

Black carbon concentrations from a Tibetan Plateau ice core

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Black carbon concentrations from a Tibetan Plateau ice core spanning 1843–1982: recent increases due to emissions and glacier melt

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Received: 23 August 2013 – Accepted: 13 September 2013 – Published: 1 October 2013

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Published by Copernicus Publications on behalf of the European Geosciences Union.

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Abstract

Black carbon (BC) deposited on snow and glacier surfaces can reduce albedo and lead to accelerated melt. An ice core recovered from Guoqu glacier on Mt. Geladaindong and analyzed using a Single Particle Soot Photometer provides the first long-term (1843–1982) record of BC concentrations from the Central Tibetan Plateau. The highest concentrations are observed from 1975–1982, which corresponds to a 2.0-fold and 2.4-fold increase in average and median values, respectively, relative to 1843–1940. BC concentrations post-1940 are also elevated relative to the earlier portion of the record. Causes for the higher BC concentrations include increased regional BC emissions and subsequent deposition, and melt induced enrichment of BC, with the melt potentially accelerated due to the presence of BC at the glacier surface. A qualitative comparison of the BC and Fe (used as a dust proxy) records suggests that if changes in the concentrations of absorbing impurities at the glacier surface have influenced recent glacial melt, the melt may be due to the presence of BC rather than dust. Guoqu glacier has received no net ice accumulation since the 1980s, and is a potential example of a glacier where an increase in the equilibrium line altitude is exposing buried high impurity layers. That BC concentrations in the uppermost layers of the Geladaindong ice core are not substantially higher relative to deeper in the ice core suggests that some of the BC that must have been deposited on Guoqu glacier via wet or dry deposition between 1983 and 2005 has been removed from the surface of the glacier, potentially via supraglacial or englacial meltwater.

1 Introduction

Black carbon (BC), an aerosol produced by the incomplete combustion of fossil and bio-fuels, can impact climate by absorbing energy directly in the atmosphere and by reducing albedo when deposited on snow and ice, potentially enhancing melt and accelerating glacial retreat (Hansen and Nazarenko, 2004). The study of BC's past and

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7, 4855–4880, 2013

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into ~5–6.5 cm segments and rinsed in ultra-pure Milli-Q water to remove the outer section of the core that can potentially be contaminated during the drilling or cutting process. Once rinsed, samples were placed in 50 mL polypropylene vials and prepared for shipment to Central Washington University in Ellensburg, WA. Due to customs issues the samples were not kept frozen. Because earlier tests showed that refreezing samples resulted in an apparent reduction in BC concentrations (Kaspari et al., 2011), the samples were stored as liquid until analyzed for BC using a Single Particle Soot Photometer (SP2) between October 2010 and July 2011.

All samples were treated and analyzed for BC according to the methods reported by Kaspari et al. (2011): samples were acidified to 0.5 mol HNO₃, sonicated for 15 min just prior to analysis, and stirred using a magnetic stirrer during analysis. Subsequent research has shown that acidification aids in recovery of BC in samples stored in polypropylene (Wendl et al., 2013), however in general we advise against acidification for samples that are kept frozen until just prior to analysis because acidification can cause a shift towards smaller BC particles (Schwarz et al., 2012). The liquid samples were nebulized using a Cetac U-5000AT+ ultrasonic nebulizer, and the resulting aerosol was coupled to the inlet of the SP2. The SP2 uses laser-induced incandescence to determine the mass of refractory BC in individual particles (Schwarz et al., 2006; Stephens et al., 2003). The SP2 at CWU was internally calibrated using Aquadag. Monitoring of liquid sample flow rate pumped into the nebulizer, fraction of liquid sample nebulized and purge airflow rate allows BC mass concentrations in the liquid sample to be determined. Schwarz et al. (2012) report that the SP2 combined with a Collison-type nebulizer can be used to measure BC mass concentration in snow with an estimated 60 % uncertainty (due to uncertainty in calibration results, and size dependent nebulization efficiency). The BC concentrations reported herein underestimate actual concentrations, and have a higher uncertainty than 60 % due to the following:

- *Loss of BC particles in the Cetac U-5000AT+ ultrasonic nebulizer.* Schwarz et al. (2012) tested nebulization efficiency based on particle size using polystyrene latex spheres ranging in size between 220 and 1537 nm, and reported that the

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Cetac nebulization efficiency was size dependent. The nebulization efficiency was 20 % or less for particles greater than 700 nm relative to particles in the 200–500 nm size range. Subsequent research at CWU confirmed the results of Schwarz for the Cetac, while Wendl et al. (2013) found that nebulization efficiency was greatest in the 300–400 nm size range, with decreased efficiency for smaller and larger particle sizes. Additionally, Schwarz et al. (2013) report larger mass size distributions of BC in snow than BC in the atmosphere, with the mass of BC cores larger than 600 nm accounting for 17 % or more of the BC mass based on snow samples in Colorado. Thus, a non-trivial mass of BC is not nebulized by the Cetac nebulizer, and thus not measured by the SP2. We correct for the mass of BC not nebulized based on gravimetric Aquadag standard solutions (assuming 70 % of the dry content of Aquadag is BC). This standard correction does not fully address the size dependent nebulization nor likely differences in the mass-size distributions of Aquadag and the snow samples. We also note that prior studies using the SP2 to measure BC concentrations in liquid samples have used other BC materials as a calibration standard, with other standards resulting in a higher corrected BC concentration than Aquadag (Wendl et al., 2013)

- *Apparent BC losses during sample storage.* As stated above, the Geladaindong samples were not kept frozen, thus we have sought to quantify changes that can occur to the sample during storage in the liquid phase. Freshly prepared Aquadag standards and environmental snow melted just prior to being measured were re-measured periodically over a 212-day period (Wendl et al., 2013). Measured BC concentrations decreased until 55 days, after which BC concentrations stabilized. Possible causes of the BC losses include BC adhering to the vial walls, and/or BC particles agglomerating above the size range at which particles are efficiently nebulized and outside of the detection range of the SP2. BC losses are proportionally greater in low concentration samples relative to higher concentration samples, however the decrease in measured BC mass is slightly larger in the higher concentration samples relative to the lower concentration samples. The implica-

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tion for the results presented herein is that relative trends are preserved in the samples that were stored in the liquid phase. However, the difference observed between low and high BC concentration samples is less than was likely preserved in the un-melted ice, and the increase factors that we report below are underestimates. This is supported by Menking et al. (2013) who used the SP2 to measure BC concentrations in samples from a Mt. Logan ice core that had been stored in the liquid state at room temperature for numerous years, and in 8 m of archived ice that was melted just prior to analysis (the liquid and frozen samples are from the same depths of adjacent slabs from the same ice core). The two BC records showed the same trends and were significantly correlated, although BC concentrations were approximately a magnitude higher in the freshly melted ice as anticipated. In addition, the relative difference between high and low concentration samples was greater in the freshly melted samples than the stored liquid samples, confirming our suggestion that increase factor analyses will underestimate actual concentration changes for samples that have been stored in the liquid phase. All of the samples presented in this study were analyzed between five and fourteen months after being melted, and BC concentrations are assumed to have reached equilibrium based on our storage experiments.

These factors lead to large uncertainties in the actual BC concentrations, with the data reported herein representing lower limit values. As a result, we report the BC concentrations as *measured* BC (MBC), and focus our interpretation on relative differences in BC rather than on absolute concentrations. Note that the concentrations reported herein predominantly represent the mass of BC particles 500 nm and smaller, which corresponds to the size range where the mass absorption cross section of BC particles is greater relative to larger particles, meaning that smaller BC particles absorb light and reduce albedo more efficiently (Schwarz et al., 2013). Despite the uncertainties in the data, the MBC data yield information that furthers our understanding of BC temporal variations and impacts on the Tibetan Plateau.

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Other sections of the ice core have been previously sampled and analyzed for soluble major ions by ion chromatography, for trace elements by inductively coupled plasma mass spectrometry, and $\delta^{18}\text{O}$ by mass spectrometry, all performed at the Climate Change Institute of the University of Maine (Grigholm et al., 2013). The ice core was dated by counting the annual layer signal from isotopes, ions, and trace elements, and verified using known age horizons from tritium that peak in 1963, ^{210}Pb and several volcanic markers (Kang et al., 2013). Although the ice core was recovered in 2005, annual mass loss of ice at this location has resulted in the loss of the most recent 22 yr of the record, and the top of the core is dated to 1982 AD (Kang et al., 2013). To account for changes in sample resolution with depth in the ice core due to glacier thinning, the high-resolution M_{BC} and Fe (used as a proxy for dust) data were re-sampled to three samples per year.

