

Rainwater chemistry at a high-altitude station, Mt. Lulin, Taiwan: Comparison with a background station, Mt. Fuji

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[1] Samples of rainwater were collected at a high-altitude site in east Asia between April 2003 and May 2005. The volume weighted mean pH value for the whole sampling period was 5.12, approaching that of typical natural water. Non-sea-salt (nss) SO_4^{2-} and NH_4^+ were the most abundant anion and cation, respectively, both existing mostly in the form of (NH₄)₂SO₄. Chloride was excessive in most of the samples. The signature of biomass burning in south and Southeast Asia was evident in the Mt. Lulin samples. Concentrations of chemical species were found to be elevated in the spring months, owing to the emissions from south/Southeast Asia and peak biomass burning activities and frequent dust storms (in the Indian Thar Desert). In the summer and fall seasons our samples are mainly influenced by marine air masses. The episodic species concentrations measured at the summit of Mt. Fuji during the spring were due to the influence of volcanic emissions from Miyake-Jima. Tropical cyclones (TC) over the western Pacific region and deep convections play important roles in the transport of boundary layer pollutants to the free troposphere, although their influence is not frequently detected at Mt. Lulin and Mt. Fuji. The rainwater chemistry at Mt. Lulin and Mt. Fuji were examined together with the aerosol chemistry obtained from the TRACE-P and ACE-Asia campaigns. The analysis of the Mt. Lulin data set demonstrates its background characteristics of the rainwater chemistry in east Asia.

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Introduction 1.

[2] The acid rain problem is a global environmental issue [Rodhe et al., 2002] because of its transboundary impact on the biogeochemical cycles. The increasing SO₂ and NO_x emission trends during the past 2 or 3 decades have intensified the acid deposition problem in Asia [Wai et al., 2005]. However, relatively few studies of the rainwater chemistry at low altitude have been conducted in east Asia and reported internationally [Chang et al., 1997; Tanner, 1999; Lin et al., 1999; Lee et al., 2000; Fujita et al., 2000; Seto and Hara, 2006]. The Acid Deposition Monitoring Network in East Asia (EANET) (http://www.eanet.cc/) is the only regional network in east Asia that is organized for continuous operation in multiple countries and only a few detailed analyses of this data set have been published [e.g., Wai et al., 2005]. There is even less understanding of the precipitation chemistry at high altitude in Asia. In contrast, acidic precipitation at high elevation has received much attention in the United States [Baron and Denning, 1993; Heuer et al., 2000]. Acidic fog and cloud water are thought to contribute to the decline of forests at high elevations

[Blank, 1985; Lin and Saxena, 1991; Kim and Aneja, 1992]. Detailed analysis and discussion of the rainwater chemistry at high altitude/free troposphere (e.g., at the summit of Mt. Fuji, Japan) in east Asia has not been reported until recently [Watanabe et al., 2006].

[3] As elsewhere in the world, SO_2 and NO_x are the major precursors of acid rain in east Asia. However, the mineral dust from Asian dust storms, which is transported by the prevailing westerlies in the spring [e.g., Zhang et al., 1997], can increase the pH value of rainwater at the downwind areas [Wang et al., 2002]. In addition, the impact of aerosols on rainwater chemistry due to the activities of biomass burning in Southeast Asia and the consumption of biofuel in China [e.g., Ma et al., 2003], although reported, has been relatively less studied [Balasubramanian et al., 1999, and references therein]. More recently, both types of aerosols have been extensively investigated in the Asian region by both aircraft and ground-based measurements, i.e., Transport and Chemical Evolution Over the Pacific (TRACE-P) [Jacob et al., 2003] and the Aerosol Characterization Experiments-Asia (ACE-Asia) [Huebert et al., 2003]. Valuable data sets were published for aerosol chemistry and physics, but no further comparison was made with precipitation chemistry at higher altitudes.

[4] In view of the unique environmental settings and the extremely scarce data set that is available at high elevations within the east Asian region, we conducted a field study at Mt. Lulin, Taiwan. We did this to characterize the back-

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Figure 1. Location of Mt. Lulin and Mt. Fuji.

ground rainwater chemistry in the free troposphere over east Asia and also to relate it to, if existing, the aerosol chemistry obtained during TRACE-P and ACE-Asia. To achieve this goal, we analyzed the data sets of Mt. Lulin and Mt. Fuji and then made a comparison of the rainwater chemistry at both sites. Finally, we compared the background rainwater chemistry at Mt. Lulin with that of other remote and highaltitude sites. It is worth noting here that the Mt. Fuji station, which is regarded as a background station in the free troposphere for northeast Asia, has been shut down since 2004 after 72 years of meteorological operation. Our present work is part of a pilot study for evaluating if the Mt. Lulin site is ready to extend its capacity in order to become a regional background station for the free tropospheric chemistry and for the study of the long-range transport of Asian pollutants associated with continental outflows.

2. Methodology

2.1. Sampling Sites

[5] Figure 1 shows the location of two sites, one at Mt. Lulin in Taiwan and the other at Mt. Fuji in Japan. Our site is located at the isolated peak of Mt. Lulin (23.51°N, 120.92°E; 2862 m AMSL), which is part of the Central Mountain Range and is located inside Jade Mountain National Park. Its high elevation assures it to be generally free from local pollution. Surrounded by other mountainous areas of relatively low altitude, only Mt. Jade (3954 m AMSL) situated to the northeast and several kilometers away, is higher than our site. Our site frequently lies in the free troposphere (especially during the winter months) and is generally free from boundary layer pollution. Since altostratus clouds in the middle layer of the troposphere alone are unlikely to account for the amounts of rainfall as recorded in our samples, other processes that lead to precipitation should also be considered, i.e., orographic precipitation and deep convections (in the summer months). Chemical species in the free troposphere are then scavenged by clouds produced by the above mentioned processes.

Meanwhile, these processes, as well as the frontal (occurring frequently in winter and spring of Taiwan) mechanisms have been reported to be able to transport pollutants from the boundary layer to the free troposphere [*Lelieveld and Detener*, 2000; *Donnell et al.*, 2001]. In view of the precipitation scavenging and the pollution transport processes at our site, our samples could be influenced by the chemical signature within both the planetary boundary layer and the free troposphere.

[6] Mt. Fuji (35.4°N, 138.7°E; 3776 m AMSL) is the highest mountain in Japan. Its summit is positioned in the free troposphere for most part of the year, serving as a suitable platform for observations of background atmospheric chemistry. It has a nice conical profile, and its summit is relatively free of mountain and valley winds. Long-range transport of pollutants from the Asian continent attributable to westerly winds in the free troposphere can be assessed at the summit. A detailed description of Mt. Fuji station can be found elsewhere [*Igarashi et al.*, 2004].

