



Deposition of anthropogenic aerosols in a southeastern Tibetan glacier

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[1] Measurements of elemental carbon (EC), water-insoluble organic carbon (WIOC), and inorganic ions from a very high resolution snow/firn core retrieved from a glacier on the southeastern Tibetan Plateau reveal increasing concentrations associated with deposition of anthropogenic aerosols during the period 1998–2005. EC, WIOC, and SO_4^{2-} concentrations in the core were 4.7, 56.0, and 4.2 ng g^{-1} in 1998, but increased to 16.8, 144.4, and 162.1 ng g^{-1} in 2005, respectively. Comparison of EC to SO_4^{2-} and K^+ concentrations indicates a relatively greater impact of fossil fuel burning on EC deposition compared to biomass burning. Significant contribution of secondary OC production is apparent from the low EC/WIOC ratios. EC concentrations show large seasonal variability, with nonmonsoon precipitation containing more than double the concentrations found in monsoon snowfall. Since snow cover on the Tibetan Plateau extends to its maximum aerial extent during the nonmonsoon season, the high EC concentrations in snowfall during this time can cause advanced snowmelt and enhanced radiative forcing.

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1. Introduction

[2] Anthropogenic aerosols may play an important role in climatic change through the worldwide acceleration of glacier melting. As the dominant fractions of fine atmospheric aerosols, carbonaceous particles and sulfate have received special attention due to their strong light-absorbing and scattering properties, respectively. Elemental carbon (EC), also referred to as black carbon (BC) or soot [Birch and Cary, 1996; Chow *et al.*, 1993] absorbs more energy from light than other aerosols. Contrarily, sulfate and organic carbon (OC) as atmospheric aerosols scatter light strongly, rather than absorbing energy. Wang [2004] believes the scattering and absorbing properties significantly alter the regional and even the global radiation budget, and consequently alter climate [e.g., Jacobson, 2001; Hansen and Sato, 2001]. In cold regions, submicron black EC either trapped in snow and ice crystals or deposited directly on the surface may reduce the surface albedo and warm snow and ice by absorbing solar radiation [Flanner *et al.*, 2007; Hansen and Nazarenko, 2004; Jacobson, 2004; Light *et al.*, 1998; Warren, 1984; Warren and Wiscombe, 1980]. Hence, the melting rate of glaciers and sea ice will accelerate with increasing EC deposition [Hansen and Nazarenko, 2004]. Melting is further reinforced by the long-term global warming trends [Barnett *et al.*, 2005].

[3] Recent decades have witnessed exceptional economic development and population increases in Asia, resulting in serious air pollution accompanying higher energy demands. For example, the term Asian Brown Cloud was coined by the international Indian Ocean Experiment (INDOEX) after its discovery during the dry season in 1999. The Asian Brown Cloud manifests as a layer of brownish haze around 3 km thick in the atmosphere, stretching from the North Indian Ocean to the Himalaya–Hindu Kush range, mainly from December to April. Air pollutants and aerosol particulates from biomass burning and industrial emissions appear to be the major cause the Asian Brown Cloud [Ramanathan *et al.*, 2005; Ramanathan and Ramana, 2005]. Direct chemical measurements show that anthropogenic sources contribute as much as 75% to the observed haze. The submicrometer aerosols in Asian haze typically contain a mixture of OC, EC, sulfates, nitrates, dust and fly ash particles [Lelieveld *et al.*, 2001; Menon *et al.*, 2002]. These aerosols significantly contribute to atmospheric solar warming and surface cooling [Ramanathan *et al.*, 2005]. Apart from Greenland and Antarctica, the high-elevation Tibetan Plateau is one of the largest snow and ice areas of the world. The Tibetan Plateau plays an important role in Asian monsoon system, and holds the headwaters of many Asia's largest rivers such as the Yangtze, the Yellow, the Ganges and the Indus. Previous studies suggest that the regional lower atmospheric warming trend associated with the thick haze has contributed to the melting of Tibetan glaciers, threatening the water supply of 60% of the world's population that live in this region [Ramanathan *et al.*, 2005, 2007a]. Furthermore, when the high loading of black carbon in the haze deposits on snow, it causes a strong radiative forcing, assessed at as much as $+20 \text{ W m}^{-2}$ during the spring [Flanner *et al.*, 2007].

