

## Characterization of airborne carbonate over a site near Asian dust source regions during spring 2002 and its climatic and environmental significance

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[1] During spring 2002, three dust storm events were monitored by filter sampling in Xi'an near an Asian dust source region of northwest China. The carbonate ( $\text{CO}_3^{2-}$ ) fraction was determined by sample acidification and thermal evolution. The  $\text{CO}_3^{2-}$  accounted for  $8.0 \pm 0.8\%$  of particles with aerodynamic diameter  $\leq 2.5 \mu\text{m}$  ( $\text{PM}_{2.5}$ ) during dust storms and  $4.7 \pm 3.0\%$  of  $\text{PM}_{2.5}$  between storms. The ratios of calcium to carbonate carbon were consistent with those of calcite ( $\text{CaCO}_3$ ). The  $\delta^{13}\text{C}$  and  $\delta^{18}\text{O}$  abundances in dust storm samples were  $-2.7 \pm 0.7\text{‰}$  and  $-5.8 \pm 1.5\text{‰}$ , which differed from  $-8.3 \pm 1.9\text{‰}$  for  $\delta^{13}\text{C}$  and  $-10.8 \pm 2.0\text{‰}$  for  $\delta^{18}\text{O}$  during normal conditions. The  $\delta^{13}\text{C}$  is positively correlated with  $\delta^{18}\text{O}$  values ( $r = 0.78$ ). This first measurement of isotopic abundance in Asian dust indicates the potential to quantify its contribution at distant locations using receptor models. By increasing the alkalinity of ocean water in the Pacific Ocean and buffering the atmospheric acidity of east Asia, the large amounts of airborne  $\text{CO}_3^{2-}$  (as high as  $44.8 \text{ Tg yr}^{-1}$ ) entrained by Asian dust may provide an important atmospheric alkaline carbon reservoir for large-scale climatic and environmental changes.

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### 1. Introduction

[2] Mineral dust plays important global roles in the land, atmosphere, and ocean [Arimoto, 2001; Ridgwell, 2002]. Within the past decade, there has been increasing interest in airborne carbonate ( $\text{CO}_3^{2-}$ ) in suspended dust because of its important roles in atmospheric chemistry, global climate, and radiative forcing [Sequeira, 1993; Buseck and Posfai, 1999; Oba and Pedersen, 1999; Li et al., 2000], presenting "carbonate mysteries" in the global climatic system [Elderfield, 2002].  $\text{CO}_3^{2-}$  can affect atmospheric chemistry and aerosol characteristics because its alkalinity favors the uptake of  $\text{SO}_2$  and  $\text{NO}_x$  and their conversion to  $\text{SO}_4$  and  $\text{NO}_3$  on the surface, as well as removal of  $\text{HNO}_3$  and  $\text{H}_2\text{SO}_4$  from the gas phase [Dentener et al., 1996]. The buffering capacity of  $\text{CO}_3^{2-}$  has been demonstrated for precipitation in China [Zhao et al., 1988] and the Northern Hemisphere [Sequeira, 1993]. Increased airborne  $\text{CO}_3^{2-}$  concentrations during glacial periods could have played an intermediary

role between the biological and alkalinity pumps in the ocean, enhancing  $\text{CO}_2$  absorption by the ocean and by rain [Oba and Pedersen, 1999]. Different mineral particles, including calcite ( $\text{CaCO}_3$ ), have different effects on Earth's radiative forcing [Li et al., 2000].

[3]  $\text{CaCO}_3$  is a prominent alkaline component of source materials in Asian dust source regions [Liu et al., 1985; Andronova et al., 1993; Gomes and Gillette, 1993]. Previous studies about  $\text{CO}_3^{2-}$  focused on Ca as an indicator [Gomes and Gillette, 1993; Sequeira, 1993], but this is not specific to  $\text{CO}_3^{2-}$  because CaO, and  $\text{Ca}(\text{OH})_2$  also contribute to ambient Ca levels [Zhang and Iwasaka, 1999].

