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 SCIENCES
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Q1 **Increasing chloride concentration causes retention**
 2 **of mercury in melted Arctic snow due to changes**
 3 **in photoreduction kinetics**

Q3 Q2 **E.A. Mann^{1,2,*}, S.E. Ziegler², A. Steffen³, N.J. O'Driscoll¹**

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1 0 A R T I C L E I N F O

12 Article history:
 18 Received 30 July 2017
 19 Revised 19 December 2017
 26 Accepted 8 January 2018
 26 Available online xxxx

22 Keywords:
 25 Mercury
 26 Photoreduction
 28 Arctic
 29 Melted snow

A B S T R A C T

Mercury (Hg) in the Arctic is a significant concern due to its bioaccumulative and neurotoxic properties, and the sensitivity of Arctic environments. Previous research has found high levels of Hg in snowpacks with high chloride (Cl⁻) concentrations. We hypothesised that Cl⁻ would increase Hg retention by decreasing Hg photoreduction to Hg(0) in melted Arctic snow. To test this, changes in Hg photoreduction kinetics in melted Alert, NU snow were quantified with changing Cl⁻ concentration and UV intensity. Snow was collected and melted in Teflon bottles in May 2014, spiked with 0–10 µg/g Cl⁻, and irradiated with 3.52–5.78 W·m⁻² UV (280–400 nm) radiation in a LuzChem photoreactor. Photoreduction rate constants (*k*) (0.14–0.59 hr⁻¹) had positive linear relationships with [Cl⁻], while photoreduced Hg amounts (Hg(II)_{red}) had negative linear relationships with [Cl⁻] (1287–64 pg in 200 g melted snow). Varying UV and [Cl⁻] both altered Hg(II)_{red} amounts, with more efficient Hg stabilisation by Cl⁻ at higher UV intensity, while *k* can be predicted by Cl⁻ concentration and/or UV intensity, depending on experimental parameters. Overall, with future projections for greater snowpack Cl⁻ loading, our experimental results suggest that more Hg could be delivered to Arctic aquatic ecosystems by melted snow (smaller Hg(II)_{red} expected), but the Hg in the melted snow that is photoreduced may do so more quickly (larger *k* expected).

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Q4 **Introduction**

47 **Mercury speciation and reactivity in the arctic**

48 Mercury (Hg), a neurotoxic and bioaccumulative environmental
 49 contaminant, exists in three main environmentally relevant
 50 forms: elemental (Hg(0)), divalent (Hg(II)) and methyl mercury
 51 (MeHg⁺) (Schroeder and Munthe, 1998; Steffen et al., 2008). Hg(0)
 52 is the main form found in the atmosphere (Lindqvist and
 53 Rodhe, 1985; Morel et al., 1998a; Wängberg et al., 2007), while

Hg(II) primarily exists in condensed phases, like water, soils and 54
 snow (Dommergue, 2003; Fain et al., 2006; Ferrari et al., 2004b; 55
 Moore and Castro, 2012; Obrist et al., 2014; Sigler and Lee, 2006). 56
 MeHg⁺ is the bioaccumulative and neurotoxic form of Hg, and as 57
 such is found in organisms (Lindberg et al., 2002; Morel et al., 58
 1998b) as well as condensed phases (Ramlal et al., 1993). Hg can 59
 move between these different species in the environment, 60
 thereby altering its environmental fate. 61

The Arctic is a Hg sensitive region, where organisms have 62
 been found to accumulate high Hg loads (Carrie et al., 2010; 63

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Kirk et al., 2012; Muir et al., 1992). Snow is one way by which Hg enters the surface environment and, once melted, may act as a vector for Hg transport into aquatic ecosystems where organisms may be exposed (Dommergue et al., 2010; Loseto et al., 2004). Hg deposited to snow by wet and dry deposition can be very labile, undergoing a number of reactions that may alter speciation in the snowpack prior to its melt (Lalonde et al., 2002a; Poissant et al., 2008; Schroeder and Munthe, 1998). The primary mechanism for Hg loss from snow prior to the snow melt period is through photochemical reduction of Hg(II) to Hg(0) (Durnford and Dastoor, 2011; Lalonde et al., 2002b; Mann et al., 2014; Mann et al., 2015b; Poulain et al., 2004), where Hg(II) bound to particulate matter in the snowpack is not thought to be susceptible to photoreduction (Brooks et al., 2006). Many factors, including incident radiation intensity (Mann et al., 2015b), temperature and snow age (Mann et al., 2015a), as well as chemical composition of the snowpack (Durnford and Dastoor, 2011; Mann et al., 2014) may be important to snowpack Hg photoreduction processes. Since Hg(0) does not sorb appreciably to ice crystals (Bartels-Rausch et al., 2008; Ferrari et al., 2004a), photoproduct Hg(0) can move from the snowpack back to the atmosphere with chemical/temperature gradients (Albert and Shultz, 2002; Anderson and Neff, 2008; Kuhn, 2001) and through snowpack ventilation (Ferrari et al., 2005). Alternatively, Hg(0) can be photooxidized back to Hg(II) and remain or subsequently react in the snowpack (Lalonde et al., 2003; Poulain et al., 2004). A full discussion of Hg(0) photooxidation is beyond the scope of this work, but further information can be found in reviews by Durnford and Dastoor (2011) and Mann et al. (2014).