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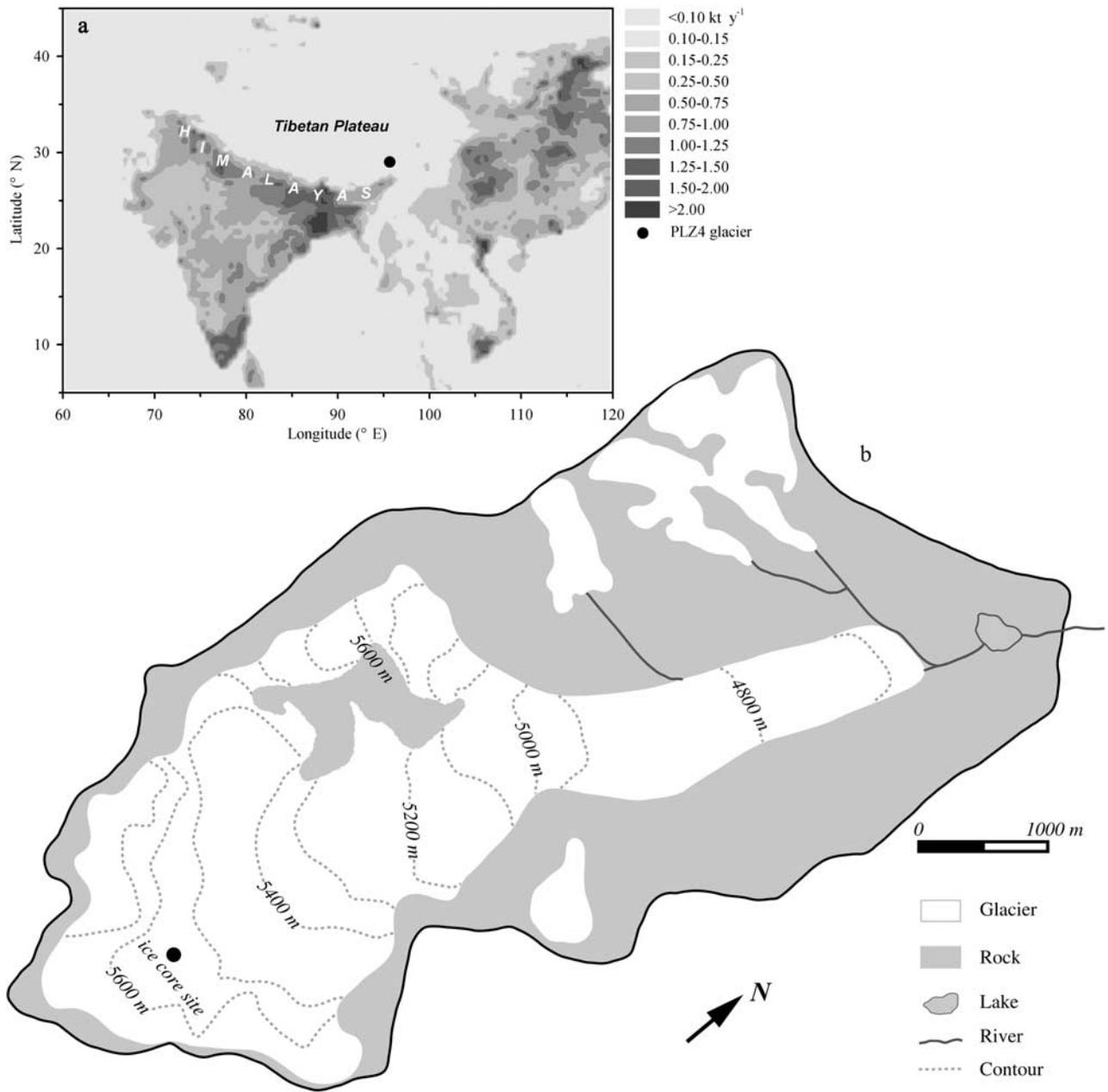


Figure 1. (a) Spatial distribution of black carbon emissions in Southeast Asia in 2000 [Ohara et al., 2007]. (b) Drill site location on the PLZ4 glacier.

[4] Despite its importance, few studies have included measurements of carbonaceous aerosol concentrations in Tibetan snow [Xu et al., 2006; Ming et al., 2008]. Consequently, few data exist regarding the deposition of the Asian Brown Cloud on snow and ice, which has precluded an accurate evaluation of the impact of anthropogenic aerosols on Himalayan glacier retreat and radiative forcing. This study presents a high-resolution ice core record of anthropogenic aerosol concentrations from the southeastern Tibetan Plateau, close to the northern limit of the Asian Brown Cloud.

