Environmental Pollution 230 (2017) 810–816

Contents lists available at ScienceDirect

Environmental Pollution

journal homepage: www.elsevier.com/locate/envpol


# Mercury risk in poultry in the Wanshan Mercury Mine, China☆

Runsheng Yin [a,*,1], Wei Zhang [b,1], Guangyi Sun [c,d], Zhaohui Feng [e], James P. Hurley [f,g], Liyuan Yang [b], Lihai Shang [c,**], Xinbin Feng [c]

[a] State Key Laboratory of Ore Deposit Geochemistry, Institute of Geochemistry, Chinese Academy of Sciences, Guiyang 550081, China
[b] School of Resources and Environment, University of Jinan, Jinan 250022, China
[c] State Key Laboratory of Environmental Geochemistry, Institute of Geochemistry, Chinese Academy of Sciences, Guiyang 550081, China
[d] University of Chinese Academy of Sciences, Beijing 100190, China
[e] Institute of Resources and Environment, Henan Polytechnic University, Jiaozuo, Henan Province 454000, China
[f] Environmental Chemistry and Technology Program, University of Wisconsin-Madison, Madison, WI 53706, USA
[g] Department of Civil and Environmental Engineering, University of Wisconsin-Madison, Madison, WI 53706, USA

ARTICLE INFO

*Article history:*
Received 24 February 2017
Received in revised form
10 July 2017
Accepted 10 July 2017
Available online 10 August 2017

*Keywords:*
Mercury
Methylmercury
Poultry
Mercury mine
Human exposure

ABSTRACT

In this study, total mercury (THg) and methylmercury (MeHg) concentrations in muscles (leg and breast), organs (intestine, heart, stomach, liver) and blood were investigated for backyard chickens, ducks and geese of the Wanshan Mercury Mine, China. THg in poultry meat products range from 7.9 to 3917.1 ng/g, most of which exceeded the Chinese national standard limit for THg in meat (50 ng/g). Elevated MeHg concentrations (0.4–62.8 ng/g) were also observed in meat products, suggesting that poultry meat can be an important human MeHg exposure source. Ducks and geese showed higher Hg levels than chickens. For all poultry species, the highest Hg concentrations were observed in liver (THg: 23.2–3917.1 ng/g; MeHg: 7.1–62.8 ng/g) and blood (THg: 12.3–338.0 ng/g; MeHg: 1.4–17.6 ng/g). We estimated the Hg burdens in chickens (THg: 15.3–238.1 μg; MeHg: 2.2–15.6 μg), ducks (THg: 15.3–238.1 μg; MeHg: 3.5–14.7 μg) and geese (THg: 83.8–93.4 μg; MeHg: 15.4–29.7 μg). To not exceed the daily intake limit for THg (34.2 μg/day) and MeHg (6 μg/day), we suggested that the maximum amount (g) for chicken leg, breast, heart, stomach, intestine, liver, and blood should be 1384, 1498, 2315, 1214, 1081, 257, and 717, respectively; the maximum amount (g) for duck leg, breast, heart, stomach, intestine, liver, and blood should be 750, 1041, 986, 858, 752, 134, and 573, respectively; and the maximum amount (g) for goose leg, breast, heart, stomach, intestine, liver, and blood should be 941, 1051, 1040, 1131, 964, 137, and 562, respectively.

© 2017 Elsevier Ltd. All rights reserved.

## 1. Introduction

Mercury (Hg), a toxic metal pollutant, is released to the environment via natural and anthropogenic sources (Pirrone et al., 2010). Mercury can be methylated via microbial processes, especially in aquatic ecosystems (Ullrich et al., 2001). Methylmercury (MeHg), the most toxic form of Hg, can be easily bioaccumulated and biomagnified in food chains. Bioaccumulation of MeHg results in high MeHg levels in fish, and fish consumption is regarded as the major exposure pathway of Hg to humans (Ullrich et al., 2001).

Mercury exposure due to non-fish food sources is thought to be limited due to the short nature of most farmed animal food chains. However, in Hg mines and other sites with extreme Hg contamination, crops at the base of the food chain can have high levels of Hg. leading to the possibility that livestock may have high levels of Hg as they forage on crops. In Wanshan Mercury Mine (WMM), which is China's largest and the world's third largest Hg mine, long-term mining activities have resulted in serious Hg contamination to the surrounding environment (Jiang et al., 2006). Rice paddies have been shown to enhance Hg methylation, and high MeHg levels were reported for rice in WMM (Rothenberg et al., 2011; Zhang et al., 2010). Rice, a staple, is considered the primary human Hg exposure source due to limited fish consumption by locals (Feng et al., 2008; Zhang et al., 2010). In recent years, due to the awareness of Hg contamination, local residents in WMM started to buy

☆ This paper has been recommended for acceptance by Dr. Jorg Rinklebe.

