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# Mercury cycling in the Arctic – does enhanced deposition flux mean net-input?

*Ralf Ebinghaus*

GKSS Research Centre Geesthacht, Institute for Coastal Research, Max-Planck-Str. 1, D-21502 Geesthacht, Germany. Email: ralf.ebinghaus@gkss.de

**Environmental context.** Mercury has unique physico-chemical characteristics that include long-range atmospheric transport, transformation into highly toxic methylmercury species, and the bioaccumulation of these compounds, especially in the marine environment. This has motivated intense international research on mercury as a pollutant of global concern. With respect to Polar regions, scientific interest and research activities were even accelerated after the discovery of the so-called atmospheric mercury depletion events (AMDEs), which are supposed to lead to enhanced mercury deposition flux into these pristine environments in the ecologically very sensitive period in polar spring.

The polar ecosystems are generally considered to be the last pristine environments of the earth. The Arctic, for example is populated by few people, has minimal commercial fishing, little industrial activity (except for some areas in the Russian Arctic[1]) and is, therefore, perceived to be relatively unaffected by human activity. In comparison, Antarctica is considered to be even less affected by any kind of anthropogenic influences.

Once contaminants reach the Polar regions, their lifetime in the troposphere depends on local removal processes. A totally unexpected finding on mercury in the Polar atmosphere was made in 1995. It was discovered that, during springtime, unexpectedly low concentrations of gaseous elemental mercury occurred in the Arctic air. This was surprising for a pollutant supposed to have a fairly long atmospheric residence time of six months to two years. This finding, the so-called atmospheric mercury depletion events (AMDEs), had significant influence on global mercury research and monitoring activities related to mercury cycling in Polar regions. Only five years after the first scientific publication of AMDEs in *Nature* by Schroeder et al.,[2] more than 200 papers dedicated to the occurrence and environmental significance of this phenomenon have been published in the peer-reviewed literature. It is now well established that AMDEs are an annually recurring polar spring-time phenomenon that result in a deposition flux. However, whether the result in total is a *net*-deposition is still not clear.

Mercury and many of its compounds exhibit unique behaviour in the environment because of their volatility, capability for methylation, and subsequent bioaccumulation, in contrast with most of the other heavy metals. Hg is emitted into the atmosphere from several natural as well as anthropogenic sources. Experimental field data and model estimates indicate that anthropogenic mercury emissions are at least as great as those from natural sources.[3–6] It is assumed that anthropogenic emissions lead to a general increase in Hg on local, regional, and global scales and that the increase in the overall global deposition since pre-industrial times is about a factor of $3 \pm 1$.[7]

Long-range atmospheric transport, the transformation into more toxic methylmercuric compounds, and their biomagnification in the aquatic foodchain have motivated intensive research on Hg as a pollutant of global concern. Hg is on the priority list of a large (and increasing) number of international agreements, conventions, and national advisories aimed at the protection of the environment including all compartments, human health, and wildlife (e.g. AMAP, UN-ECE, HELCOM, OSPAR, and many others).

In the atmospheric environment the most important species are gaseous elemental Hg, divalent reactive gaseous mercury (RGM), which consists of various oxidised $Hg^{II}$ compounds, and particle-bound Hg, which consists of various Hg compounds. It should be noted that information on the speciation/fractionation of these different chemical and physical forms is largely operationally defined.

Conversions between these different forms provide the basis of Hg's complex distribution pattern on local, regional, and global scales.

AMDEs were initially considered to result in an important net input of atmospheric mercury into the polar ecosystems during the spring period. But more recent studies, including the work

*Ralf Ebinghaus is an analytical and environmental chemist and heads the Department for Environmental Chemistry of the Institute for Coastal Research at GKSS Research Centre. He is a Professor at the University of Lüneburg and teaches Atmospheric and Marine Chemistry. Since 2003, Dr Ebinghaus has been an invited lecturer at the annual European Research Course on Atmospheres (ERCA). He has published more than 60 scientific papers and has co-edited two books and a special journal issue on mercury and related fields. Presently, Dr Ebinghaus is a member of the Steering Committee of the 9th International Conference on Mercury as a Global Pollutant, to be held in Guiyang, China, June 7–12 2009.*





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in this Research Front, point out that complex environmental processes take place after deposition that may result in less significant net-inputs from the atmosphere, since a certain fraction (or even the entire mass loading?) of deposited mercury may be recycled.

