Chemosphere 187 (2017) 294–301

Contents lists available at ScienceDirect

Chemosphere

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

Pb concentrations and isotopic record preserved in northwest Greenland snow

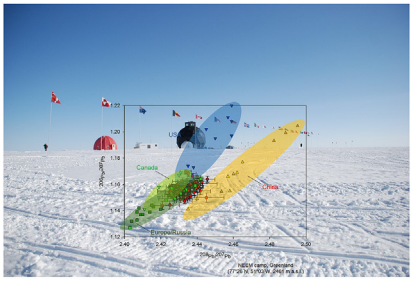
Jung-Ho Kang [a, *], Heejin Hwang [a], Changhee Han [a, b], Soon Do Hur [a], Seong-Joong Kim [c], Sungmin Hong [b, **]

[a] Division of Polar Paleoenvironment, Korea Polar Research Institute (KOPRI), 26 Songdomirae-ro, Incheon, 21990, Republic of Korea
[b] Department of Ocean Sciences, Inha University, Incheon, 22212, Republic of Korea
[c] Division of Polar Climate Sciences, Korea Polar Research Institute (KOPRI), 26 Songdomirae-ro, Incheon, 21990, Republic of Korea

HIGHLIGHTS

- Pb records investigated from a 3.2-m snow pit in northwest Greenland.
- Northwest Greenland snow is highly enriched with anthropogenic Pb.
- Pb concentrations exhibit seasonal peaks in winter–spring layers.
- $^{206}Pb/^{207}Pb$ ratios have gradually increased between 2003 and 2009.
- Contribution of Pb from China was very large through winter 2005–spring 2006.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 14 April 2017
Received in revised form
18 July 2017
Accepted 18 August 2017
Available online 30 August 2017

Handling editor: Martine Leermakers

Keywords:
Lead concentration
Lead isotope ratio
Enrichment factor
Greenland snow

ABSTRACT

We present high-resolution lead (Pb) concentrations and isotopic ratios from a northwest Greenland snow pit covering a six-year period between 2003 and 2009. Pb concentrations ranged widely from 2.7 pg g^{-1} to 97.3 pg g^{-1}, with a mean concentration of 21.6 pg g^{-1}. These values are higher than those recorded for the pre-industrial period. Pb concentrations exhibit seasonal spikes in winter–spring layers. Crustal Pb enrichment factors (EF) suggest that the northwest Greenland snow pit is highly enriched with Pb of predominantly anthropogenic origin. The $^{206}Pb/^{207}Pb$ ratios ranged from 1.144 to 1.169 with a mean value of 1.156, which fall between less radiogenic Eurasian-type and more radiogenic Canadian-type signatures. This result suggests that several potential source areas of Pb impact on northwest Greenland. Abrupt changes in Pb concentrations and Pb isotope ratios were observed and related to seasonal shifts in source regions of aerosol transport. The $^{206}Pb/^{207}Pb$ isotope ratio increased gradually between 2003 and 2009. The similarity of the three-isotope plot ($^{206}Pb/^{207}Pb$ versus $^{208}Pb/^{207}Pb$) between some of our samples and Chinese urban aerosols suggests a steadily increasing contribution of Chinese Pb to northwest Greenland snow.

© 2017 Published by Elsevier Ltd.

1. Introduction

Lead (Pb) concentrations in the Greenland ice sheet have altered dramatically as a result of anthropogenic Pb, which is influenced by civilization, industrialization, and environmental policy. The first

* Corresponding author.
** Corresponding author. Department of Ocean Sciences, Inha University, Incheon, 22212, Republic of Korea.
E-mail addresses: jhkang@kopri.re.kr (J.-H. Kang), smhong@inha.ac.kr (S. Hong).

http://dx.doi.org/10.1016/j.chemosphere.2017.08.101
0045-6535/© 2017 Published by Elsevier Ltd.

record of Pb pollution in the Greenland ice sheet dates from 2500 years ago, during the peak period of Greco-Roman civilization (Hong et al., 1994). Hong et al. (1994) measured Pb concentrations in ice sections of the 3028 m GRIP ice core dated from 7760 years–470 years ago. Natural Pb concentrations in the Greenland ice sheet were about 0.5 pg g^{-1} between 7700 and 3000 years ago, increasing to 3 pg g^{-1} at the apogee of the Roman Empire (i.e., six times higher than natural values) owing to Pb and Ag production during the Greek and Roman periods. Pb concentrations declined to 0.7 pg g^{-1} around 1500 years ago with the fall of the Roman Empire, and then increased continuously during Medieval and Renaissance times.

Pb concentrations in the Greenland ice sheet continued to increase steadily from the 1750s (~10 pg g^{-1}) to the 1900s (~50 pg g^{-1}) as a consequence of anthropogenic Pb emissions mainly from nonferrous metal smelting production and coal combustion in the Northern Hemisphere (Murozumi et al., 1969). Starting from the 1930s, a rapid increase was then observed in Greenland snow and ice, reaching peak concentrations in the late 1960s (~200-fold above the natural values) (Murozumi et al., 1969; Candelone et al., 1995; McConnell and Edwards, 2008). This enormous increase was mainly linked to the very extensive use of Pb additives in gasoline from the early 1920s (Nriagu, 1990). Increased Pb pollution, as recorded from the Greenland ice sheet, contributed to a change in environmental perception and to the adoption of environmental policies; for example, the Clean Air Act of 1970, which sought to reduce air pollution in the United States, focusing on reduced use of leaded gasoline. Policies to phase out leaded gasoline have been effective in reducing atmospheric Pb concentrations, with evidence for this found in Greenland snow. Following the decline in use of leaded gasoline in the United States and other countries, Pb concentrations in the Greenland ice sheet dropped significantly from the late 1960s onward (Boutron et al., 1991).