* Corresponding author.
** Corresponding author.
*E-mail addresses:* yinrunsheng2002@163.com (R. Yin), shanglihai@vip.skleg.cn (L. Shang).
[1] These authors contributed equally to this work.

http://dx.doi.org/10.1016/j.envpol.2017.07.027
0269-7491/© 2017 Elsevier Ltd. All rights reserved.

imported food supplies. Local crops were more and more used to feed poultry (e.g., chickens, ducks and geese) and domestic animals (e.g., pigs), to increase the economic value of local crops. Theoretically, Hg in crops can be bioaccumulated in meat products, and if at high concentrations pose risks to the population who consume them.

Birds have been considered as bio-indicators of environmental Hg contamination (Bond and Diamond, 2009; Peterson et al., 2017). Total Hg (THg) concentrations cannot fully represent the environmental risk of Hg in poultry, and measurement of MeHg concentrations is necessary due to its high toxicity. Previous studies showed that >95% of the MeHg in food items is likely taken up by birds and mammals, whereas the corresponding proportion for inorganic Hg species is only about <15% (WHO, 1993; Mori et al., 2012). THg and MeHg concentrations have shown to be largely variable among different tissues of different bird species, while organs have higher THg and MeHg concentrations compared to muscles (DesGranges et al., 1998; Ruelas-Inzunza et al., 2009; Kalisinska et al., 2014).

In China and many other countries, there is a long history and culture of consumption of organs, and elevated Hg levels in organs may induce a risk of human Hg exposure. However, Hg levels and risks in poultry meat products in the WMM has been still poorly understood so far, and therefore warrants investigation. In this study, THg and MeHg burdens in muscles (leg and breast), organs (intestine, heart, stomach, liver) and blood of chickens, ducks and geese in the WMM were investigated. We further evaluated the risk of Hg exposure though consumption of poultry by local residents.

## 2. Methods

### 2.1. Study area and sample collection

WMM is located in Guizhou Province, Southwestern China (Fig. 1). As the “Mercury Capital” of China, Hg mining in WMM has lasted for few thousand years, and Hg was intensively extracted before WMM was officially closed at the beginning of this century (Jiang et al., 2006; Feng et al., 2008). Long-term Hg mining activities have resulted in several large Hg-rich calcine piles, which are mainly located at the headwaters of major rivers (Yin et al., 2013a, 2016a). The Wukeng (WK) pile, which is one of the largest piles at WMM, is located on the Dashuixi River (DSX). Runoff of Hg from the WK pile led to extremely high Hg levels in the DSX river which is an important water resource for nearby farmlands (Yin et al., 2016b). In recent years, illegal Hg mining was revived near the Gouxi (GX) site, and resulted in extremely high Hg concentrations in ambient air (Li et al., 2009). At both DSX and GX sites, elevated Hg levels were reported for vegetables, rice and other crops (Qiu et al., 2008; Yin et al., 2013b).

In December 2015, backyard poultry were collected from 2 farms at DSX and 2 farms at GX. A number of about 5–10 poultry were fed in each farm, and we purchased all poultry from the farm. Poultry were fed with local crops and vegetables for approximately 1 year. A total of 35 poultry were collected at DSX (chicken: 3 males and 7 females; duck: 3 males and 2 females; goose: 2 males) and GX (chicken: 3 males and 3 females; duck: 8 males and 4 females). Each live bird was weighed before processing. Whole blood samples were collected in PTFA tubes and weighted. Then 2 g of fresh blood sample was immediately pipetted into two 15 mL centrifuge tubes (1 g blood in each tube) and stored at −20 °C for further THg and MeHg analysis (Section 2.2). No anticoagulant was used because anticoagulant has the potential to break up the original Hg species (Pirrone and Mahaffey., 2005). In this study, only edible portions including muscles such as leg and breast (or pectoralis) and organs such as intestine, heart, liver and stomach (or proventriculus) were collected. Non-edible tissues (e.g., feather, skin, fat, head and feet, etc), which were dirty and hard to wash, were not investigated in our study although we acknowledged that some of these tissues are often consumed by the local people. Leg, breast, intestine, heart, stomach and liver of each bird were separated, washed with 18.2 MΩ cm water (Millipore water system, Germany), weighed (wet weight, ww), packaged in sealed polyethylene bags, and stored at −20 °C until chemical analysis.

### 2.2. Total mercury and MeHg concentration analysis

Approximately 50 g of leg and breast muscle pieces were randomly used, and were further minced to ensure homogenization, prior to being digested for THg and MeHg analysis. Each organ sample (heart, stomach and liver) was entirely minced. A stainless steel knife and a plastic cutting board were used, and were washed with EtOH prior to preparing each sample.

For THg analysis, approximately 1 g (ww) of each sample was digested with 5 mL of $HNO_3$ at 95 °C for 3 h until the sample was completely dissolved in an open system. 0.5 mL of BrCl was added to the digest for overnight, to convert all Hg species to Hg(II). Excessive BrCl was reduced by 0.2 mL of $NH_2OH \cdot HCl$. The digestate was analyzed by cold vapor atomic fluorescence spectrometry (CVAFS) (Tekran 2500, Canada) according to USEPA (2002). Accuracy was assessed using certified reference material (TORT-2, lobster), sample duplicates, method blanks, matrix spikes. The limit of detection (3σ) was 10 pg/g for THg. The measured THg for TORT-2 was 275 ± 11 ng/g (SD, n = 6), comparable with the certified value of 270 ± 20 ng/g. Recoveries of THg were 96–106%, and the relative standard deviation of sample duplicates were within 10%.

