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Industrial-era lead and mercury contamination in southern Greenland implicates North American sources

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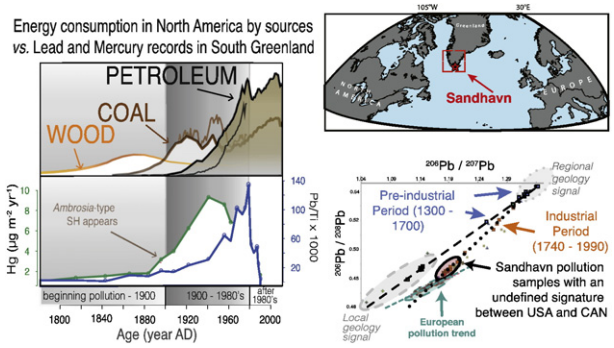
HIGHLIGHTS

- Lead (Pb) isotope and mercury (Hg) data are presented from southern Greenland.
- Two Pb geological sources (local and regional) and a pollution source were found.
- Atmospheric Hg and Pb pollution increase from the mid-18th century.
- Hg and Pb pollution show a similar trend to combustible consumption in North America.
- Pb isotopic data does not allow separation between sources in the USA and CA.

GRAPHICAL ABSTRACT



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ABSTRACT

To study the long-range transport of atmospheric pollutants from lower latitude industrial areas to the Arctic, we analysed a peat core spanning the last ~700 cal. yr (~1300–2000 CE) from southern Greenland, an area sensitive to atmospheric pollution from North American and Eurasian sources. A previous investigation conducted in the same location recorded atmospheric lead (Pb) pollution after ~1845, with peak values recorded in the 1970s, and concluded that a North American source was most likely. To confirm the origin of the lead, we present new Pb isotope data from Sandhavn, together with a high-resolution record for mercury (Hg) deposition. Results demonstrate that the mercury accumulation rate has steadily increased since the beginning of the 19th century, with maximum values of 9.3 μg m^{-2} yr^{-1} recorded ~1940. Lead isotopic ratios show two mixing lines: one which represents inputs from local and regional geogenic sources, and another that comprises regional geogenic and pollution sources. Detrending the Pb isotopic ratio record (thereby extracting the effect of the geogenic mixing) has enabled us to reconstruct a detailed chronology of metal pollution. The first sustained decrease in Pb isotope signals is recorded as beginning ~1740–1780 with the lowest values (indicating the highest pollution signature) dated to ~1960–1970. The $^{206}Pb/^{207}Pb$ ratio of excess Pb (measuring 1.222, and reflecting pollution-generated Pb), when compared with the Pb isotopic composition of the Sandhavn peat record since the 19th

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century and the timing of Pb enrichments, clearly points to the dominance of pollution sources from North America, although it did not prove possible to further differentiate the emissions sources geographically.

1. Introduction

The Arctic, including Greenland (Fig. 1), has experienced significant human impacts through the effects of long-range atmospheric transport of pollutants since ancient times. The oldest evidence of atmospheric metal pollution in Greenland dates back to the Carthaginian and Roman periods, and is attested to by an increase in lead (Pb) concentrations between 680 BCE and 193 CE measured in the Summit ice core (Hong et al., 1994). This was accompanied by a change in the Pb isotopic composition that suggested the source of pollution was from Spanish ores (Rosman et al., 1997). Evidence of medieval lead pollution in Greenland, dating to the 15th century AD, has been proposed using inverse modeling on data obtained from Lake Igaliku in southern Greenland (Massa et al., 2015). Before that study was conducted, no significant changes in the modern levels of atmospheric metal deposition had been identified prior to the 18th century AD in Greenland (Bindler et al., 2001b; Candelone et al., 1995; Michelutti et al., 2009; Murozumi et al., 1969; Rosman et al., 1994; Silva-Sánchez et al., 2015).

Most investigations into long-range atmospheric pollution in Greenland consider the Pb content and isotopic ratios in ice cores and lake sediments. By contrast, there are fewer records available for the accumulation of other heavy metals such as mercury (Hg). An increase in Hg content since the Industrial Revolution has been demonstrated in Greenlandic marine and lake sediments, as well as ice cores (Asmund and Nielsen, 2000; Bindler et al., 2001a; Dommergue et al., 2016; Faïn et al., 2009; Lindeberg et al., 2006), and the associated risks for Arctic wildlife and human populations have been highlighted (AMAP, 2011). Very few of these investigations have produced Hg records that extend into the pre-industrial era. Consequently our understanding of the long-term accumulation of Hg in the Greenlandic environment remains relatively poor.

