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Human exposure to trace elements in central Cambodia: Influence of seasonal hydrology and food-chain bioaccumulation behaviour

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ABSTRACT

Exposure to mercury and other trace elements remains an important public health concern, worldwide. The present study involved a comprehensive field study to determine concentrations of fourteen trace elements (Al, As, Cr, Co, Cd, Cu, Fe, Hg, Mn, Ni, Pb, Se, V and Zn) in surface water and different fish species from Tonlé Sap Lake in central Cambodia, during both the dry and wet seasons. Total arsenic (tAs) and Mn in surface water during the dry season exceeded WHO drinking water guidelines. Total mercury (tHg) concentrations (µg/g wet wt.) in fish during the wet season (GM = 0.055; CI_{95} = 0.01–0.26) were approximately 15 times higher ($P < 0.05$) compared to those during the dry season (GM = 0.0035; CI_{95} = 0.0004–0.033). Mean target hazard quotients (THQs) for inorganic arsenic (iAs), methyl mercury (MeHg), Mn and Pb were > 1, with estimated maximum values greatly exceeding 1. Mean THQs of Zn, Cd, Ni and Se were very near 1, with estimated maximum values exceeding 1. The MeHg THQ (min-max range: 0.16–9.09) during the wet season was 7 times higher than in the dry season (min-max range: 0.05–1.35). Concentrations of Hg and other trace elements varied widely between fish species. The findings suggest that exposure of some trace elements via water and food is of concern in this region. High consumption rates of fish and rice key factors related to trace element exposure. Seasonal hydrology and species-specific bioaccumulation behaviour in the Tonlé Sap Lake watershed also play an important role. The generated information will be useful to better mitigate trace element exposure in this region.

1. Introduction

Contamination of land and water resources and related human health risks remains a major concern, worldwide (de Souza Machado et al., 2016; Fairbrother et al., 2007). Some trace elements are considered essential or beneficial and can be necessary for daily nutrition and physiology (Bender et al., 2009). Others such as Pb, Hg, Cd and As are non-essential and can be considered potentially toxic xenobiotic substances. Environmental exposure of trace elements via inhalation, ingestion or dermal absorption may result in high internal concentrations and adverse health impacts such as disruption of the central nervous system, reproductive failure, genotoxicity, and gastro-intestinal problems (Luoma and Rainbow, 2008; Wu et al., 2010).

In aquatic environments, trace elements can be present in a variety of chemical forms (species) and may be dissolved in the water column or bound to sediments (Namieśnik and Rabajczyk, 2010). The toxicity of trace elements can vary between different chemical species. For example, the organometallic form of Hg, methylmercury (MeHg) is much more toxic than inorganic Hg (NRC, 2000). Conversely, inorganic arsenic (iAs) is the toxicologically relevant As species, compared to organic forms such as dimethylarsinic acid (DMA) and monomethlyarsonic acid (MMA), (Clarkson, 1987; Jomova et al., 2011; Rahman and Hasegawa, 2011; Rasheed et al., 2017; Schoof et al., 1999; Xue et al., 2010).

Numerous studies have reported on the bioaccumulation of trace elements in aquatic organisms and food chains, worldwide (Bandowe et al., 2014; Bo et al., 2015; Cano-Sancho et al., 2015; Chi et al., 2007; Dural et al., 2007; Heshmati et al., 2017; Jonathan et al., 2015; Kalay et al., 1999; Lewi et al., 2002; Monikh et al., 2015; Rashed, 2001; Taweel et al., 2013; Velusamy et al., 2014; Zhang et al., 2015). It is well established that Hg can biomagnify in aquatic food chains, with concentrations in top predatory fish greatly exceeding those in lower trophic organisms. The high degree of Hg bioaccumulation in aquatic organisms is attributed to increased uptake and persistence of the

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organometallic species, methylmercury (MeHg), (Li et al., 2015). Thus, Hg is typically the trace element of greatest concern in terms of health risks associated with fish consumption. This is particularly true for populations or communities with diets containing a significant proportion of fish, especially high trophic-level fish.

In central Cambodia, the Tonlé Sap Lake watershed has experienced increasing pollution in recent years, mainly due to increasing human settlements in the region (IUCN, 2013; MRC, 2010a; Sarkkula et al., 2003). Previous environmental monitoring investigations, utilizing dissolved oxygen, ammonium, total phosphorus, chemical oxygen demand, pH and nitrate as pollution indicators, found that approximately 25% of water samples from Tonlé Sap Lake were classified as having poor water quality, with only 6% of samples categorized as high quality (MRC, 2007, 2008). Another recent water quality study of the Lower Mekong Basin recently reported the Tonlé Sap Lake exhibits moderate to poor water quality and is characterized by high nutrient and dissolved solid levels (Chea et al., 2016). The unique hydrological cycle between wet and dry seasons elicits high biodiversity and productivity in this watershed. During the wet season (June to October), the excess water from the Mekong River enters Tonlé Sap Lake, which causes intensive flooding that extends the lake over a large floodplain. At the end of the wet season, the flow reverses and the lake empties (Matsui et al., 2006; MRC, 2010b). The depth of Tonlé Sap Lake is approximately 1–2 m in the dry season and 8–11 m during the wet season, with an estimated water volume ranging from 418.3 to 2223.9 million m^3, respectively (Arias et al., 2013; Siev et al., 2016).

