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## Uptake of nitrate and sulfate on dust aerosols during TRACE-P

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[1] Aerosol data collected near Asia on the DC-8 aircraft platform during TRACE-P has been examined for evidence of uptake of  $\text{NO}_3^-$  and  $\text{SO}_4^{2-}$  on dust surfaces. Data is compared between a sector where dust was predominant and a sector where dust was less of an influence. Coincident with dust were higher mixing ratios of anthropogenic pollutants.  $\text{HNO}_3$ ,  $\text{SO}_2$ , and CO were higher in the dust sector than the nondust sector by factors of 2.7, 6.2, and 1.5, respectively. The collocation of dust and pollution sources allowed for the uptake of  $\text{NO}_3^-$  and  $\text{nss-SO}_4^{2-}$  on the coarse dust aerosols, increasing the mixing ratios of these particulates by factors of 5.7 and 2.6 on average. There was sufficient  $\text{nss-SO}_4^{2-}$  to take up all of the  $\text{NH}_4^+$  present, with enough excess  $\text{nss-SO}_4^{2-}$  to also react with dust  $\text{CaCO}_3$ . This suggests that the enhanced  $\text{NO}_3^-$  was not in fine mode  $\text{NH}_4\text{NO}_3$ . Particulate  $\text{NO}_3^-$  (p- $\text{NO}_3^-$ ) constituted 54% of the total  $\text{NO}_3^-$  (t- $\text{NO}_3^-$ ) on average, reaching a maximum of 72% in the dust sector. In the nondust sector, p- $\text{NO}_3^-$  contributed 37% to t- $\text{NO}_3^-$ , likely due to the abundance of sea salts there. In two other sectors where the influence of dust and sea salt were minimal, p- $\text{NO}_3^-$  accounted for <15% of t- $\text{NO}_3^-$ . **INDEX TERMS:** 0305 Atmospheric Composition and Structure: Aerosols and particles (0345, 4801); 0345 Atmospheric Composition and Structure: Pollution—urban and regional (0305); 0365 Atmospheric Composition and Structure: Troposphere—composition and chemistry; 0368 Atmospheric Composition and Structure: Troposphere—constituent transport and chemistry; **KEYWORDS:** heterogeneous uptake of nitrate, Asian dust, TRACE-P

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### 1. Introduction

[2] The two primary objectives of NASA's Transport and Chemical Evolution over the Pacific (TRACE-P) mission were to characterize the sources and chemical composition of Asian outflow and to study the evolution of this outflow. As part of this mission, measurements of aerosol chemical and physical properties were made aboard the DC-8 aircraft [see Jordan *et al.*, 2003; Dibb *et al.*, 2003]. Samples clearly influenced by dust were obtained. Recent modeling and laboratory studies [e.g., Zhang *et al.*, 1994; Dentener *et al.*, 1996; Xiao *et al.*, 1997; Goodman *et al.*, 2000; Phadnis and Carmichael, 2000; Song and Carmichael, 2001a, 2001b; Underwood *et al.*, 2001] have suggested that alkaline dust particles can take up acids resulting in increased coarse mode  $\text{NO}_3^-$  and  $\text{SO}_4^{2-}$ . In this paper, we report in situ evidence collected regionally aboard the DC-8 that supports prior indications that such uptake occurs in the atmosphere.

[3] Using 40 years of dust storm records, Sun *et al.* [2001] describe how dust is transported out of China. These

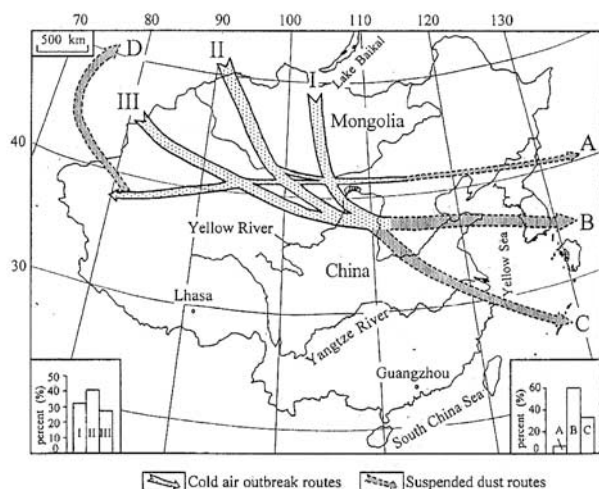
dust storms occur predominantly in spring [Liu, 1985; Sun *et al.*, 2001] and are due to cold air outbreaks which cause frontal systems and the Mongolian cyclonic depression. About 78% of dust storms are associated with the Mongolian cyclone with the remainder associated solely with the passage of cold fronts [Sun *et al.*, 2001]. There are two dominant source regions of dust in East Asia, the gobi deserts in Mongolia and northern China, and the Taklimakan Desert in western China. Owing to the mountains surrounding the Taklimakan, dust can only be exported out of this region when strong easterlies loft dust above 5 km in altitude (the height of the mountains). At these heights, this dust becomes entrained in the jet stream and may then be transported long distances over the Pacific Ocean and to North America. Dust from the gobi deserts of Mongolia and northern China on the other hand, is generally lofted to altitudes <3 km and then transported in a southeasterly direction, depositing dust on the Loess Plateau, eastern Asia, and the western Pacific (Figure 1 [Sun *et al.*, 2001]).

[4] The mineralogy of loess at its source consists primarily of quartz ( $\text{SiO}_2$ ), feldspars, micas, clays, carbonates (primarily  $\text{CaCO}_3$ ), and several minor minerals [Pye, 1987; Gao and Anderson, 2001]. The carbonate content is relatively constant at 12% by weight in loess, dust, and ground surface samples [Derbyshire *et al.*, 1998]. Nishikawa *et al.* [1991] found that weight fractions of  $\text{SO}_4^{2-}$  in soils of arid regions of China ranged from <0.01% to 0.46%, insufficient to account for the high fraction of  $\text{SO}_4^{2-}$  in dust downwind

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**Figure 1.** Map of cold air outbreaks and suspended dust routes in East Asia, from Sun *et al.* [2001].

of these regions. Individual dust particles from five dust storms in Beijing examined by Zhang and Iwasaka [1999] showed very little water-soluble  $\text{SO}_4^{2-}$  on the surface of these particles. Only  $\sim 15\%$  of the particles had a sulfur peak; even fewer, 11%, showed any  $\text{NO}_3^-$ . This suggests that little chemical alteration takes place prior to reaching Beijing. Yet, samples collected in Qingdao, 500 km southeast of Beijing, showed 50–80% of the coarse particles ( $>2 \mu\text{m}$  diameter) were coated with  $\text{SO}_4^{2-}$  [Parungo *et al.*, 1995]. This chemical alteration then must occur downstream from Beijing.

[5] The presence of  $\text{CaCO}_3$  in Asian dust is important because it reacts with acids such as sulfuric and nitric acid. In this way, dust particles may be chemically altered from their original composition at their source. Assuming the dust particles travel along a path where anthropogenic S and N sources are important, the surface area provided by the dust particles may lead to significant alteration of the air mass chemistry as  $\text{SO}_2$  and  $\text{HNO}_3$  transfer from gas to particulate phase [Zhang *et al.*, 1994; Xiao *et al.*, 1997; Song and Carmichael, 2001a, 2001b]. The presence of these acidic ions on dust can change the solubility of the aerosol from hydrophobic to hydrophilic [Song and Carmichael, 2001a].