3 Results and discussion

3.1 Results

Six-hundred and twenty-five samples were analyzed for BC from the upper 42 m of the core, providing a record of M_{BC} concentrations from the central Tibetan Plateau from 1843–1982 (Fig. 2). The record is characterized by an increase in M_{BC} concentrations during the most recent period of the record: the highest concentrations are observed from 1975–1982, which corresponds to a 2.0-fold increase in average and a 2.4-fold increase in median values relative to 1843–1940 (Table 1). Causes for the higher M_{BC} concentrations include increased emissions and subsequent deposition, and/or increased glacier melt. These factors are considered in more detail below.

3.2 Increased BC emissions and deposition

BC fossil fuel sources include the burning of coal, diesel, and gasoline for manufacturing, transportation, heating and cooling, and power generation (Bond et al., 2004;

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2012; Conway et al., 1996). Xu et al. (2012) monitored BC concentrations in the snowpack above the superimposed ice layer on a Tien Shan glacier over a year, and found that relative to freshly fallen snow, during the melt season BC was enriched in the surface snow and to an even greater extent in the snow/firn directly above the superimposed ice layer. Xu et al. propose that meltwater can flush BC through the upper ~ 1 m of the snowpack. This results in lower BC concentrations in the snow between the surface and bottom depths of snow/firn, with BC unable to flow below the superimposed ice layer, resulting in build-up of BC at this horizon.

To assess if melt induced enrichment in BC may have contributed to the increased occurrence in layers of relatively high M_{BC} preserved in the Geladaindong ice core since the 1940s, we examine glacier variation and temperature data from the Geladaindong region. We also further consider the Geladaindong ice core record, as BC and other light absorbing impurities can not only become concentrated during melt, but also contribute to accelerated melt.

Glacier retreat in recent decades is widespread across the Tibetan Plateau (Yao et al., 2012). Specific to the Geladaindong region, Kang et al. (2013) show that Geladaindong (5750 m) and nearby Nyainqentanglha (5850 m) have not received net accumulation since the 1980s and 1950s, respectively. This indicates widespread melt in the region in recent decades, consistent with results previously reported in the Himalaya (Kehrwald et al., 2008). Ye et al. (2006) reconstructed glacier area in the Geladaindong region using topographic maps from 1969 and Landsat images from 1973–1976, 1992 and 2002. While some glaciers advanced during the time period investigated, the area of glacier retreat was much larger. During the period of overlap with the Geladaindong ice core, glacier area decreased by 4.7 km^2 from 1969 to 1976, followed by accelerated glacier retreat during the 1980s and 1990s.

Causes of glacier retreat are commonly attributed to rising temperatures (Barnett et al., 2005; Ye et al., 2006) reduction in accumulation due to atmospheric circulation changes (Yao et al., 2012), and deposition of light absorbing impurities including BC (Hansen and Nazarenko, 2004). Surface temperatures on the Tibetan Plateau have in-

aerosol optical depth over the Tibetan Plateau, which could counter the increased irradiance from cloud cover reductions. This is supported by observational data from the Tibetan Plateau showing a decrease in surface solar radiation from 1960–2009, with aerosol emissions identified as a plausible cause (You et al., 2013).

BC and dust in snow and ice reduces the surface albedo, which heats the snowpack by conducting energy from the heated BC and dust to snow grains, resulting in accelerated snow metamorphism. This in turn leads to coarser snow grains, which further reduces the snow albedo (Painter et al., 2012). At Geladaindong, positive feedback cycles may exist in which (1) greater BC deposition has lowered the surface albedo and increased melt, and/or (2) warmer temperatures have increased glacier melt, resulting in higher BC concentrations at the surface of the glacier from mechanical trapping, lower surface albedo, and increased melt. Thus, the higher M_{BC} concentrations observed since the 1940s may be caused by the combined effects of higher BC deposition on the glacier from increasing regional BC emissions, and melt induced enrichment of BC, with the melt potentially accelerated due to the presence of BC.