Pb concentrations in the Greenland ice sheet remain high compared to those of the pre-industrial period (Barbante et al., 2003). From 1981 to 1990, Pb concentrations in snow from central Greenland varied widely, from 4.3 pg g^{-1} to 160 pg g^{-1} (Sherrell et al., 2000). Following the global phase out of leaded gasoline (i.e., since the 1980s), Asian industrial emissions of Pb from coal combustion and metal smelting have become the principal source of Pb in the North Pacific atmosphere (Osterberg et al., 2008). Asia has become the largest anthropogenic source of trace metals in the atmosphere and China-emitted aerosols account for a substantial and growing fraction of anthropogenic Pb deposited in Greenland (Bory et al., 2014); however it is still unclear how much of anthropogenic Pb is deposited in Greenland and there are limited data indicating recent variations in Pb concentrations in Greenland snow.

The four stable isotopes of Pb (^{204}Pb, ^{206}Pb, ^{207}Pb and ^{208}Pb) can act as a powerful geochemical tracer (Komárek et al., 2008). In snow and ice, Pb isotope ratios can be used not only to distinguish natural Pb from anthropogenic Pb, but also to identify specific potential source areas. The Pb isotopic composition of ores and industrial Pb varies globally but is regionally consistent. By comparing Pb isotope ratios in natural archives to those of regional emission sources, atmospheric transport pathways and mixing processes can be determined. For example, the isotope characteristics of a three-isotope plot (^{206}Pb/^{207}Pb versus ^{208}Pb/^{207}Pb) in Greenland ice samples identified the Southern Spanish mining region as the dominant sources of anthropogenic Pb emissions between 680 BCE and 193 AD (Rosman et al., 1997). Similarly, a steep increase in the ^{206}Pb/^{207}Pb isotope ratio of Greenland snow core sections indicates that the United States became a significant source between 1967 and 1975, with the subsequent decrease in the ^{206}Pb/^{207}Pb isotope ratio after 1976 attributed to lower but constant input of Pb from Eurasia and Canada (Rosman et al., 1993, 1994).

In this study, we investigated temporal profiles of Pb concentrations and isotopic ratios from a 3.2 m depth snow pit near the North Greenland Eemian Ice Drilling (NEEM) camp in northwest Greenland. We investigated Pb concentrations and isotopic ratios to expand our understanding of recent variations in Pb deposition and Pb sources.

2. Material and methods

2.1. Field sampling

As a part of the NEEM project, we excavated a 3.2-m depth snow pit on 26 June 2009 at the NEEM study site (77°26′N, 51°03′W, 2461 m a.s.l.) in northwestern Greenland (Kang et al., 2015). We dug a snow pit using metal shovels and then shaved away about 10 cm of snow from the wall of the pit using pre-cleaned low density polyethylene (LDPE) shovels. A continuous series of 70 snow samples were collected by horizontally pushing an acid-cleaned polytetrafluoroethylene (PTFE) tube and hammer. The snow samples were placed in acid-cleaned 1 L LDPE bottles. Precautions were taken in the field to minimize the possibility of snow contamination (Hong et al., 2000). All sample bottles were transported frozen from the NEEM camp to the laboratory at the Korea Polar Research Institute (KOPRI) and kept frozen at −20 °C until analysis.

2.2. Sample preparation and instrumental analysis

Since contamination significantly affects the measurement of trace elements and Pb isotopes, we prepared the samples in a class 10 laminar airflow clean bench within class 1000 clean room facilities at KOPRI, and followed executed clean protocols to prevent contamination (Kang et al., 2015). Samples were thawed and aliquoted into 15 mL LDPE bottles. Bottles were cleaned prior to use, following a rigorous acid rinsing procedure (Hong et al., 2000).

All aliquots for trace element analysis were acidified to 1% “Optima” grade nitric acid (HNO_3) (Fisher Scientific, Canada). Concentrations of Pb and other trace elements were determined using Inductively Coupled Plasma-Sector Field Mass Spectrometry (ICP-SFMS; Element2, Thermo Scientific, Germany) equipped with an Apex HF high efficiency sample inlet system (Apex HF, Elemental Scientific, USA). Analytical instrumentation and ultra clean working conditions are described in detail by Hong et al. (2009). Special attention was given to daily optimization of instrumental parameters in order to obtain high intensities and minimize possible interference. Detection limits, defined as three times the standard deviation of 10 measurements of the blank (1% “Optima” grade HNO_3 solution in sub-boiled water) were 0.09 and 0.24 pg g^{-1}, for Pb and Ba, respectively. Data quality was estimated by analyzing the SLRS-5 riverine water certified reference material (National Research Council, Canada).

Full analytical procedures for Pb isotopic measurement are described in Han et al. (2015). Briefly, an approximately 10 mL aliquot was weighed into a preconditioned Teflon vial (Savillex, USA) containing 10 μL HNO_3 (Fisher “Optima” ultrapure grade), 20 μL hydrofluoric acid (HF, Merck “Ultrapur” grade), and 4 μL dilute phosphoric acid (H_3PO_4) (Merck “Suprapur” grade; approximately 5 wt% solution). The mixture was evaporated to dryness at sub-boiling temperature in a class 10 clean hood. The evaporated residue was mixed with a droplet of silica-gel activator and then transferred onto a degassed zone-refined rhenium filament. A

Download English Version:

https://daneshyari.com/en/article/5746424

Download Persian Version:

https://daneshyari.com/article/5746424

Daneshyari.com