2.2. Sampling and Analysis

[7] Sampling procedures were similar to those in the work of Lin et al. [1999]. Weekly samples were collected by a wet-only collector every Tuesday similar to the weekly samplings of the U.S. National Atmospheric Deposition Program. Samples were collected during the period from April 2003 to May 2005. Each sample was labeled with the date of collection (see the dates on the abscissa in Figure 4). Field-sampling buckets and laboratory-used polyethylene sampling bottles were all rinsed by deionized water prior to use. The rainwater sample was first filtered through a 0.45 μ m pore size mixed cellulose ester filter and then collected directly into the sampling bottle. The bottle was then sealed and kept in a refrigerator at 4°C prior to chemical analysis. The pH and conductivity of the rainwater sample was measured by a coupled pH/conductivity meter (Mettler Toledo, Model MPC 227) with the check reference of pH 4.0 and 7.0 and KCl solution, respectively. The species concentrations were measured by ion chromatography (Dionex Model DX-100). The analytical columns CS-12A 4 mm (with self-regenerating suppresser and eluent of 18 mM MSA at a flow rate of 0.9 mL min⁻¹) and AS012A 4 mm (with self-regenerating suppresser and eluent of 2.1 mM Na₂CO₃ + 0.8 mM NaHCO₃ at a flow rate of 1.6 mL min⁻¹) were used for cations and anions, respectively. Since no direct measurement of HCO_3^- was made, it was estimated based on the pH level [World Meteorological Organization (WMO), 1994].

[8] Laboratory quality assurance and quality control (QA/QC) included the routine running of blanks and control samples, as well as replicate samples. They were based on the protocols of the EPA NIEA-PA101, NIEA-PA104, and NIEA-PA105. External calibration was made using standard stock solutions and the r^2 of linear regression for each ion species was constrained to be >0.995. All tests were taken for every 15 samples. The lowest control limit was defined as 3 standard deviations (σ) of multiple analysis, while 6 σ was adopted for very low concentrations as by *Winiwarter et al.* [1998]. The detection limits for cations (NH₄⁴, Na⁺, K⁺, Mg²⁺, Ca²⁺) and anions (Cl⁻, NO₃⁻, SO₄²⁻), for instance, were 0.92, 0.38, 0.36, 0.64, 1.05 μ eq l^{-1} and 0.33, 0.50, 0.58 μ eq l^{-1} , respectively.



Figure 2. Relationship between Cl⁻ (μ eq l^{-1}) and Na⁺ (μ eq l^{-1}) in (a) Mt. Lulin and (b) Mt. Fuji.

[9] In view of the possibility that only trace quantities of species concentrations existed in the rainwater samples, the Global Atmospheric Watch (GAW) ion balance criteria [*WMO*, 1994] was adopted as the first data quality check of our samples. The well-known ion balance, defined as the ratio of the equivalent concentrations of total anions to total cations was adopted (1.00 ± 0.25 here) as the second measure. The samples were rejected only when the above two criterion were not satisfied. About 72% of the data

(39 samples) were retained after the data quality check was performed. The comparison between theoretically calculated and measured conductivities of rainwater samples was further made, based on *WMO* [1994] criteria, although few outliers were found that did not substantially affect the above results.

[10] HYSPLIT-4 (Hybrid Single-Particle Lagrangian Integrated Trajectory-Version 4) model developed by NOAA Air Resources Laboratory (http://www.arl.noaa.gov/ready/ hysplit4.html) was applied in the present study to determine the transport of air parcels at Mt. Lulin and Mt. Fuji. The settings of the model are mentioned in the sections that follow and elsewhere [*Wai and Tanner*, 2004]. Surface weather maps from the Japanese Meteorological Agency (JMA) and satellite images from GMS-5 were adopted whenever appropriate.

3. Results and Discussion

3.1. Rainwater Chemistry at Mt. Lulin

[11] Table 1 shows the volume-weighted mean (VWM) species concentrations for the entire sampling period. The pH value over the entire period was 5.12 (volume-weighted H⁺ concentration = 7.6 μ eq l^{-1}), which approaches the pH value of typical natural rainwater of 5.6. Acidic and alkaline species nss-SO₄²⁻ and NH₄⁺ (VWM concentrations: 15.3 and 12.3 μ eq l^{-1} , respectively) were the most abundant anions (24% of the total concentration) and cations (19% of the total concentration), followed by NO₃⁻ (VWM concentration: 10.8 μ eq l^{-1} ; 17% of the total concentration). This more or less explains the low acidity of the rainwater chemistry at the monitoring site, since the strong acidic anions are neutralized by alkaline cations. Sea-salt species concentrations were low, due to the high altitude of the site.

[12] Despite a relatively high correlation (R = 0.76, p < 0.001, N = 39) between Cl⁻ and Na⁺ concentrations (Figure 2a), the ratio of Cl⁻/Na⁺ of 1.65 (compared to the seawater ratio 1.16) suggests excessive Cl⁻ in most of the samples. Only five of the samples had ratios similar to the sea-salt value. Excessive Cl⁻ was also reported in the snow samples collected at high-altitude sites (\geq 3000 m) in the Alps [*Winiwarter et al.*, 1998] and the Himalayas [*Shrestha et al.*, 1997]. It is worth noting that the Cl⁻/Na⁺ ratio reported in several rainwater chemistry studies near the surface level was similar to that of seawater [*Wai et al.*, 2005, and references therein]. It is well-known that HCl is volatilized after the reaction of strong mineral acids with sea-salt aerosols. However, the scavenging of HCl can lead to a Cl⁻/Na⁺ ratio similar to that of seawater and cannot

Table 1. Species Concentrations, $\mu eq l^{-1}$, Measured at Mt. Lulin^a

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	Ν	Rainfall	pН	Cl-	NO_3^-	SO_4^{2-}	nss- SO4 ²⁻	H^+	$\mathrm{NH_4}^+$	Na ⁺	K^+	Mg^{2+}	Ca ²⁺
					Apr 20	03 to May	2005						
VWM	39	-	5.12	7.2	10.8	15.7	15.3	7.6	12.3	2.9	1.4	1.7	3.9
					Seasonal V	WM Conce	entration						
Spring (Mar to May)	14	52.7	5.11	5.8	13.1	17.5	17.2	7.8	18.9	2.3	1.7	2.2	5.9
Summer (Jun to Aug)	15	61.2	5.15	9.5	9.7	15.0	14.5	7.0	9.6	3.9	1.2	2.2	3.8
Fall (Sep to Nov)	7	65.7	5.24	5.1	8.3	11.5	11.2	5.7	4.8	2.7	0.9	0.9	1.4
Winter (Dec to Feb)	3	91.7	4.91	6.4	12.2	20.1	20.0	12.4	16.1	1.5	1.8	0.0	3.5

^aRainfall shown as per sample basis, in units of mm. Values of pH are volume weighted.