[6] Evidence of  $\text{SO}_4^{2-}$  and  $\text{NO}_3^-$  on dust is found in studies of individual particles [e.g., Wu and Okada, 1994; Parungo *et al.*, 1995; Gao and Anderson, 2001] as well as studies of bulk aerosol composition [e.g., Choi *et al.*, 2001; Kim and Park, 2001] and precipitation [e.g., Minoura *et al.*, 1998]. Individual particles show both  $\text{NO}_3^-$  and  $\text{SO}_4^{2-}$  inclusions along with  $\text{CaCO}_3$ , as well as particles where all of the  $\text{CO}_3^{2-}$  has been replaced [Wu and Okada, 1994; Parungo *et al.*, 1995; Gao and Anderson, 2001]. Bulk aerosols from dust events show that the dominant water-soluble ions are  $\text{SO}_4^{2-}$ ,  $\text{NO}_3^-$ ,  $\text{Ca}^{2+}$ , and  $\text{Mg}^{2+}$  [Choi *et al.*, 2001; Kim and Park, 2001]. Where size-resolved data is available,  $\text{NO}_3^-$  and  $\text{SO}_4^{2-}$  are found primarily in the coarse fraction associated with  $\text{Ca}^{2+}$  during dust events [Kim and Park, 2001]. In the absence of dust,  $\text{SO}_4^{2-}$  is primarily in the fine mode, while  $\text{NO}_3^-$  can be associated with either  $\text{NH}_4^+$  in the fine mode [Kim and Park, 2001; Song and Carmichael, 2001a] or  $\text{Na}^+$  in the coarse mode [Song and Carmichael,

2001a]. In precipitation in Japan the highest average concentrations of  $\text{NO}_3^-$ ,  $\text{SO}_4^{2-}$ , and  $\text{Ca}^{2+}$  are observed during Kosa (dust storm) events [Minoura *et al.*, 1998]. The low pH of precipitation during these dust events is attributed to the large amounts of acidic ions on the Kosa particles scavenged by the precipitation [Minoura *et al.*, 1998]. By the time dust particles reach Korea and Japan,  $\sim 75\%$  of the carbonate has been displaced by  $\text{SO}_4^{2-}$  and  $\text{NO}_3^-$  [Nishikawa *et al.*, 1991].

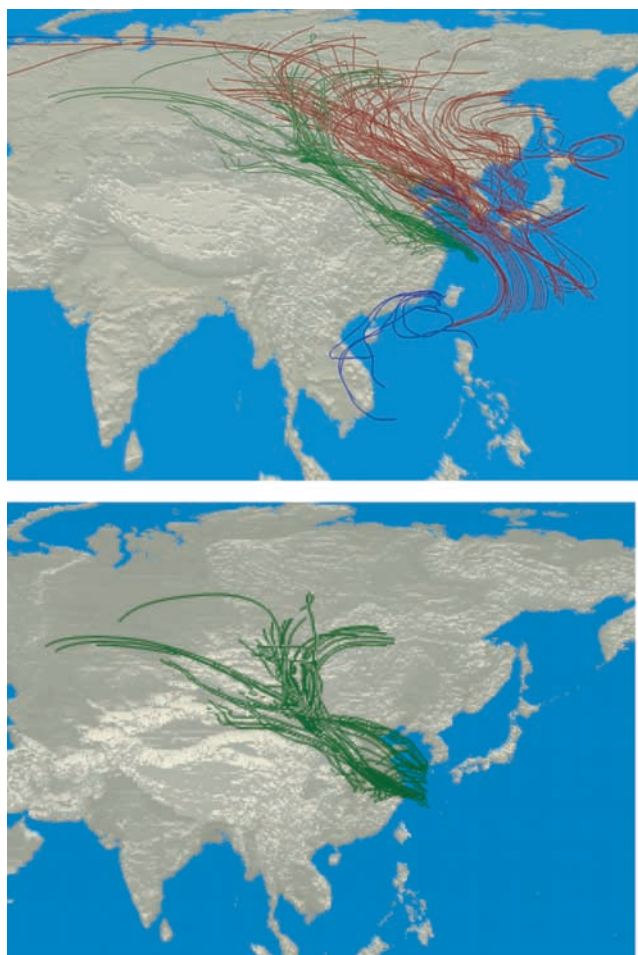
[7] Various modeling studies have estimated the degree to which heterogeneous reactions contribute to the production of coarse mode  $\text{SO}_4^{2-}$  and  $\text{NO}_3^-$ . Chameides and Stelson [1992] estimated  $>70\%$  of  $\text{SO}_4^{2-}$  formed by heterogeneous reactions. Using a regional three-dimensional (3-D) model for the period 1–14 March 1994, Xiao *et al.* [1997] estimated chemical conversion of  $\text{SO}_2$  in the presence of dust contributed 20–40% to total  $\text{SO}_4^{2-}$  production in East Asia. Song and Carmichael [2001a], also using a regional 3-D model, found heterogeneous reactions account for 10–40% of  $\text{SO}_4^{2-}$  production in dust plumes and 10–80% in sea-salt-dominant regions. They attribute the greater production of  $\text{SO}_4^{2-}$  on sea salt versus dust to the dust mass distribution having a larger coarse fraction than sea salt.

[8] Dentener *et al.* [1996] looked at irreversible reactions of  $\text{HNO}_3$ ,  $\text{N}_2\text{O}_5$ ,  $\text{NO}_3$ ,  $\text{HO}_2$ ,  $\text{O}_3$ , and  $\text{SO}_y$  on dust surfaces. They predicted that a substantial fraction of  $\text{SO}_4^{2-}$  is associated with mineral aerosol. They also found that an even larger fraction of gas phase  $\text{HNO}_3$  may be neutralized by mineral aerosol. Phadnis and Carmichael [2000] predict  $>70\%$  of gas-phase  $\text{HNO}_3$  is partitioned onto dust over the gobi deserts, while 10–70% of  $\text{HNO}_3$  ends up in particulate phase over the rest of East Asia. Song and Carmichael [2001a] predict 10–50% of  $\text{HNO}_3$  partitioned into  $\text{NO}_3^-$  in the boundary layer, with this partitioning exceeding 70% in dust and sea-salt plume centers. In the free troposphere, they predict 10–30% of  $\text{HNO}_3$  partitioned into particulate phase, increasing to  $>50\%$  in dust plume centers. Song and Carmichael's model [2001a] also suggests that while  $\text{NO}_3^-$  resides primarily in coarse mode, in regions with abundant  $\text{NH}_3$  and  $\text{HNO}_3$ , fine mode  $\text{NH}_4\text{NO}_3$  may be found. The region where this is most likely to occur is around the lower Huang River, where there is high population density and agricultural activity [Song and Carmichael, 2001a].

[9] A laboratory study of the uptake of  $\text{HNO}_3$  on  $\text{CaCO}_3$  particles [Goodman *et al.*, 2000] found the reaction was limited to the particle surface in the absence of water. However, in the presence of water vapor the reaction was not limited to the surface of the particle. Underwood *et al.* [2001] studied the uptake of  $\text{NO}_2$  and  $\text{HNO}_3$  in the laboratory coupled with model calculations. They found that while uptake of  $\text{NO}_2$  does not appear to be significant, uptake of  $\text{HNO}_3$  is significant, with mixing ratios reduced by 30%. They note that this fraction is likely to increase under humid in situ conditions versus the dry conditions of their experiment.

[10] A companion paper [Jordan *et al.*, 2003] presents a thorough discussion of the DC-8 aerosol observations during TRACE-P in the context of four source regions based on back trajectories. These four sectors are NNW, Channel, WSW, and SE Asia (moving roughly counterclockwise from north to south). WSW and SE Asia samples were





**Figure 2.** Back trajectories of the dust sector (Channel, green lines) and the nondust sector (NNW, red lines).

generally collected above 2 km altitude and represent relatively long range transport. NNW and Channel included many low altitude samples (<2 km) which were heavily influenced by sea salts (both sectors) and dust (Channel). Here we focus on these latter two sectors to look for evidence of gas to particle transfer of  $\text{NO}_3^-$  and  $\text{SO}_4^-$ .