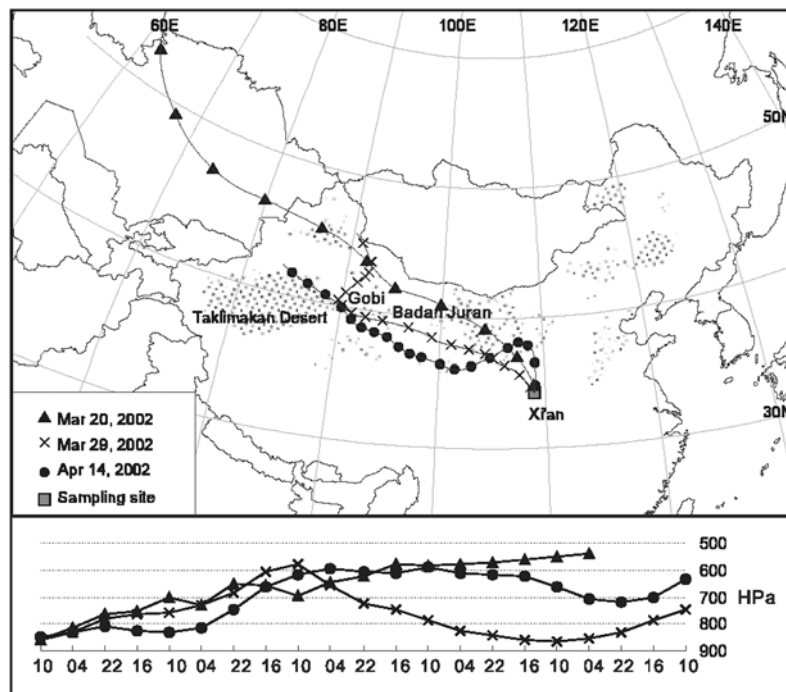
[4] Large amounts of dust from Central Asia are carried by midlatitude prevailing winds and deposited in east Asia and beyond. Portions of this Asian dust are found in the near-surface atmosphere, in deep-sea sediments of the remote Pacific [Duce et al., 1980; Hovan et al., 1989], and in Greenland ice cores [Biscaye et al., 1997]. Asian dust has been found also in the atmosphere of Korea and Japan [Okada et al., 1990; Ro et al., 2001; Kawamura et al., 2004] and in the northern United States [Watson, 2002; VanCuren and Cahill, 2002; VanCuren, 2003]. In spite of many Asian dust studies, information about  $\text{CO}_3^{2-}$  in the atmosphere near Asian dust source regions is scarce. Soil-derived  $\text{CO}_3^{2-}$  from Asia is of interest for a better understanding of alkaline matter contribution to climatic and environmental effects at local, regional, and global scales [Dentener et al., 1996]. This paper reports the first measurements and comparisons of  $\text{CO}_3^{2-}$  in Asian dust storm (DS)

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**Figure 1.** Five-day isentropic back trajectories from Xi'an for 20 and 29 March and 14 April 2003 at 0000 UTC. Each segment denotes the transport pathway every 6 hours, and trajectories are shown for 5 days. Shaded regions indicate desert source regions.

and non-dust-storm (NDS) periods at a continental site in Asian dust source regions.

## 2. Sampling and Analyses

### 2.1. Sampling

[5] Xi'an ( $33^{\circ}29' - 34^{\circ}44'N$ ,  $107^{\circ}40' - 109^{\circ}49'E$ , population 5 million), the largest city in northwestern China, lies in the south margin of the Loess Plateau, China. During the spring, desert dust entrained by winter monsoonal winds from western and northwestern Chinese deserts such as the Taklimakan and Badari/Juran reaches Xi'an, with high ambient particulate matter (PM) concentrations. PM collected in Xi'an during dust storms has a composition similar to that of source regions [Zhuang *et al.*, 1992; Zhang *et al.*, 1993; Gao *et al.*, 1997]. The Xi'an monitoring site (400 m above the sea level) is situated  $\sim 15$  km south of downtown Xi'an. The site has an urban-scale zone of representation [Chow *et al.*, 2002] with no major industrial activities or local fugitive dust sources in the surrounding area, which is mostly residential. Aerosol samples were obtained from the rooftop of the Institute of Earth Environment, Chinese Academy of Sciences, at 10 m above ground level.

[6] Particles with aerodynamic diameter  $\leq 2.5 \mu\text{m}$  ( $\text{PM}_{2.5}$ ) samples were collected from 20 March to 17 April 2002 with a minivolume [Baldauf *et al.*, 2001] sampler (Airmetrics, Springfield, Oregon, USA) operating at flow rates of  $5 \text{ L min}^{-1}$  [Cao *et al.*, 2003] on 47 mm Whatman quartz fiber filters (QM-A). Filters were preheated at  $800^{\circ}\text{C}$  for 3 hours to remove any residual carbon content. Twenty-four-hour sampling was conducted each day. Additional and shorter-duration samples were collected during the 20 March and 14 April dust storms to obtain a characterization of

pure Asian dust particles. Thus 31 aerosol samples were collected in 29 days.

[7] Three dust storms occurred in spring 2002 in Xi'an: on 20 March (DS I), 29 March (DS II), and 14 April (DS III). On the basis of 5-day three-dimensional back trajectories (NOAA HYSPLIT model), all of the dust storms originated from the northwest deserts and Gobi regions (Figure 1). DS I was the most intense in recent years [Zhang *et al.*, 2005]. Figure 1 shows strong winds over the northwest dust source regions and transport through 18 provinces in northern China. DS I lasted more than 50 hours (from  $\sim 6:00$  a.m., 20 March to  $\sim 9:00$  a.m., 22 March). The impact of this dust storm was detected in Beijing [Sugimoto *et al.*, 2003], and Chongwon-Chongju in Korea [Chung *et al.*, 2003]. The effects of DS III also were observed at the Zhenbeitai supersite of ACE-Asia [Alfaro *et al.*, 2003], which is located  $\sim 600$  km north of Xi'an.

### 2.2. Mass and Elemental Analyses

[8] The aerosol mass was determined gravimetrically using an electronic microbalance with a  $1 \mu\text{g}$  sensitivity (Mettler M3, Switzerland). Quartz filters were weighed after being equilibrated for 24 hours in a silica gel desiccator and at a constant (within  $\pm 2^{\circ}\text{C}$ ) temperature between  $20^{\circ}\text{C}$  and  $23^{\circ}\text{C}$ . Each filter was weighted at least three times before and after sampling and the net mass sampling was obtained by subtracting the average preweight data from the average postweight data. The difference among the three repeated weightings was less than  $10 \mu\text{g}$  for a blank filter and less than  $20 \mu\text{g}$  for a sampled filter.