2. Ice Core and Methods

[5] The Palong-Zanbu No. 4 Glacier (PLZ4; 96°55.04'E, 29°12.75'N) is located in the Kangri Garpo Range, south-

eastern Tibetan Plateau (Figure 1a), along the windward margin of the Indian monsoon. Under the influence of the monsoon, heavy precipitation occurs in this range, making it the largest glacierized area (approximately 28,000 km²) in the low latitudes of the Northern Hemisphere. During the summer of 2006, a 29 m ice core was retrieved at an elevation of 5500 m above sea level (asl) on the PLZ4 glacier (Figure 1b) using an electromechanical drill. The 9.5 cm diameter ice core was packed in precleaned plastic bags (40 to 100 cm length) immediately after drilling, and then stored in a temporary snow grotto at a temperature below -5°C. The core was transported from the drill site in insulated tubes to generator-powered freezers aboard vehicles, which immediately transferred the core to the cold storage (-20°C) facilities in Lhasa. Ice core sections were cut lengthways into

3 columns for analysis of oxygen isotopes, carbonaceous aerosols (EC and WIOC), and inorganic ions. Postdepositional alteration is assumed to be minimal based on the lack of melt layers, indicating below-freezing temperatures year-round, and the lack of annual chemical signal dampening with ice core depth. The possibility of postdepositional alteration is also reduced with increasing annual accumulation, since seasonal snowfall will be more rapidly covered by greater amounts of accumulation.

[6] The ice samples for measurement of carbonaceous aerosols were prepared at a temperature of -20°C in a class 100 clean room. The outer approximately 1.0 cm portion of each section was pared away with a precleaned scalpel. The remaining inner section measured ~ 3 cm in diameter and ~ 40 cm in length. Between 150 and 300 g was used for each analytical sample, with a total of 72 samples prepared. Prefired quartz fiber filters (Tissuquartz 2500QAT-UP 47 mm, Pall) were used for analysis. The original diameter of each 4.7 cm filter was cut into 6 smaller pieces measuring 1.5 cm in diameter. The smaller pieces were heated in an oxygen stream for 5 h in a tube oven (800°C), after being cut from the larger filter size to minimize possible contamination. Ice samples were weighed and then allowed to melt at room temperature in precleaned glass containers. To ensure the complete transfer of carbonaceous particles to the filters, the water samples were filtered twice using a stainless steel unit (400 mL volume). This resulted in a 1.0 cm diameter circle on each of the quartz fiber filters, which were dried in glass vacuum desiccators. To avoid possible positive EC artifacts, carbonates were removed prior to analysis by dripping 50 μL 0.1M HCl onto the sample spot three times, as suggested by Lavanchy *et al.* [1999].

[7] EC and WIOC analysis was achieved using a Desert Research Institute (DRI) Model 2001 Thermal/Optical Carbon Analyzer (Atmoslytic Inc., Calabasas, California). Each filter was analyzed for eight carbon fractions in a 0.5 cm^2 punch, following the Interagency Monitoring of Protected Visual Environments (IMPROVE) thermal/optical reflectance protocol [Cao *et al.*, 2003; Chow *et al.*, 2004]. The applied working conditions permitted the separation of four OC fractions (OC1, OC2, OC3, and OC4 at 120°C , 250°C , 450°C , and 550°C in a helium atmosphere, respectively), one PC fraction (a pyrolyzed carbon fraction determined when reflected laser light attains its original intensity after oxygen is added to the analysis atmosphere), and three EC fractions (EC1, EC2, and EC3 at 550°C , 700°C , and 800°C in a 2% $\text{O}_2/98\%$ He atmosphere, respectively).

[8] The second column of ice core was cut at ~ 10 cm intervals into 289 sections and allowed to melt at room temperature. Oxygen isotope ratios were determined via the classical headspace equilibration technique [Epstein and Mayeda, 1953] using a MAT-253 isotope mass spectrometer. Results are reported in standard delta (δ) notation versus standard mean ocean water (SMOW). The $\delta^{18}\text{O}$ precision was less than $\pm 0.2\%$, based on the maximum deviation of external standards for all sample runs.

[9] The third column of ice core was cut into ~ 35 cm length sections (86 samples), with the outer portion again pared away with a precleaned scalpel in a cold, class 100 clean room. Samples were allowed to melt at room temperature in precleaned plastic bottles immediately prior to ion chromatographic analysis. Dionex ICS-2500 and ICS-2000

ion chromatographs measured the anions (SO_4^{2-} , NO_3^- , Cl^-) and cations (Na^+ , NH_4^+ , K^+ , Mg^{2+} , Ca^{2+}), respectively. Anions were separated with an AS11-HC 4-mm analytical column, ASRS-ULTRA II 4-mm suppressor, 0.5 mL sampling loop and 25 mmol/L KOH eluent. Cations were analyzed with a CS12 4-mm analytical column, CSRS-ULTRA II 4-mm suppressor, 25 μL sampling loop and 25 mmol/L methanesulfonic acid eluent. The analytical precision for NH_4^+ concentration was less than 3%, with less than 1% concentration precision for other ions. The detection limit for all ions is less than 1 ng g^{-1} .