Table 2. Principal Factor Loadings of Varimax Rotation ObtainedFrom Rainwater Samples at Mt. Lulin During 2003–2005^a

Chemical Ions	Factor I	Factor II	Factor III
Cl ⁻	0.24	-0.04	0.94
NO_3^-	0.91	0.12	0.30
nss-SO42-	0.89	0.25	0.31
H^+	0.12	0.97	-0.09
NH4 ⁺	0.89	0.19	0.26
Na ⁺	0.50	-0.15	0.76
K ⁺	0.70	-0.01	0.40
Mg ²⁺	0.77	-0.08	0.23
Ca ²⁺	0.87	-0.10	0.23
Percent of variance	63.41	14.33	7.43
Eigenvalue	5.71	1.29	0.67

^aFactor loadings exceeding 0.7 are bold-faced.

satisfactorily explain the excess of Cl^- in our samples. A possible explanation to enrich the Cl^- concentration in rainwater sample is the reaction of O_3 at sea-salt aerosol surfaces, generating Cl_2 , followed by rapid photochemical conversion of Cl_2 to HCl which is then eventually scavenged by cloud and rain droplets [*Talbot et al.*, 1992, and reference therein]. Such conversion of Cl_2 to HCl is enhanced by intense photochemical processes aloft in the air rather than at surface level. The emission of HCl from volcanoes [*Orlando and Schauffler*, 1999] may also contribute to the Cl^- enrichment.

[13] Throughout the entire sampling period there was a high correlation coefficient (R = 0.92, p < 0.001, N = 34) between nss-SO₄²⁻ and NO₃⁻ concentrations. This suggests that they have a common transport mechanism, rather than there being a reaction between their precursors [*Lee*, 1993; *Báez et al.*, 1997]. All samples have similar ratio of nss-SO₄²⁻/NO₃⁻ (1.40). This ratio is also comparable to the value in remote Arctic regions (1.16 [*Talbot et al.*, 1992]). In contrast, this ratio was observed to be 2.29 in Taipei, the capital of Taiwan [*Lin et al.*, 1999]. The excessive nss-SO₄²⁻ was attributed to the long-range transport of sulfur compounds. The species of Mg²⁺ was correlated (R = 0.77, p < 0.001, N = 39) with Ca²⁺, suggesting its nonmarine and crustal origin. Only four samples with higher Mg²⁺ but low Ca²⁺ concentrations indicated a marine origin.

[14] Species with high correlations also included NH₄⁺ and nss- SO_4^{2-} (R = 0.87, p < 0.001, N = 39; NH₄⁺/nss- SO_4^{2-} = 1.06), suggesting that NH_4^+ and $nss-SO_4^{2-}$ existed in the form of (NH₄)₂SO₄, due to complete neutralization of H_2SO_4 and NH_3 . The NH_4^+ - NO_3^- pair also had a high correlation between them (R = 0.86, p < 0.001, N = 39), indicating that NH₄NO₃ could coexist with (NH₄)₂SO₄. The species K^+ was correlated with NH_4^+ (R = 0.77, p < 0.001, N = 31) and NO_3^- (R = 0.74, p < 0.001, N = 31), suggesting that K⁺ mainly came from the biomass burning. NH₃ emissions from biomass burning were reported [Hegg et al., 1988]. Ma et al. [2003] reported that K⁺ highly correlated (r > 0.95) with NH₄⁺ and NO₃⁻ in the plumes dominated by biomass burning in the nearby region (20°N, 120-130°E; >2000 m AMSL) during the TRACE-P campaign.

[15] Factor analysis of Varimax rotation was further carried out to assess the possible sources of rainwater contamination. Table 2 shows that three factors are identified and the loadings exceeding 0.7 are highlighted. Factors I and II accounted for 63 and 14% of total variance, respectively. The former is a mixing factor including the anthropogenic species (nss-SO₄²⁻, NO₃⁻), natural decomposition/biomass burning species (NH₄⁺ and K⁺) and soil/dust species (Mg²⁺ and Ca²⁺) [*Momin et al.*, 2005]. The latter loads on remarkable H⁺ (0.97) but insignificant nss-SO₄²⁻ (0.25), are indicative of an acidic factor. The third factor shows the loadings for Na⁺ and Cl⁻, in the form of sea-salt, which only accounts for 7.4% of the total variance.

3.2. Seasonal Variations of VWM Concentrations at Mt. Lulin

[16] In order to analyze the data set in more detail, it was further divided based on different seasons, and seasonal VWM species concentrations were tabulated (Table 1, with the definition of the seasons). Five-day backward trajectories at Mt. Lulin, classified by different seasons are shown in Figure 3. The starting heights of the trajectories were set at 2860 and 4000 m AMSL in order to investigate the air motion aloft the site. Whenever rainfall was recorded on the corresponding day, the trajectory sets at two mentioned levels were computed.

[17] In the spring the VWM pH was 5.11. The concentrations for most of the species (except Na⁺ and Cl⁻) were elevated. It should be pointed out that spring is the season when Asian exports of natural and anthropogenic pollution aerosols to the Pacific are at its maximum, due to biomass burning peaks during this time in Southeast Asia. In addition, the cold fronts are responsible for the strong seasonal outflow to the Pacific [*Jordan et al.*, 2003; *Jacob et al.*, 2003, and references therein]. The plots (Figure 4) of the species concentrations show that abnormally high concentrations of all crustal and anthropogenic species appeared mainly in the spring months. However, the high concentrations of such samples are mostly due to exceptionally low amounts of rainfall.

