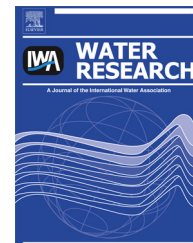




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Extremely elevated methyl mercury levels in water, sediment and organisms in a Romanian reservoir affected by release of mercury from a chlor-alkali plant

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ABSTRACT

We examined mercury (Hg) biogeochemistry and biomagnification in the Babeni Reservoir, a system strongly affected by the release of Hg from a chlor-alkali plant. Total mercury (THg) concentrations in river water reached 88 ng L⁻¹ but decreased rapidly in the reservoir (to 9 ng L⁻¹). In contrast, monomethylmercury (MMHg) concentrations increased from the upstream part of the reservoir to the central part (0.7 ng L⁻¹), suggesting high methylation within the reservoir. Moreover, vertical water column profiles of THg and MMHg indicated that Hg methylation mainly occurred deep in the water column and at the sediment–water interface. The discharge of Hg from a chlor-alkali plant in Valcea region caused the highest MMHg concentrations ever found in non-piscivorous fish worldwide. MMHg concentrations and bioconcentration factors (BCF) of plankton and macrophytes revealed that the highest biomagnification of MMHg takes place in primary producers.

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1. Introduction

Mercury (Hg) is considered a priority hazardous substance because of its toxicity (WFD, 2008). Although the toxic effects of Hg depend on the chemical species, all forms may damage the central nervous system (Harada, 1995). Human exposure to Hg takes place mainly via the intake of the organic form monomethylmercury (MMHg) through fish consumption (Clarkson, 1993, 1998; WHO, 2004; UNEP, 2013). Anthropogenic emission sources of Hg covered by Greenhouse Gas and Air Pollution Interactions and Synergies model constitute about 30% of total atmospheric Hg emissions, while the remainder is attributed to the natural sources and re-emissions (Amann et al., 2011; UNEP, 2013). Although atmospheric emissions dominate Hg inputs on a global scale, the direct release of Hg into terrestrial and aquatic ecosystems from industrial processes have impacted numerous sites (Stewart et al., 2011). Localized Hg release can occur as a result of industrial processes that use Hg such as chlor-alkali facilities. The European chlor-alkali industry accounts for 18% of global Hg consumption, with 50% of European consumption used for Hg-cells (UNEP, 2012). In 2011, 40% of chlorine and caustics production in Europe still used Hg-cell technology (Eurochlor, 2013). Hg released from chlor-alkali plants can enter aquatic systems where it may be converted to MMHg. The formation of MMHg in freshwater systems is influenced by a wide variety of environmental factors, including temperature, pH, redox potential, the presence of inorganic and organic complexing agents, total microbial activity and the concentration of bioavailable Hg rather than the total Hg pool (Jonsson et al., 2012). Hg methylation is mediated mainly by sulphate-reducing bacteria (SRB) and iron-reducing bacteria (FeRB) (Pak and Bartha, 1998; Fleming et al., 2006; Parks et al., 2013). The highest Hg methylation occurs mostly at oxic/anoxic transition zones in sediments (Compeau and Bartha, 1985; Gray and Hines, 2009) or in hypolimnetic water (Eckley and Hintelmann, 2006).

Hg forms must be bioavailable to be methylated, accumulated and retained within the tissues of organisms. The enrichment of MMHg in plankton and consequently in the food chain has been explained by a greater concentration of MMHg in the algal cytoplasm compared to inorganic-Hg (IHg), which is typically associated with cellular membranes (Bloom, 1992; Mason et al., 1996; Wu and Wang, 2011). The behaviour of Hg in aquatic systems and Hg accumulation in biota differ from one system to another and MMHg concentration in fish may be 10^7 times higher than in water (Qureshi et al., 2009; Riva-Murray et al., 2013).

Previous research in the Babeni Reservoir revealed that sediments and fish are highly Hg contaminated due to discharge from a chlor-alkali plant that uses Hg-electrodes (Bravo et al., 2009, 2010). The Olt River is an important source of fish for the local population, thus Hg-contamination poses a severe health risk to those consuming large amounts of fish from the Babeni Reservoir, i.e. anglers (Bravo et al., 2010). The main aim of this study was to determine the fate of Hg in a reservoir affected by release of Hg from a chlor-alkali plant. We therefore investigated (i) Hg methylation in water column and sediments (ii) downstream transport of Hg as well as (iii) MMHg biomagnification in food chains. This knowledge is critical for

the development of adequate remediation and risk management strategies aimed to minimize the impact of MMHg on consumers. The present study is of interest not only because it describes a severe case of contamination, but also because it traces the fate of Hg from the sources to the consumers by analysing the processes that may be applicable for similar Hg contaminated systems worldwide.

2. Materials and methods

2.1. Study area

The largest chlor-alkali production plant in Central and Eastern Europe is located close to the Olt River in Rm Vâlcea, an urban area of about 90 km² and 110,000 inhabitants. Characteristics of the Babeni Reservoir (Fig. 1) have been described previously (Bravo et al., 2010). Briefly, the reservoir has a mean volume of $35.3 \cdot 10^6$ m³, a surface area of $90.5 \cdot 10^5$ m², a mean depth of 3.9 m and a maximum depth of 16 m. The Babeni Reservoir is eutrophic with average concentrations (measured between 2000 and 2004) of total phosphorus of 1.2 ± 1.5 μ M and total nitrogen of 85 ± 2 μ M. Concentrations of SO_4^{2-} and Cl^- ranged from 343 μ M to 552 μ M and 3.5 mM–7.2 mM, respectively. Electrical conductivity and pH at 1.5 m depth ranged from 519 μ S cm⁻¹ to 1142 μ S cm⁻¹ and from 7.8 to 8.7, respectively (Data of Apele Romane Olt, see acknowledgements).

The Ionesti Reservoir (Fig. 1) has a surface area of $42.7 \cdot 10^4$ m² and a maximum depth of 12 m. The Valcea Reservoir is located upstream of the industrial platform and was used to determine the local background levels of Hg and MMHg concentrations. This study presents the results of two sampling campaigns in the three reservoirs during October 2007 (autumn) and August 2009 (summer).

2.2. Sampling

2.2.1. Water column

Surface waters (1.5 m depth) were collected in summer 2009 with a GoFlo[®] bottle (PVC). Water samples were collected in the Valcea Reservoir (VWC1, VWC2, VWC3, VWC4) and the Babeni Reservoir: 6 along a longitudinal transect (BWC1, BWC2, BWC3, BWC4, BWC5, BWC6) and 2 more samples (BWC7 and BWC8) in a vertical profile at location BWC6 (Table 1, Fig. 1). For THg and MMHg measurements water of 0.5 L per sample was immediately filtered (0.45 μ m, Sterivex[®]-HV) on the boat, placed in Teflon[®] bottles and acidified (1% v/v final concentration; HCl Ultrex II, J. T. Baker[®]). Water column suspended particulate THg and MMHg samples were collected by filtering 0.5 L through hydrophilic PTFE membranes with 0.45 μ m pore size (LCR, Millipore) (Cossa and Gobeil, 2000).

2.2.2. Sediment cores

Sediment cores were collected using a 60-mm gravity corer (Uwitec, Austria). Two cores were collected in October 2007 (VC1B and BC1B). Cores with about 40 cm of overlying water were sectioned within 12 h in a glove-bag under a N₂-atmosphere. For porewater extraction, sediment subsamples were filled into centrifuge tubes under anoxic conditions and

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