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Investigation of the relationship between Hg speciation in soil and human health and ecological risk assessment

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Abstract Mercury (Hg) pollution in soils can have major effects on human health and ecological systems. Concentrations, toxicological behaviour and bioavailability of different Hg species, both in the environment and in biological systems differ greatly, and are significant in the estimation of both human health and ecological risk assessment. Herein the significance of appropriate selection of species in both human health and ecological risk assessments is considered.

Keywords: mercury species - total mercury - human health risk assessment - ecological risk assessment-reference dose

1. Introduction

Mercury (Hg) pollution in soils is a problem of major significance, which has severe impacts on human health (Greenwood, 1985; Takaoka et al., 2014) and the environment (Cavoura et al., 2019; Vermeer, Armstrong and Hatch, 1973). Mercury can exist in the environment and in biological systems as metallic mercury (Hg^0), inorganic and organic mercuric (Hg^{2+}) Hg, and mercurous (Hg$^+$) forms (Liu, Cai and O'Driscoll (Eds), 2012). The specific form, or species, of Hg is a critical parameter in Hg toxicokinetic and toxicodynamic studies (ATSDR, 1999; Counter and Buchanan, 2004). For example consumption of MeHg in fish can affect the nervous system and cause severe neurological problems (Nabi, 2014). Hg^0 inhaled as vapor, can be rapidly absorbed through bloodstream and transmitted to all body tissues due to its high solubility. Both Hg^0 and inorganic Hg^{2+} species can induce renal effects and mercury chloride compounds have been associated with Acrodynia disease (Kim, Kabir and Jahan, 2016). Environmental systems are also affected by Hg exposure. In the aquatic environment, for example, water biota tend to bioaccumulate and biomagnify MeHg with severe health impacts for organisms higher in the food chain and untimely on human health (Beckers and Rinklebe, 2017). Invertebrates of soil ecosystem, like earthworms, also accumulate MeHg and participate in Hg methylation (Zhang et al., 2009; Rodríguez Álvarez et al., 2014; Dang et al., 2015). This paper explores the role of speciation in Hg in human health and ecological risk assessment.

2. Methods

Data from studies on human health risk assessments and ecological risk assessments, based on soil concentrations of Hg species, was aggregated and assessed. The search was limited to range between 2010 and 2021. Searches were performed using the search engine PubMed of the database MedLine, the bibliographic database of Scopus, the search engine of Google Scholar and the single search engine of Association of Greek Academic Libraries (HEAL-LINK).

3. Results and Discussion

Estimating the potential impact of a hazard on a specified human population was generally based on the four basic steps involving the identification of issue, hazard assessment, exposure assessment and risk characterization. Identification of the particular species involved was the most significant step in the process since this dictated the concentrations to be determined in assessments and the relevant toxicological parameters of exposure. The vast majority of studies on human health risk assessment and ecological risk assessment for Hg in contaminated soils used models such as US Environment Protection Agency (US EPA), the Ministry of Ecology and Environment of the People's Republic of China human health risk assessment models, and the ecological index bioaccumulation factor (BAF), and were almost exclusively based on total soil Hg concentrations (Ordóñez et al., 2011; Liu et al., 2016; Le Faucheur et al., 2016). Speciation was considered only in a few studies (Jia et al., 2018; Jiang et al., 2021; Rodríguez Álvarez et al., 2014; Zhang et al., 2009) in the calculation of risk assessment.