[18] Most of the trajectories originate, same as in the winter as discussed later, from Southeast or south Asia (Figure 3a). This is especially true for the trajectories that started at 4000 m AMSL, and suggests that in the spring strong westerlies prevail in the middle troposphere over east Asia. Therefore as expected, the high concentrations of K⁺, NH_4^+ , and NO_3^- can be explained by the peak activities of biomass burning in Southeast and south Asia. Some of the trajectories point to India, where the emission of the precursor gas NH₃ is very high [Streets et al., 2003]. The high emission of NH₃ in southern China may also have contributed to the elevated NH_4^+ concentration of our rainwater samples. The concentrations of $nss-SO_4^{2-}$ and NO_3^- were half of those measured at our remote monitoring site at Peng Jia Yu (25.65°N, 122.07°E, 100 m AMSL) near the surface layer. This can be explained by the fact that the low-level trajectories from Peng Jia Yu originate from northeastern China where the emissions of precursor gases SO₂ and NO_x are intense [Streets et al., 2003]. However, the high-level trajectories traverse over southern China and then over Southeast or south Asia. The paths of these trajectories suggest that the impacts of Asian dust storms on our samples are insignificant. Therefore the high Ca^{2+} content (5 out of 14 samples with concentrations >10 μ eq l^{-1} and one sample has a Ca²⁺ concentration of 99.9 μ eq l^{-1}) in samples taken in the spring is not due to the impacts of Asian dust storms on northwestern China. Ma et al. [2003]









Figure 4. Plots of (a) rainfall amount (mm; bar) and pH value (solid line); concentrations (μ eq l^{-1}) of (b) Cl⁻ (green), Na⁺ (dark blue), K⁺ (light blue), Mg²⁺ (violet), and Ca²⁺ (yellow); and (c) NO₃⁻ (red), nss-SO₄²⁻ (green) and NH₄⁺ (dark blue) versus sampling date at Mt. Lulin.

reported little influences of mineral dust or sea salt on the plumes dominated by biomass burning in the nearby region mentioned at 3000 m AMSL during the TRACE-P campaign (9 March 2001). On the contrary, TOMS images (http://toms.gsfc.nasa.gov/) show high values of aerosol index (AI) from the Sahara/Arabian Deserts to India/Southeast Asia corresponding to most samples with high Ca²⁺ concentrations. High dust concentrations are also evident from the global aerosol simulation SPRINTARS (http:// cfors.riam.kyushu-u.ac.jp/~toshi/SPRINTARS/) from Africa to India. Momin et al. [2005] attributed the high SO_4^{2-} and Ca^{2+} concentration in their rainwater samples in India (similar to our cases) to the transport from the African continent. In our case the trajectory air masses associated with two samples with high Ca²⁺ concentrations intercepted the areas of high AI over the South China Sea/Philippine Sea. The trajectories for the rest of these samples originated from the Indian desert Thar (about 17-27°N, 70-75°E) with their starting height being at 4000 m AMSL in general. This implies the significant influence of the chemical signatures of free tropospheric air on our samples, at least in the spring time. Owing to the impact of the Thar Desert, the frequency of dust storms from northern and northwestern India is maximum during the premonsoon season (March to May) [Dey et al., 2004, and reference therein].

[19] In summer the VWM concentrations for all species are similar (except H⁺ concentration) to those of the entire monitoring period. It is worth noting that concentrations for these species are not the lowest, contrary to the findings of the site as mentioned in Peng Jia Yu and despite the dilution effects due to the highest amount of rainfall per sample. More than 95% of trajectories come from the clean marine atmosphere and from Southeast Asia where low pollution emissions are found during the summer (Figure 3b). Therefore emissions from Taiwan are the dominant source of pollution. The increase in the thickness of the planetary boundary layer during summer months, due to the intense heating up of the ground, leads to the frequent impact of boundary layer pollution on our site. The highest rate of conversion of HCl from Cl₂ by intense incoming solar radiation as mentioned in the previous section is evident from the highest ratio of Cl⁻/Na⁺ (2.8) during the summer months. A small amount of HCl is believed to come from incineration processes. The highest seasonal VWM pH value of 5.15 approaches the value of typical natural rainwater.

[20] The rest of our data set for the summer and fall months, when TCs (tropical cyclones) appear frequently over the western Pacific Ocean, has been checked for the possibility of the mentioned impacts of the TC. However, the TCs located far from Taiwan and their influences are not significant in some cases. The trajectories show their origins as being marine or they traverse from regions with a recorded noticeable rainfall. This implies that the impact of high pollution loadings associated with the TC on our samples is infrequent.

[21] Another sample that started on 5 August 2003 also shows peaks of species concentrations (Figure 4). Rainfall was recorded as 10 mm per week on 5-11 August 2003. Unstable air occurs frequently in the summer months due to the intense solar heating of the surface. The infrared satellite images for these days show local clouds (not shown here) appearing over Taiwan in the afternoon (but not in the morning). This indicates that a deep convection developed during these days, which is the main engine for transporting the boundary layer pollutants to the upper level air and resulted in elevated species concentrations in this sample.

[22] In the fall the trajectory plots starting at 2862 m AMSL indicated that most of the trajectories traverse over a marine atmosphere and the coastal areas of China and Korea, even though very few of the trajectories originated from Southeast Asia (Figure 3c). At 4000 m AMSL, the trajectories originated from Southeast and south Asia, and westerly flows reappeared at the middle troposphere. The species concentrations for this season were the lowest. This is explained by the facts that (1) most of the air masses are of oceanic origin; (2) the direct influence of boundary layer pollution is much less in the fall compared to the summer months because of less surface heating; (3) export of boundary layer pollutants from deep convective (other than TC) and frontal lifting processes are less common compared to those in summer and winter/spring months, respectively; (4) the average surface wind speed is higher in the fall than in the summer because of the smaller influence of the subtropical high-pressure system on Taiwan, resulting in a higher ventilation of local pollution in the fall within the boundary layer in general (The average ventilation coefficients on the upwind side of the Mt. Lulin for the summer and fall of 2004 were estimated to be 1430 and 2403 $m^2 s^{-1}$, respectively.) The VWM pH value in the fall was 5.24, which is less than that in the summer. This indicates that rainwater acidity is controlled by the balancing effects of acidic and alkaline species concentrations but not by the absolute concentrations of these species or by the emission strengths of their precursors [Wai et al., 2005].

[23] In winter the VWM concentrations for most of the species (except Mg^{2+}) are relatively high. The volume weighted pH value (4.91) is the lowest among all seasons. The average rainfall amount per sample (91.7 mm per week) is the highest. Generally, the trajectory paths (Figure 3d) are to the west of our site pointing to Southeast or south Asia, indicating the prevailing westerly flows in the middle troposphere in winter. However, only three weekly samples were taken in winter and the data set should be analyzed with caution. No further discussion is therefore made.