3. Results and Discussion

3.1. Seasonal Variations of Carbonaceous Aerosols

[10] A number of studies have verified that the precipitation “amount effect” causes more negative $\delta^{18}\text{O}$ values during the summer monsoon precipitation, and less negative values during nonmonsoon precipitation on the southern and eastern Tibetan Plateau [Tian *et al.*, 2003; Aizen *et al.*, 1996, 2006; Thompson *et al.*, 2000]. The resulting regular $\delta^{18}\text{O}$ annual cycles (Figure 2) established the ice core chronology. Figure 2 shows eight well-preserved annual cycles from 1998 to 2005 in the PLZ4 ice core.

[11] The very high accumulation (3.5 m annual average) at the drill site provides a good opportunity to observe the seasonal variations of aerosol concentrations. We applied a normalization technique [Barlow *et al.*, 1993; Shuman *et al.*, 1995; Yao *et al.*, 1999; Aizen *et al.*, 2004, 2006] to determine the monthly accumulation and corresponding seasonal means for isotopes and other constituents. Annual accumulation of each sample in the ice core was normalized by the ratio of seasonal precipitation to the annual total, as measured at the Bomi meteorological station, about 130 km from the ice core site. Stable isotopes ratios in precipitation samples collected at the Bomi weather station for a 1-year period reveal an annual maximum during the premonsoon season an annual minimum during the summer monsoon season. For this study, the peak $\delta^{18}\text{O}$ value was used to divide accumulation at the beginning April, based on the annual isotope maximum recorded at the Bomi station. The annual water equivalent precipitation from 1998 to 2005 averaged 2450 mm at the drill site. A two-season approach was used based on the annual accumulation regime, with the monsoon season from June through September and the nonmonsoon season from October through May. Each annual layer was divided by water equivalent thickness into portions associated with monsoon accumulation (50.5% of the annual total), and nonmonsoon accumulation (49.5% of the annual total). These percentages are based on long-term records of monthly mean precipitation from 1970 to 2003 at the Bomi weather station.

[12] Table 1 summarizes the seasonally averaged EC and WIOC concentrations, which are plotted together with annual average concentrations in Figure 3. As expected, the EC concentrations show regular seasonal cycles, similar to those revealed by the $\delta^{18}\text{O}$ seasonal variation (Figure 2). The highest average EC concentrations were associated with nonmonsoon precipitation, reaching 20.8 ng g^{-1} in 2005 (Table 1). Relatively lower average EC concentrations were observed for monsoon precipitation, ranging from 3.3 to 10.3 ng g^{-1} . Annual average EC concentrations revealed a

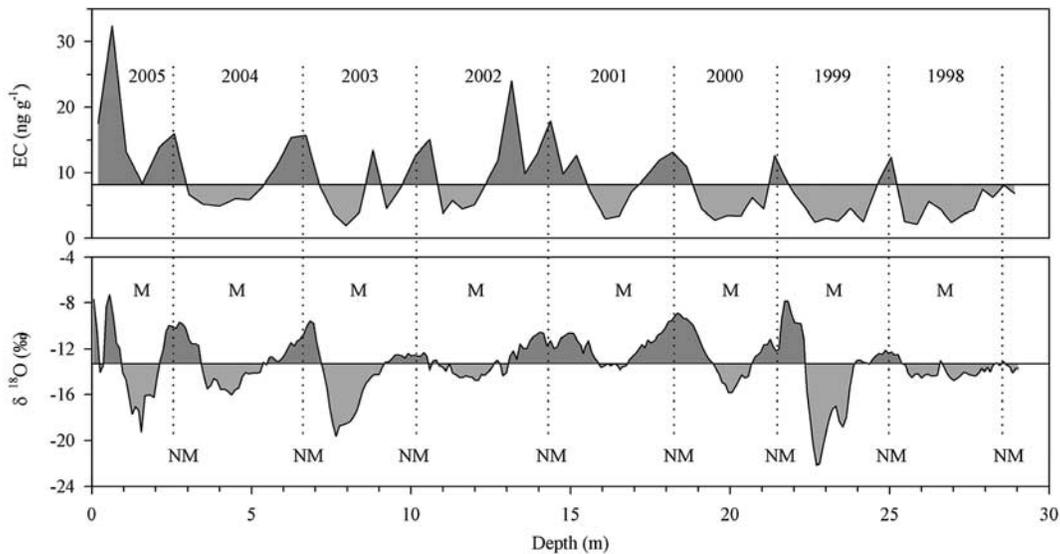


Figure 2. Ice core dating based on seasonal variations of $\delta^{18}\text{O}$. Elevated elemental carbon (EC) concentrations are apparent during nonmonsoon (NM) times of the year.

3.5-fold increase from 1998 to 2005 (Table 1). Annual WIOC fractions are given in Table 2 as a percent of total WIOC. It is apparent that during melting of the ice core samples, oxygenated species constituting the PC fraction are partially removed. The annual average PC fraction ranged from 11.1 to 15.0% (12.5% average) of the WIOC concentration (Table 2). Relatively higher PC/OC ratios (13–57%, 42% average) are reported for atmospheric aerosols over Europe [Pio *et al.*, 2007].