## 2. Approach

[11] Back trajectories coupled with aerosol chemical data were used to classify the DC-8 TRACE-P aerosol data into four groups [Jordan *et al.*, 2003]. The back trajectories were calculated using a kinematic model based on global meteorological analyses from the European Centre for Medium-Range Weather Forecasts (ECMWF) [Fuelberg *et al.*, 2003]. The chemical samples were collected over periods ranging from ~5 to 35 min during level flight legs. The back trajectories were calculated every 5 min during level flight legs. Thus an individual aerosol sample might have anywhere from one to seven back trajectories associated with it. Samples that had multiple trajectories which emanated from more than one of the four sectors were classified as mixed and were not used any further in the analysis. The idea was to obtain a set of samples that represented a given sector with as little interference from other sectors as

possible. In this way, 77 samples were obtained for the NNW sector, 38 for Channel, 31 for SE Asia, and 47 for WSW.

[12] NNW and Channel trajectories both emanate from northern Asia, with the Channel trajectories bounding the NNW trajectories to the south and west (Figure 2, top). The Channel trajectories themselves are bounded to the south by the Tibetan Plateau. WSW trajectories (not shown here) typically reflect high altitude, long range transport from the west as far away as Africa. SE Asia trajectories tend to circle over the western Pacific and southeast Asia. Both of these latter sectors have much lower mixing ratios of aerosols in general than the two former sectors, making them less appropriate for a comparison which tries to isolate the dust component. For a more extensive discussion of the sampling method and back trajectory calculations, please refer to the work of Jordan *et al.* [2003]. A complete discussion of the aerosol properties of each sector appears in that paper. Here the intention is to use just the Channel and NNW sectors to examine the influence of dust on aerosol chemistry, particularly, the uptake of  $\text{NO}_3^-$  and  $\text{SO}_4^-$  on dust surfaces. Note that mixing ratios (pptv) were converted to moles or mass per unit volume using standard cubic meters rather than volumetric cubic meters (e.g.,  $\mu\text{g m}^{-3}$  STP or  $\mu\text{mol SCM}^{-1}$ ).

## 3. Discussion

[13] Sun *et al.* [2001] described the pathways of the cold air outbreaks which lead to dust storms, as well as the pathways the suspended dust particles follow once suspended (Figure 1). Pathway I is followed by cold air masses near Lake Baikal which move southward across central Mongolia and China. Pathway II involves cold air masses in the northwest which result in dust storms along the Hexi Corridor (a northwest-southeast trending geographic region which connects the Xinjiang and Gansu provinces) and gobi deserts in northern China. Pathway III results in dust storms both in the Hexi Corridor and Taklimakan Desert. Most of the cold air outbreaks follow pathway II (41%), followed by pathway 3 (37%), and pathway I (32%). Sun *et al.* [2001] then break down the routes the suspended dust travels. Routes A, B, and C (Figure 1) carry dust from the gobi deserts in Mongolia and China depositing it en route to Korea, Japan, and the Pacific Ocean. Route B is most common (60%) followed by C (33%), then A (7%). Route D carries dust out of the Taklimakan Desert to high altitudes where it becomes entrained in the jet stream and is transported long distances downwind [Sun *et al.*, 2001].

[14] During TRACE-P, using aerosol chemistry coupled with back trajectories, we determined the path followed by the majority of the dust observed by the DC-8 [Jordan *et al.*, 2003]. The back trajectories for these dust samples (Figure 2, bottom) closely resemble the C and B routes described by Sun *et al.* [2001] (Figure 1).

[15] In order to investigate the role of dust in heterogeneous uptake of  $\text{NO}_3^-$  and  $\text{SO}_4^-$ , we compare the dust sector (Channel) to the adjacent sector (NNW). The primary distinction between these sectors is the presence of dust. However, Channel also has higher pollution inputs as well, as evidenced by gas phase species such as  $\text{HNO}_3$  and CO (Table 1). This results in all aerosol species showing higher

**Table 1.** Means, Standard Deviations, Medians, Minima, Maxima, and Number of Samples for Various Aerosol and Gas Phase Species<sup>a</sup>

	NO <sub>3</sub> <sup>-</sup> , nmol/m <sup>3</sup>	nss-SO <sub>4</sub> <sup>=</sup> , nmol/m <sup>3</sup>	nss-Ca <sup>2+</sup> , nmol/m <sup>3</sup>	Na <sup>+</sup> , nmol/m <sup>3</sup>	NH <sub>4</sub> <sup>+</sup> , nmol/m <sup>3</sup>	HNO <sub>3</sub> , nmol/m <sup>3</sup>	SO <sub>2</sub> , nmol/m <sup>3</sup>	CO, ppbv	Ethyne/CO, pptv/ppbv
<i>Channel</i>									
Mean ± st. dev.	74 ± 110	91 ± 91	168 ± 177	64 ± 91	104 ± 115	44 ± 49	147 ± 173	258 ± 138	3.3 ± 1.1
Median (min.-max.)	40 (1–60)	78 (5–491)	115 (2–622)	26 (1–314)	92 (6–639)	33(6–264)	115(1–931)	256 (109–830)	3.2 (1.9–8.0)
No. of samples	38	38	38	38	38	38	33	38	38
<i>NNW</i>									
Mean ± st. dev.	13 ± 16	35 ± 26	10 ± 12	57 ± 70	38 ± 29	17 ± 13	24 ± 36	176 ± 49	3.0 ± 0.8
Median (min.-max.)	6 (1–90)	31 (3–96)	5 (0–59)	12 (1–255)	26 (1–101)	17(3–61)	8(1–174)	185 (43–266)	3.1 (0.9–4.6)
No. of samples	77	77	77	77	77	76	62	77	75

<sup>a</sup>St. dev., standard deviation; min., minimum; max., maximum; no., number.

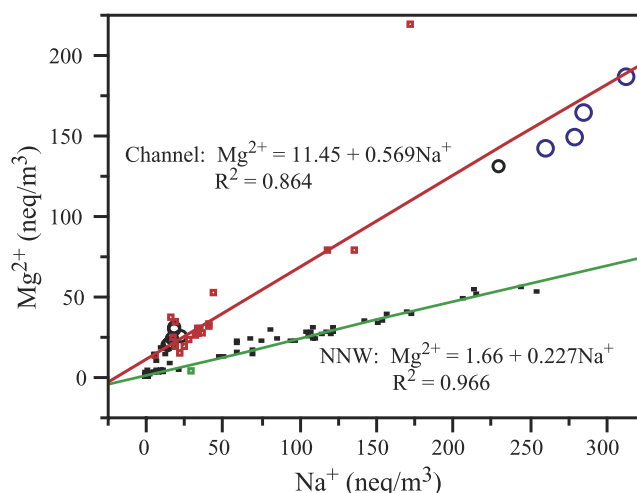
means in Channel than NNW except for Na<sup>+</sup>. The primary source of Na<sup>+</sup> in the DC-8 measurements is sea salt; hence there is little difference between the mean mixing ratios for these groups.

[16] As discussed by Keene *et al.* [1986], Na<sup>+</sup> and Mg<sup>2+</sup> are generally considered the best tracers of sea salt. However, this can become complicated when crustal sources add a significant amount to these species in coastal environments. Comparing the measured ratio of Mg<sup>2+</sup>/Na<sup>+</sup> to the equivalence ratio of 0.227 found in bulk seawater [Wilson, 1975; Keene *et al.*, 1986], one can determine which species to use as the reference sea-salt species. A ratio in excess of 0.227 indicates a crustal Mg<sup>2+</sup> influence, while a ratio less than this suggests a crustal Na<sup>+</sup> component. Here the data closely adheres to the sea-salt ratio for the NNW group (Figure 3). However, Mg<sup>2+</sup> is clearly enhanced in Channel, altering the slope significantly from what one would expect for sea salt (Figure 3). Thus for this study, Na<sup>+</sup> is used as the reference species for sea salt, since it appears less likely to suffer from major deviations from the marine source. However, it is known that Na<sup>+</sup> is a component of Asian dust [Song and Carmichael, 2001b]. Hence in calculating the sea-salt and non-sea-salt components of the aerosol species reported here, the sea-salt component will be somewhat overestimated, leading to an underestimate of non-sea-salt species for the Channel group.