3.3. Rainwater Chemistry of Mt. Fuji

[24] Rainwater chemistry studies have been conducted and reported at the summit of Mt. Fuji (35.35°N, 138.73°E, 3776 m AMSL) in Japan since 1990 [*Dokiya et al.*, 1995; *Hayashi et al.*, 2001]. Only recent data sets from 2001 to 2004 are included for discussion here, as well as that of 2002 adopted by *Watanabe et al.* [2006]. The sampling durations were mostly 4 h (ranging from 1.5 to 14 h depending on whether the rainwater amount was enough for chemical analysis) for these studies, compared to the weekly samplings of our work at Mt. Lulin. Therefore it is possible that several samples were collected on the same day, depending upon the actual meteorological conditions of that day.

[25] About 75% of data were retained after the same data quality check was performed as mentioned in section 2. The rejected data were likely due to possible contamination of the samples during the sampling procedure [*Dokiya et al.*,

	Station	Ν	pН	Cl^{-}	NO_3^-	SO_4^{2-}	nss- SO ₄ ²⁻	H^+	NH_4^+	Na^+	K^+	Mg^{2+}	Ca^{2+}
						(2001-20	004)						
μ	Mt. Fuji	41	4.66	18.9	8.6	26.1	23.8	22.1	11.6	18.5	9.8	1.5	6.4
Maximum	5	-	-	148.0	34.4	320.6	316.1	398.1	52.3	141.9	51.8	7.7	32.7
Minimum		-	-	0.0	1.0	0.9	0.2	0.4	0.8	0.0	0.4	0.0	0.6
						(2003-20	005)						
μ	Mt. Lulin	39	5.12	8.6	15.8	22.5	22.0	7.5	21.8	4.0	2.4	2.4	7.8
Maximum		-	-	19.0	83.7	100.3	98.2	27.5	102.5	17.3	12.2	13.0	99.9
Minimum		-	-	0.0	4.6	6.9	6.8	1.2	0.0	0.0	0.0	0.0	0.0
					Seaso	nal Mean C	oncentration						
Spring (Mar to May)	Mt. Fuji	15	4.37	9.7/3.7	8.3/5.7	41.8/16.3	40.8/15.6	42.9/11.2	10.9/6.6	8.0/5.9	4.1/3.6	3.0/0.6	7.9/1.4
Summer (Jun to Aug)	5	11	4.88	0.5	3.0	7.8	7.7	13.1	2.5	1.2	0.7	0.8	3.3
Fall (Sep to Nov)		13	5.54	45.2	13.6	22.7	17.0	2.9	18.3	47.4	16.5	1.5	8.2
Winter (Dec to Feb)		2	4.70	0.0	8.8	30.1	29.8	20.0	18.1	2.8	2.7	-	-
29-30 May 2001		2	3.7	48.7	25.1	207.5	204.9	249.1	38.7	22.0	5.6	4.2	21.8
6 Jun 2001		6	4.9	-	2.5	5.3	5.1	12.6	1.7	1.3	-	1.2	1.9
					Ba	seline Conc	entration						
	Mt. Fuii	-	5.01	10.6	5.3	16.8	16.0	9.8	7.3	7.6	6.1	1.0	5.0
	Mt. Lulin	-	5.23	7.3	12.4	15.9	15.5	5.9	15.4	2.8	2.2	0.8	5.3

Table 3. Species Concentrations, $\mu \neq l^{-1}$, Measured at Mt. Fuji Compared With Those at Mt. Lulin^a

^aReported as nss-Ca²⁺ concentration at Mt. Fuji. Mean values for the whole data set/two samples removed as mentioned in the text are shown for spring at Mt. Fuji. Episodic species concentrations are also listed for comparison.

2001] under extreme conditions at the summit. The rainfall amount measured by standard rain gauge is not available since the precipitation comes not only from aloft but also from every direction at the summit [Dokiya et al., 2001]. Therefore VWM species concentrations cannot be calculated instead average concentrations at Mt. Fuji are presented here.

[26] Table 3 summarizes the rainwater chemistry of Mt Fuji from 2001 to 2004. A smaller number of samples is available for species K⁺, Ca²⁺, and Mg²⁺ (26, 30, and 21 samples, respectively). The average pH value for the whole period was 4.66. The acidic species $nss-SO_4^{2-}$ and the H^+ ions (mean concentrations: 23.8 and 22.1 μ eq l^{-1} , respectively) were the most abundant anions and cations (19% of the total concentration for each), followed by the species Na^+ and Cl^- (15% of the total concentration for each).

[27] The species Cl^- and Na^+ were highly correlated (R = 0.87, p < 0.001, N = 39) with a slope of about one (Figure 2b). This indicates minimal chloride depletion in the rainwater samples and a sea-salt origin of these species. It is worth noting that there is a very high correlation between K^+ and these species (R > 0.82), with a K^+/Na^+ equivalent ratio of about 0.34. This ratio is about 15 times higher than that of sea water, and the excess K^+ is thought to originate from biofuel consumption as will be discussed later. The Varimax rotated factor analysis also indicates very high loadings (>0.92) of K⁺, Na⁺, and Cl⁻ species with eigenvalue 5.59. This suggests that these loadings are brought together through long-range transport by the westerly flows (refer to the trajectory analysis in the following section) from east Asia.

[28] The species Mg^{2+} shows a high correlation (R = 0.79, p < 0.001, N = 21) with Ca^{2+} , suggesting its nonmarine and crustal source origin. When removing one sample of extreme values, $nss-SO_4^{2-}$ and NO_3^{-} are associated with each other (R = 0.81, p < 0.001, N = 40) with a slope of about 2.44.

[29] If two samples with an exceptionally high nss- SO_4^{2-} concentration (316.1 and 93.6 μ eq l^{-1}) are removed from the data set of Mt. Fuji, NH₄⁺ becomes well correlated with $nss-SO_4^{2-}$ (r = 0.76, p < 0.001, N = 40) and the average slope decreases from 2.37 to 1.06. This indicates that NH_4^+ and nss-SO₄²⁻ existed in the form of $(NH_4)_2SO_4$. The NH₄⁺ and NO₃⁻ correlation was very high (R = 0.87, p < 0.001, N = 40).

[30] The species K^+ was correlated with NH_4^+ (R = 0.71, p < 0.001, N = 26). A relatively large value of correlation coefficient (0.76) between K^+ and NH_4^+ was also found during ACE-Asia aircraft measurement over northeastern Asia [Lee et al., 2003]. It is believed that NH_4^+ sampled within such areas originated from the biofuel consumption in China [Lee et al., 2003; Ma et al., 2003].