[13] Similar seasonal cycles of increased aerosol and EC loading during the cold seasons (and lesser loading with the return of the monsoon) have also been observed in the atmosphere over the Nepal Himalayas from 1998 to 2002 [Carrico *et al.*, 2003], and during October 2004 to December 2005 by the Maldives Climate Observatory–Hanimaadhoo [Ramanathan *et al.*, 2007b]. In South Asia, precipitation shows distinct seasonal distributions, with high precipitation from summer to autumn and low precipitation from winter to spring. This explains the occurrence of the Asian Brown Cloud from November through April, when the weather is dry and atmospheric aerosols are not as easily eliminated. Our drill site lies within the observed northern limit of the

Asian Brown Cloud. The spatial distribution of black carbon emissions in Southeast Asia is depicted in Figure 1a [Ohara *et al.*, 2007]. The pollutants in the brown haze can be blown into the southeastern Tibetan Plateau region by the south branch of the westerlies, swept over the south side of the Himalayas, and deposited on the PLZ4 glacier resulting in relatively high EC concentrations.

[14] EC-bearing snow can reduce albedo, causing a strong radiative forcing that accelerates snow/glacier melt by absorbing more solar radiation [Hansen and Nazarenko, 2004; Jacobson, 2004; Light *et al.*, 1998; Warren, 1984; Warren and Wiscombe, 1980]. Recent simulations show that a reduction in albedo from soot exceeds the “dimming” effect on solar radiation when EC concentrations are on the order of 10 ng g^{-1} or more [Flanner *et al.*, 2008]. Nonmonsoon concentrations exceeded this value during 2001–2005; annual average concentrations exceeded 10 ng g^{-1} during 2002 and 2005 (Table 1). During the nonmonsoon season, the Asian Brown Cloud forms as a 3-km-thick haze present to the south of the Himalayas from a height of 2 to 5 km. The associated high EC concentrations in nonmonsoon snowfall are also coincident with the time of maximum

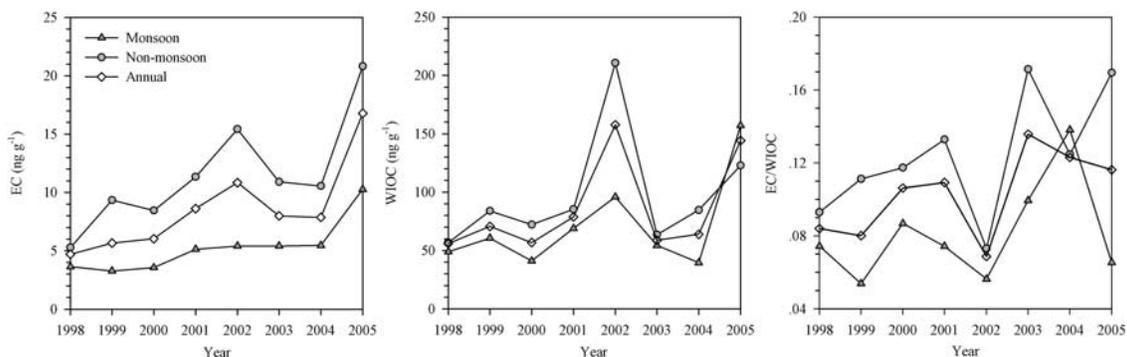


Figure 3. Annual and seasonal variation of EC, WIOC, and EC/WIOC concentrations in the PLZ4 ice core, 1998–2005.

Table 1. Annual and Seasonal Average Elemental Carbon and Water-Insoluble Organic Carbon Concentrations Recorded in the PLZ4 Ice Core, Southeastern Tibetan Plateau, 1998–2005^a

Year	EC (ng g ⁻¹)		WIOC (ng g ⁻¹)			
	Monsoon	Nonmonsoon	Annual	Monsoon	Nonmonsoon	Annual
2005	10.3	20.8	16.8	157.1	122.8	144.4
2004	5.5	10.6	7.9	39.5	84.6	63.9
2003	5.4	10.9	8.0	54.4	63.6	58.9
2002	5.4	15.4	10.8	96.0	210.8	157.7
2001	5.1	11.3	8.6	69.0	85.3	78.8
2000	3.6	8.5	6.0	41.0	72.1	56.8
1999	3.3	9.3	5.7	60.7	83.9	70.7
1998	3.6	5.3	4.7	48.9	56.8	56.0

^aAbbreviations are as follows: EC, elemental carbon; WIOC, water-insoluble organic carbon.

aerial extent of snow cover, further exacerbating melting due to increasing surface albedo.