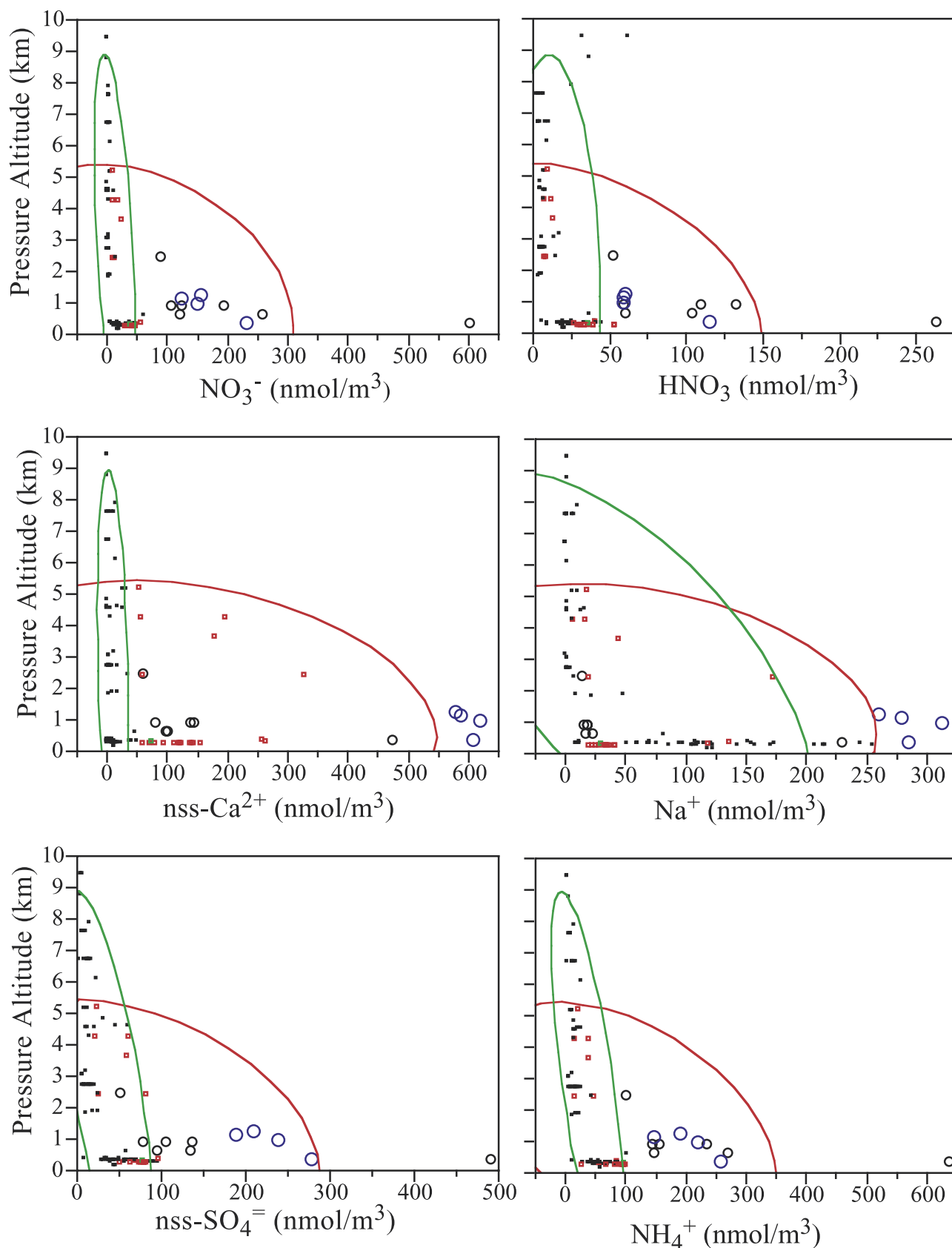
[17] The distribution of NO<sub>3</sub><sup>-</sup>, HNO<sub>3</sub>, nss-Ca<sup>2+</sup>, Na<sup>+</sup>, nss-SO<sub>4</sub><sup>=</sup>, and NH<sub>4</sub><sup>+</sup> as a function of altitude show that most species are enhanced at low altitudes (Figure 4). Density ellipses which enclose 90% of the points within each group show Channel mixing ratios enhanced compared to those of NNW. The enhancement of NO<sub>3</sub><sup>-</sup> may be due to three possible reaction routes: (1) uptake of HNO<sub>3</sub> by sea salt, (2) uptake of HNO<sub>3</sub> by alkaline dust, primarily by reaction with CaCO<sub>3</sub>, or (3) formation of fine mode NH<sub>4</sub>NO<sub>3</sub>. The first pathway may be eliminated since there is little difference in the availability of Na<sup>+</sup> between Channel and NNW (Figure 4). Further, several of the high NO<sub>3</sub><sup>-</sup> samples (circles in Figure 4) appear at low concentrations of Na<sup>+</sup>. Four of the high NO<sub>3</sub><sup>-</sup> samples (blue circles in Figure 4) do have high Na<sup>+</sup> concentrations; however, these samples also have the highest nss-Ca<sup>2+</sup> measured (Figure 4). This suggests the high Na<sup>+</sup> in these samples is due to crustal dust rather than to sea salt. All of the high NO<sub>3</sub><sup>-</sup> samples are also high nss-Ca<sup>2+</sup> samples. In addition to the high NO<sub>3</sub><sup>-</sup> markers (circles), there are many samples with enhanced dust (red squares in Figure 4). These samples do not have enhanced HNO<sub>3</sub>, which suggests that both HNO<sub>3</sub> and nss-Ca<sup>2+</sup> need

to be enhanced to yield enhanced NO<sub>3</sub><sup>-</sup> via uptake on dust. Another possible explanation for the enhanced NO<sub>3</sub><sup>-</sup> may be the formation of fine mode NH<sub>4</sub>NO<sub>3</sub> rather than coarse mode uptake of NO<sub>3</sub><sup>-</sup>. All of the enhanced NO<sub>3</sub><sup>-</sup> samples are observed to have enhanced NH<sub>4</sub><sup>+</sup> as well (Figure 4). Most of the markers also show enhanced nss-SO<sub>4</sub><sup>=</sup> (Figure 4). Hence both mechanisms 2 and 3 may be important.

