

NSF OPP 1854462: Collaborative Research: Constraining the role of chemical transformations on the cycling of mercury at the Arctic Ocean air-sea interface.

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Air-sea exchange is the dominant source and the major sink for ocean mercury (Hg). The loss of Hg as elemental Hg (Hg⁰) from the surface ocean to the atmosphere prolongs the lifetime of Hg in the biosphere, but also mitigates the buildup of Hg in ocean waters. Mercury inputs into the atmosphere are from natural and predominantly anthropogenic sources. The changing level of methylmercury (MeHg), the most bioaccumulative form of Hg in seafood, and that of greatest health concern, is directly related, with some lag in response, to the changing inputs of Hg into the atmosphere from anthropogenic activities. In the Arctic, the role of air-sea exchange in mitigating inputs of Hg to the Arctic Ocean is less well understood. Furthermore, there is disagreement over the relative importance of atmospheric inputs relative to rivers and other coastal inputs, and the role of Hg⁰ evasion, both from the ocean and from the surrounding terrestrial environment, especially during the substantial snow and ice melt of the Arctic spring. Compared to the global ocean, where ~90% of the Hg inputs are returned to the atmosphere by gas evasion of Hg⁰, the modeled evasion losses for the Arctic Ocean ranges from 5 to 56% of total inputs suggesting a lesser role for gas evasion in this region. Overall, there is substantial uncertainty over the magnitude of Hg sources and sinks to the Arctic Ocean, and fluxes, particularly the gas evasion rate of Hg⁰, are not well constrained by measurement. Estimating the magnitude of Hg⁰ evasion is complicated by the existence of significant transformations between the oxidized and reduced forms of Hg (Hg^{II} and Hg⁰) in the marine boundary layer and in the ocean mixed layer.

In this work, we will address this lack of experimental evidence to gain further insights into Hg cycling across the air-sea interface of the Arctic Ocean and improve our estimates of the gas exchange flux. If the evasion of Hg⁰ from the surface waters of the Arctic could be accurately assessed, it would then be possible to estimate the impact of redox reactions and microbial processes on the relative concentration and distribution of the oxidized and reduced forms of Hg in the Arctic surface waters. We will use an independent gas-exchange tracer based on the ²²²Rn-²²⁶Ra pair to accomplish this goal. In addition, as

more studies on the photochemical and dark oxidation and reduction of Hg in Arctic surface waters are required, we will perform such experiments at sea. The field work will take place in late May-June 2021 along a transect from open water, through the



Ice cover map for May 1 (lighter shading) and June 20, 2018 (grey shading) illustrating the change in ice cover in spring 2018. The likely extent of the cruise into the ice with station locations (circles) are shown. Ice coverage from NSIDC (2018) website.

marginal ice zone and into completely covered ice regions (see Figure).the USCGC Healy. The on-board experiments will be augmented by post-cruise experiments on land.