3.2. Chemical Variability, 1998–2005

[15] Annual and seasonal concentrations of EC, WIOC, and inorganic ions in the ice core increased from 1998 to 2005 (Tables 1 and 3), concurrent with a decrease in precipitation reported by *Sontakke et al.* [2008]. For carbonaceous matter, the increase was more moderate than for inorganic ions (a factor of 3 for EC and 2.2 for WIOC). Although the proportion of increase for most inorganic ions is large, actual concentrations remain relatively low compared to other Tibetan Plateau sites such as the East Rongbuk glacier in the central Himalayas [*Hou et al.*, 2002], the Guliya ice cap on the northwest Tibetan Plateau [*Thompson et al.*, 1997], and the Dundee ice cap on the northeast Tibetan Plateau [*Thompson et al.*, 1989]. However, the increase in concentrations is cause for concern, as an accelerating increase since 2001 was observed for all constituents (Tables 1 and 3).

[16] For the period 1998–2005, northern India precipitation was lowest during 2002 and 2005–2006 [*Sontakke et al.*, 2008], concurrent with years of relatively higher chemical concentrations in the PLZ4 ice core (Tables 1 and 3). Severe drought conditions occurred in 2002, with one of the shortest recorded monsoons in history. July rainfall was 51% of the normal amount in 2002, surpassing all previously recorded droughts [*Poorest Areas Civil Society Programme*, 2004]. As expected, all concentrations in the ice core increased dramatically in conjunction with the drier periods of 2002 and 2005–2006. Most notable increases were observed for Na⁺, Cl⁻, K⁺ and WIOC (Tables 1 and 3). Indian monsoon failures have been previously associated with high dust and Cl⁻ concentrations in the Dasuopu ice core, central Himalayas [*Thompson et al.*, 2000]. This suggests that droughts in India intensify transport of dust and pollutants to the Tibetan Plateau glaciers due to increased dust loading and longer atmospheric suspension during dry times compared to wet. The seasonal variation of EC concentrations in the PLZ4 ice core, as well as the seasonal eruption of the Asian Brown Cloud, are now linked closely to the seasonal wet and dry cycles in India.

3.3. Discussion of Sources

[17] Comparisons of ionic concentrations in ice can be used to trace the sources of atmospheric aerosols at remote sites. Different chemical compositions in the Tibetan Plateau ice core derive from different origins. Ca²⁺, a major con-

stituent in Tibetan Plateau ice, originates primarily from soil dust [*Thompson et al.*, 1989, 1997]. The aerosol SO₄²⁻ in Himalayan ice is formed by oxidation of SO₂, chiefly arising from fossil fuel combustion [*Duan et al.*, 2007], with lesser contributions from dust and seawater. NO₃⁻ is often found on the surface of aged dust due to heterogeneous deposition of HNO₃ and NO_x [*Underwood et al.*, 2001], and has been associated with combustion processes [*Mayewski et al.*, 1990; *Legrand and Mayewski*, 1997; *Legrand et al.*, 1999]. NH₄⁺ comes mainly from agricultural fertilizers [*Kang et al.*, 2003; *Hou et al.*, 2003] and biomass burning [*Legrand et al.*, 1992; *Whitlow et al.*, 1994; *Fuhrer et al.*, 1996].

[18] On average, Ca²⁺ contributed the most to the total ionic concentration in the ice core (over 30%), followed by NO₃⁻ (~20%), SO₄²⁻ (~14%), and NH₄⁺ (~10%). Cl⁻ and Na⁺ contributed about 11% and 7%, respectively. K⁺ and Mg²⁺ concentrations were relatively lower, accounting for ~4% and ~1% of the total, respectively. The annual average ion concentrations in the PLZ4 ice core ranked in the following order: Ca²⁺ > NO₃⁻ > SO₄²⁻ > Cl⁻ > NH₄⁺ > Na⁺ > K⁺ > Mg²⁺ (Table 3). Sodium and chloride are likely influenced mainly by seawater contributions given the close proximity to the Indian Ocean and the similarity of the average Cl⁻/Na⁺ ratio in the ice core (1.74) to the seawater ratio (1.80). Ratios of Na/Ca (0.26) and Cl/Ca (0.34) in the ice core are similar to ratios reported by *Legrand et al.* [2002] for the free troposphere over Europe (0.16 and 0.26, respectively), indicating the inorganic ions are also impacted by atmospheric dust at this study location.