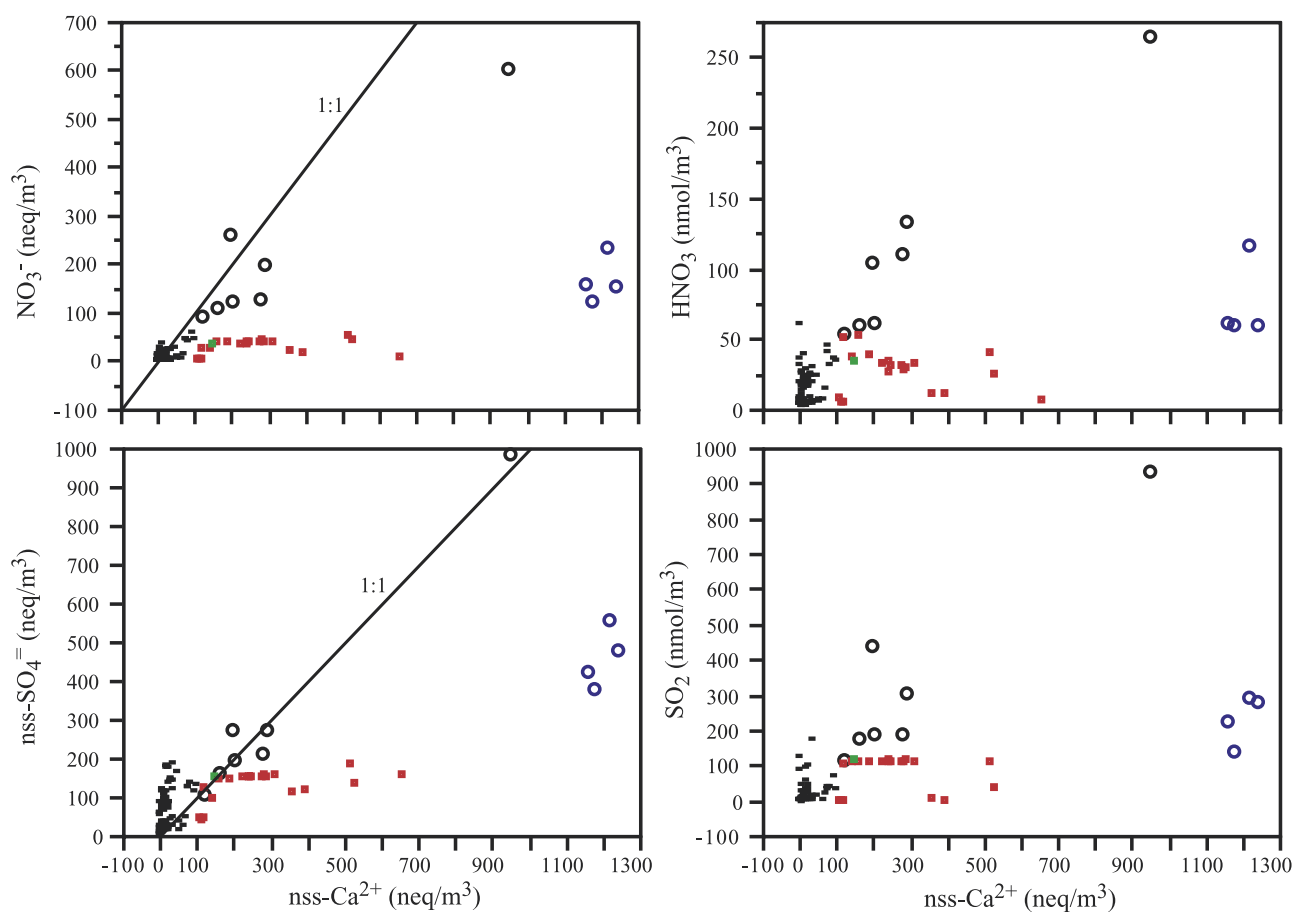
[18] Taking nss-Ca<sup>2+</sup> values >100 neq/m<sup>3</sup> to indicate enhanced dust, there are clearly two populations of NO<sub>3</sub><sup>-</sup> (Figure 5). Samples where NO<sub>3</sub><sup>-</sup> is  $\approx$  about 100 neq/m<sup>3</sup> (circles in Figure 5) tend to fall near the 1:1 line, although generally there is excess nss-Ca<sup>2+</sup> compared to NO<sub>3</sub><sup>-</sup>. The other population is denoted by red squares. For these samples, although nss-Ca<sup>2+</sup> indicates an abundance of dust, there is little appreciable increase in NO<sub>3</sub><sup>-</sup>. Note that the one green square indicates a sample that is likely to be predominantly sea salt, since it lies on the seawater slope shown in Figure 3. The blue circles in Figure 5 have NO<sub>3</sub><sup>-</sup> in excess of 100 neq/m<sup>3</sup>, yet they are well away from the 1:1 line. Given their very high amount of nss-Ca<sup>2+</sup>, it is possible that either the sample was obtained prior to the air mass reaching chemical equilibrium between the dust and HNO<sub>3</sub>, or there was insufficient HNO<sub>3</sub> to produce more particulate NO<sub>3</sub><sup>-</sup>. The correlation between the black circles and nss-Ca<sup>2+</sup> is



**Figure 3.** Mg<sup>2+</sup> versus Na<sup>+</sup>. Red linear fit to Channel data points; green fit to NNW data points. Circles indicate dust samples with enhanced NO<sub>3</sub><sup>-</sup> and nss-SO<sub>4</sub><sup>=</sup>. Red squares indicate dust samples without enhanced pollution species.



**Figure 4.** Aerosol and HNO<sub>3</sub> mixing ratios shown as a function of altitude. The density ellipses enclose 90% of the points within their respective groups, Channel (red ellipse) and NNW (green ellipse). Circles indicate dust samples with enhanced NO<sub>3</sub><sup>-</sup> and nss-SO<sub>4</sub><sup>-</sup>. Red squares indicate dust samples without enhanced pollution species.



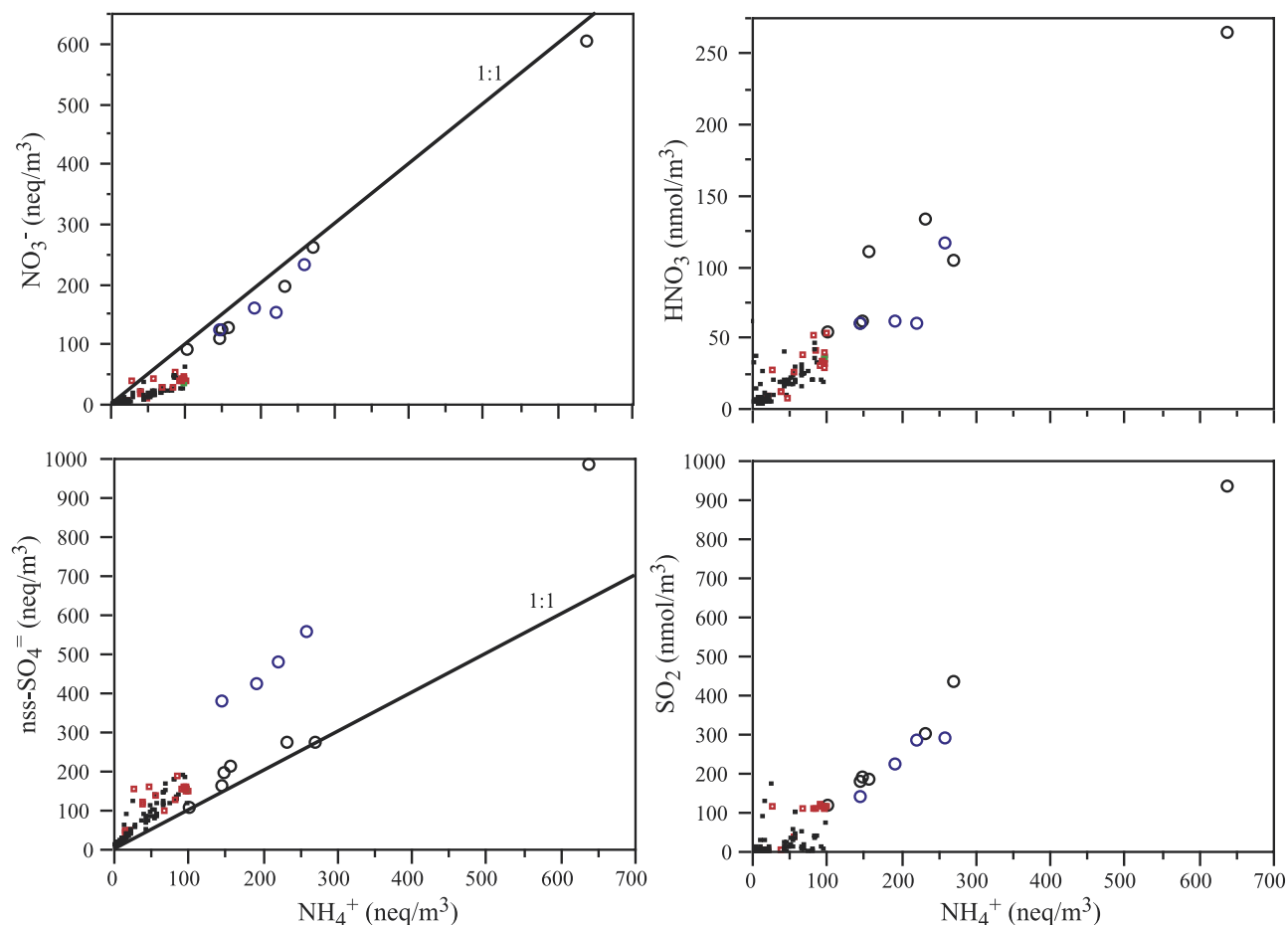
**Figure 5.**  $\text{NO}_3^-$ ,  $\text{nss-SO}_4^-$ ,  $\text{HNO}_3$ , and  $\text{SO}_2$  shown as a function of dust ( $\text{nss-Ca}^{2+}$ ). Circles indicate dust samples with enhanced  $\text{NO}_3^-$  and  $\text{nss-SO}_4^-$ . Red squares indicate dust samples without enhanced pollution species.