[9] The samples were analyzed for Ca concentrations by proton-induced X-ray emission spectroscopy (PIXE) using

2.5 MeV protons with a 10 nA beam current from a  $1.7 \times 2$  MV accelerator at Beijing Normal University. The PIXE system was calibrated using standards produced by Micro-Matter Co. [Zhang *et al.*, 1997]. Blank filter background spectra were subtracted prior to peak integration. Analysis of eight samples of standard reference material from the *National Bureau of Chemical Exploration Analysis* [1984], China, showed that satisfactory precision (<10%) and accuracy (<15%) were achieved.

### 2.3. Carbonate Carbon Analysis

[10] Samples were analyzed for carbonate carbon (CC) using a DRI Model 2001 Thermal/Optical Carbon Analyzer [Chow *et al.*, 2001, 2004]. To release and quantify CC, the  $0.526 \text{ cm}^2$  punch was first bathed in pure helium (He) at  $40 \text{ cm}^3 \text{ min}^{-1}$  for 90 s to attain equilibrium with the environment. Twenty microliters of 25% v/v Ortho phosphoric acid ( $\text{H}_3\text{PO}_4$ ) was then deposited onto the punch with a microliter syringe. The amount of  $\text{H}_3\text{PO}_4$  is sufficient to convert up to  $800 \mu\text{g m}^{-3}$  of CC to  $\text{CO}_2$ . The quartz fiber filter acts as a wick to distribute the  $\text{H}_3\text{PO}_4$  solution evenly throughout the filter and its particle deposit. Evolved  $\text{CO}_2$  is entrained in the He carrier gas, passes through oxidizing and reducing ovens, and is measured as  $\text{CH}_4$  by a flame ionization detector (FID). The detection limits for CC were below  $1.0 \mu\text{g m}^{-3}$ . The analyzer was calibrated with known quantities of  $\text{CH}_4$  every day. Replicate analyses were performed on one out of 10 samples and differences were smaller than 5% for CC.

[11] CC (12 amu) can be converted to the equivalent  $\text{CO}_3^{2-}$  (60 amu) concentration by a factor of 5 (60/12) multiplier.

### 2.4. Stable Carbon and Oxygen Isotope Analysis

[12] The  $^{13}\text{C}$  and  $^{18}\text{O}$  isotopic abundances were measured on a Finnigan MAT 252 mass spectrometer fitted with an online Finnigan automatic  $\text{CO}_3^{2-}$  reaction system ("Kiel device"). The filters were cut into few small pieces and put into the glass test tube, and then baked about 1 hour at  $70^\circ\text{C}$ . When the vacuum reached  $<150 \mu\text{b}$ ,  $\text{H}_3\text{PO}_4$  was added to react with  $\text{CO}_3^{2-}$ . The reaction was stabilized under a constant temperature ( $70^\circ\text{C}$ ) for 600 s; impurity gases were eliminated in another 90 s. The evolved  $\text{CO}_2$  was transferred to a liquid nitrogen dewar controlled at  $-170^\circ\text{C}$  liquid nitrogen. Impurity gases were removed at their gas condensation temperature to purify the  $\text{CO}_2$ . The dewar was heated to release the  $\text{CO}_2$  to another dewar, and their  $\text{CO}_2$  was transferred to the ion source of MAT 252 mass spectrometer for  $^{13}\text{C}$  and  $^{18}\text{O}$  analysis. Isotopic results are presented in the usual  $\delta$  notations as the permil (‰) deviation of the sample  $\text{CO}_2$  from the Pee Dee Belemnite (PDB) standard.

$$\delta_{\text{sample}} = R_{\text{sample}}/R_{\text{standard}} - 1 \times 1000\text{‰} \quad (1)$$

where  $R = ^{13}\text{C}/^{12}\text{C}$  or  $^{18}\text{O}/^{16}\text{O}$ . The reproducibility of the system is  $\pm 0.1\text{‰}$  for  $\delta^{13}\text{C}$  and  $\pm 0.2\text{‰}$  for  $\delta^{18}\text{O}$  on the basis of replicate measurements of an internal  $\text{CO}_3^{2-}$  standard. Eighteen samples with high  $\text{CO}_3^{2-}$  contents were measured for the stable C-O isotopes. Replicate analyses of three samples with high  $\text{CO}_3^{2-}$  content showed that analytical

errors in the carbon and oxygen isotope ratios were within 1% and 5%, respectively.