While many recent studies focus on the role of BC in reducing snow albedo and accelerating snow and glacier melt in the region (e.g. Ming et al., 2008, 2009; Xu et al., 2009, 2012), less attention has been given to the efficacy of BC to reduce albedo and accelerate melt in the presence of other absorbing impurities. Albedo reductions from BC will be less in the presence of other absorbing impurities because the other impurities will capture some of the solar absorption that the BC would receive in the absence of other impurities (Kaspari et al., 2011). This is particularly relevant for glaciers in this region where the presence of other impurities can be high. In a modeling study Flanner et al. (2009) estimated that the Eurasian spring radiative forcing from BC and dust in snow is 2.7 and 1.2 $W m^{-2}$, respectively. Kaspari et al. (2011) used BC and dust mass concentration and size distributions from an Everest ice core to estimate that the radiative forcing from BC exceeded that of dust, however large uncertainties in the optical properties of the dust along with the same BC analytical issues described in the methods above put these results in question. Wang et al. (2013) measured light-

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absorbing particles in snow across Northern China using a spectrophotometer, and found that snow particulate light absorption was dominated by BC in the northeast, whereas it was dominated by local soil and desert dust on the northern boundary of the Tibetan Plateau. Kaspari et al. (2013) measured BC and the total impurity load in snow samples from Mera glacier in Nepal, and based on model results found that while BC reduces albedo, dust dominates albedo reduction and the impact of BC is diminished in the presence of dust.

Because the M_{BC} concentrations reported herein underestimate actual BC concentrations in addition to uncertainties in the optical properties and absorption efficiency of dust in the Geladaindong ice core, we do not attempt to assess albedo reductions due to BC and dust, nor the albedo reduction of BC relative to dust. Instead, we examine trends in the M_{BC} and Fe records, using Fe as a proxy for dust because iron oxides dominate light absorption by mineral dust. From 1843 to 1982 M_{BC} and Fe display a close relationship, with peak concentrations commonly coinciding (Fig. 2). This is likely due to: (1) the shared seasonality of BC and dust, both of which display peak atmospheric concentrations during the winter/spring (Qian et al., 2009; Xu et al., 2009; Kaspari et al., 2011); (2) the mixing of aerosols and dust during long-distance transport; and (3) the mechanical trapping of impurities at the ablation surface during conditions of melting and sublimation (Conway et al., 1996; Xu et al., 2012). Although M_{BC} and Fe concentrations peak coincidentally, the Fe record does not display the same increase in maximum concentrations post-1940 as the M_{BC} record (Fig. 2). Rather, average concentrations of Fe decrease from 1843–1982 (Table 1). Because dust would undergo similar enrichment under melt conditions as BC, the lower Fe concentrations in the recent portion of the record suggest that either Fe deposition has decreased, or that BC deposition has increased associated with rising emissions. The different trends between the BC and Fe records further suggests that if changes in the concentrations of absorbing impurities have influenced recent glacial melt, it may be due to the presence of BC rather than dust. This conclusion is in agreement with the findings of Kaspari

et al. (2011), who documented stable dust concentrations from ~ 1860–2000 in the presence of increasing BC concentrations in a Mt. Everest ice core.

3.4 Comparison to other Asian BC records

There is interest in comparing the Geladaindong M BC ice core record with other BC records from the region in regards to BC concentrations and trends, as this information has the potential to further understanding of the sources, transport, deposition and impacts of BC in the region. Here we focus on a comparison of the Geladaindong M BC ice core record with a BC record from a Tibetan Plateau lake sediment core (Cong et al., 2013), and with an Everest BC record (Kaspari et al., 2011). We focus on these two records because they are the only BC records from the region that span back to ~ 1850, and the Everest ice core was analyzed for BC by the same methods employed here.