In addition to some contribution from local sources, a long-distance origin is accepted for atmospheric pollutants recorded in Greenland (Skov et al., 2016). While the existence of a wide range of pollution origins existing simultaneously is likely (such as North America, Europe and Asia), strong control on atmospheric pollutant transport is exerted by seasonal Arctic and Subarctic air masses (Sturges and Barrie, 1989). The polar front is not zonally symmetrical and can extend as far south as ~40°N over Eurasia in January, thus making northern Eurasia the major source region for air pollution transport into the Arctic (Law and Stohl, 2007). As such, results from recent snow and atmospheric aerosols collected from the Canadian High Arctic (Shotyk et al., 2005a;

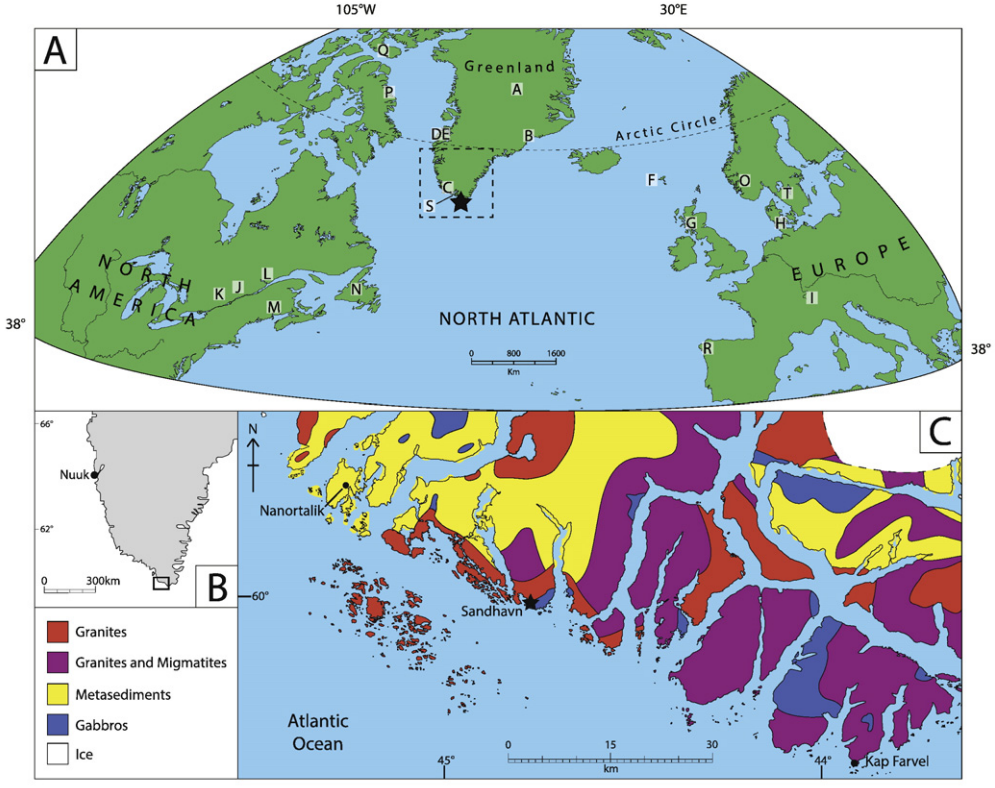

Fig. 1. (A) The location of Sandhavn (black star) and other places mentioned in the text. Key to labels: A Summit ice cores (Rosman et al., 1997, 1994, 1993); B rock samples from Kangerlussuaq (Taylor et al., 1992); C peat record in Tasiusaq, Greenland (Massa et al., 2015; Shotyk et al., 2003); D Akilia rocks, Greenland (Whitehouse et al., 2005); E lake sediments, Lake 53 and Lake 16, SW Greenland (Bindler et al., 2001a, 2001b); F peat record from the Faroe Islands (Shotyk et al., 2005a, 2005b, 2005c); G *Sphagnum* moss samples from Scotland (Farmer et al., 2002); H peat record from Denmark (Shotyk et al., 2003); I peat core from Schöpfenwaldmoor, Switzerland (Weiss et al., 1999); J & K lake sediments from Lake Ontario and Lake Erie (Graney et al., 1995); L & N aerosols samples from Chicoutimi and Newfoundland, Canada (Bollhöfer and Rosman, 2001); M peat record from Caribou bog, US (Roos-Barraclough et al., 2006); O peat records, Norway (Dunlap et al., 1999); P lake sediments, Lake CF8 Baffin Island (Michelutti et al., 2009); Q snow samples from Devon Island (Shotyk et al., 2005b); R peat records from NW Spain (Martínez Cortizas et al., 2012); S rock samples from south Greenland (Andersen, 1997; Colville et al., 2011; Kalsbeek and Taylor, 1985); T peat record from southern Sweden (Bindler, 2003). (B) Location of the study area (boxed) within Greenland. (C) Simplified geological map of the area around Sandhavn. (Data from GEUS, 2017)