Marine Chemistry 182 (2016) 38–50

Contents lists available at ScienceDirect

Marine Chemistry

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

Determining the pathways, fate, and flux of atmospherically derived trace elements in the Arctic ocean/ice system

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ARTICLE INFO

Article history:
Received 29 December 2015
Received in revised form 21 April 2016
Accepted 25 April 2016
Available online 30 April 2016

Keywords:
Atmospheric deposition
Aerosols
Arctic
Trace elements

ABSTRACT

Aerosol deposition is an important pathway for delivering trace elements, including those of anthropogenic origin, into the Arctic. Assessment of this process is difficult in the harsh Arctic environment, and limited field studies have forced a reliance on poorly constrained models. Here we use the cosmic ray produced radioisotope, ^{7}Be, to trace the atmospheric deposition of elements within the Arctic water/ice/snow system, and link aerosol concentrations to flux. Seawater, ice, snow, melt pond, and aerosol samples were collected during late summer 2011 as part of the RV Polarstern's ARK-XXVI/3 campaign. From the measured ^{7}Be inventories we determined an average ^{7}Be flux of 109 dpm/m^{2}/d, which is consistent with results from previous studies in the region. Snow, ice and melt ponds represent significant reservoirs of ^{7}Be, and the relative ^{7}Be inventory in ice increased through late August, as melt pond inventories decreased with onset of freezing. The total (water/ice/snow system) inventory was relatively constant across our transect, but mixed layer inventories increased towards lower latitudes as ice-free, open water was approached. The latter gradient drives transport of ^{7}Be, and presumably other atmospherically-derived species, towards the ice-covered ocean mixed layer. This is modeled by advective transport along the Transpolar Drift. The average ^{7}Be aerosol concentration was 0.0182 dpm/m^{3}. None of the lithogenic aerosol elements showed any significant enrichment above crustal composition, while the pollution-derived elements (Cr, Ni, Cu, Zn, Cd, Sb, Pb) showed varying degrees of enrichment relative to crustal values. Historical aerosol ^{7}Be data was used to derive a seasonal cycle in the ^{7}Be inventory that was calibrated to the inventory measured in this study, using an effective bulk (wet plus dry) deposition velocity of 1350 m/day. This deposition velocity was then used to estimate the seasonal atmospheric flux of aerosol trace elements.

1. Introduction

Atmospheric transport and deposition of aerosols is an important delivery mechanism of natural and contaminant trace elements to the Arctic (e.g. Davidson et al., 1981, 1985; Barrie, 1986; Macdonald et al., 2005; Shaw, 1995; Stohl, 2006; Hov et al., 2007; Law and Stohl, 2007; Frossard et al., 2011; Zhan and Gao, 2014). Existing data show that atmospheric deposition of contaminant elements like Hg, Pb, and Se may be a major input of these elements to the Arctic, with likely sources being anthropogenic — industrial or power plant emissions associated with fossil fuel combustion in Europe, Russia, and Asia (e.g. Barrie and Hoff, 1985; Maenhaut et al., 1989; Sirois and Barrie, 1999; Shevchenko et al., 2000, 2003; Gong and Barrie, 2005; Outridge et al., 2008; Dietz et al., 2009, Durnford and Dastoor, 2011). The atmospheric input of trace elements plays a key role in controlling biogeochemical processes in the ocean (e.g., Martin et al., 1990, Falkowski, 1997; Coale et al., 1996, Morel et al., 2003; Morel and Price, 2003; Krishnamurthy et al., 2009), and recent work suggests that this might be true in the Arctic as well (Taylor et al., 2013). These inputs have strong implications for the ecosystem, and even human health (e.g. Grebmeier et al., 2006; Garza, 2001; Pungowiyi, 2000; Parkinson and Berner, 2009). Assessment of this input is difficult however because measurements of deposition rates in remote ocean regions are rare, and particularly daunting in the Arctic because harsh conditions and limited research platforms make it difficult to obtain quality-controlled precipitation and aerosol chemistry measurements on a routine basis. Furthermore, the Arctic is complicated by the existence of different catchments (water/ice/snow) that partition the atmospheric input such that elements will have circuitous paths to the oceanic ecosystem. The seasonality of this partitioning between compartments (as a function of melt and sea ice extent) likely affects ocean chemistry and the ecosystem. Understanding the seasonality of aerosol trace element (TE) partitioning will provide insight into how the distribution of TEs will change as the Arctic ice-cover continues to evolve over the coming years.