As snow melts, the contaminants it contains can be transported with meltwater. Ionic components, including Hg, leave primarily in the first meltwater fractions (Dommergue et al., 2010; Kuhn, 2001), effectively becoming concentrated in the aqueous phase during first days of the snow melt period (Dommergue et al., 2010). In the Arctic, depending on the location of the snowpack, and the environmental conditions at the time of melt, this meltwater may collect into melt water ponds (Aspmo et al., 2006) and sit over sea ice for some time where photoreactions may potentially occur, or move directly into lakes, wetlands and the Arctic Ocean, transporting the contained Hg to these ecosystems for further reactions and/or incorporation into food webs.

Melt water ponds, which can form on Arctic sea ice during the polar melt period, are a transient environmental compartment, and are likely to contain snow melt water (Aspmo et al., 2006). Melt water ponds can be 1 to 10 m in diameter, with typical depths from 10 to 30 cm (Aspmo et al., 2006). The overall area covered by melt ponds is highly variable, though at their peak, these ponds may cover 20%–50% of the Arctic sea ice surface (Eicken et al., 2002). Relatively few studies have reported melt water pond evolution or coverage through time; however, Polashenski et al. (2012) concluded that evolution of melt water ponds in northern Alaska occurred as a three stage process. These stages included an initial rapid increase of melt water pond coverage (stage I), a stage of melt pond draining where areal coverage declined (stage II), and a final gradual increase in melt water pond coverage with the end of the melt season (stage III) (Polashenski et al., 2012). Since a portion of the Hg remaining in the snow pack until melt has

been found to be transported to receiving bodies with melting snow (Dommergue, 2003; Dommergue et al., 2010), these melt water ponds would also contain Hg from the melted snowpack. While water sits in these melt water ponds, photoreactive contaminants, like Hg, may have the potential to undergo reaction and change speciation. The presence of Hg in such melt water pond systems has been observed (Aspmo et al., 2006); however, the behaviour of contained Hg over the lifetime of these melt ponds has not been considered in detail. As a result, while the data presented in this study has potential use as a first approximation of the kinetic changes that may be expected to occur for Hg in frozen snow with changing Cl⁻ concentrations, they also provide information regarding Hg behaviour in this environmental compartment that is currently underrepresented in the general consideration of Arctic Hg cycling.

An accurate description of the photochemical dynamics of Hg reactions in both frozen and melted snow is clearly important for correctly describing and predicting Hg behaviour in Arctic environments. Interactions of Hg with ions are important, but presently some are poorly quantified with regard to the effects on Hg photochemical reaction kinetic parameters; specifically, chloride (Cl⁻) is suspected to affect Hg photochemical reactions in Arctic snow, but the magnitude of the effect on Hg photoreaction kinetics has not been fully quantified (Mann et al., 2014), nor have the effects of Cl⁻ on Hg in Arctic snow melt been investigated.

Mercury and chloride interactions

Elevated Cl⁻ concentrations have been found to co-exist with high Hg levels in Arctic snow, which has led to the conclusion that Cl⁻ may help retain Hg in these environments; however, an investigation of the effects of Cl⁻ on the kinetics of Hg reduction in frozen and/or melted snow has not been undertaken. Chloride concentrations of 120 mg/g have been measured in frost flowers near a lead in Arctic sea ice (Douglas et al., 2005), and Domine et al. (2004) determined that wind deposition of Cl⁻ resulted in Cl⁻ concentrations of ~16 mg/g in a Ny Alesund snowpack. Several groups have measured increasing Hg in Arctic snow with decreasing distance from oceanic coasts, or leads/polynyas (Constant et al., 2007; Douglas et al., 2005; Garbarino et al., 2002a), and higher Hg concentrations have been observed in snow over sea ice, as compared to inland snow (Poulain et al., 2007a; Steffen et al., 2013). These high Hg concentrations observed have been proposed to be due to Cl⁻ content in one of two ways: either by enhancing the photooxidation of Hg(0) within or above the snowpack, or by decreasing the extent of photochemical reduction of Hg(II) through the formation of a Hg form that is more resistant to photoreduction (chlorocomplexes, particle bound Hg); in either case, the net result is a decrease in the relative loss of Hg as Hg(0) from the snowpack. Photooxidation of snowpack Hg(0) has been proposed to be more favourable, or to occur to a greater extent in snow with higher Cl⁻ concentrations (Amyot et al., 2003; Lahoutifard et al., 2006; Lalonde et al., 2003), and lower fluxes of Hg(0) evasion have been measured in snow over sea ice compared to inland snow (Steffen et al., 2013). Lalonde et al. (2003) found that photoreduction was suppressed in snow samples containing a higher Cl⁻ concentration, and St. Louis 181

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