3.4. Seasonal Differences of Species Concentrations at Mt. Fuji

[31] The data set of Mt. Fuji is further divided based on different seasons (Table 3). Owing to the 4-h sampling protocol, a larger sample size was obtained although only 9, 6, 6, and 2 sampling days were available in spring, summer, fall, and winter, respectively. Therefore these results should be interpreted with caution and the seasonal differences in species concentrations at Mt. Fuji are only discussed briefly with some case studies, as follows.

[32] The starting heights of the trajectories were set at 4000 m and 5000 m AMSL, similar to those adopted by Watanabe et al. [2006]. The trajectories are computed on a daily (rather than every 4 h) basis according to the sampling days. Many trajectories (Figure 5) originate from the west side of the summit, indicating that the westerly flows prevail over the site. This agrees with the year-round measurements of wind speed and direction reported by Igarashi et al. [2004].

[33] The mean concentrations of NH_4^+ and $nss-SO_4^{2-}$ were elevated during the winter months (Table 3). However, the





(**b**) VIS, 00UTC, 2 June 2001



Figure 6. (a) Surface pressure map and (b) its corresponding visible satellite image showing the Mei-yu front located over southern China at 0000 UTC, 2 June 2001.

sample size is too small to support further meaningful discussion. The transport pathways of the trajectory in spring (Figure 5a) were more or less similar to those reported by Igarashi et al. [2004] for the middle of February 2003. Although most of them traversed over north/northeastern China with high pollution emissions at the boundary layer as noted earlier, no elevated species concentration were found when two samples with episodic concentrations (as discussed below) were removed. A closer examination of the heights of the trajectories indicates that most of them were above 3000 m AMSL, well above the polluted boundary layer. Lee et al. [2003] also summarized that most of the species $(NH_4^+, SO_4^{2-}, NO_3^-, K^+, Na^+, and Cl^-)$ concentrations in fine aerosols above this level were at least a factor of 5 less than those in the boundary layer. Asian dust storms occur most frequently in springtime and their impact on the upper level air is expected to be significant, particularly for enhancing the crustal Mg^{2+} and Ca^{2+} .

However, the lowest Mg^{2+} and Ca^{2+} concentrations among other seasons are believed to be due to the fewer samples available for capturing the dust storm signatures as discussed later in section 3.5.

[34] The abnormally high concentrations for most of the species on 29-30 May 2001 are believed to be due to the influence of the volcanic plume of Miyake-Jima (34.08°N, 139.53°E) located 170 km southeast of Mt. Fuji. The differences in mean concentrations in spring before and after the removal of these two samples are shown in Table 3. On the basis of the forward and backward trajectory plots (not shown here) the influence of the plumes from Miyake-Jima on Mt. Fuji starts less than 12 h after leaving the volcano. Episodic concentrations of SO_2 and aerosol SO_4^{2-} were reported to be due to the influences of the plumes of Miyake-Jima [Igarashi et al., 2004; Naoe et al., 2003]. However, Igarashi et al. [2004] commented that the volcanic episodes occurred very infrequently and that the local wind direction and the existence of local convection played important roles in such episodes. Lee et al. [2003] reported coemission of NH₃, SO₂, and aerosol SO_4^{2-} of Miyake-Jima. The elevated NH_4^+ and nss-SO₄²⁻ concentrations (52.3 and 316.1 μ eq l^{-1} , respectively) on 30 May 2001 are the highest in the data set of Mt. Fuji.

[35] On the basis of the trajectory plots in the summer months (Figure 5b), the air masses are mostly resident above 3000 m AMSL. At the same time, some of them originate from the marine boundary layer where clean air masses are expected, and they result in the lowest concentrations for most species except Mg²⁺ and Ca²⁺ which can be attributed to a marine origin. *Igarashi et al.* [2004] also reported the lowest SO₂ concentration being measured during the summer months at Mt. Fuji. They argued that a Pacific High covering the Japanese islands can transport clean marine air to the summit.

[36] The concentrations for all the species were low on 6 June 2001. For instance, the concentrations of Ca^{2+} and nss- SO_4^{2-} were 1.9 and 5.1 μ eq l^{-1} , respectively. However, the corresponding trajectories pointed to the boundary layer of southern China, where high pollution emissions were expected from the Pearl River Delta (PRD). Closer examination of the surface weather maps from 1 to 6 June (Figure 6) shows that a quasi-stationary (Mei-yu) front was located over southern China which was associated with heavy rainfall, thereby reducing the pollution levels. Meanwhile, no TC was found near the South China Sea during those days and the associated high pollution loadings mentioned in section 3.2 did not appear over the coastal areas of southern China.

[37] In the fall the concentrations for most species are the highest among the seasons. The species concentrations in the fall were averaged from the data set available in September 2002 only, which was analyzed in detail by *Watanabe et al.* [2006]. The backward trajectories in the fall are plotted in Figure 5c. They argued that the high concentrations of Na⁺ and Cl⁻ in the rainwater were due to the rising motion associated with a strong TC (Sinlaku, central pressure 960 hPa) approaching Taiwan and which then struck the southern part of China. After these species were lifted from the boundary layer to the free troposphere, they were transported to the summit through long range transport. It should be pointed out that other anthropogenic and

crustal species can also be transported to the summit along with the sea salt species. It is reflected by the elevated concentrations of NH_4^+ , SO_4^{2-} , NO_3^- , K^+ , and Ca^{2+} , which are always associated with the elevated sea salt species concentrations in the samples. The elevated anthropogenic and crustal concentrations are believed to come from the high pollution loadings in the boundary layer over eastern China, which is similar to the case of Mt. Lulin discussed in section 3.2.

[38] The rest of the data set has been checked for the possible impact of the TC on the middle troposphere rainfall chemistry. This was done by checking for any TC located in the western Pacific region from the surface weather map, starting 10 d prior to each sampling day. However, no TC existed in this region, or if it did its influence was unlikely to affect the rainwater chemistry at Mt. Fuji, as indicated by the paths of backward trajectories.