## 3. Results

### 3.1. Temporal Variations of $\text{PM}_{2.5}$ Mass and Carbonate Carbon Concentrations

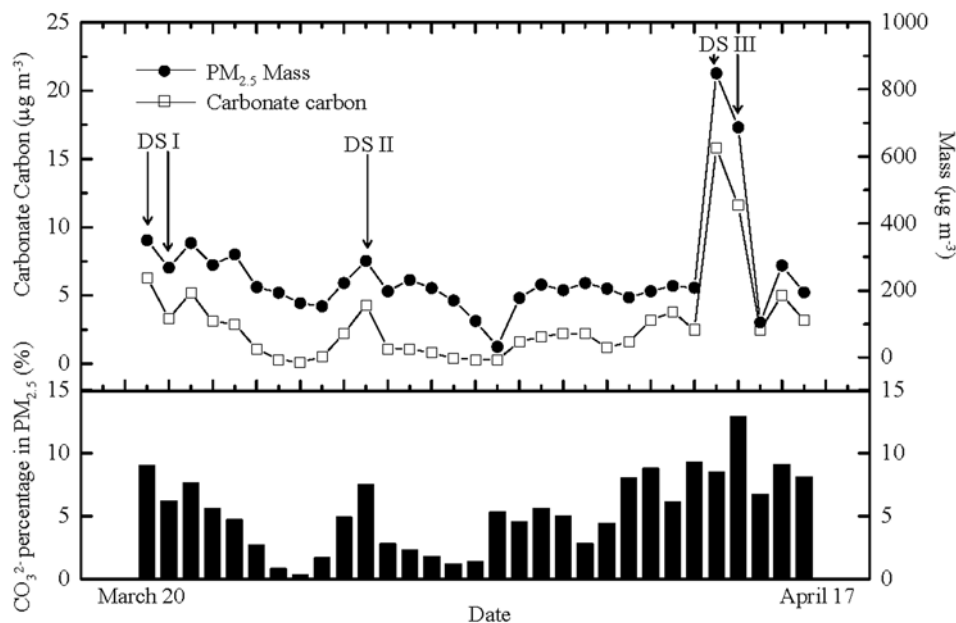
[13] Daily  $\text{PM}_{2.5}$  and CC concentrations from 20 March to 17 April in Xi'an are shown in Figure 2.  $\text{PM}_{2.5}$  mass and CC concentrations in the DS samples increased because of the large contribution of crustal matter. The highest  $\text{PM}_{2.5}$  concentrations was  $740 \mu\text{g m}^{-3}$  on 14 April, which was more than 3 times the daily average concentration of  $200 \mu\text{g m}^{-3}$  during NDS period. The peak concentrations of daily average CC reached  $13 \mu\text{g m}^{-3}$  on the same day, seven times the average CC concentration of  $1.9 \mu\text{g m}^{-3}$  during the NDS period. Compared with the  $\text{PM}_{2.5}$  concentration of  $350 \mu\text{g m}^{-3}$  at the 20 March episode in Xi'an, the TSP (total suspended particles) at Beijing was as high as  $11,000 \mu\text{g m}^{-3}$  on 20 March [Sugimoto *et al.*, 2003], and the  $\text{PM}_{2.5}$  at Chongwon-Chongju Korea was  $331 \mu\text{g m}^{-3}$  on 23 March [Chung *et al.*, 2003]. The peak hourly concentration of  $\text{PM}_{2.5}$  during DS III achieved  $848 \mu\text{g m}^{-3}$  (Figure 2). In contrast,  $\text{PM}_{10}$  hourly mass concentrations at Zhenbeitai reached  $4650 \mu\text{g m}^{-3}$  [Alfaro *et al.*, 2003]. Between 20 March and 17 April, two precipitation events (25 March and 4 April) occurred and  $\text{PM}_{2.5}$  and CC levels decreased. Because few measurements are available elsewhere for CC, it is difficult to compare concentrations at different locations for the same dust storm events. Kawamura *et al.* [2004] reported a TSP CC of  $6.3 \mu\text{g m}^{-3}$  at the Kosan ( $126^\circ 10' \text{E}$ ,  $33^\circ 17' \text{N}$ ) supersite of ACE-Asia during DS I, compared with the  $\text{PM}_{2.5}$  CC at Xi'an of  $4.3 \mu\text{g m}^{-3}$ .

[14]  $\text{PM CO}_3^{2-}$  has not been measured very often, as shown in Table 1.  $\text{CO}_3^{2-}$  is measured by the acidification method [Chow and Watson, 2002] (i.e., acid releases the  $\text{CO}_3^{2-}$  in the filters), or by X-ray diffraction [Sturges *et al.*, 1989; Esteve *et al.*, 1997]. Compared with other measurements, the average airborne  $\text{CO}_3^{2-}$  in Xi'an during NDS periods was higher than those reported in Table 1.  $\text{CO}_3^{2-}$  levels during Asian dust storms ( $36 \mu\text{g m}^{-3}$ , average value of five dust storm samples) are the highest reported anywhere.

[15] The fractions of  $\text{PM}_{2.5}$  composed of  $\text{CO}_3^{2-}$  are also given in Figure 2.  $\text{CO}_3^{2-}$  on average accounted for 8.0% of  $\text{PM}_{2.5}$  during DS and 4.7% of  $\text{PM}_{2.5}$  under NDS conditions. The average fraction of  $\text{CO}_3^{2-}$  in DS samples is similar to that in deep China loess (6.7% [Wen, 1989]) deposit. It is also similar to the  $\text{CO}_3^{2-}$  mass fraction of 7.1% from surface samples taken from the Taklimakan desert [Wang *et al.*, 2004], which is the most important source of Asian dust.