For MeHg analysis, about 1 g (ww) of each sample was digested with 5 mL of 25% KOH solution at 75 °C for 3 h in a close system (pressure decomposition). Then the extract was buffered with sodium acetate at pH 4.9 and ethylated in a borate glass bottle with a Teflon-lined cap. Quantification of MeHg was performed by gas chromatographic separation and pyrolysis, followed by CVAFS detection (MERX, Brooks Rand) according to USEPA (2001a). Accuracy was assessed by method blanks, certified reference materials (TORT-2), and sample duplicates. The limit of detection (3σ) was 2 pg/g for MeHg. Measured MeHg concentration of TORT-2 was 143.4 ± 6.7 ng/g (SD, n = 6), consistent with the certified value (152 ± 20 ng/g). MeHg recoveries ranged from 90 to 102%, and the relative standard deviation for sample duplicates were <10%.

### 2.3. Total mercury and MeHg concentration burdens

THg and MeHg burdens of each tissue were estimated based on THg and MeHg concentrations multiplied by the weight of the tissue. The sum of THg and MeHg burdens of all edible tissues, were evaluated to roughly represent the total burden of poultry. The proportion of THg and MeHg burdens of each tissue was further estimated to understand the relatively distribution of Hg.

### 2.4. Statistical analyses

Data were analyzed in SPSS (version 13.0). Distributions of Hg concentrations (THg and MeHg) were not normal, and $Log_{10}$ transformation of the data produced normal distributions for the data, and therefore geometric mean concentrations were reported in this study. Correlation coefficients ($r$) and significance probabilities ($p$) were computed for the linear regression fits according to Pearson correlation analysis. One-way analysis of variance (ANOVA) was carried out to compare whether concentrations varied significantly between poultry species, and among different tissues.