BC flux in a sediment core from Nam Co Lake, Central Tibetan Plateau was relatively constant from 1857–1900, increased gradually from 1900–1960, and increased rapidly between 1960 and 2009 (Cong et al., 2013). The authors suggest that the rise in BC flux was most likely due to BC transported from South Asia. That the BC flux in the Nam Co core increases in recent decades supports that an increase in BC concentrations at Geladaindong are due at least in part to higher BC emissions during the latter portion of the 20th century.

Samples from Geladaindong and Everest ice cores were stored for extended periods of time in the liquid phase. Based on the storage tests described previously, here we assume that BC losses during vial storage stabilized in both cores, and that a comparison of concentrations between the records provides a qualitative assessment of differences in BC concentrations. For the period of overlap between the two records (1860–1982), M BC concentrations are more than threefold higher at Geladaindong than Everest, which may be due to differences in snow accumulation, atmospheric BC concentrations, elevation or melt (Table 2). The snow accumulation rate affects the resulting concentration of BC in snow. If two ice core sites differed only in their annual

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Xu et al. (2012) noted that due to the coupled impacts of greenhouse-gas warming and BC enrichment in surface snow, dirty ice that can at present form in the accumulation zone underlying the snowpack can be exposed in the future as the glacier equilibrium line altitude (ELA) increases. The ELA at Guoqu glacier has increased in recent decades, thus this site is a potential example of the process that Xu et al. projects already occurring.

BC deposition on snow and glaciers remains a concern both for its radiative forcing effect, and its ability to affect glacier mass balance and the timing and availability of water resources (e.g., Hansen et al., 2004; Ramanathan and Carmichael, 2008). For example, using a numerical model applied to BC observed on a Tibetan glacier, Yasunari et al. (2010) estimated that a 2.0–5.2 % albedo reduction could result in a 11.6–33.9 % increase in annual discharge if the reduced albedo snow layer remained at the glacier surface. While this increase in discharge likely overestimates actual increases in discharge since snow with a lower BC concentration and higher albedo can be deposited during the summer monsoon, it nevertheless highlights that BC has the potential to accelerate snow/ice melt, affecting the availability of water resources. Increased melt can thus lead to an increased supply of glacier runoff in the short term, but the concern for the future is extensive glacier retreat, resulting in a decreased availability of water resources.

4 Conclusions

Here we presented the first long-term (1843–1982) record of BC concentrations from the Tibetan Plateau reconstructed from a Mt. Geladaindong ice core. The record is characterized by an increase in BC concentrations during the most recent period of the record, with median BC concentrations from 1975–1982 more than twofold greater relative to 1843–1940. The higher BC concentrations in recent decades are likely caused by a combination of increased emissions from regional BC sources, and melt induced

enrichment of BC, with the melt potentially accelerated due to the presence of BC at the glacier surface.

Guoqu glacier on Mt. Geladaindong serves as a potential example of a glacier where an increase in the ELA is already exposing buried high impurity layers. Because the most recent 22 yr (1983–2005) of accumulation were not preserved at the drill site on Guoqu glacier, it is notable that BC concentrations in the uppermost layers of the Geladaindong ice core are not markedly higher. This suggests that some of the BC that must have been deposited on Guoqu glacier via wet or dry deposition between 1983 and 2005 has been removed from the surface of the glacier, potentially via supraglacial or englacial meltwater.

The available BC and dust data are not sufficient to quantify the relative absorption of BC versus dust to albedo reductions. However, that dust concentrations are lower in recent decades suggest that if changes in the concentrations of absorbing impurities have influenced recent glacial melt, the melt may be due to the presence of BC rather than dust. Further observational studies are needed to assess the relative contribution of different absorbing impurities (e.g., BC, dust, colored organics, humic-like substances, snow algae) to snow and glacier melt.

Acknowledgements. This research was funded by the National Science Foundation (OISE-0653933 and EAR-0957935) and by a Geological Society of America Graduate Research Grant.

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Table 1. Geladaindong average and (median) Fe and M_{BC} concentrations ($\mu\text{g L}^{-1}$) and increase factors based on data resampled to 3 samples/year.