### 3.2. Elemental Composition

[16] The maximum possible CC was estimated as 0.3 times the calcium (Ca) concentration, which is the ratio of the atomic weights of carbon (12 amu) and Ca (40 amu) in calcium carbonate (the most common geological form). This is an upper limit for actual  $\text{CO}_3^{2-}$  levels, since Ca in surface soils is often present in oxide forms such as CaO. Generally, calcium carbonate in the forms of calcite ( $\text{CaCO}_3$ ), orthorhombic aragonite ( $\text{CaCO}_3$ ), and hexagonal



**Figure 2.** Concentrations of  $\text{PM}_{2.5}$  mass and carbonate carbon, and the  $\text{CO}_3^{2-}$  fraction of  $\text{PM}_{2.5}$  at Xi'an during 20 March to 17 April 2002. (The first sample of DS I was collected for 8 hours, and the remainder was collected for 16 hours. The first sample of DS III was collected for 10 hours, and the remainder was collected for 14 hours. Other samples were collected for 24 hours.)

dolomite ( $\text{CaMg}(\text{CO}_3)_2$ ) constitute  $\sim 4\%$  of the Earth's crust, but this abundance is higher in Asian dust source regions [Liu *et al.*, 1985; Gomes and Gillette, 1993]. China loess samples are characteristically  $\text{CO}_3^{2-}$  rich in calcite ( $\sim 100\%$ ) rather than dolomite [Wen, 1989]. The results of PIXE analysis for Ca are presented in Table 2, which shows that elevated Ca concentrations are associated with elevated CC concentrations, and vice versa. For 11 of the 12  $\text{PM}_{2.5}$  samples, measured CC is more than half of the Ca-estimated CC, consistent with Ca being present mostly in  $\text{CO}_3^{2-}$  minerals, such as calcite. Only the sample collected on 30 March measured CC less than 25% of the Ca-estimated CC, implying that most of the Ca in this sample was in a noncarbonate form.

### 3.3. Stable Carbon and Oxygen Isotopic Characteristics

[17] Samples from Asia, Africa, America, and Europe demonstrated that carbon and oxygen isotopic abundance in pedogenic  $\text{CO}_3^{2-}$  varies with local ecosystem and climatic

conditions [Cerling, 1984]. Stable carbon and oxygen isotopic compositions of  $\text{CO}_3^{2-}$  are useful indicators of paleoecologic and paleoclimatic variations [Wen, 1989; Li *et al.*, 1995]. Carbonates from different areas of North China have distinct  $^{13}\text{C}$  and  $^{18}\text{O}$  isotopic compositions [Wen, 1989; Li *et al.*, 1995]. These isotopic abundances might be useful for identifying a quantitative Asian dust contribution to ambient  $\text{PM}_{2.5}$ .

[18] Figure 3 shows that the  $\delta^{13}\text{C}$  and  $\delta^{18}\text{O}$  abundance in  $\text{PM}_{2.5}$   $\text{CO}_3^{2-}$  fall within the range of fresh water  $\text{CO}_3^{2-}$  and resemble the isotopic characteristics of eolian sand and loess. These samples separate into two distinct groups (Figure 3) from which information can be gained about different source contributions. For  $\delta^{13}\text{C}$ , the maximum value ( $-1.4\text{‰}$ ) occurred in the 20 March dust storm sample, while the minimum value ( $-9.3\text{‰}$ ) occurred in the non-dust-storm samples of 12 and 13 April. The average  $\delta^{13}\text{C}$  for dust storm samples (five samples in total, two from DS I, one from DS II, and two from DS III (Figure 3)) was  $-2.7\text{‰}$ , which is close to the  $\delta^{13}\text{C}$  value of  $-3.5\text{‰}$  in

**Table 1.** Examples of Carbonate ( $\text{CO}_3^{2-}$ ) Concentrations in Suspended Particles

Reference	Study Location	Study Period	Carbonate Concentration (Size Fraction), $\text{ng m}^{-3}$
Mueller <i>et al.</i> [1971]	Pasadena, California, USA	summer 1969	<20–880 (PM of 0.1–17 $\mu\text{m}$ )
Sturges <i>et al.</i> [1989]	Toronto, Canada	January and July 1986	920–1470 ( $\text{PM}_{2.5}$ ) (calcite)
Clarke and Karani [1992]	Leeds, UK	winter 1989–1990	20–30 ( $\text{PM}_{2.5}$ )
Esteve <i>et al.</i> [1997]	Castellon, Spain	July 1993	550 (quartz, calcite, gypsum)
Zappoli <i>et al.</i> [1999]	Great Hungarian Plain and central Sweden	summer 1996	<100–200 (PM < 1.5 $\mu\text{m}$ ) (CC)
Zappoli <i>et al.</i> [1999]	Po Valley, Italy	fall 1996	400–3000 ( $\text{PM}_{2.5}$ ) (CC)
Chow <i>et al.</i> [2002]	58 sites of IMPROVE network, USA	1996–1999	from <100 to 420 ( $\text{PM}_{2.5}$ ) (CC)
Venkataraman <i>et al.</i> [2001]	Mumbai, India	1999	1300–2500 ( $\text{PM}_{10}$ ) (CC)
This study	DS, Xi'an, China	spring 2002	36,000 ( $\text{PM}_{2.5}$ )
This study	NDS, Xi'an, China	spring 2002	9700 ( $\text{PM}_{2.5}$ )

**Table 2.** Comparison of the Upper Limit of Carbonate Carbon With Measured Carbonate Carbon for Selected Samples<sup>a</sup>

Date	Concentration, $\mu\text{g m}^{-3}$		Measured CC	Measured CC/UL CC Mass Ratio, %
	Ca	UL CC		
20 March 2002	21.8	6.5	6.3	96
21 March 2002	12.2	3.7	5.2	141
22 March 2002	12.2	3.6	3.1	85
23 March 2002	11.3	3.4	2.9	85
24 March 2002	7.5	2.3	1.1	50
29 March 2002	9.9	3.0	4.3	146
30 March 2002	14.3	4.2	1.1	23
14 April 2002	7.4	2.2	2.5	114
14 April 2002	43.3	13.0	15.8	122
15 April 2002	7.5	2.3	2.4	108
16 April 2002	13.4	4.0	5.0	123
17 April 2002	15.2	4.6	3.2	69

