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A modified QWASI model for fate and transport modeling of mercury between the water-ice-sediment in Lake Ulansuhai

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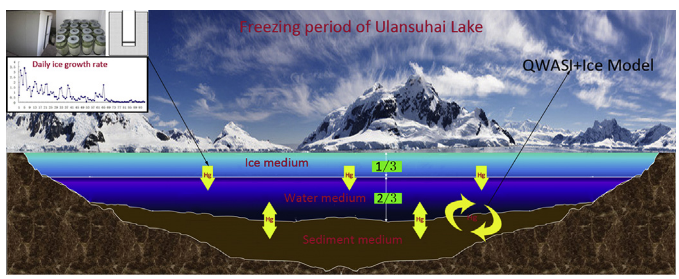
HIGHLIGHTS

- Mercury contamination in lakes a growing concern in northern China.
- Hg distribution differs between ice growth and ice free periods.
- Ice as an environmental medium was added to develop a new QWASI + ice model.
- Model results compared to experimental tests and within acceptable margin of error.
- QWASI + ice offers potential for more accurate prediction of Hg contamination in winter.

GRAPHICAL ABSTRACT



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ABSTRACT

Mercury contamination from industrial and agricultural drainage into lakes and rivers is a growing concern in Northern China. Lake Ulansuhai, located in Hetao irrigation district in Inner Mongolia, is the only sink for the all industrial and agricultural drainage and sole outlet for this district to the Yellow River, which is one of the main source of drinking water for the numerous cities and towns downstream. Because Ulansuahi is ice-covered during winter, the QWASI model was modified by adding an ice equation to get a more accurate understanding of the fate and transport of mercury within the lake. Both laboratory and field tests were carried out during the ice growth period. The aquivalence and mass balance approaches were used to develop the modified QWASI + ice model. The margins of error between the modelled and the measured average concentrations of Hg in ice, water, and sediment were 30%, 26.2%, and 19.8% respectively. These results suggest that the new QWASI + ice model could be used to more accurately represent the fate and transport of mercury in the seasonally ice-covered lakes, during the ice growth period.

1. Introduction

Mercury (Hg) pollution is a serious problem due to its toxicity and subsequent accumulation in aquatic habitats. Mercury contamination is characterized by persistence, easy mobility, and high toxicity. Because of its ease of mobility, Hg can be found far

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from its original point of contamination. The major sources of mercury in aquatic systems today are: direct mercury contaminated industrial effluent discharges, surface water runoff from pesticides, atmospheric deposition, and sediment carried by streams that flow through the slag from mine smelting activities (Francesconi et al., 1997; Matthews et al., 1995; Qiu et al., 2004).

Many species of mercury can translate into the organometallic cation methylmercury (MeHg), which is bioaccumulative and highly poisonous (Björnberg et al., 2005; Rodrigues et al., 2013; Zahir et al., 2005). Because Hg^0 has a high vapor pressure and barely dissolves in water (the solubility is 0.06 mg/L at 25 °C), it can stay in the atmosphere for up to a year, spreading across the planet through the global mercury cycle. Eventually, in the presence of O_3, H_2O_2, halogens, and oxidizers in the atmosphere, Hg^0 oxidizes into water-soluble Hg^{2+} and returns to earth through both wet and dry deposition.

This atmospheric mobility results in mercury contamination being found far from its originating sources. In the 1980s, for example, significant levels of mercury were discovered in North American and northern European lakes that were far from any sources of mercury pollution. In Sweden alone, an estimated 10,000 lakes were deemed to exceed the blacklisting limit of 1 mg Hg/kg in a l kg pike. Lindqvist et al. (1991) also measured MeHg levels in fish that could only be explained by atmospheric contamination (Lindqvist et al., 1991; Sakata et al., 2006).

Contaminated sediment carried by streams and runoff is another significant secondary source of mercury in aquatic systems. Through the sedimentation process, mercury redistributes between different environmental media. As the metal associates with the surface of particles, it is preferentially transported, deposited, and eventually buried with fine-grained, high surface area sediments (Conrad and Chisholm-Brause, 2004). As a result of hydrodynamics (e.g., tidal forces, waves, wind, tsunamis, earthquakes, and disturbance created by shipping and dredging projects) and bioturbation, this sediment becomes re-suspended, releasing contaminants into the aquatic system as secondary pollution (Chapman, 1997; Gambrell et al., 1991; Riedel et al., 1999).