even closer to 1:1 for  $\text{nss-SO}_4^-$  (Figure 5) than for  $\text{NO}_3^-$ . Again, there is little increase in  $\text{nss-SO}_4^-$  with increasing  $\text{nss-Ca}^{2+}$  for the red square population. The blue circles are still set apart with respect to  $\text{nss-SO}_4^-$  and  $\text{nss-Ca}^{2+}$ .  $\text{HNO}_3$  and  $\text{SO}_2$  (Figure 5), suggest the circles reflect the presence of higher levels of gas phase pollution than is present for the red squares (or the non-dust-enhanced black squares). This suggests that only dust intermingled with a sufficient amount of gas phase pollution will show significant uptake of  $\text{NO}_3^-$  and  $\text{SO}_4^-$ . Note that where  $\text{nss-Ca}^{2+}$  is low, there are still appreciable amounts of  $\text{nss-SO}_4^-$ . Presumably, this  $\text{nss-SO}_4^-$  is due to uptake of  $\text{nss-SO}_4^-$  on sea salts (coarse mode) or the presence of fine mode  $\text{NH}_4\text{HSO}_4$  and  $(\text{NH}_4)_2\text{SO}_4$ . These results are similar to those reported by Choi *et al.* [2001]. They also reported cases in Seoul where some dust events were enhanced in pollution species, while other events were not.

[19] Next consider the possibility that the observed  $\text{NO}_3^-$  resides in fine mode  $\text{NH}_4\text{NO}_3$  particles. There is a stunning correlation between  $\text{NO}_3^-$  and  $\text{NH}_4^+$  (Figure 6), with the data points lying along the 1:1 line indicative of  $\text{NH}_4\text{NO}_3$ . There is some excess  $\text{NH}_4^+$  compared to the  $\text{NO}_3^-$ , but not much. This suggests that particulate  $\text{NO}_3^-$  is in fine rather than coarse mode. However, the relationship between  $\text{nss-SO}_4^-$  and  $\text{NH}_4^+$  (Figure 6) argues against this scenario. There is clearly more than enough  $\text{nss-SO}_4^-$  to accommodate all of

the  $\text{NH}_4^+$ . Given the preferential formation of  $\text{NH}_4\text{HSO}_4$  and  $(\text{NH}_4)_2\text{SO}_4$  to that of the more volatile  $\text{NH}_4\text{NO}_3$  for typical atmospheric temperatures and pressures, it seems unlikely that the bulk of  $\text{NH}_4^+$  is associated with  $\text{NO}_3^-$  in this region. The correlation observed here may be coincidental. There are very good correlations between  $\text{NH}_4^+$  and both  $\text{HNO}_3$  and  $\text{SO}_2$  (Figure 6) suggesting that  $\text{NH}_4^+$  is simply a good tracer for pollutants. Investigating this possibility further, there are excellent correlations between  $\text{CO}$  and  $\text{NH}_4^+$  ( $R^2 = 0.94$ ) and Ethyne/ $\text{CO}$  and  $\text{NH}_4^+$  ( $R^2 = 0.92$ ) for the dust sector data (Figure 7). Both of these,  $\text{CO}$  and Ethyne/ $\text{CO}$ , are used as tracers for pollution. Neither one plays any role in the formation of  $\text{NH}_4^+$ , which supports the assertion that the good correlation between  $\text{NH}_4^+$  and  $\text{NO}_3^-$  seen here is coincidental. Although modeling work by Song and Carmichael [2001a] suggests that  $\text{NH}_4\text{NO}_3$  may form along the “lower courses of the Huang River” (which underlies the path of the dust flows), there is evidence suggesting that  $\text{NO}_3^-$  is associated with the coarse mode. Wu and Okada [1994] found coarse  $\text{NO}_3^-$  in all of their samples collected in Nagoya, Japan, including on the surface of dust particles during a Kosa event. Further, Kim and Park [2001] comparing samples during a dust storm and nondust periods in Seoul, Korea, found that the maximum concentration of  $\text{NH}_4^+$  was in the fine mode ( $<2 \mu\text{m}$  diameter) under both conditions. Meanwhile, both  $\text{SO}_4^-$  and  $\text{NO}_3^-$  were enhanced





**Figure 6.**  $\text{NO}_3^-$ ,  $\text{nss-SO}_4^-$ ,  $\text{HNO}_3$ , and  $\text{SO}_2$  shown as a function of  $\text{NH}_4^+$ . Circles indicate dust samples with enhanced  $\text{NO}_3^-$  and  $\text{nss-SO}_4^-$ . Red squares indicate dust samples without enhanced pollution species.

during the dust storm in the coarse fraction and were well correlated with coarse mode  $\text{Ca}^{2+}$ . In plots of *Kim and Park's* [2001] size-resolved data, there is no evidence of a fine mode peak in  $\text{NO}_3^-$ . In the absence of size-resolved aerosol chemical measurements during TRACE-P, these observations argue against  $\text{SO}_4^-$  and  $\text{NO}_3^-$  being primarily associated with  $\text{NH}_4^+$  in the presence of dust.

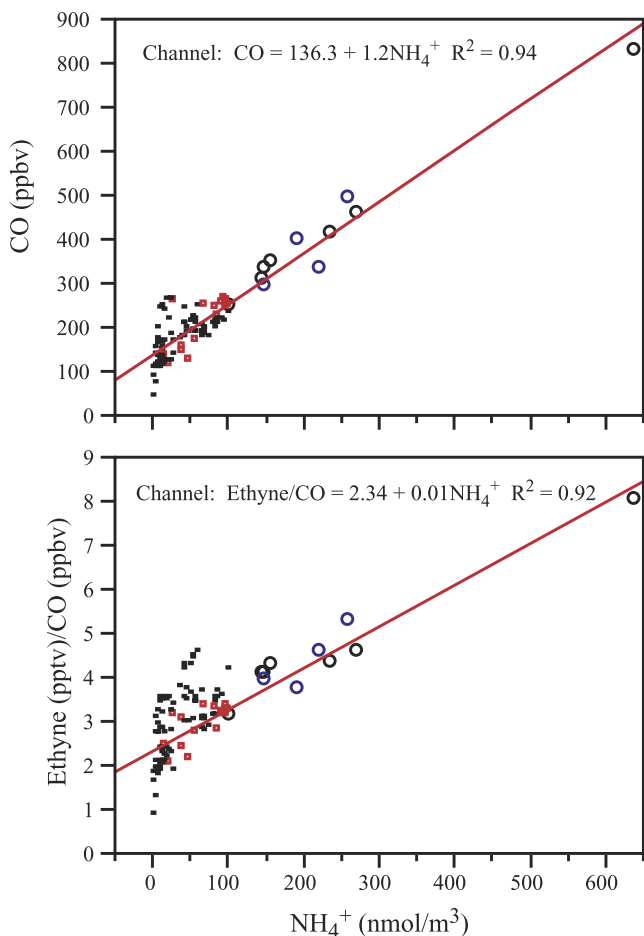
[20] To estimate how much  $\text{nss-SO}_4^-$  remains available for uptake on  $\text{nss-Ca}^{2+}$ , assume all particulate  $\text{NH}_4^+$  is in the form of  $(\text{NH}_4)_2\text{SO}_4$ . Then, measured  $\text{NH}_4^+$  equivalence is subtracted from measured  $\text{nss-SO}_4^-$  equivalence to yield excess  $\text{nss-SO}_4^-$  (Table 2). Further,  $\text{NO}_3^-$  is added to this excess  $\text{nss-SO}_4^-$ , and the sum is compared to  $\text{nss-Ca}^{2+}$  (Table 2). On average, 57% of the available  $\text{nss-SO}_4^-$  can be taken up by  $\text{NH}_4^+$ , with the remainder available for uptake on dust and sea-salt surfaces. Even including  $\text{NO}_3^-$ , there is ample  $\text{nss-Ca}^{2+}$  to take up the measured  $\text{NO}_3^-$  and excess  $\text{nss-SO}_4^-$ . Indeed, even if there were no  $\text{NH}_4^+$ , the available  $\text{nss-SO}_4^-$  and  $\text{NO}_3^-$  would only react with 76% of the available  $\text{nss-Ca}^{2+}$ .