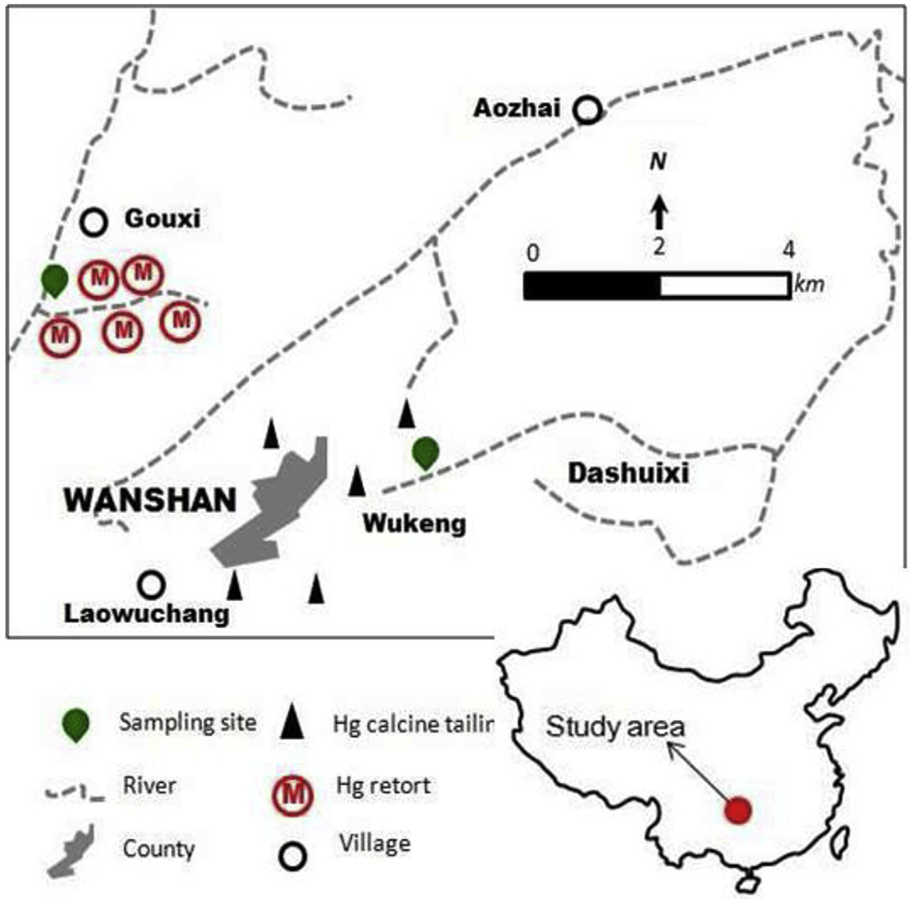


**Fig. 1.** Study area and sampling sites.

## 3. Results and discussion

### 3.1. THg and MeHg distributions

Only a few studies have reported Hg concentrations in poultry meat products, and previous results mainly showed very low concentrations for THg and MeHg in poultry meat products, in general less than 10 ng/g (ww) for THg and less than 4 ng/g (ww) for MeHg (Table 1). Several studies showed relatively higher Hg concentrations but they were reported in dry weight (Table 1). Therefore, consumption of meat is not likely a major concern for human MeHg exposure in most areas, especially in comparison to fish and rice (USEPA, 1997; Zhang et al., 2010). However, in certain areas (e.g., Hg mines and gold mining sites), poultry meat products have shown much higher Hg concentrations of tens to thousands ng/g (Table 1). THg and MeHg concentrations in muscles and organs of this study, reported in wet weight, are summarized in Fig. 2(A–F). Our concentration values were more consistent with that reported for Hg pollution sites (Ji et al., 2006; Bortey-Sam et al., 2015), suggesting pollution of Hg in WMM poultry. At DSX, 90% of livers and 40% of blood samples for chickens, and 20% of legs, 20% of breasts, 20% of intestines, 100% of livers and 20% of blood samples for ducks, and 100% of blood samples for goose, exceeded the Chinese national standard limit for THg in meat (50 ng/g). At GX, much higher proportions for chickens (breast: 17%; intestine: 50%; heart: 17%; stomach: 34%; liver: 100%; blood; 83%) and ducks (leg: 50%; breast: 58%; intestine: 75%; heart: 42%; stomach: 33%; liver: 92%; blood: 58%) exceeded the Chinese national standard limit for meat THg (Fig. 2A–C). All poultry species showed similar THg distribution patterns (Fig. 2A–C), with higher THg concentrations in liver and blood, and relatively lower concentrations in muscles and other organs (e.g., intestine, heart, stomach).

MeHg concentrations showed a similar distribution patterns to THg (Fig. 2D–F). Liver and blood had the highest geometric mean MeHg concentrations of 19.6 ng/g (range: 3.6–62.8 ng/g; ww) and 8.6 ng/g (range: 1.4–17.6 ng/g; ww), respectively. These values were higher than previous results on MeHg concentrations in the WMM rice (mean: 8.5 ng/g; range: 1.9–27.6 ng/g; dry weight) (Feng et al., 2008). The observation of high MeHg levels in liver and blood indicates that these dietary sources may also be important MeHg exposure sources to WMM residents. Other tissues (e.g., leg, breast, intestine, heart and stomach) showed relatively lower mean MeHg concentrations (3.7–5.4 ng/g, ww), lower than in WMM rice. However, rice MeHg were reported in dry weight concentrations in previous work (Feng et al., 2008).

### 3.2. Understanding THg and MeHg differences among poultry species

THg and MeHg concentrations in tissues were largely variable among poultry species (Fig. 2A–F). At both sites, chickens showed the lowest THg and MeHg levels in tissues, whereas ducks and geese showed relatively higher THg and MeHg. The variability of THg and MeHg levels among poultry species may be determined by their differing food sources. Poultry in the WMM were backyard-fed, and typically prey on field food sources other than field crops from farmers. Chickens mainly feed in arid farmlands, whereas

**Table 1**
Previous results on THg and MeHg concentrations in muscles and organs in poultry.

| Species | Muscle | THg (ng/g) | MeHg(ng/g) | Organs | THg (ng/g) | MeHg(ng/g) | Study area | Reference |
|---|---|---|---|---|---|---|---|---|
| Chicken (ww) | Muscle | N/A | 0.1–3.1[b] | Liver | N/A | 0.5–16.5[b] | Northern Greece | Kambamanoli-Dimou et al., 1989 |
| Chicken (ww) | Muscle | 1.8 ± 1.0[a] (0.8–5.6[b]) | N/A | Liver | 2.1 ± 0.5[a] (1.6–2.8[b]) | N/A | Northern Poland | Falandysz et al., 1994 |
| Turkey (ww) | Muscle | 1.2 ± 0.6[a] (0.6–2.9[b]) | N/A | Liver | 2.9 ± 1.8[a] (1.3–6.2[b]) | N/A | | |
| Duck (ww) | Muscle | 1.9 ± 0.6[a] (0.8–3.4[b]) | N/A | Liver | 3.4 ± 1.5[a] (1.0–7.0[b]) | N/A | | |
| Geese (ww) | Muscle | 1.3 ± 0.8[a] (0.5–4.0[b]) | N/A | Liver | 2.3 ± 1.0[a] (1.0–5.9[b]) | N/A | | |
| Chicken (ww) | Muscle | 0.6 ± 0.1[a] | N/A | | | | Jakarta, Indonesia | Surtipanti et al., 1995 |
| Chicken (ww) | Muscle | 5.0 ± 0.3[a] | N/A | Liver | 5.0 ± 0.4[a] | N/A | Assiut and Beni-Suef Cities, Egypt. | Sharkawy and Ahmed, 2002 |
| Duck (ww) | Muscle | 3.8 ± 0.2[a] | N/A | Liver | 6.1 ± 0.4[a] | N/A | | |
| Geese (ww) | Muscle | 2.0 ± 0.1[a] | N/A | Liver | 4.0 ± 0.3[a] | N/A | | |
| Chicken (ww) | Muscle | 8.4 ± 8.3[a] (0.5–30[b]) | N/A | | | | Brazil | Batista et al., 2012 |
| Chicken (dw) | Leg | 1.3–3.2[b] | N/A | Liver | 1.6–5.5[b] | N/A | Hyderabad, Pakistan | Shah et al., 2010a; Shah et al., 2010b |
| | Breast | 1.4–3.9[b] | N/A | Heart | 1.4–3.3[b] | N/A | | |
| Chicken (dw) | Muscle | 9–11[b] | N/A | | | | Northern Saudi Kingdom | Alturiqi and Albedair., 2012 |
| Chicken (dw) | | | | Liver | 0–2.8 | N/A | Bucharest, Romania | Ghimpeteanu et al., 2012 |
| Chicken (dw) | Muscle | 15 ± 50[a] | N/A | | | | Northern Algeria | Badis et al., 2014 |
| Chicken (dw) | Muscle | 9 ± 10[a] | N/A | | | | Southern Algeria | Badis et al., 2014 |
| Duck (ww) | Muscle | 8±3[a] | N/A | Liver | 22±8[a] | N/A | Shanghai, China | Ji et al., 2006 |
| | | | | Stomach | 22 ± 17[a] | N/A | | |
| Chicken (ww) | Muscle | 10 ± 10[a] (10–20[b]) | N/A | Liver | 110 ± 70[a] (50–250[b]) | N/A | Tarkwa gold mining, Ghana | Bortey-Sam et al., 2015 |
| Duck (ww) | Muscle | 158 ± 34[a] | N/A | Liver | 4465 ± 1567[a] | N/A | Wanshan Mercury Mine, SW China | Ji et al., 2006 |
| | | | | Stomach | 96 ± 13[a] | N/A | | |

