## **Supporting Information**

## **Xu et al. 10.1073/pnas.pnas.0910444106**

## **SI Text**

AS

**Materials and Methods.**Ice cores were cut lengthways into 4 columns by using a band saw in a cold room with temperature of  $-20$  °C. One column was cut at intervals 10–25 cm into segments and used for black soot measurements. The outer  $\approx$  1.0-cm portion of each segment was pared away with a precleaned scalpel at a temperature of  $-20$  °C in a class-100 laminar flow cabinet. The remaining inner section in a weight range of 150–300 g was then allowed to melt at room temperature with a precleaned glass container in a class-100 clean room. Immediately after melting, the liquid sample was filtered through a prefired quartz fiber filter, which was heated in an oxygen stream for 5 h in a tube oven with temperature of 800 °C. The water samples were filtered twice, and the containers and filtration unit were rinsed three times with ultrapure water (Milli-Q, 18.2  $M\Omega$ ; Millipore) to ensure complete transfer of carbonaceous particles to the filters. Filtration efficiency was tested in several studies using similar procedures, with filtration efficiency found to be about 95% (1–4). To avoid possible positive black carbon (BC) artifacts, carbonates were removed before analysis by dripping  $50 \mu L$  of 0.1 M HCl onto the sample spot three times, as suggested by Lavanchy et al. (3).

BC and organic carbon (OC) on the filters were measured by using the Interagency Monitoring of Protected Visual Environments (IMPROVE) thermal/optical reflectance protocol. The applied working conditions permitted the separation of four OC fractions  $(OC<sub>1</sub>-OC<sub>4</sub>)$  in a helium atmosphere and three BC fractions (BC<sub>1</sub>–BC<sub>3</sub>) in a 2% O<sub>2</sub>/98% He atmosphere in response to the thermal program. Pyrolized carbon split from BC fractions was determined when reflected laser light attained its original intensity after oxygen was added to the analysis atmosphere. Detailed procedures and quality assurance can be found in the Cao et al. (5).

We note that the OC concentration yield on the quartz filter was not total organic carbon, but the water-insoluble organic

- 92:9801–9809. 3. Lavanchy V, et al. (1999) Elemental carbon (EC) and black carbon (BC) measurements with a thermal method and an aethalometer at the high-alpine research station Jungfraujoch. *Atmos Environ* 33:2759 –2769.
- 4. Lavanchy V, et al. (1999) Historical record of carbonaceous particle concentrations from a European high-alpine glacier (Colle Gnifetti, Switzerland).*J Geophys Res* 104:21227– 21236.

carbon and part of water-soluble organic carbon that were adsorbed by the quartz filter during the filtering. The influence of dust particles on laser reflection and the split point of pyrolized carbon were quantitatively evaluated for the possible carbon artifacts in the IMPROVE analysis, as was the analysis reproducibility. A shallow ice core of 5.47 m in length loaded with high dust was cut lengthways into four parallel samples, with the dust portion analyzed in four different filtering approaches involving removing dust and remaining dust, and the use of single and double filters. Considering all of the possible effects on BC/OC measurements addressed above, the uncertainty was assessed to be 16% for OC and 15% for BC.

**Tibetan Air Temperature Change During 1951–2007.** [Fig. S1](http://www.pnas.org/cgi/data//DCSupplemental/Supplemental_PDF#nameddest=SF1) shows the surface air temperature anomaly on the Tibetan Plateau relative to the base period 1951–1980 based on the temperature analysis of Hansen et al. (6). We defined the Tibetan Plateau as the area above 4,000-m height at latitudes between 35 °N and 50 °N and longitudes between 70 °E and 105 °E.

**Ice Core Dating.** Ice core dating was achieved via the annual layer counting that is made possible by an annual cycle of  $\delta^{18}O$  isotopic ratios and BC concentration [\(Fig. S2\)](http://www.pnas.org/cgi/data//DCSupplemental/Supplemental_PDF#nameddest=SF2). The layer-counted age was verified to be accurate within 1 year by using the maximum in ice core  $\beta$ -activity concentration, corresponding to the 1963 atmospheric maximum associated with open-air nuclear weapons testing.

Data for Fig. 3 are given in [Table S1.](http://www.pnas.org/cgi/data//DCSupplemental/Supplemental_PDF#nameddest=ST1) The monsoon period was defined as June through September, and the nonmonsoon period was the remaining 8 months of the year. Seasonal concentrations were calculated by comparing annual ice core accumulation to the seasonal percent share of total annual precipitation by using the method defined by Aizen et al. (7).

- 5. Cao JJ, et al. (2003) characteristics of carbonaceous aerosol in Pearl River Delta Region China during 2001 winter period. *Atmos Environ* 37:1451–1460.
- 6. Hansen J, et al. (2001) A closer look at United States and global surface temperature change. *J Geophys Res* 106:23947–23963.
- 7. Aizen VB, et al. (2006) Climatic and atmospheric circulation pattern variability from ice-core isotope/geochemistry records (Altai, Tien Shan and Tibet). *Ann Glaciol* 43:49 – 59.

<sup>1.</sup> Cachier H, Pertuisot MH (1994) Particulate carbon in arctic ice. *Analusis* 22:M34 –M37. 2. Chylek P, et al. (1987) Aerosol and graphitic carbon content of snow. *J Geophys Res*



**Fig. S1.** Surface air temperature anomaly on the Tibetan Plateau, relative to 1951–1980 mean, averaged over the area with altitude greater than 4,000 m above sea level.

 $\overline{a}$ 



Fig. S2. BC and OC concentrations and  $\delta^{18}O$  versus depth and ice core dating for the Zuoqiupu ice core.

## **Table S1. Data used for Fig. 3 and annual snow accumulation on the Zuoqiupu glacier**

PNAS PNAS