1.1. The need for a QWASI + ice model

Given mercury's highly mobile nature, quantifying its migration between different environmental media is an effective way to predict contamination and evaluate the hazard to humans and the environment. An accurate model would be useful for simplifying and simulating mercury's behaviour in the natural environment, and predict its fate and distribution. The results could contribute to environmental pollution prediction, environmental quality assessment, pollution control, risk assessment of optimization, and environmental management etc.

In recent years, many researchers have been applying multimedia models to study the distribution of mercury and other contaminants in natural bodies of water (i.e. lakes and rivers), and have made considerable progress with respect to the accuracy of their predictions. Because fugacity is not suitable as an equilibrium criterion for non-volatile matter, a new approach based on an equilibrium criterion of aquivalence was developed to analyze the fate of lead in Lake Ontario. The results demonstrated that the modelled average concentrations of lead were approximately consistent with the measured results, establishing a new way to study heavy metal dynamics in water bodies (Mackay and Diamond, 1989). Woodfine et al. (2000) subsequently applied the qualified Quantitative Water, Air, and Sediment Interaction (QWASI) model to represent the distribution of Cu and Ni in Baby and Alice lakes, Ontario. The results not only successfully described the processes of contamination in the lakes over 20 years but also successfully forecasted the fate of Cu and Ni, suggesting that using aquivalence is appropriate for describing the fate of heavy metals (Woodfine et al., 2000).

Although the aforementioned QWASI model has been used to model the distribution of heavy metals in air, water, and sediment, it is rarely applied to lakes with an ice cover medium. We argue that doing so is critical for bodies of water, such as Ulansuhai, that experience a freezing period with an ice cover because the ice cover significantly affects the distribution and migration of heavy metals during ice season.

Research suggests that chemicals concentrate in water during the freezing process. Zhang et al. (2012) studied the migration mechanism of the total dissolved solids (TDS) between ice and water during the freezing process of Lake Ulansuhai, and found that approximately 80% (3.602×10^8 kg) of the TDS migrated from the ice cover to the water body beneath, during the ice growth period (Zhang et al., 2012). Shafique et al. (2012) determined that the separation efficiency of inorganic ions, soluble organic compounds, and dyes between ice and water in both natural streams and synthetic solutions depended on different factors, such as rate of cooling, pH, and concentration. Furthermore, the treatment of potable water by freezing to remove inorganic (Mn(II), In(III), and Au(I)) impurities showed that the zone freezing procedure is most efficient at low ice front velocities (i.e., no greater than 0.15 cm h^{-1}) (Skorobogatov et al., 2009).

From these studies, it can be concluded that chemicals indeed concentrate in water bodies during the freezing process. This is of particular importance in lakes as the concentration of chemicals in the water increases during the freezing process, and the dynamic equilibrium of the chemicals between the water and sediment is upset. The chemicals migrate to the sediment due to the difference in the aquivalence gradient. The freezing process may therefore play a very important role in the migration and distribution of mercury in lakes that freeze seasonally. Adding ice cover effects to modify the QWASI model offers one approach to more accurately model and predict the fate and transport of mercury in the water body, sediment, and ice of Lake Ulansuhai during the freezing period.[1]

2. Materials and methods

2.1. Brief overview of Lake Ulansuhai

Lake Ulansuhai is one of the eight biggest fresh water lakes in China. It is located in the cold and arid region in central Inner Mongolia (40°36′–41°03′N, 108°43′–108°57′E), where it is the only sink for the industrial and agricultural drainage area of the Hetao irrigation district. The lake has a total open water area of 290 km^2, of which 119 km^2 is covered by reeds. The water residence time in the lake is 240–290 days. The region has four distinct seasons with a mean annual air temperature of 7.6 °C, and an average temperature of −15 °C in the winter. The temperature varies dramatically from as cold as −23 °C in winter to as high as 32 °C in summer.

In winter, the lake is typically frozen for about five months (from November to April) with a maximum ice cover thickness of one third to two thirds the depth of the lake. The ice as an environment medium, therefore, may play a very important role in the lake water environment, especially in the distribution and migration process of contaminants between the ice cover (mainly solid phase), the water body (mainly liquid phase), and the sediment (mix of solid and liquid phases).

[1] The 'freezing period' refers to the moment the ice starts to form on the lake until it reaches its maximum thickness.