[a] Mean±SD.
[b] Range.

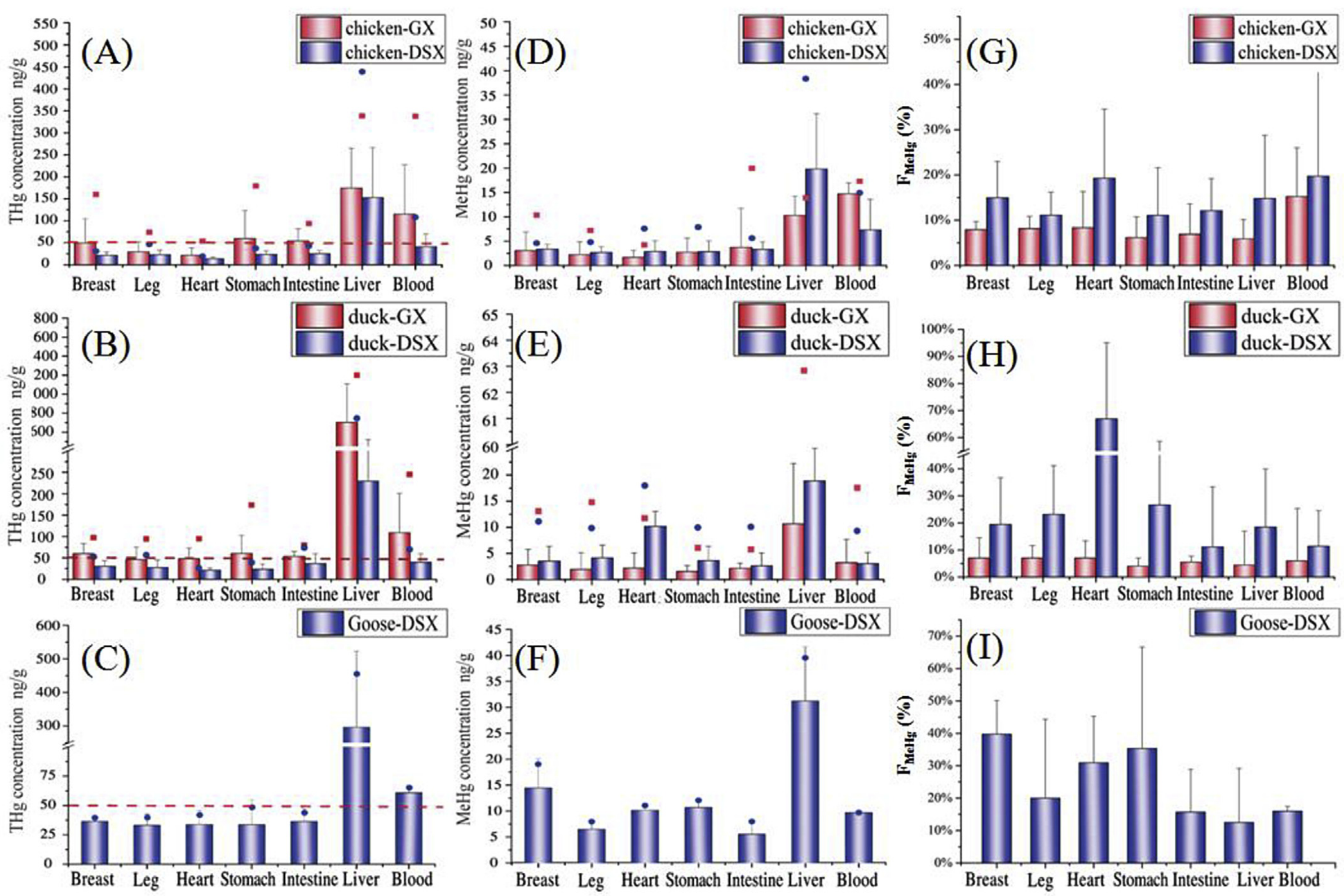


**Fig. 2.** Variations of THg concentration (A–C), MeHg concentration (D–F), and MeHg fraction (G–I) in poultry tissues in the WMM. Red dash lines represent the Chinese national standard limit for THg in meat (50 ng/g). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