<sup>a</sup>“UL CC,” upper limit of carbonate carbon (determined as 0.3 times the corresponding calcium concentration). “Measured CC,” measured carbonate carbon.

modern eolian sand from the northwest deserts [Li *et al.*, 1995] and to the  $\delta^{13}\text{C}$  value of 0.7‰ in surface samples from the Taklimakan desert [Wang *et al.*, 2004]. Asian dust in PM<sub>2.5</sub> can remain suspended for more than one week or can be resuspended after deposition. The local Xi’an atmosphere showed Asian dust contributions during several NDS periods. The average  $\delta^{13}\text{C}$  for NDS periods is  $-8.3\text{‰}$ , close to  $\delta^{13}\text{C}$  value of  $-7.5\text{‰}$  for surface soil in Xi’an [Wen, 1989]. The variations of  $\delta^{18}\text{O}$  show a similar relationship with the  $\delta^{13}\text{C}$  from DS and NDS samples. As shown in Figure 3,  $\delta^{13}\text{C}$  values were strongly correlated with  $\delta^{18}\text{O}$  values ( $r = 0.78$ ,  $p < 0.001$ ), but the relationship appears stronger for DS than for NDS samples.

[19] The isotopic composition of  $\text{CO}_3^{2-}$  during NDS periods (Figure 3) reflects the mixed contributions from Asian and local dust sources. The relative contributions from Asian dust and local sources can be apportioned as follows [Langmuir *et al.*, 1978; Watson *et al.*, 2002]:

$$\delta\text{M} = X_A\delta_A + X_B\delta_B \quad (2)$$

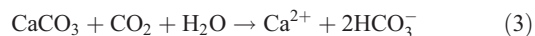
where  $\delta\text{M}$  denotes  $\delta^{13}\text{C}$  of ambient samples with Asian and local dust mixture (middle group of points in Figure 3),  $\delta_A$  denotes the  $\delta^{13}\text{C}$  of  $\text{CO}_3^{2-}$  in DS samples ( $-2.7\text{‰}$ , upper group in Figure 3) and  $\delta_B$  denotes the  $\delta^{13}\text{C}$  of  $\text{CO}_3^{2-}$  in NDS local samples ( $-9.5\text{‰}$ ) (the lower left group in Figure 3). The relative contributions from dust storm source accounted for 32–89% of ambient samples during different NDS periods with carryover from the DS periods. The highest Asian dust contributions occurred on 22 March, two days of DS I (Figure 2). PM<sub>2.5</sub> entrained by this intense dust storm remained in the air for several days because there was no precipitation and limited deposition. The lowest Asian dust contribution (32%) to mixed samples occurred on 28 March.

## 4. Discussion

### 4.1. Climatic Effects

[20] Dust inputs cause biological and geochemical response in oceans [Ridgwell, 2002]. The capacity of the oceanic “biological pump” is large enough to have con-

tributed to the reduction of atmospheric  $\text{CO}_2$  levels during glacial periods [Sarnthein *et al.*, 1988; Ridgwell, 2002]. Eolian  $\text{CO}_3^{2-}$  would have deposited on the sea surface and increased the alkalinity of subsurface waters in the ocean via

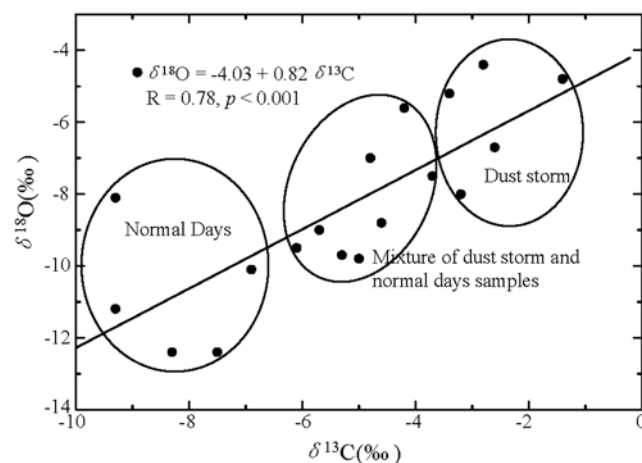


[21] The result is the Pacific Ocean “alkalinity pump” proposed by Boyle [1988] and Broecker and Peng [1989].