	1843–1940	1940–1982	1975–1982	1940–1982/ 1843–1940	1975–1982/ 1843–1940
M_{BC}	1.7 (1.2)	2.9 (1.5)	3.4 (2.9)	1.7 (1.2)	2.0 (2.4)
Fe ($\mu\text{g L}^{-1}$)	289.9 (188.8)	253.4 (155.7)	199.2 (145.6)	0.9 (0.8)	0.7 (0.8)

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Table 2. Geladaindong and Everest M_{BC} and Fe ($\mu\text{g L}^{-1}$) average and (median) concentrations based on data resampled to 3 samples/year from 1860–1982 (the period of overlap between the two records), average snow accumulation rate at each site (cm water equivalent/year), and BC and Fe flux (accumulation rate \times average (or median) concentration). The Everest data is from Kaspari et al. (2011), however concentrations reported here have been corrected for nebulizer losses based on Aquadag standards.

	Geladaindong (5750 m)	Everest (6518 m)	Geladaindong/Everest
M_{BC} ($\mu\text{g L}^{-1}$)	2.1 (1.3)	0.6 (0.3)	3.5 (4.3)
Fe ($\mu\text{g L}^{-1}$)	255 (176)	107 (73)	2.4 (2.4)
accumulation rate (cm w.e. yr^{-1})	30	50	0.60
M_{BC} flux ($\text{ng } M_{BC} \text{ cm}^{-2} \text{ yr}^{-1}$)	63 (39)	30 (15)	2.1 (2.6)
Fe flux ($\text{ng Fe cm}^{-2} \text{ yr}^{-1}$)	7650 (5280)	5350 (3650)	1.4 (1.5)

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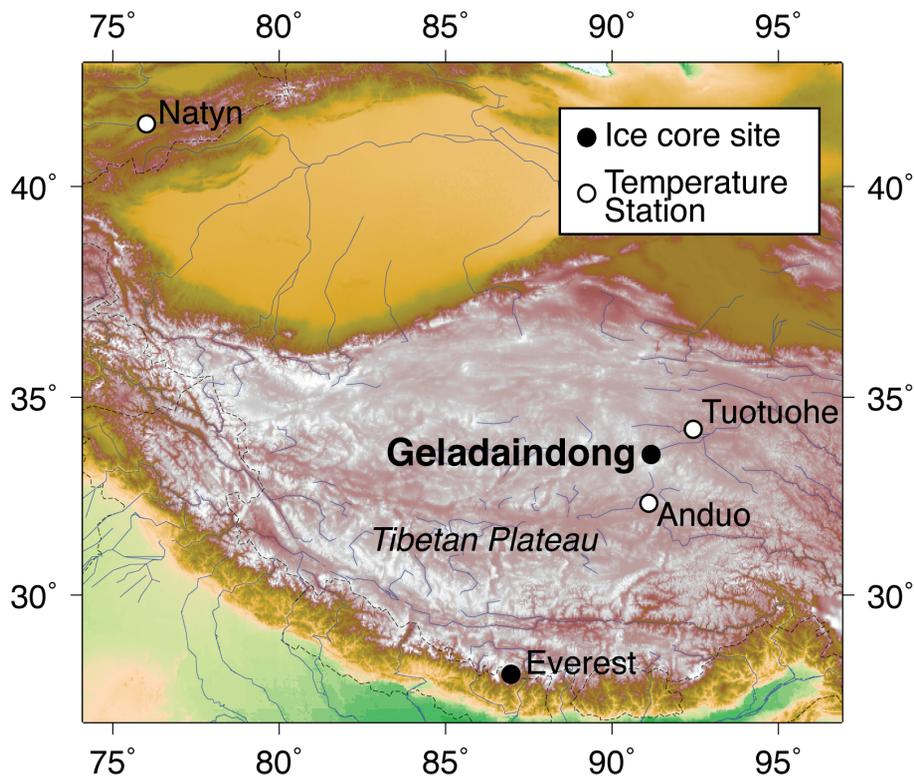


Fig. 1. Map showing the location of the Geladaindong and Everest ice core sites, and temperature stations discussed in the text.