Tonlé Sap Lake, also known as the Great Lake, is the largest freshwater lake in Southeast Asia. This watershed comprises an important food fishery, supporting approximately four million people living within the six surrounding provinces (IUCN, 2013; Matsui et al., 2006). The total fish catch of the watershed is approximately 230,000 t/year, nearly half of Cambodia's total fish production. It is estimated that about a half of the Cambodian population is directly dependent on the watershed for water and food resources (MRC, 2010b). It is important to note that the flood cycle is important for fish migration and population dynamics within this watershed (Lamberts, 2001). During the extensive flood period, passive migration of eggs, fry, juvenile and adult fish into the Tonlé Sap occurs, as water from the Mekong river flows north and fills the floodplain. During the dry season, as water recedes and flows south, back toward the Mekong, many fish migrate out of the lake due to poor water quality, while others may remain year-round residents of the lake. The seasonal hydrology of this system may therefore influence contaminant loadings, as well as fate and bioaccumulation patterns of trace elements and other contaminants.

Information regarding trace elements and other chemical contaminants of concern in Tonlé Sap Lake are limited (MRC, 2007). One previous study, investigating the degree of trace element contamination in drinking water, found relatively high concentrations of As and Mn that may pose serious exposure risks to the local population (Buschmann et al., 2007, 2008). Studies of trace element bioaccumulation and related human exposure risks in this region are also limited. Recent studies have documented the levels and exposure risks of organochlorine pesticides, Hg and As in fish and other food items in three Cambodian provinces, namely Kampong Cham, Kratie and Kandal provinces (Cheng et al., 2013; Wang et al., 2013, 2011). The findings indicated that the daily dietary intake of MeHg in Kratie and Kampong Cham were greater than the United States Environmental Protection Agency (U.S. EPA) recommended reference dose (RfD), while daily intake for Kratie exceeded the provisional tolerable weekly intake (PTWI) imposed by World Health Organization (WHO). Chanpiwat et al. recently reported concentrations and exposure risks of several trace elements in southern Vietnam (Chanpiwat et al., 2016). The results indicated that daily consumption of local fish in this region may result in adverse health impacts, due to elevated intake of Zn, As, and Cu.

Currently, there is a lack of information regarding trace element concentrations and exposure risks in the Tonlé Sap Lake region of Cambodia, a critical freshwater system that provides drinking water and food resources to a large portion of the country's population. The objective of the present study was to conduct a comprehensive field investigation to measure concentrations of several trace elements in water and fish from different locations in Tonlé Sap Lake. Samples of water and different fish species collected during the dry season (March) and wet season (October) in Tonlé Sap Lake were analyzed for a range of trace elements, including Al, As, Cr, Co, Cd, Cu, Fe, Hg, Mn, Ni, Pb, Se, V and Zn. The generated concentration data were utilized to assess human health risks related to daily intake via water and food consumption in this region. The influence of seasonal hydrology and species-specific bioaccumulation on trace element intake and exposure risks are further evaluated and discussed.

2. Methods

2.1. Field sampling

Field sampling was conducted within Tonlé Sap Lake, which is connected to the Mekong River via the Tonlé Sap River (Supplementary information, Fig. S1). Water and fish samples were collected in the dry (March) and wet (October) seasons in 2011, at four sites distributed along the Tone Sap Lake (12°31′-13°12′ N, 103°53′-104°27′ E), (Supplementary information, Table S1 and Fig. S1). At each site, water samples (n = 5) were collected below the surface (depth between 20 and 30 cm) using 500 mL polyethylene bottles, which were pre-conditioned with 5% nitric acid (HNO_3) and rinsed with Milli-Q water. Water samples were acidified with 10% HNO_3 immediately after collection, stored on ice during transportation from the field sites to laboratory facilities at the University of Health Sciences, Phnom Penh. In the laboratory, water samples were filtered using a 0.45 µm micropore membrane filter (Merck Millipore Ltd., Ireland). Filtrate samples were stored at 4 °C until analysis. A total of forty-one different fish species were collected during the field study (Supplementary information, Table S2). Tonlé Sap Authority fisheries biologists helped to coordinate fish sampling and provided the scientific and common names for the collected fish species at the time of collection. For many species, multiple individuals were collected and analyzed separately. For other species (typically small fish), several individual fish (3–10 individuals) were pooled to comprise composite samples for analysis (Supplementary information, Table S2). The weight (kg) and length (cm) of individual fish were measured within 2 h after collection. Fish tissues (liver and muscle) were excised and stored at −20 °C prior to digestion.

Microwave digestion of fish tissue samples followed previously reported methods (Sivaperumal et al., 2007). Briefly, 0.2–0.5 g of dry fish muscle tissue (excluding skin) was placed in TFM Teflon vessels pre-soaked with 10% HNO_3 and washed with MiliQ water. 7 mL of 65% (w/v) HNO_3 and 1 mL of 30% (w/v) H_2O_2 were added, mixed thoroughly and left at room temperature for 30 min. Samples were then digested using a microwave digester (Milestone ETHOS PLUS, Italy). The operating temperature, power and digestion time settings of the microwave digestion were 190 °C, 1000 W and 15 min, respectively. Digested samples were allowed to cool to room temperature and then filtered using 0.45 µm nylon syringe filters (Whatman® Puradisc 25 syringe filters). Samples were then diluted to 20 mL with MiliQ water. All samples were processed in batches of 15, each consisting of 1 procedural blank.

2.2. Chemical analysis

The concentrations of individual trace elements in the processed samples were determined using Inductively Coupled Plasma Mass Spectrometry (ICP-MS, Agilent 7700 Series). A standard addition approach was employed to quantify trace element concentrations in water