Atmospheric Environment 176 (2018) 91–102

Contents lists available at ScienceDirect

Atmospheric Environment

journal homepage: www.elsevier.com/locate/atmosenv

Atmospheric depositions of natural and anthropogenic trace elements on the Guliya ice cap (northwestern Tibetan Plateau) during the last 340 years

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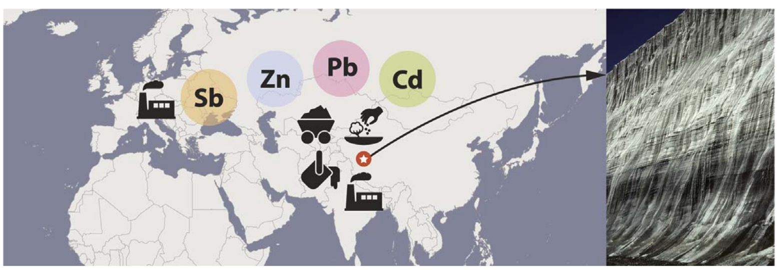
GRAPHICAL ABSTRACT



ARTICLE INFO

Keywords:
Atmospheric pollution
Coal combustion
Heavy metals
Fossil fuel
Tibet
Ice core

ABSTRACT

A continuous record of 29 trace elements (TEs) has been constructed between 1650 and 1991 CE (Common Era) from an ice core retrieved in 1992 from the Guliya ice cap, on the northwestern Tibetan Plateau. Enrichments of Pb, Cd, Zn and Sb were detected during the second half of the 19th century and the first half of the 20th century (~1850–1950) while enrichments of Sn (1965–1991), Cd and Pb (1975–1991) were detected during the second half of the 20th century. The EFs increased significantly by 20% for Cd and Sb, and by 10% for Pb and Zn during 1850–1950 relative to the pre-1850s. Comparisons of the Guliya TEs data with other ice core-derived and production/consumption data suggest that Northern Hemisphere coal combustion (primarily in Western Europe) is the likely source of Pb, Cd, Zn, and Sb during the 1850–1950 period. Coal combustion in Europe declined as oil replaced coal as the primary energy source. The European shift from coal to oil may have contributed to the observed Sn enrichment in ~1965 (60% EF increase in 1975–1991), although regional fossil fuel combustion (coal and leaded gasoline) from western China, Central Asia, and South Asia (India, Nepal), as well as Sn mining/smelting in Central Asia, may also be possible sources. The post-1975 Cd and Pb enrichments (40% and 20% EF increase respectively in 1975–1991) may reflect emissions from phosphate fertilizers, fossil fuel combustion, and/or non-ferrous metal production, from western China, Central Asia, and/or South Asia. Leaded gasoline is likely to have also contributed to the post-1975 Pb enrichment observed in this record. The results strongly suggest that the Guliya ice cap has recorded long-distance emissions from coal combustion since the 1850s with more recent contributions from regional agriculture, mining, and/or fossil fuel combustion. This new Guliya ice core record of TEs fills a geographical gap in the reconstruction of the pollution history of this region that extends well beyond modern instrumental records.

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https://doi.org/10.1016/j.atmosenv.2017.11.040
Received 5 August 2017; Received in revised form 20 November 2017; Accepted 23 November 2017
Available online 08 December 2017
1352-2310/

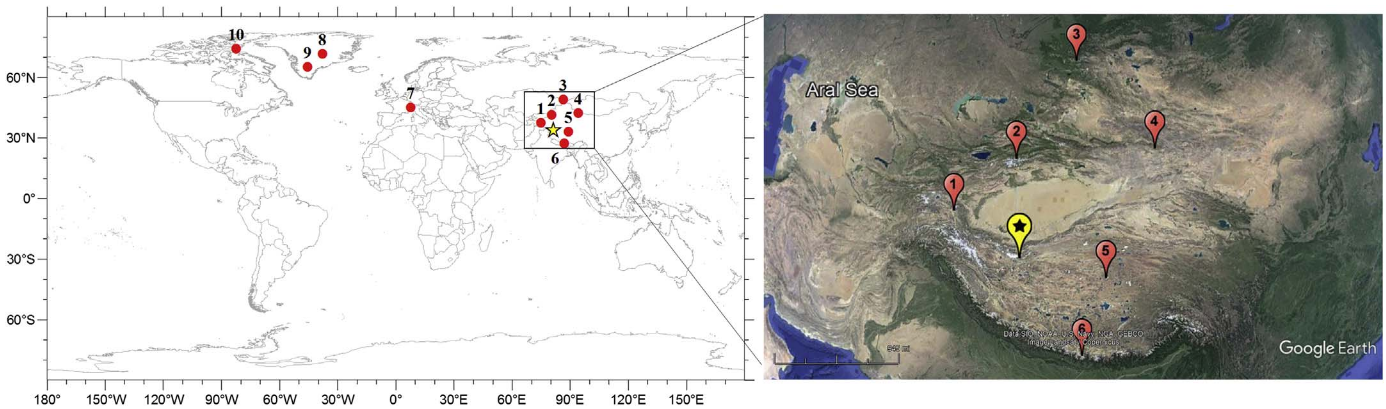

Fig. 1. Left: Map showing the locations of the Guliya Ice Cap (star) and other ice cores discussed in the text: 1. Muztagh Ata (East Pamirs); 2. Inilchek (Tien Shan); 3. Belukha (Siberian Altai); 4. Miaoergou (Eastern Tien Shan); 5. Puruogangri (Central Tibet); 6. Everest (Himalayas); 7. Colle Gnifetti (Alps); 8. Summit (Greenland); 9. ACT2 (Greenland); 10. Devon Island (Canadian High Arctic). Right: inset of the TP region showing the different ice core sites.