3.5. Comparison Between Sampling Sites and the Aerosol Chemistry During TRACE-P and ACE-Asia

[39] Apart from the intercomparison of the rainwater chemistry between the two sites, the data sets of the aerosol chemistry (in the fine mode) obtained from the TRACE-P and ACE-Asia campaigns are included in this study to examine if there is consistency in the findings between aerosol and rainwater chemistry as mentioned in section 1. The insights and relationship between aerosol and rainwater chemistry are then to be investigated. The findings from Ma et al. [2003] regarding the measurements of the plumes from the biomass burning in Southeast Asia by TRACE-P P-3B aircraft and the findings of Lee et al. [2003] regarding the measurements over the East China Sea and around Japan and the Korean Peninsula by ACE-Asia C130 aircraft are mostly used in the present study. Detailed flight tracks and sampling procedures of these experiments can be found in the two literatures and references therein. However, the comparison should be made with caution since these campaigns were conducted in late winter/spring only and the chemical properties of the aerosols may vary from season to season.

[40] Table 3 shows that the acidity of the rainwater at Mt. Lulin (5.12) is lower than that at Mt. Fuji (4.66). While the species concentrations for nss- SO_4^{2-} , Ca^{2+} , and Mg^{2+} at Mt. Lulin and Mt. Fuji were relatively comparable, the sea-salt species Na⁺ and Cl⁻ and the K⁺ concentrations were more than double at Mt. Fuji, but they were about a factor of two less for NH₄⁺ and NO₃⁻. Only three samples had a ratio of Cl⁻/Na⁺ larger than 1.6 at Mt. Fuji. No excess chloride was found for most of the samples, which differs from the finding at Mt. Lulin. The reasons for that might be (1) the photochemical reactions were not rigorously compared to those at the lower latitude where Mt. Lulin is located; (2) Cl⁻ emitted from incineration processes at the boundary layer is unlikely to reach the summit of Mt. Fuji which is located higher than our site by about 1000 m.

[41] The average ratio of nss-SO₄²⁻/NO₃⁻ was 1.38 at Mt. Lulin, which is within the typical range obtained from the fine aerosol measurements during TRACE-P [*Ma et al.*, 2003]. This may imply that the contribution of trace gases SO₂ and HNO₃ to nss-SO₄²⁻ and NO₃⁻, respectively, in rainwater are insignificant, compared to the contributions of aerosol nss-SO₄²⁻ and NO₃⁻. The higher ratio (2.44) at

Mt. Fuji indicates a higher contribution of $nss-SO_4^{2-}$ to the rainwater acidity than that of NO_3^- , which may be due to the influence of the pollution plumes (enriched by SO_2) from north/northeast China and the volcanic plumes from Japan.

[42] The ratio of Mg^{2+}/Ca^{2+} was 0.15 at Mt. Lulin, which is in excellent agreement with the value 0.16 measured by the TRACE-P aircraft P-3B when the high Mg^{2+} samples of marine origin were removed [*Lee et al.*, 2003]. The flight tracks of the aircraft covered from 8°N to 40°N, taking some measurements near Taiwan. The agreement may suggest that these species were scavenged directly by cloud or rain droplets from the aerosol phase to the aqueous phase. The ratio was 0.20 at Mt. Fuji as compared to the value of 0.12 measured by aircraft NCAR C130 with flight tracks mainly around Japan, eastern China, and Korea [*Lee et al.*, 2003]. They also pointed out that the high correlation between these species was due to the crustal origin of Mg^{2+} for most of the samples.

[43] The species NH_4^{+} and $nss-SO_4^{2-}$ appeared as (NH₄)₂SO₄ at both sites. Lee et al. [2003] concluded that in most conditions NH3 was emitted in quantities comparable to or greater than H₂SO₄ and its precursor SO₂ throughout the flight tracks of the TRACE-P and ACE-Asia campaigns. This more or less explains the complete neutralization of H_2SO_4 and NH_3 . The molar ratio of $NH_4^+/$ NO_3^- obtained in Mt. Lulin (1.39) and in Mt. Fuji (1.49) showed good agreement. The larger-than-one ratio may be explained by the higher emission quantities of NH₃ than NO_x over China and Southeast/south Asia [Streets et al., 2003]. The molar ratio of $NH_4^+/(NO_3^-+SO_4^{2-})$ showed an altitude dependence in the TRACE-P and ACE-Asia campaigns. This ratio decreased with the altitude from 2000 to 5000 m in both campaigns. A similar variation is found in the present study when comparing the data sets of Mt. Lulin (median value of 0.58) with those of Mt. Fuji (median value of 0.20) in the spring.

[44] Mineral particles were reported to be important carriers and sinks for NO₃⁻ [*Matsumoto et al.*, 2004; *Dentner et al.*, 1996] and thus a high correlation should exist between Ca²⁺ and NO₃⁻. However, the correlation was only moderate (r = 0.62) at Mt. Lulin and Mt. Fuji (r = 0.63).

3.6. Comparison of Baseline Concentrations

[45] In general, the baseline concentration of a specific pollutant is determined on the basis of large volume of data set, while only 3-year data are available for Mt. Lulin. Instead, the comprehensive statistic approaches and comparisons with other sites are adopted. The baseline species concentrations (Table 3) are defined as the average concentrations which are free from the effects of episodic influences from the source regions, and their concentrations are temporally more uniform [Al-Momani et al., 1998]. While the definition of episodic concentration was not mentioned clearly by these authors, it was defined as two standard deviations plus the average concentration [Wai and Tanner, 2005; Chung et al., 1999]. However, too few data can be removed from the data sets in our case, and therefore such criteria is relaxed to one standard deviation plus the average concentration here, resulting in at most 20% of data being removed for both sites. While it is worth noting that Ca^{2+} , Mg^{2+} , nss- SO_4^{2-} concentrations and pH agree so well at

Table 4. A Comparison of VWM Species Concentrations, $\mu eq l$, at Our Site with Those at Other
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Locations	Period	Elevation, m	Cl^{-}	NO_3^-	SO_4^{2-}	nss- SO ₄ ²⁻	H^+	NH_4^+	Na^+	K^+	Mg^{2+}	Ca ²⁺
Mt. Lulin	2003-2005	2860	7.2	10.8	15.7	15.3	7.6	12.3	2.9	1.4	1.7	3.9
Lhasa, China	1998 - 2000	3700	21.6	7.3	6.1	4.9	<1	8.7	9.8	9.8	11.8	198.8
Mt. Fuji, Japan	2001 - 2004	3780	18.9	8.6	26.1	23.8	22.1	11.6	18.5	9.8	1.5	6.4
Malaysia	1996-1997	1545	5.8	4.7	11.4	10.8	7.4	5.6	4.7	2.7	1.0	3.8
Happo, Japan	2003	1850	7.3	6.6	15.0	14.4	12.4	6.4	4.8	<1	2.4	3.8
Mt. Rocky, US	2004	2920	1.0	9.5	6.1	6.0	4.8	6.3	1.0	0.3	0.0	5.8
Hedo, Japan	2003	50	451	9.5	70.4	22.6	12.5	10.9	430	10.1	98.6	7.0
Ogasawara, Japan	2003	230	286	4.7	36.0	8.1	9.1	3.6	251	5.6	62.2	16.0
Arctic region	1988	ground	9.4	3.7	8.6	8.0	20.2	5.1	4.6	1.6	1.8	4.2