[19] Non-sea-salt (nss) contributions of SO₄²⁻, K⁺, Mg²⁺ and Ca²⁺ were calculated using the standard mean seawater ratios for SO₄²⁻/Na⁺ (0.252), K⁺/Na⁺ (0.03595), Mg²⁺/Na⁺ (0.1206), and Ca²⁺/Na⁺ (0.03791). Calculation of nss concentrations were used for a general comparison among ions, although a portion of the Na and Cl concentrations undoubtedly arrive with nss dust as well. Results show nss-SO₄²⁻, nss-K⁺ and nss-Ca²⁺ account for 87%, 93% and 99% of their total concentrations, respectively, while nss-Mg²⁺ accounted for only 20% of its total concentration. The high average nss-Ca²⁺/nss-SO₄²⁻ ratio of 2.5 provides further evidence of dust influence at the remote southeastern Tibetan Plateau region. Concentrations of nss-SO₄²⁻ are mainly due to fossil fuel emissions [*Lelieveld et al.*, 2001], and ionic compositions dominated by nss-SO₄²⁻, NH₄⁺, and NO₃⁻ (over 43% of the total inorganic ion concentration) in the PLZ4 ice core all implicate combustion sources [*Carrico et al.*, 2003]. The total sea salt contribution (the sum of Na⁺, Cl⁻, sea salt SO₄²⁻, K⁺, Mg²⁺ and Ca²⁺) accounts for less than 22% of the total ionic concentration.

Table 2. WIOC Fractions as a Percentage of Total WIOC

Year	OC1/OC (%)	OC2/OC (%)	OC3/OC (%)	OC4/OC (%)	PC/OC (%)
2005	0.9	21.2	49.7	16.3	12.0
2004	1.0	15.3	46.0	22.7	15.0
2003	0.7	21.9	48.8	15.9	12.7
2002	2.7	24.8	51.2	10.2	11.1
2001	1.6	26.2	47.1	12.8	12.4
2000	0.4	23.9	49.7	14.0	12.0
1999	0.4	25.2	48.7	12.7	13.1
1998	0.4	27.4	47.9	12.8	11.5

Table 3. Annual Average Inorganic Ion Concentrations in the PLZ4 Ice Core^a

Year	Na ⁺	NH ₄ ⁺	K ⁺	Mg ²⁺	Ca ²⁺	Cl ⁻	SO ₄ ²⁻	NO ₃ ⁻
2005	48.5	83.1	28.8	10.4	194.9	68.7	162.1	181.3
2004	14.2	35.6	6.5	2.3	84.5	17.4	50.2	63.9
2003	12.7	20.4	7.7	1.2	103.1	21.3	19.6	31.7
2002	32.4	19.8	16.1	1.7	91.1	53.8	32.8	63.4
2001	17.7	15.0	7.1	3.4	52.4	26.5	11.3	33.2
2000	6.7	2.1	3.9	1.9	40.5	11.9	7.6	21.4
1999	4.7	6.3	2.8	0.8	32.5	8.0	3.1	8.2
1998	6.3	18.5	2.5	0.2	34.4	7.4	4.2	12.4

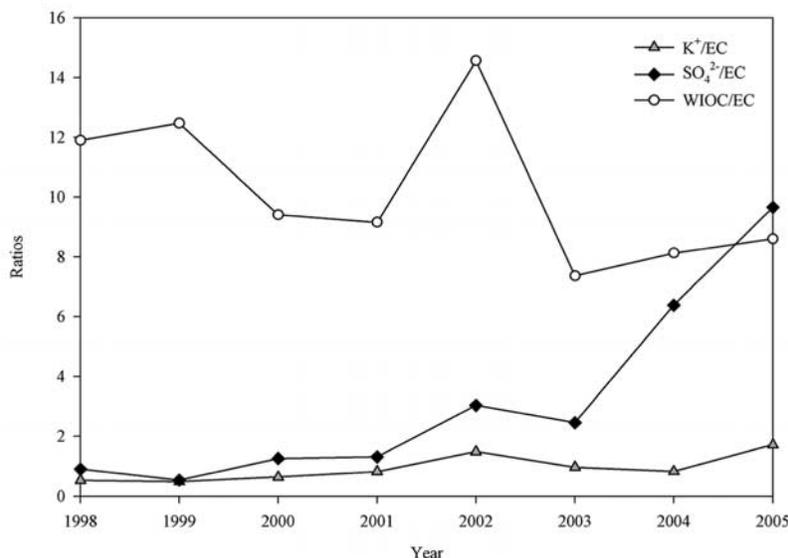
^aUnits are nanogram per gram.

[20] The main source of EC is predominantly from combustion emissions, although no major pollution sources exist adjacent to the remote location of the high-elevation drill site (5500 m asl). Consequently, the EC deposited on the glacier was transported over medium to long distances. The drill site, located at the southeastern margin of the Tibetan Plateau, is mainly under the influence of equatorial-maritime air masses originating in the Indian Ocean between June and September [Bryson, 1986]. During winter and spring, the Tibetan Plateau splits the westerly jet stream into two currents, the south and north branches. The south branch moves eastward along south side of the Himalayas and flows over the southeastern Tibetan Plateau. Atmospheric pollutants emitted in South Asia can be transported year-round to the southeastern Tibetan Plateau glacier region under monsoon conditions and by the westerlies.