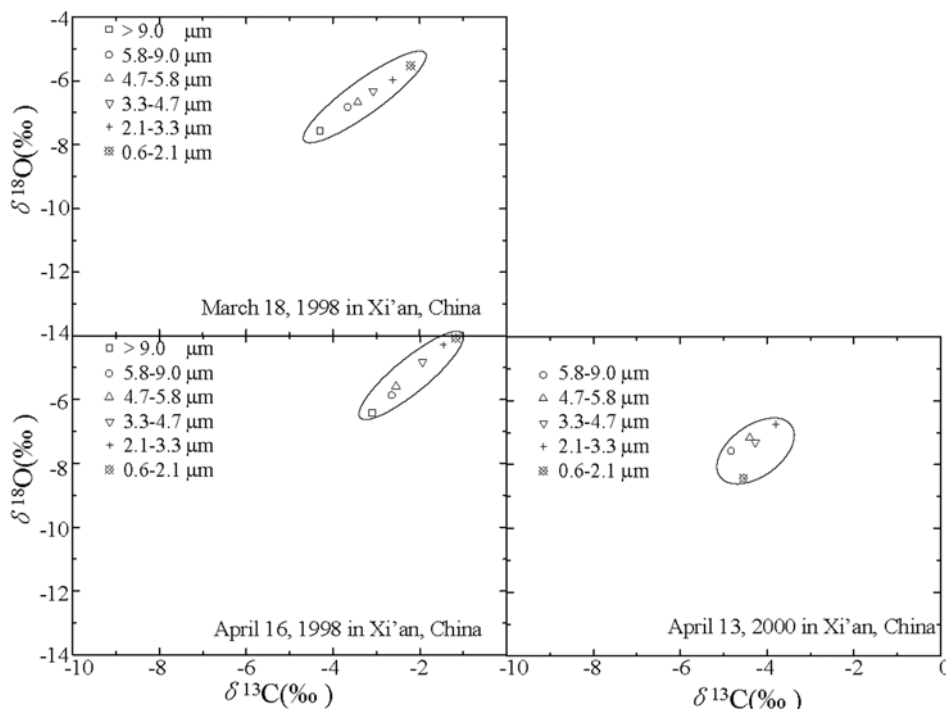
[22] The deserts of northern and northwestern China release about  $\sim 800$  Tg of dust into the atmosphere each year [Zhang *et al.*, 1997]. About 20% of this dust is transported over continental China, and 50% of this amount is subject to long-range transport to the Pacific Ocean and beyond. Since  $\text{CO}_3^{2-}$  accounts for 8.0% of the mass, the amount of airborne  $\text{CO}_3^{2-}$  deposited in the Pacific Ocean could be 32 Tg  $\text{yr}^{-1}$ ; i.e.,  $3.2 \times 10^{13}$  g  $\text{yr}^{-1}$  (1 Tg =  $10^{12}$  g). Keir [1995] noted that an additional input of  $1.3 \times 10^{13}$  mol  $\text{yr}^{-1}$  of dissolved  $\text{CO}_3^{2-}$  from glacial sources could account for half ( $\sim 40$  ppmv) of the atmospheric  $\text{CO}_2$  difference between the last glacial maximum and the Holocene period. Thus eolian  $\text{CO}_3^{2-}$  deposited at the rate of  $3.2 \times 10^{13}$  g  $\text{yr}^{-1}$  in the Pacific Ocean may have decreased atmospheric  $\text{CO}_2$  by  $\sim 1.6$  ppmv ( $5.0 \times 10^{16}$  mol of carbon (280 ppmv) is the estimated global concentration in the preindustrial global atmosphere [Siegenthaler and Oeschger, 1987]; 1.6 ppmv of  $\text{CO}_2$  is equal to  $2.8 \times 10^{14}$  mol of carbon).  $\text{CO}_3^{2-}$  from Asian dust deposited in the Pacific Ocean could lead to the sequestration of  $\text{CO}_2$  at the rate of  $1.0 \times 10^{15}$  g  $\text{yr}^{-1}$ ; i.e., 1.0 Pg  $\text{yr}^{-1}$  (1 Pg =  $10^{15}$  g, or 0.3 Pg carbon). For comparison, the total emission of anthropogenic  $\text{CO}_2$  from China in 1997 was 8.9 Pg [Gao, 2002]. If Oba and Pedersen’s [1999] “raindrop alkalinity pump” is real, eolian  $\text{CO}_3^{2-}$  will also decrease atmospheric  $\text{CO}_2$ . The important role of eolian  $\text{CO}_3^{2-}$  from Asian dust originating in China has been underestimated in previous studies.

### 4.2. Environmental Effects

[23] About 20% of the suspended dust (800 Tg  $\text{yr}^{-1}$ ) is transported over continental China [Zhang *et al.*, 1997];

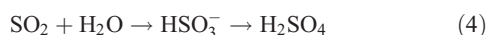


**Figure 3.** Stable C-O isotopic compositions of carbonate in dust storm and non-dust-storm samples.



**Figure 4.** The  $^{13}\text{C}$  and  $^{18}\text{O}$  isotopic abundances for different size fractions during dust storms. Samples with values below lower quantifiable limits are omitted.

therefore about  $12.8 \text{ Tg yr}^{-1}$  of  $\text{CO}_3^{2-}$  could be added to the atmosphere of eastern China on the basis of a 8.0%  $\text{CO}_3^{2-}$  content. Chemical reactions take place among  $\text{CO}_3^{2-}$ ,  $\text{SO}_2$ , and sulfur in the atmosphere, via