ducks and geese are both arid farmlands and rice paddy feeders. Rice paddies are potential zones of Hg methylation enhancement compared to arid farmlands, and rice in the WMM has been shown to exhibit higher MeHg levels than arid land crops such as corn, wheat, etc (Qiu et al., 2008). Invertebrates in rice paddies also have high MeHg, which can be eaten by ducks and geese (Abeysinghe et al., 2016). In general, chickens and ducks in the GX have higher THg concentrations, but lower MeHg concentrations than the DSX chickens and ducks (Fig. 2A and B, D, F), which may due to food sources. For instance, higher THg concentrations and lower MeHg concentrations have been reported for rice at GX (THg: 199 ± 143 ng/g; MeHg: 12 ± 11 ng/g; n = 14; dw) than at DSX (59 ± 40 ng/g; MeHg: 14.6 ± 4.7 ng/g; n = 25; dw) (Feng et al., 2008; Zhang et al., 2010). Overall, we suggest that food source is an important factor affecting THg and MeHg levels in poultry. We also did not find clear differences in THg and MeHg between males and females of any poultry species ($p > 0.05$, *t*-test), although we acknowledge a limited sample size.

The fraction of MeHg ($F_{MeHg}$-calculated in percent of THg concentration) in different tissues of poultry groups at two sites are presented in Fig. 2(G–I). $F_{MeHg}$ values in this study range from 4 to 71%, which is a smaller range and mean than in wild birds. MeHg fractions of 20–100% have been reported for wild bird tissues (Ruelas-Inzunza et al., 2009; Aazami et al., 2012; Campbell et al., 2005; Houserova et al., 2007; Scheuhammer et al., 2007). Differences in MeHg fractions among wild birds and poultry may due to food source differences. Wild birds prey on a variety of food items in the field, whereas poultry feeds on both field food items and crops, and some field food sources (e.g., invertebrates) likely have higher MeHg fractions than crops (Abeysinghe et al., 2016). Food chain length is shorter for poultry than wild birds, and therefore less Hg bioaccumulation. Poultry at DSX have higher $F_{MeHg}$ levels than at GX, which may be explained by the fact that the DSX poultry consumed more field food items than GX poultry. Indeed, due to less intake of field food items, no difference in $F_{MeHg}$ was observed between the GX chickens and ducks. Unlike GX, $F_{MeHg}$ was largely variable among different poultry species at DSX. Geese and ducks in general, have higher $F_{MeHg}$ than chickens (Fig. 2G–I), and this further supports our hypothesis that the DSX poultry consumed more field food items. The higher $F_{MeHg}$ in geese and ducks is consistent with the fact that they feed more extensively in rice paddies.

### 3.3. Understanding THg and MeHg differences in tissues

Higher THg and MeHg concentrations in liver and blood (Fig. 2A–F) is consistent with previous observations (summarized in Table 1). A positive correlation (Fig. 3A, $r = 0.4953$, $p<0.01$) was observed between liver and blood $\log_{10}$ (THg) values, whereas little to no correlations can be observed between other tissues for both THg and MeHg values. It is noteworthy that a higher $F_{MeHg}$ was found in heart and muscles. Particularly, livers show the lowest $F_{MeHg}$ values. Similar trends were also reported by previous studies (DesGranges et al., 1998; Ruelas-Inzunza et al., 2009; Aazami et al., 2012; Teraoka et al., 2012; Kalisinska et al., 2014). Detoxifying MeHg, whereby MeHg is demethylated to inorganic Hg, has shown to take place in bird organs and tissues, especially in liver (Bond and Diamond., 2009; Houserova et al., 2007; Scheuhammer et al., 2007; Ikemoto et al., 2004), as studies have reported negative correlations between $F_{MeHg}$ and THg in livers of wild birds (Thompson and Furness., 1989; Kim et al., 1996). It is interesting to note in our study that the mean $F_{MeHg}$ decreases with an increase of $\text{Log}_{10}$ (THg) in liver of all poultry examined (Fig. 3B, $r = 0.7072$, $p<0.01$). This too, is consistent with previous observations that Hg demethylation processes occur in bird livers.