[21] This suggests the uptake of  $\text{nss-SO}_4^-$  and  $\text{NO}_3^-$ , in the heavily dust-impacted Channel samples, is limited by the amount of  $\text{HNO}_3$  and  $\text{SO}_2$  present, not by dust. Although, it is possible that all the available  $\text{Ca}^{2+}$  on the surface of the particles is associated with  $\text{NO}_3^-$  and  $\text{SO}_4^-$ , while the

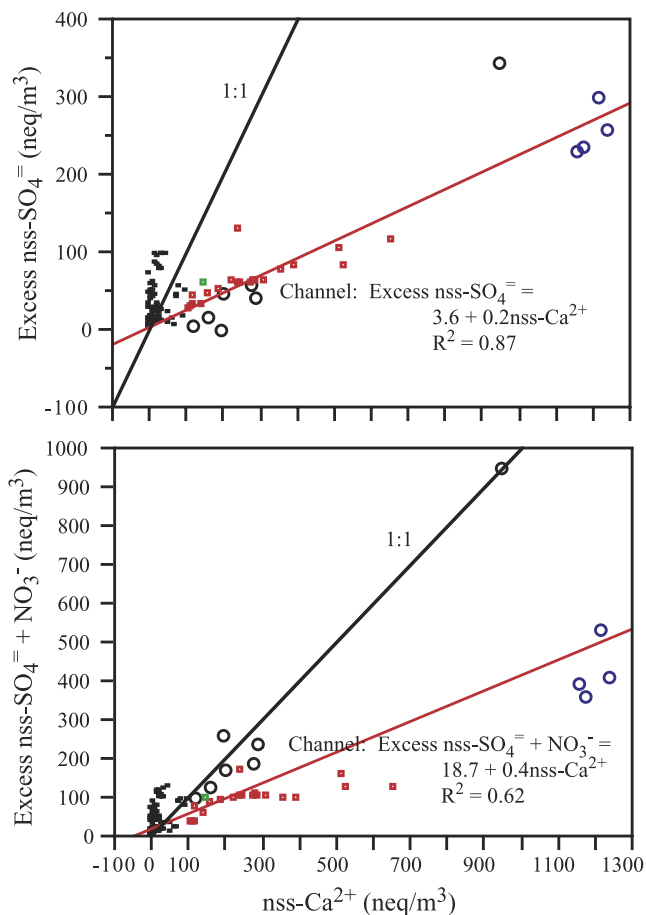
remaining  $\text{Ca}^{2+}$  is in the core of the particles where it cannot react with external gas phase species, this scenario is unlikely given the study by *Goodman et al.* [2000]. They found that in the presence of water vapor, the reaction of  $\text{HNO}_3$  can proceed into the bulk of the particles driving off all of the  $\text{CO}_3^-$ . This makes all of the observed  $\text{Ca}^{2+}$  potentially available for reaction. Given this argument, the uptake observed during TRACE-P was probably limited by the availability of acidic gases, rather than dust.

[22] In comparison, in the NNW sector where dust is not so prevalent, there is insufficient  $\text{nss-Ca}^{2+}$  available to take up the available excess  $\text{nss-SO}_4^-$  and  $\text{NO}_3^-$  (Table 2). Here these species are more likely to be associated with sea salt and are present at much lower levels than observed in the dust sector. To further illustrate this, Figure 8 shows excess  $\text{nss-SO}_4^-$  versus  $\text{nss-Ca}^{2+}$ . If there was sufficient  $\text{nss-SO}_4^-$  to react with all of the  $\text{nss-Ca}^{2+}$  present, the data would fall along the 1:1 line. In fact, where dust is not enhanced, there is more  $\text{nss-SO}_4^-$  than can be accounted for by dust (black squares in Figure 8). This suggests this  $\text{SO}_4^-$  is associated with fine mode  $\text{NH}_4^+$  or perhaps with coarse mode sea salt. However, where there is plenty of dust (circles and red squares in Figure 8), excess  $\text{nss-SO}_4^-$  is well correlated with  $\text{nss-Ca}^{2+}$  ( $R^2 = 0.87$ ) with a slope of 0.2. Further, when  $\text{NO}_3^-$  is included in this analysis, the most polluted samples





**Figure 7.** CO and Ethyne/CO versus  $\text{NH}_4^+$ . All data from NNW and Channel are plotted, but only those from Channel are used for the linear fits. Circles indicate dust samples with enhanced  $\text{NO}_3^-$  and  $\text{nss-SO}_4^{2-}$ . Red squares indicate dust samples without enhanced pollution species.



**Figure 8.** Excess  $\text{nss-SO}_4^{2-}$  and excess  $\text{nss-SO}_4^{2-} + \text{NO}_3^-$  versus  $\text{nss-Ca}^{2+}$  show how much of these species are available for uptake by dust after allowing  $\text{NH}_4^+$  to take up as much  $\text{nss-SO}_4^{2-}$  as possible. Circles indicate dust samples with enhanced  $\text{NO}_3^-$  and  $\text{nss-SO}_4^{2-}$ . Red squares indicate dust samples without enhanced pollution species.

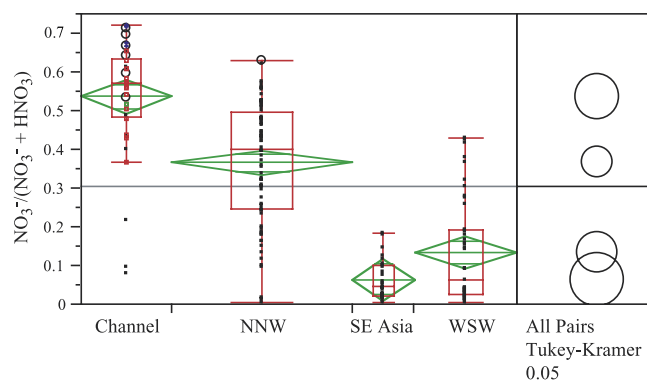
are seen to lie along the 1:1 line (Figure 8), suggesting most of the  $\text{CO}_3^{2-}$  in the dust has been displaced. For the remainder of the dust enhanced samples, the uptake appears to be limited by the availability of  $\text{HNO}_3$  and  $\text{SO}_2$ .

[23] This exercise probably underestimates the amount of excess  $\text{nss-SO}_4^{2-}$ . As noted earlier, the non-sea-salt fractions have been underestimated, since some component of the  $\text{Na}^+$  present is due to dust, even though it is all assumed to

be sea salt. Thus the total amount of species in non-sea-salt fractions is somewhat higher than that shown here. Another issue to bear in mind comes from a study of individual particle composition. *Gao and Anderson* [2001] reported that fly ash particles (distinguishable from dust by their spherical rather than irregular shape) also show aggregation with  $\text{CaSO}_4$ . This is attributed to the use of  $\text{CaCO}_3$  to scrub  $\text{SO}_2$  from stack emissions. This can confound the interpre-

**Table 2.** Means, Standard Deviations, Medians, Minima, Maxima, and Number of Samples for Excess  $\text{nss-SO}_4^{2-}$  and  $\text{NO}_3^-$  Compared to Available Dust