Given the dearth of direct measurements, the ocean community has relied on atmospheric transport and deposition models (e.g., Gao et al., 2003; Hand et al., 2004; Jickells et al., 2005; Mahowald et al., 2005,

http://dx.doi.org/10.1016/j.marchem.2016.04.006
0304-4203/

2009) to assess aerosol fluxes. However these models are themselves poorly constrained as to the amounts of precipitation delivered to the ocean and the parameterization of aerosol removal processes. Alternatively, natural radionuclides delivered to the earth surface from the atmosphere serve as useful tracers of the input of atmospherically derived chemical species as the source terms are definable and measurements are not readily contaminated (Turekian et al., 1983; Kadko and Prospero, 2011). The information obtained from the use of such tracers can be used to characterize the deposition of trace elements. For example, the global distribution of the cosmogenic isotope ^{7}Be in the atmosphere and its deposition to the ocean has been modeled (Brost et al., 1991; Koch et al., 1996, 2006), and parameters developed in such models can be extended to the modeling of other species including those from anthropogenic sources (Kadko and Prospero, 2011; Kadko et al., 2015).

As described here, ^{7}Be is particularly well suited for studying the deposition of chemical species into the Arctic as the inventory of ^{7}Be, partitioned between the ice/ocean, is a direct measure of its rate of atmospheric input to these catchments (Cámara-Mor et al., 2011; Galfond et al., 2014). We use the inventory and distribution of ^{7}Be within the water column, ice, snow, and melt ponds to trace the partitioning and pathways of atmospherically-deposited elements within the Arctic system. By coupling the ^{7}Be and trace element aerosol concentrations, estimates of the atmospheric deposition flux of aerosol trace elements are also derived.

2. Background

Be-7 is a cosmic ray produced isotope (half-life = 53.3 d) that is deposited upon the ocean surface primarily by precipitation and subsequently homogenized within the surface mixed layer (e.g. Silker, 1972; Aaboe et al., 1981; Young and Silker, 1980; Kadko and Olson, 1996; Kadko, 2000; Kadko and Prospero, 2011; Kadko and Johns, 2011). The ^{7}Be flux and water column inventory vary as a function of rainfall, and over broad oceanic regions are relatively constant. This is manifested by the observation that water column inventories are inversely related to surface salinity (Young and Silker, 1980; Kadko and Olson, 1996; Kadko and Johns, 2011). In the low particle environment of the open ocean, numerous studies have shown that ^{7}Be is quite soluble thus allowing particle scavenging losses to be ignored (e.g. Silker, 1972; Andrews et al., 2008). Therefore, in the absence of physical removal processes other than radioactive decay, the water column inventory of the isotope represents an integration of the atmospheric input flux over approximately the previous mean-life (77 d) of the isotope, making it relevant to studies encompassing seasonal timescales.

Atmospheric deposition of ^{7}Be has been routinely measured for many years by precipitation collection on land, and long-term systematic studies of precipitation have shown that temporal variation of ^{7}Be deposition occurs over timescales ranging from single precipitation events to seasons (e.g. Olsen et al., 1985; Dibb, 1989; Todd et al., 1989; Baskaran et al., 1993). In Bermuda for example, a correlation between monthly precipitation and ^{7}Be flux was observed over the course of an 8 month study (Turekian et al., 1983), and more recently over a nearly two year period at the same location (Kadko and Prospero, 2011).