1. Introduction

Anthropogenic emissions (e.g., greenhouse gases, trace elements) to the atmosphere have dramatically increased since the Industrial Revolution in the 19th century. Trace elements (TEs) are emitted to the atmosphere from both natural sources including wind-borne dust particles, sea salt, volcanoes, forest fires and biogenic emissions (Nriagu, 1989a) and anthropogenic processes such as fossil fuel combustion, mining, ferrous and non-ferrous metal production, and waste incineration (Pacyna and Pacyna, 2001; Marx and McGowan, 2010). It was estimated that during the mid-1990s fossil fuel combustion (primarily coal and oil) was the dominant source of atmospheric TEs worldwide followed by metal production processes and mining (Pacyna and Pacyna, 2001; Marx and McGowan, 2010). Currently, coal, oil, and natural gas, are still the primary sources of global energy (BP, 2016). Coal has been an important fossil fuel since the onset of the Industrial Revolution and is expected to continue in wide use due to its abundant reserves around the world (Shafiee and Topal, 2009; BP, 2016).

High temperature processes such as fossil fuel combustion and pyrometallurgy generate fumes and fine particles (< 0.1 μm–10 μm) containing toxic metals (e.g., Cd, Zn, Pb) (Sobanska et al., 1999; Ohmsen, 2001) that if not captured by emission controls can have an atmospheric residence time of ~10 days, giving them sufficient time to be transported over long distances and subsequently deposited far from their emission sources (Quinn and Ondov, 1998; Pacyna and Pacyna, 2001; Marx and McGowan, 2010). Trace elements can have adverse effects on humans and other terrestrial and aquatic organisms depending on their toxicity and bioavailability (e.g., physical and chemical form).

Atmospheric TE monitoring programs, along with emission inventories, have been conducted in recent decades (Nriagu, 1989a; Olendrzynski et al., 1996; Pacyna and Pacyna, 2001; Tian et al., 2015). However, they lack pre-1900 information which is necessary to contextualize current atmospheric changes. Natural archives that receive only atmospheric input and that are geomorphically stable such as lake sediments, peat bogs and ice cores are best to reconstruct records of past atmospheric metal depositions (Cooke and Bindler, 2015; Gabrielli and Vallelonga, 2015; Hansson et al., 2015; Marx et al., 2016). These archives provide different advantages/disadvantages as explained below.

Lakes and ombrotrophic peat bogs are widespread around the world whereas ice fields are confined to polar and high altitude regions. Lakes and peat bogs are easier to access and sample. Because of their location, lake sediments and peat bogs provide local/regional information allowing the study of small and medium scale variability while glaciers receive long-range inputs recording large-scale variability. Glaciers are fed exclusively by atmospheric inputs which is a great advantage to study atmospheric metal depositions compared to peat bogs and lake sediments which can be subjected to both atmospheric and terrestrial inputs. Peat bogs, as opposed to ice cores, are dated based on indirect methods such as ^{14}C, decreasing their chronology precision. Other than TEs, several additional parameters (e.g., dust particles concentrations, soluble ions concentrations) can be measured in ice cores in co-registered samples to have a better understanding of the possible sources. Moreover, much higher temporal resolution can be achieved in ice cores (seasonal/annual) compared to peat bogs (decadal to centennial) and to lake sediments (decadal/centennial to millennial) (Shotyk, 1996; Shotyk et al., 1998; Martínez Cortizas et al., 2002; Kylander et al., 2010; Ferrat et al., 2012; Bradley, 2015; Cooke and Bindler, 2015; Gabrielli and Vallelonga, 2015; Hansson et al., 2015; Shotyk et al., 2017) making ice cores an extremely useful tool to study past atmospheric pollution.

Glaciers and ice sheets preserve atmospheric species that are deposited as snow accumulates over time, thus creating valuable records of past climatic/environmental conditions. Glaciers act as sinks of atmospheric species, but they can also serve as sources by releasing TEs in meltwater (Zhang et al., 2015a) which may affect the people who depend on glacial meltwater. Ice cores from polar glaciers and ice sheets have been used to obtain TE records. However, only a few non-polar ice core records (Hong et al., 2009; Kaspari et al., 2009; Lee et al., 2011; Eichler et al., 2014; Gabrielli et al., 2014; Uglietti et al., 2015; Wang et al., 2016; Beaudon et al., 2017) provide continuous information back to pre-industrial times. Thus, ice core records of TEs from mid- and low-latitudes are needed to assess the spatial and temporal extent and levels of pollution in the environment. This information can be used by modelers to assess pollution transport at local, regional, and global scales and by policy makers to develop strategies and policies to reduce their emissions.

The Tibetan Plateau (TP) comprises ~46,000 glaciers in an area of ~100,000 km^2 (Yao et al., 2012). These glaciers collectively contain the largest natural reservoir of ice outside of the Polar Regions and are the primary source for major rivers in Asia. The Guliya ice cap (35°17′N, 81°29′E; 6200 m asl) located in the western Kunlun Mountains on the Qinghai-Tibetan Plateau, China (Fig. 1), is the largest (> 200 km^2) ice cap in the subtropical zone. Guliya resembles a "polar" ice cap with low temperatures and low precipitation (Thompson et al., 1995, 1997).

The atmospheric circulation over the TP is controlled by the East Asian and South Asian summer monsoons, the westerlies that ultimately originate over the North Atlantic, and their interactions (Schiemann et al., 2009; Yao et al., 2013; Maussion et al., 2014). The pronounced seasonality on the TP affects the intensity and position of the westerly jet. During winter (December–February), the westerlies are strong and dominate over the TP as the result of a geopotential height