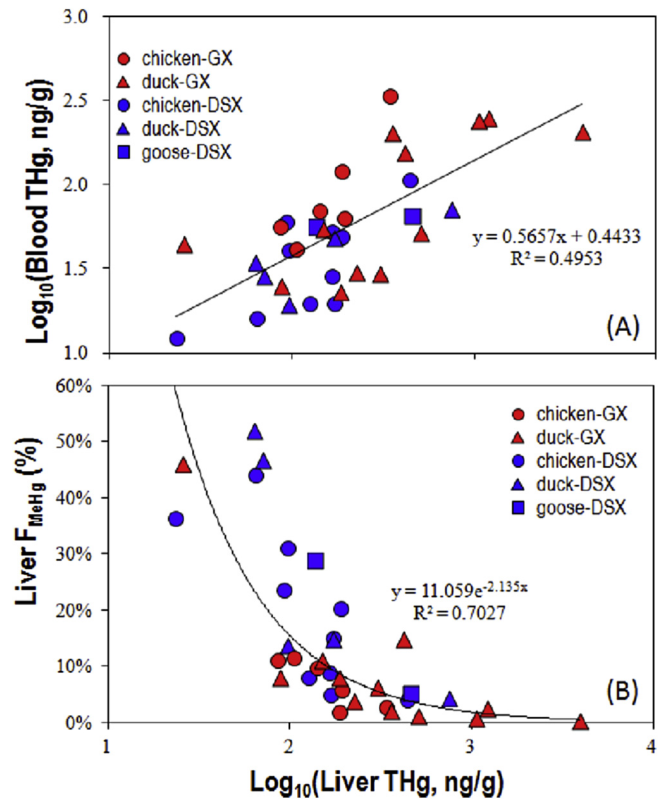


**Fig. 3.** Relations between liver and blood THg concentrations (A), and between liver THg concentration and liver $F_{MeHg}$ fraction (B) for the WMM poultry.

Studies hypothesized that enzyme systems for demethylating MeHg exist in the liver, and this hypothesis has been shown in laboratory feeding experiments using rats and mice (Yasutake and Hirayama, 2001; Roos, 2011).

### 3.4. THg and MeHg burdens in tissues of poultry and their risks to local inhabitants

The average body weight (bw) of 60 kg has been previously proposed for adults from southern China (Zhang et al., 2010; Li et al., 2012). The reference dose of THg and MeHg are 0.57 μg/kg bw/day and 0.10 μg/kg bw/day, according to the World Health Organization and the U.S. Environmental Protection Agency, respectively (USEPA, 2001b; JECFA, 2003). In other words, daily intake of no more than 34.2 μg and 6.0 μg for THg and MeHg, respectively, are recommended for WMM adults. Total burdens of THg in chickens, ducks and geese range from 15.3 to 238.1 μg, 39.9–275.2 μg, and 83.8–93.4 μg, respectively. Total burdens of MeHg in chickens, ducks and geese range from 2.2 to 15.6 μg, 3.5–14.7 μg, and 15.4–29.7 μg, respectively. A substantial amount of poultry in the WMM, especially ducks and geese, have total burdens of THg and MeHg that exceed the daily intake limits for THg and MeHg.

Relative distribution of THg and MeHg burdens in tissues show that for most poultry species, muscles (leg and breast) have less THg and MeHg burdens than the sum of other tissues (Fig. 4). Organs and blood account, in general, for >50% of total burdens for THg and MeHg in poultry, and according to our study, these tissues only account about 20% of the total weight. High THg and MeHg burdens in organs and blood, therefore, poses a risk to the local population in the WMM. In China and many other countries, eating poultry organs is common and liver and blood in this region