[24] Accordingly,  $12.8 \text{ Tg yr}^{-1}$  of  $\text{CO}_3^{2-}$  may neutralize about  $13.4 \text{ Tg yr}^{-1}$  of  $\text{SO}_2$ . Compared with the total emission of  $\text{SO}_2$  (25.2 Tg) from China in 1995 [Streets and Waldhoff, 2000], the  $\text{SO}_2$  buffered by eolian  $\text{CO}_3^{2-}$  could absorb more than half of  $\text{SO}_2$  and  $\text{CO}_3^{2-}$  to be similarly distributed in space and time. Zhao *et al.* [1988] observed that high levels of windblown dust dominate the atmospheric buffering capacity in southwestern China. Bulk aerosol data [Winchester and Wang, 1989] from the Asia-Pacific region provide evidence for a statistical relationship between sulfate and dust. Larssen and Carmichael [2000] also observed impacts in Europe and North America associated with high levels of strong acids in precipitation that had not been widely detected in China. This may be partly due to the fact that northern China acid deposition was heavily modified by natural soil dust from desert and semiarid areas. Mineral particles that contained sulfuric acid and sulfate, which formed by heterogeneous processes, were found in the troposphere (4–21 km altitude) in northern China [Xu *et al.*, 2001]. Kawamura *et al.* [2004] observed that carbonates most likely react with acidic species (such as  $\text{H}_2\text{SO}_4$ ,  $\text{HNO}_3$ , and organic acids) in the atmosphere during long-range transport (atmospheric titration). Dentener *et al.* [1996] showed that for major portions

of the globe, more than 50% of the sulfate was associated with dust. He mentioned that the reaction of  $\text{SO}_2$  on Ca-rich mineral aerosol was likely to play an important role downwind of arid source regions, which was especially important for regions in Asia. As a result, the eolian  $\text{CO}_3^{2-}$  that originates from Asian dust source regions has great buffering capacity for neutralizing regional acid rain in Asia.

#### 4.3. Isotopic Marker for Eolian Sources During Long-Range Transport

[25] Isotopic methods also have been used to address the provenance of marine sediments and atmospheric dust particles [Arimoto, 2001]. Much of the early isotopic work on provenance involved studies of strontium (Sr), but more recently samarium (Sm) and neodymium (Nd) isotopes have been used. Rubidium (Rb)–Sr isotope ratios also have been utilized to explain the transport of Asian dust [Kanayama *et al.*, 2002].  $^{13}\text{C}$  and  $^{18}\text{O}$  in  $\text{CO}_3^{2-}$  offer another marker that can be used in conjunction with trace element and other isotopic abundances to better elucidate source origins.

[26]  $\text{PM}_{2.5}$  were measured in this experiment, in order to determine the extent to which  $^{13}\text{C}$  and  $^{18}\text{O}$  abundances vary with size, three previous size-separated dust storm samples from Xi'an were used to analyze the stable C–O isotopic composition. These samples were collected by an Anderson 8-stage sampler (Thermo Electron Co., USA) from three heavy dust storms on 18 March 1998, 16 April 1998, and 13 April 2000 [Cao, 2001]. The relationships of  $\delta^{13}\text{C}$  versus  $\delta^{18}\text{O}$  for the three samples are shown in Figure 4. There are no notable differences among various sizes for the three samples. Differences of  $\delta^{13}\text{C}$  abundances between the largest and smallest size fractions are less than  $-2\text{‰}$ . This difference is much lower than the  $-5.6\text{‰}$  difference between the average  $\delta^{13}\text{C}$  abundance in DS and NDS  $\text{PM}_{2.5}$

samples. The  $\delta^{13}\text{C}$  values appear to decrease with increasing size (Figure 4). Since  $\text{PM}_{2.5}$  has a longer lifetime, and transports farther than larger size fractions, the  $\text{PM}_{2.5}$   $\delta^{13}\text{C}$  are most useful for source apportionment of Asian dust at distant sites.

[27] The  $\text{CO}_3^{2-}$   $\delta^{13}\text{C}$  abundance at Kosan on 21 March 2002 was  $-1\text{‰}$  [Kawamura et al., 2004], close to the Xi'an  $-1.4\text{‰}$  abundance. The similarity between the stable carbon in  $\text{CO}_3^{2-}$  at Xi'an and at Kosan (2000 km away) implies that  $^{13}\text{C}$  in  $\text{CO}_3^{2-}$  could distinguish Asian dust from other dust events of great distances from the source.

## 5. Conclusion

[28] Large amounts of airborne  $\text{CO}_3^{2-}$  from Asian dust can affect climatic and environmental changes in the Asian-Pacific region.  $\text{CO}_3^{2-}$  accounts for about 8.0% of Asian dust mass on the basis of the analysis of three dust storm events in Xi'an. There are also high  $\text{CO}_3^{2-}$  contents (4.7%) in the normal atmosphere in Xi'an.  $\text{CaCO}_3$  is major form of this  $\text{CO}_3^{2-}$ . Asian dust is characterized by  $-2.7\text{‰}$  for  $\delta^{13}\text{C}$  and  $-5.8\text{‰}$  for  $\delta^{18}\text{O}$ . The supply of airborne  $\text{CO}_3^{2-}$  to the Pacific Ocean could facilitate the sequestration of  $\text{CO}_2$  at the rate of  $1.0 \text{ Pg yr}^{-1}$  (0.3 Pg carbon). The  $\text{SO}_2$  buffered by eolian  $\text{CO}_3^{2-}$  could be as high as  $13.4 \text{ Tg yr}^{-1}$ , which exceeds more than half of the total emission of  $\text{SO}_2$  from China in 1995. Airborne  $\text{CO}_3^{2-}$  from Central Asia may provide the most important atmospheric alkaline carbon reservoir for sequestering  $\text{CO}_2$  and buffering acid rain in the Asian-Pacific region because of high emission of mineral dust and its high content of  $\text{CO}_3^{2-}$ .

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