^aMean concentrations reported in Mt. Fuji. Nss-SO₄²⁻ concentration only reported in Happo, Hedo, Ogasawara, Mt. Fuji, and our work, calculated nss-SO4 concentration based on Na⁺ concentration are reported at other locations. H⁺ concentration in Lhasa reported within 1998–1999. Only estimated species concentrations at Mexico are reported. Arctic region data are only available in July to August. References for various data sets are Mt. Lulin (present work); Lhasa [*Zhang et al.*, 2002]; Mt. Fuji (present work); Malaysia [*Ayers et al.*, 2002]; Happo, Hedo, and Ogasawara (http://www.eanet.cc/); Mt. Rocky (http://nadp.sws.uiuc.edu/); Arctic region [*Talbot et al.* 1992].

both sites, the NH_4^+ and NO_3^- concentrations at Mt. Lulin are about double. On the other hand, the K⁺ and Na⁺ concentrations at Mt. Fuji are higher than those at Mt. Lulin by a factor of two. Therefore the baseline species concentrations of Mt. Lulin are found to be comparable in magnitude to those of Mt. Fuji. The summit of Mt. Fuji was reported as an excellent platform for atmospheric background monitoring [Igarashi et al., 2004, and references therein]. In view of the above, it is suggested that the measurements of Mt. Lulin are representative of the background characteristics in rainwater in east Asia, after removal of the influencing episodic periods. However, there are drawbacks in such a comparison due to the limited samples mentioned and the short sampling durations at Mt. Fuji, which may influence the reliability of the findings here. Therefore the Mt. Lulin data set is also compared with those of other remote sites at high elevations as follows.

3.7. Comparison With Data Sets Measured in Other Areas

[46] Rainwater chemistry data sets from other remote or background monitoring sites located at high altitudes and remote islands as well as those in the arctic region are compared here (Table 4). It should be noted that a few measurements were conducted more than 10 years ago, so the chemical properties in the rainwater may have already been changed. Nevertheless, these data sets can act as some reference information regarding the rainwater chemistry of our site. The comparison is also complicated by the fact that the distributions of pollution sources and their strengths are highly inhomogeneous in a global sense. Continental-scale pollution transport is another complicated factor. Therefore Asian measurements are more important for our comparison here.

[47] A comparison of the data sets (Table 4) obtained by sites at high altitude (>1000 m AMSL) indicates that sea salt species and K⁺ in our site ranks at the low side, in the sense of Asian measurements or on a global comparison. Sea salt concentrations are even higher in Lhasa and Mt. Fuji than those in sites of lower altitude in Asia. A similar K⁺ concentration, as a biomass burning tracer, was measured at Malaysia and at our site. The concentrations of anthropogenic species such as nss-SO₄²⁻, NO₃⁻, and NH₄⁺ tend toward the high side when they are compared with those of Asian measurements, but they rank at the middle in a global

sense. On the other hand, the opposite holds true for the observation on H⁺ concentration. The H⁺ concentration is less than 1μ eq l^{-1} in Lhasa, due to the large amount of alkaline species Ca²⁺ existing in the rainwater samples. A low Ca²⁺ concentration was measured in our site, based on Asian or global measurements. When comparing the results at our site with those at remote islands and the arctic region, the sea salt species concentrations are comparable to the arctic values, while the concentrations on Cape Hedo (Okinawa Island) and Ogasawara Island are expected to be very high because the monitoring sites are close to the ocean. The anthropogenic species concentrations mentioned at our site rank at the high side. However, H^+ , Ca^{2+} , and K^+ concentrations at our site are low. To summarize, the concentrations of all species measured at Mt. Lulin rank within the range of concentrations recorded at other highaltitude or remote sites. Combined with previous discussions, we suggest that Mt. Lulin can be used as a regional site in Southeast Asia for the study of background rainwater chemistry and long-range transport of atmospheric pollutants.

4. Concluding Remarks

[48] The very first determinations of major ion concentrations in rainwater from Mt. Lulin, a high-elevation site in east Asia, have been reported in this paper, as well as a comparison with those at the Mt. Fuji background site. The following conclusions are drawn:

[49] 1. The volume-weighted mean pH value of the rainwater at Mt. Lulin was 5.12 for the whole sampling period of 2003'2005, approaching that of the typical natural water. The average pH value was higher than the value at Mt. Fuji (4.66).

[50] 2. In the rainwater samples, $nss-SO_4^{2-}$ was the most abundant anion at both Mt. Lulin and Mt. Fuji, while NH_4^+ and H^+ was the most abundant cation at Mt. Lulin and Mt. Fuji, respectively.

[51] 3. The species $nss-SO_4^{2-}$ and NO_3^{-} were highly correlated with each other at both sites. However, the higher ratio of $nss-SO_4^{2-}/NO_3^{-}$ at Mt. Fuji indicates a higher contribution of $nss-SO_4^{2-}$ to the rainwater acidity compared to that of NO_3^{-} .

[53] 5. The trajectory studies at both sites suggested that Mt. Lulin and Mt. Fuji are mostly influenced by crustal and anthropogenic sources at low ($< 25^{\circ}$ N) and middle ($>25^{\circ}$ N) latitudes, respectively.

[54] 6. The baseline species concentrations at both Mt. Lulin and Mt. Fuji are comparable. Compared with other high-altitude and remote sites, the concentrations of all species measured at Mt. Lulin rank within the range of concentrations recorded at those sites. Therefore it is suggested that apart from the summit of Mt. Fuji, the Lulin site is a high-altitude background monitoring site for rainwater chemistry in east Asia. While Mt. Fuji has its importance to identify the impacts of the anthropogenic activities (and the Asian dust storms) on the free troposphere over the middle latitude, Mt. Lulin plays an important role in the identification of the impacts of biomass burning from south and Southeast Asia and dust storms from the Indian Thar Desert on the upper air/free troposphere over the low latitude.

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