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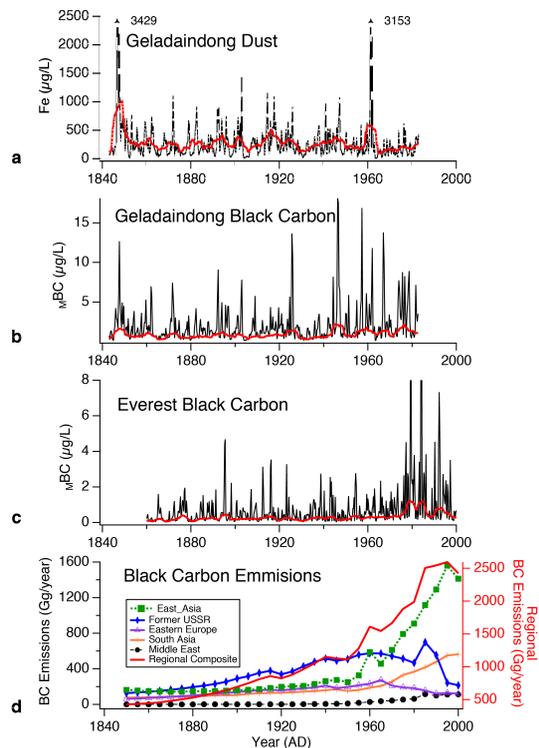


Fig. 2. (a) Geladaidong Fe (used as a proxy for dust), (b) Geladaidong $m\text{BC}$ and (c) Everest $m\text{BC}$ (Kaspari et al., 2011; note the concentrations plotted here have been corrected for nebulizer losses) smoothed with a 15 point boxcar (red). (d) Black carbon emissions from energy related combustion (Bond et al., 2007). The regional black carbon emissions (red) are a summation of emissions from East Asia, the Former USSR, Eastern Europe, South Asia, and the Middle East.

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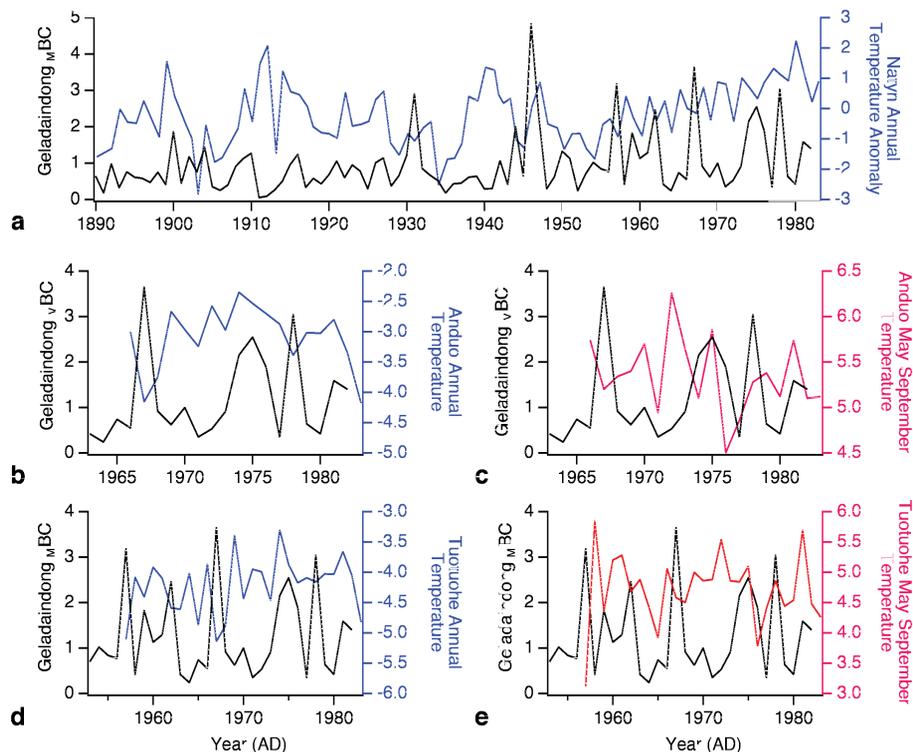


Fig. 3. Geladaindong_MBC and **(a)** Natyn annual mean temperature anomaly (relative to 1950–1979 reference period) from Liu and Chen (2000), **(b)** Anduo annual temperature, **(c)** Anduo May–September temperature, **(d)** Tuotuohe annual temperature, and **(e)** Tuotuohe May–September temperature.

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