The vast majority of studies focused on human exposure used a reference dose (RfD) for inorganic Hg, specifically data for $HgCl_2$ (Tvermoes et al., 2014; Reis et al., 2014). This can be justified, since essentially this is a compound on which toxicological data is available (SCOEL, 2007), while for other compounds there are gaps in knowledge (SCOEL, 2007). It is worth noting however, that this form of Hg in fact contributes only a small extent to the total content of soil. Often in soil, Hg is encountered bound to organic matter (Różański, Castejón and Fernández, 2016). In the environment this is certainly a less mobile form, but if, for example, dermal contact with contaminated soil was being assessed, a lack of mobility would not limit exposure. Of course, there are no toxicological studies on Hg bound to organic matter, since this is an ill-defined compound with no standard reference material available for toxicological studies. Exposure to MeHg directly from contaminated soil was not considered in any of the literature. Considering that the concentration of MeHg rarely exceeds 2% of the total Hg content (Horvart and Kotnik., 2019), this may be generally appropriate, however, certain speciation studies have identified concentration of MeHg at concentrations up to 46.52 μg kg^{-1} (Jia et al. 2018) and it is prudent that this species be considered in risk assessment studies since the RfD is lower than that of $HgCl_2$ (RfD $_{HgCl2}$=3x10^{-4} mg m^{-3}, RfD$_{MeHg}$=1x10^{-4} mg kg^{-1} day^{-1}) (US EPA, 1995).

An exposure assessment based on daily intakes of both THg and MeHg via consumption of vegetables was undertaken by Jia et al., (2018) based on the Technical guidelines for risk assessment of contaminated sites" (HJ 25.3-2014). The mean PDI$_{THg}$ values was 0.82 μg/kg bw/d for adults and 1.21 μg/kg bw/d for children while the average PDI of MeHg was 0.34 ng/kg bw/d and 0.49 ng/kg bw/d for adults and children. Both values were lower than the reference dose (RfD) for MeHg (0.1 μg/kg bw/d) established by US EPA (2001) and the average daily intake (ADI) of MeHg (0.23 μg/kg bw/d) (JEFCA, 2006). Environmental exposure to Hg^0 from soils through inhalation is little studied even although Hg^0 is particularly volatile, reemission is a common occurrence all soil surfaces and the absorption of inhaled Hg^0 vapor is estimated to 70-80% (Jiang et al., 2021). Additionally, as a result of Hg^0 oxidation to Hg^{2+} inside the body, health effects extend to the central nervous system, skin and kidneys (Flavia Ruggieri et al., 2017). Only two (2) studies were found (Nakazawa et al., 2016; Jiang et al., 2021). Jiang et al. (2021) estimated both the hazard quotient of oral ingestion (HQ$_{ing}$) and the hazard quotient of inhalation of soil Hg^0 vapor (HQ$_{inh}$). The HQ$_{ing}$ was based on THg concentration and the HQ$_{inh}$ was based on the modeled soil Hg^0 vapor using the three-phase partitioning model. While HQ$_{ing}$ was 1.57, HQ$_{inh}$ was 1168.

Ecological risk similarly was based primarily on total Hg concentrations (Crnić et al., 2016; Egwu et al., 2019). One study estimating Hg bioaccumulation based on both total Hg and MeHg concentration (Rodriguez Alvarez et al, 2014) concluded that while BAF$_{THg}$ ranged from 0.02 to 0.11, BAF$_{MeHg}$ was significantly higher and ranged from 1.7 to 5.9. Similarly, Zhang et al (2009) calculated BAF$_{THg}$ between 0.04 and 0.539 while BAF$_{MeHg}$ ranged from 10.163-31.387. Despite the mean MeHg concentration of 6.96 μg kg^{-1} being well below the soil guideline values of Environment Agency (2009) (410 mg kg^{-1} DW MeHg for industrial land), this massive increase in BAF$_{MeHg}$, was a result of the lipophilic nature of MeHg facilitating absorbed in earthworms (ATSDR, 2013; Hirano and Tamae, 2011),

4. Conclusions

Although human health risk assessment models are based on toxicokinetic and toxicodynamic data of individual species, the vast majority of human health risk assessments are based on total Hg soil concentrations and toxicological data for $HgCl_2$. Caution must be exercised where discrete species such as MeHg and Hg^0 are present, since these species can significantly affect exposure. Similarly, in ecological risk assessments, the BAF for specific species should be species-specific, as species behaviours in the soil and biological media differs greatly and can greatly affect assessment.

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