

# Isotopic Attribution of Atmospheric Species and Sources of Mercury to the Arctic Ocean Ecosystem

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The contribution of atmospheric species ( $\text{Hg}^0$ ,  $\text{Hg}^{2+}$ ) and sources of mercury (Hg) to the Arctic Ocean have been subject to scientific debate. Mercury stable isotopes reveal the relative importance of various atmospheric species and their sources in marine zooplankton, sampled from coastal to open ocean (Chukchi Sea, Beaufort Sea) at two different depths (0-surface, 0-chlorophyll maxima) and in three size fractions (0.2-1 mm, 1-5 mm, >5 mm). The Hg concentrations in zooplanktons are similar between the two size fractions of 0.2-1 mm ( $62.2 \pm 36.0 \mu\text{g/kg}$ ,  $n=14$ ) and 1-5 mm ( $67.9 \pm 46.1 \mu\text{g/kg}$ ,  $n=18$ ), but much lower in the >5 mm fraction ( $39.1 \pm 26.1 \mu\text{g/kg}$ ,  $n=9$ ). The Hg isotope ratios exhibit significant relationships with the Hg concentrations in the smallest size fraction of zooplankton (0.2-1 mm;  $\delta^{202}\text{Hg} = -0.26 \pm 0.41\text{‰}$ ,  $\Delta^{199}\text{Hg} = -0.15 \pm 0.22\text{‰}$ ) but not in other fractions (1-5 mm;  $\delta^{202}\text{Hg} = -0.13 \pm 0.44\text{‰}$ ,  $\Delta^{199}\text{Hg} = -0.06 \pm 0.18\text{‰}$ , >5 mm;  $\delta^{202}\text{Hg} = -0.36 \pm 0.38\text{‰}$ ,  $\Delta^{199}\text{Hg} = 0.21 \pm 0.17\text{‰}$ ), indicating that the small zooplanktons are effective in integrating Hg sources from the surrounding environment. We apply Hg isotope ratios of the small size zooplankton to an isotopic mixing model, previously established to quantify the contribution of atmospheric  $\text{Hg}^0$  and  $\text{Hg}^{2+}$  to the global ocean seawater and biota (Jiskra et al 2021<sup>[1]</sup>). The zooplanktons ( $\Delta^{200}\text{Hg} = 0.00 \pm 0.04\text{‰}$ ) reveal large proportions of  $\text{Hg}^0$  ( $71.4 \pm 18.6\%$ ) relative to  $\text{Hg}^{2+}$  ( $28.6 \pm 18.6\%$ ), consistent with the results modelled by Jiskra et al (2021) and recent results suggesting that  $\text{Hg}^0$  contribution is underestimated in the global ocean. Furthermore, we use the measured Hg isotope ratios of sampled total gaseous Hg ( $\text{Hg}^0$ ) and riverine dissolved Hg ( $\text{Hg}^0$ ,  $\text{Hg}^{2+}$ ) as well as snow/rainfall ( $\text{Hg}^{2+}$ ), reported by Campeau et al (2022)<sup>[2]</sup> and Obrist et al (2017)<sup>[3]</sup>, as end-members to estimate the sources of  $\text{Hg}^0$  to marine zooplanktons. We find that total gaseous Hg ( $43.4 \pm 24.4\%$ ) and riverine dissolved Hg ( $34.9 \pm 22.8\%$ ) are equally important as a source of  $\text{Hg}^0$  to the zooplanktons. Future analyses will be conducted in zooplanktons in relation to oceanic depth and distance from coastal region to provide deeper understanding of Hg sources and exposure pathways to the Arctic Ocean ecosystems.

[1] Jiskra (2021), Nature 597(7878), 678-682.

[2] Campeau (2022), Sci. Total Environ. 806, 150808.

[3] Obrist (2017), Nature 547(7662), 201-204.