	$\text{nss-SO}_4^{2-}$ , $\text{neq/m}^3$	$\text{NH}_4^+$ , $\text{neq/m}^3$	Excess $\text{nss-SO}_4^{2-}$ , $\text{neq/m}^3$	$\text{NO}_3^-$ , $\text{neq/m}^3$	Excess $\text{nss-SO}_4^{2-} + \text{NO}_3^-$ , $\text{neq/m}^3$	$\text{nss-Ca}^{2+}$ , $\text{neq/m}^3$
<i>Channel</i>						
Mean $\pm$ st. dev.	$182 \pm 183$	$104 \pm 115$	$78 \pm 84$	$74 \pm 110$	$152 \pm 178$	$335 \pm 353$
Median (min.-max.)	156 (11–982)	92 (6–639)	58 (0–343)	40 (1–60)	103 (6–945)	229 (4–1245)
No. of samples	38	38	38	38	38	38
<i>NNW</i>						
Mean $\pm$ st. dev.	$70 \pm 52$	$38 \pm 29$	$32 \pm 28$	$13 \pm 16$	$45 \pm 37$	$19 \pm 24$
Median (min.-max.)	62 (6–191)	26 (1–101)	23 (3–98)	6 (1–90)	39 (3–128)	12 (0–119)
No. of samples	77	77	77	77	77	74



**Figure 9.** Comparison of  $\text{NO}_3^-$  partitioning between the dust sector (Channel), a sector with lots of sea salt (NNW), and two sectors where the influence of dust and sea salt are minimal (SE Asia and WSW). Green diamonds show means (center horizontal line) with the 95% confidence interval indicated by the upper and lower points of the diamonds, while the width of the diamond is proportional to the group size. Red rectangles show quantiles, with upper and lower bars indicating the 90th and 10th percentiles, respectively, top and bottom of box are at the 75th and 25th percentiles, respectively, and the red line in the box is the median. Circles on the right indicate whether the means are statistically different. The center of each circle is aligned with the mean of its respective group; the diameter of the circle spans the 95% confidence level. When the circles do not overlap, the means are significantly different. When they do overlap, the means may not be significantly different.

tation of non-size-resolved aerosol chemistry, because there will be a component of the  $\text{nss-Ca}^{2+}$  in the fine fraction which is not related to dust. However, in the presence of large dust outbreaks it is expected that this contribution to the total  $\text{nss-Ca}^{2+}$  will be small.

[24] Finally, the partitioning between particulate  $\text{NO}_3^-$  ( $\text{p-NO}_3^-$ ) and gas phase  $\text{HNO}_3$  is shown for dust (Channel) versus nondust (NNW) sectors (Figure 9). For comparison the partitioning is also shown for the other two sectors studied during TRACE-P, SE Asia and WSW. The samples from these sectors have minimal sea salt and dust influences (see Jordan *et al.* [2003] for a complete description of these sectors). Here we see that while the means for SE Asia and WSW may not be significantly different from each other, they are both significantly different from the other two, which are also significantly different from each other (Figure 9). For the groups least affected by the presence of sea salt and dust (SE Asia and WSW), the amount  $\text{p-NO}_3^-$  accounts for is only 6–13% of the total  $\text{NO}_3^-$  ( $\text{t-NO}_3^- = \text{p-NO}_3^- + \text{HNO}_3$ ) present, on average (Table 3). Where dust is not a dominant component of the atmosphere (NNW),  $\text{p-NO}_3^-$  accounts for 37% of the  $\text{t-NO}_3^-$  on average, probably due to the influence of sea salts in this sector. For the dust sector,  $\text{p-NO}_3^-$  makes up 54% of the total on average, with maximum values of 72% observed (Table 3). These numbers agree well with those predicted by Song and Carmichael [2001a] where 10–50% of  $\text{t-NO}_3^-$  would be particulate in the presence of dust and sea salts, with >70% in dust and sea salt plume centers in

the boundary layer, while in the free troposphere, 10–30% would be  $\text{p-NO}_3^-$  on sea salts and >50% in dust plume centers.

#### 4. Summary

[25] Dust storms are prevalent in East Asia, with the dust being carried downwind along well-defined routes [Sun *et al.*, 2001]. This dust starts out with very little  $\text{NO}_3^-$  or  $\text{SO}_4^{2-}$  in the dust grains [Nishikawa *et al.*, 1991; and Zhang and Iwasaka, 1999]. However, in the presence of pollution, these dust particles can take up  $\text{NO}_3^-$  and  $\text{SO}_4^{2-}$ , altering both the partitioning between particulate and gas phases of these species, as well as their size distributions in the atmosphere (especially,  $\text{SO}_4^{2-}$ ). During the TRACE-P mission, dust was found to be well confined to a particular sector emanating from Asia in good agreement with the routes described by Sun *et al.* [2001].

[26] This dust sector also contained the highest mixing ratios of pollutant species observed, with gas-phase  $\text{HNO}_3$  2.7 times higher on average than in the adjacent nondust sector. Similarly,  $\text{SO}_2$ , CO, and the ratio of Ethyne/CO were 6.2, 1.5, and 1.1 times higher in the dust samples. Sea salt could not account for the enhancements in  $\text{NO}_3^-$  and  $\text{SO}_4^{2-}$  (factors of 5.7 and 2.6, respectively), since the mean mixing ratios of  $\text{Na}^+$  in the dust and nondust regions were comparable (64 and 57  $\text{nmol/m}^3$ , respectively). Although a good correlation between  $\text{NO}_3^-$  and  $\text{NH}_4^+$  was observed, it appears that this was coincidental. As a tracer of anthropogenic species,  $\text{NH}_4^+$  also exhibited excellent correlations with CO and the Ethyne/CO ratio (0.94 and 0.92, respectively). These species are not involved in any way with  $\text{NH}_4^+$  formation. Prior observations in East Asia where size-resolved data was available did not show fine mode peaks of  $\text{NO}_3^-$  when dust was present. Further, calculations showed that there is ample  $\text{nss-SO}_4^{2-}$  to occupy the available  $\text{NH}_4^+$  and still have enough left over to react with dust.

[27] During TRACE-P, the uptake of  $\text{NO}_3^-$  and  $\text{SO}_4^{2-}$  appeared to be limited by the availability of gas phase species. Where there is ample  $\text{HNO}_3$  and  $\text{SO}_2$  along with dust particles, the uptake of  $\text{NO}_3^-$  and  $\text{SO}_4^{2-}$  can be substantial and in some cases may entirely drive off the  $\text{CO}_3^{2-}$  in the dust aerosol. The partitioning between particulate and gas-phase  $\text{NO}_3^-$  shows 54% of the total  $\text{NO}_3^-$  ( $\text{t-NO}_3^-$ ) in the dust sector is in particulate ( $\text{p-NO}_3^-$ ) phase on average. In some samples,  $\text{p-NO}_3^-$  exceeded 70%. This is in good agreement with model predictions [Song and Carmichael, 2001a]. In the nondust sector,  $\text{p-NO}_3^-$  constituted 37% of  $\text{t-NO}_3^-$ , likely due to the heavy influence of sea salt. Two other sectors with little influence of dust or sea salt had  $\text{p-NO}_3^-$  contributing a small fraction to the total, <15%.

[28] These results provide evidence of in situ uptake of acidic gases on alkaline dust particles in support of model predictions and laboratory studies. This uptake may result in

**Table 3.** Means, Standard Deviations, Medians, Minima, Maxima, and Number of Samples for Nitrate Partition Comparison  $\text{NO}_3^-/(\text{NO}_3^- + \text{HNO}_3)$

NNW	Channel	WSW	SE Asia
$0.37 \pm 0.16$	$0.54 \pm 0.15$	$0.13 \pm 0.13$	$0.06 \pm 0.05$
0.40 (0.00–0.63)	0.57 (0.08–0.72)	0.06 (0.00–0.43)	0.04 (0.00–0.18)
76	38	43	26

enhanced removal of nitrogen from the atmosphere, which may be important on regional scales. The presence of  $\text{NO}_3^-$  and  $\text{SO}_4^-$  on coarse particles may lead to enhanced acid deposition along with its detrimental effects to terrestrial ecosystems than would be observed in regions without dust outbreaks. Enhanced deposition of  $\text{NO}_3^-$  to marine ecosystems may make more of this limiting nutrient available to the coastal biota. These effects may be magnified, if emissions of pollutant N and S increase with further industrialization in East Asia.

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