Jaffe and Strode[8] provide further evidence that Asian emissions account for more than 50% of the global anthropogenic input into the atmosphere. Major source categories are coal combustion and smelting, and observations indicate that they are significantly larger than accounted for in emission inventories. Mercury is an element and a natural constituent of the Earth's system, but once removed from the Earth's surface crust by anthropogenic activities this fraction will diversify and add to the natural biogeochemical cycling and pattern. Mercury that is not deposited locally is available for transport to the global atmosphere.

Choi and Grandjean[9] review human methylmercury (MeHg) toxicology and the effects of MeHg exposure on neurobehavioural and cardiovascular outcomes, based on epidemiological data.

Mercury health risks are mainly related to MeHg, which is globally found in seafood and freshwater fish, and constitutes the dominant source of human exposure. Especially in Arctic marine wildlife, MeHg concentrations can be very high, and pose a possible risk to northern indigenous communities who use these animals for food, often extensively. It is known that the developing brain and the brain function of children is adversely affected by this contaminant. More recent results indicate that MeHg may also promote the development of heart disease. On the other hand the authors clearly point out that certain essential nutrients in fish and seafood may provide beneficial effects on brain development, and may protect against the development of heart disease. One major conclusion is that future studies should assess both beneficial and adverse effects of fish and seafood and that regulatory agencies should develop balanced risk communication strategies that include these two sides of a consumer diet rich in seafood and fish.

Studies on AMDEs have previously revealed evidence (based on snow samples) that deposited Hg enters the Arctic ecosystem where it is potentially harmful. Hedgecock et al.[10] now question whether this fear is grounded. They report a photochemical box model that reveals evidence that the net deposition in fact appears to be minimal.

A major environmental and health concern is whether atmospheric mercury deposited during AMDEs leads to enhanced production and uptake of highly toxic MeHg species in polar ecosystems.

Hammerschmidt and Fitzgerald[11] show that, unexpectedly, the magnitude of deposition rates and the occurrence of MeHg in biota samples (here mosquitoes) are not directly linked. Deposition rates directly at the Arctic coast are estimated to be 20-fold higher than 200 km inland; however, the authors conclude that the ecosystem impact is comparable, possibly due to photochemically induced reemission of mercury previously deposited during AMDEs.

This study supports model estimates (such as those presented by Hedgecock et al.[10]), that the net mercury input is lower than the gross deposition after AMDEs.

Outridge et al.[12] present a detailed and comprehensive mass balance study to provide insight into sources, sinks, and processes that may be responsible for observed mercury concentrations and trends in Arctic biota. They conclude that the total mercury input on an annual basis, including wet deposition, coastal erosion, seawater import, and seasonally occurring AMDEs, is almost in balance with the annual outputs, mainly driven by shelf sedimentation and seawater export.

In general it can be concluded that, during the last decade, significant progress in our understanding of the global biogeochemical cycling of mercury has been made by intensified international research activities, promoted by the discovery of AMDEs in polar environments. New knowledge has been generated by field measurements, laboratory studies, and modelling exercises and scientific consensus has been achieved on several points, including chemical processes and environmental boundary conditions as prerequisites for AMDEs.

It is widely accepted that AMDEs lead to a deposition flux; however, the ultimate fate of deposited mercury in Arctic environments is much less clear.

The papers in this Research Front issue provide valuable new information and opinion on Asian mercury emissions, mercury cycling in Polar regions, and the toxicological relevance. While these papers not only provide new findings and answers they also pose several important questions and provide a snapshot of current research directions to set a useful scene for future research.

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