The standing crop (inventory) of ^{7}Be in the surface ocean (or snow/ice) also affords a way to determine the depositional flux of this isotope; at steady state the input flux of ^{7}Be (atoms min/m^{2}) should be balanced by the ^{7}Be inventory, or decay rate, integrated over the water column (dpm/m^{2}). Note that the standing crop of ^{7}Be in the ocean/ice system is insensitive to short-term (daily-weekly) variability of ^{7}Be input since ^{7}Be has a seasonal-time scale radioactive mean life, lending to the system an inertia with respect to short term input events. Aaboe et al. (1981) calculated open ocean standing crops of ^{7}Be in the Sargasso Sea and found the results comparable to that expected from previously collected precipitation data, indicating that the ^{7}Be flux for an oceanic location can be estimated by utilizing water column inventories. Kadko and Prospero (2011) made the first concurrent measurements of ^{7}Be deposition from rain (bulk) collectors on Bermuda with ocean profile measurements of ^{7}Be inventories at the nearby US JGOFS Bermuda Atlantic Time-Series study site (BATS) and Hydrostation S in the Sargasso Sea. The average oceanic ^{7}Be flux determined from the ocean inventory was nearly identical at the two sites (0.048 ± 0.010 dpm/cm^{2}/d^{1}), and matched the flux determined at Bermuda by precipitation (bulk) collection (weighted average 0.048 ± 0.027 dpm/cm^{2}/d^{1}). The 1-dimensional approach assumed here (no lateral effects) appears valid due to the relatively short half-life of ^{7}Be and the constancy of ^{7}Be deposition over broad latitudinal bands. Of relevance to this work, the ability to derive the atmospheric flux of ^{7}Be from its ocean/ice

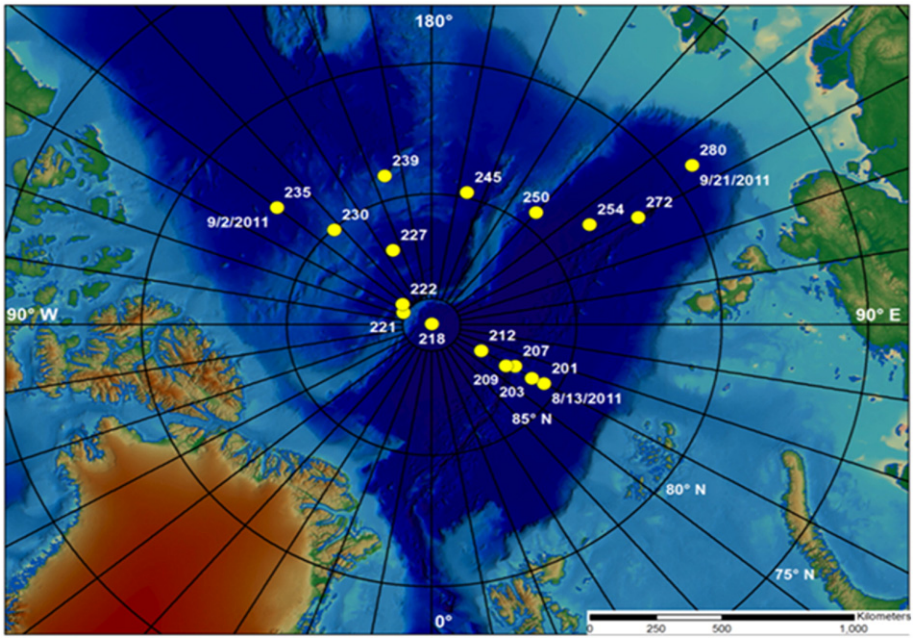

Fig. 1. Cruise track of ARK-XXVI/3, with start and termination dates indicated. Station locations marked by yellow circles with station numbers shown.