[21] Unlike EC, some portion of OC can dissolve in water, resulting in EC/WIOC ratios melted ice samples that should be higher than EC/OC ratios in the atmosphere. EC/OC ratios have commonly been used to distinguish combustion sources of anthropogenic aerosols [Wolff *et al.*, 1982; Turpin *et al.*, 1990; Turpin and Huntzicker, 1991; Chow *et al.*, 1996; Gillier and Gertler, 2000; Gillies *et al.*, 2001; Andreae and Merlet, 2001]. Relatively high EC concentrations and low OC concentrations were found in atmospheric particulate matter sampled at two Maldives climate observatories during

the dry season, with seasonal differences in EC/OC ratios attributed to high contributions from biofuel burning and fossil fuel combustion during the more polluted dry season [Stone *et al.*, 2007]. Although EC/OC ratios in the atmosphere can vary substantially depending on the source [Mazzera *et al.*, 2001], lower EC/OC ratios are often associated with biomass burning [Andreae and Merlet, 2001; Cao *et al.*, 2005; Han *et al.*, 2008]; EC/OC ratios lower than 0.10 suggest significant contribution from secondary OC during transport. In the PLZ4 ice core, the average EC/WIOC ratio is 0.11 ± 0.06 . If we assume there is approximately two times as much water-soluble OC than water-insoluble [Pio *et al.*, 2007], we can estimate an EC/OC ratio in the ice core of 0.04. This ratio may be higher by a factor of two (0.08) compared to atmospheric concentrations, due to less scavenging of EC compared to OC. This estimate is significantly lower compared to the BC/OC ratios reported from INDOEX (0.90) [Novakov *et al.*, 2000] and lower than EC/OC ratios resulting from primary emissions associated with either biomass burning (0.17) or fossil fuel emissions (1.72) in the European free troposphere [Gelencsér *et al.*, 2007]. Thus, the low EC/WIOC ratios in the Tibetan Plateau ice core are clearly explained by origins other than combustion, such as formation of secondary OC during transport. Secondary OC production was previously reported as the major contribution (89–95%) to the OC concentrations in ice from the Alps [Legrand *et al.*, 2007].

[22] K⁺/EC and SO₄²⁻/EC ratios have also been used to investigate potential sources of carbonaceous compounds in the extensive haze layers observed in the Indian subcontinent [Novakov *et al.*, 2000; Salam *et al.*, 2003]. Owing to the long transport distance and possible soil dust alteration, the ratios of K⁺/EC and SO₄²⁻/EC may not represent the original chemistry of the combustion emissions. However, higher SO₄²⁻/EC ratios indicate greater contributions from fossil fuel burning, while relatively higher K⁺/EC ratios indicate greater contribution from biomass burning [Novakov *et al.*, 2000]. In the PLZ4 ice core, the greater increase in the SO₄²⁻/EC ratio compared to the K⁺/EC ratio (Figure 4) indicates a greater

**Figure 4.** Changes in K⁺/EC, SO₄²⁻/EC, and WIOC/EC ratios in the PLZ4 ice core from 1998 to 2005.

contribution from fossil fuel emissions compared to biomass burning, results that are in accordance with previous findings over the tropical Indian Ocean [Novakov et al., 2000].

4. Summary and Conclusion

[23] A high-resolution ice core record from a southeastern Tibetan Plateau glacier reveals increasing concentrations in anthropogenic aerosol deposition. A persistent decrease in precipitation and increase in pollutant emissions, as well as dry years recently recorded in India, have amplified the contributions of anthropogenic aerosols to Tibetan Plateau glaciers. Carbonaceous aerosol concentrations show distinct seasonal variability, with highest concentrations during the nonmonsoon season, corresponding temporally with occurrences of the Asian Brown Cloud, and lowest concentrations during the monsoon season. Since snow cover reaches its largest aerial extent during the nonmonsoon season, particulate deposition on the Tibetan Plateau snow has an important impact on melting and radiative forcing. An increase in melting can also impact the seasonal water supply for South and East Asia.

[24] On the basis of the analysis of inorganic ions, EC/WIOC, $\text{SO}_4^{2-}/\text{EC}$, and K^+/EC ratios we find dust and combustion to be the two major aerosols sources in the ice core records. Low EC/WIOC ratios indicate secondary OC contributions. On the basis of the greater increase in the $\text{SO}_4^{2-}/\text{EC}$ ratio compared to the K^+/EC ratio, carbonaceous aerosols contributed from fossil fuel emissions are dominant over biomass burning, and have become increasingly significant in the southeastern Tibetan Plateau.

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