching the North Pacific Ocean: 1 sampling and analysis. Journal of Geophysical Research 99, 21017–21023.
- Liu, C.L., 1997. Transport of particulate heavy metals to the China Sea via river and atmosphere. M.Sc. Dissertation, Ocean University of Qingdao, Qingdao, 105pp. (in Chinese).
- Martin, J.H., Gordon, R.M., 1988. Northeast Pacific iron distribution in relation to phytoplankton productivity. Deep-Sea Research 35, 177–196.
- Parungo, F., Nagamoto, C., Zhou, M.Y., Hansen, A.D.A., Harris, J., 1994. Aeolian transport of aerosol black carbon from China to the ocean. Atmospheric Environment 28, 3251–3260.
- Taylor, S.T., McLennan, S.M., 1995. The geochemical evolution of the continental crust. Review of Geophysics 33, 241– 265.
- Uematsu, M., Duce, R.A., Prospero, J.M., Chen, L.Q., Merrill, J.T., McDonald, R.L., 1983. Transport of dust from Asia

over the North Pacific Ocean. Journal of Geophysical Research 88, 5343–5352.

- Uematsu, M., Duce, R.A., Prospero, J.M., 1985. Deposition of atmospheric mineral particles in the North Pacific Ocean. Journal of Atmospheric Chemistry 3, 123–138.
- Xu, X.H., Yao, L.K., Li, J.L., 1996. Acid and base characteristics of ambient aerosols in Qingdao area. Research of Environmental Sciences 9 (5), 5–8 (in Chinese).
- Yao, X.H., Hao, J.M., Wang, W., Cui, P., 1998. Chloride depletion in coastal aerosol at Xiamen. China Environmental Science 18 (1), 34–38 (in Chinese).
- Zhang, J., Huang, W.W., 1992. Potential chemical link between continental wind deposits, marine aerosol and pelagic sediments—An example of Chinese loess. Deep-Sea Research 39, 1809–1816.
- Zhang, J., Huang, W.W., Liu, S.M., Liu, M.G., Yu, Q., Wang, J.H., 1992. Transport of particulate heavy metals towards China Sea: a preliminary study and comparison. Marine Chemistry 40, 161–176.
- Zhang, D.Z., Iwasaka, Y., 1999. Nitrate and sulfate in individual Asian dust-storm particles in Beijing, China in spring of 1995 and 1996. Atmospheric Environment 33, 3213–3223.
- Zhang, J., Liu, S.M., Lu, X., Huang, W.W., 1993a. Characterising Asian wind-dust transport to the Northwest Pacific Ocean: direct measurements of dust flux for two years. Tellus 45B, 335–345.
- Zhang, X.Y., Arimoto, R., An, Z.S., Chen, T., Zhang, G.Y., Zhu, G.H., Wang, X.F., 1993b. Atmospheric trace elements over source regions for Chinese dust: concentrations, sources and atmospheric deposition on the Loess Plateau. Atmospheric Environment 27, 2051–2067.
- Zhang, J., Yu, Z.G., Liu, S.M., Xu, H., Wen, Q.B., Shao, B., Chen, J.F., 1997. Dominance of terrigenous particulate organic carbon in the high-turbidity Shuangtaizihe Estuary. Chemical Geology 138, 211–219.