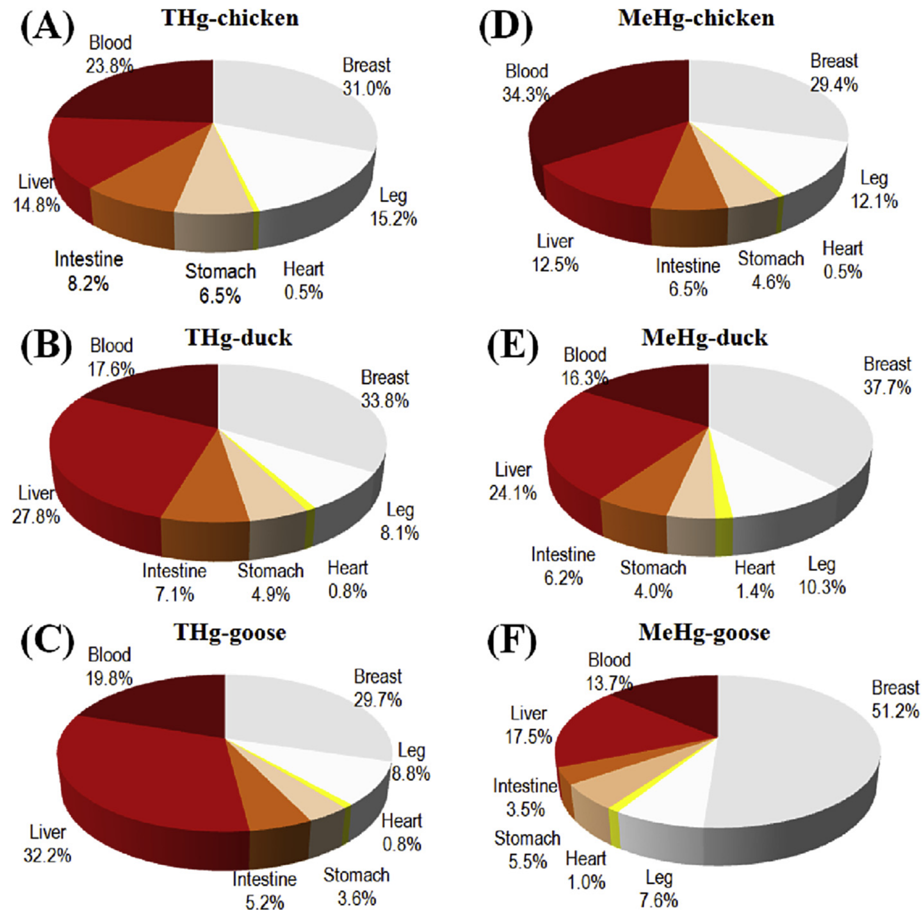


**Fig. 4.** Relative distribution of THg (A–C) and MeHg (D–F) burdens in poultry tissues at the GX and DSX.

have high THg and MeHg concentrations that result in high risk from human Hg exposure. Many cuisines that contain more than one liver are also served in restaurants. The highest THg burdens in liver (74.3 μg) is about 2.2 times than the daily intake limit for THg (34.2 μg), and the highest MeHg burdens in liver (5.3 μg) is also close to the daily intake limit for MeHg (6.0 μg). Considering this information, eating livers and blood of local poultry is strongly not recommended for WMM residents.

According to their geometric mean THg and MeHg concentrations, we further estimated the maximum amount of muscles, organs and blood that should be eaten to not exceed the daily intake limit for THg and MeHg. To not exceed the daily intake limit for THg, our results suggest that the maximum amount for chicken leg, breast, heart, stomach, intestine, liver, and blood are 1384 g, 1498 g, 2315 g, 1214 g, 1081 g, 257 g, and 717 g, respectively; the maximum amount for duck leg, breast, heart, stomach, intestine, liver, and blood are 750 g, 1041 g, 986 g, 858 g, 752 g, 134 g, and 573 g, respectively; and the maximum amount for goose leg, breast, heart, stomach, intestine, liver, and blood are 941 g, 1051 g, 1040 g, 1131 g, 964 g, 137 g, and 562 g, respectively. To not exceed the daily intake limit for MeHg, our results showed that the maximum amount for chicken leg, breast, heart, stomach, intestine, liver, and blood are 2062g, 2657 g, 2886 g, 2396 g, 1932g, 432 g, and 703 g, respectively; the maximum amount for duck leg, breast, heart, stomach, intestine, liver, and blood are 1495 g, 1830g, 1278 g, 2314 g, 1936g, 346 g, and 1387 g, respectively; and the maximum amount for goose leg, breast, heart, stomach, intestine, liver, and blood are 415 g, 922 g, 590 g, 561 g, 1077 g, 192 g, and 617 g, respectively.

## 4. Conclusions

As a product of human population growth and industry, environmental Hg contamination continues to be a global issue with increasing risk to wildlife and human populations in many parts of the world. Mercury exposure due to non-fish food sources is thought to be limited due to the short nature of most farmed animal food chains. In this study however, we observed high levels of Hg (especially liver and blood) can be associated with poultry surrounding mercury mines. A substantial amount of chickens, ducks and geese in the WMM have total burdens of THg and MeHg that exceed the daily intake limits for THg (34.2 μg) and MeHg (6 μg). The enrichment of Hg in poultry is likely resulted from bioaccumulation of Hg from local food stuff (e.g., crops and vegetables). This study demonstrated poultry meat products (especially liver and blood) can be an important Hg exposure source for the WMM residents.

## Acknowledgments

Runsheng Yin was fund by the Chinese Academy of Sciences through the Hundred Talent Plan. This work was also supported by the National Basic Research Program of China (No: 2013CB430004), the Natural Science Foundation of China (41303014, 41173024), and the CAS Youth Innovation Promotion Association (2011280, 2013256). Four anomalous reviewers and editor Joerg Rinklebe were acknowledged for their thoughtful comments that have largely improved the quality of this work.

## References


Aazami, J., Esmaili-Saria, A., Bahramifar, N., Savabieasfahani, M., 2012. Total and organic mercury in liver, kidney and muscle of waterbirds from wetlands of the caspian sea, Iran. Bull. Environ. Contam. Toxicol. 89 (1), 96–101.

Abeysinghe, K., Yang, X., Goodale, E., 2016. Total and methylmercury concentrations over a gradient of contamination in earthworms living in rice paddy soil. Environ. Toxicol. Chem. http://dx.doi.org/10.1002/etc.3643.

Alturiqi, A.S., Albedair, L.A., 2012. Evaluation of some heavy metals in certain fish, meat and meat products in Saudi Arabian markets. Egypt. J. Aquatic Res. 38 (1), 45–49.

Badis, B., Rachid, Z., Esma, B., 2014. Levels of selected heavy metals in fresh meat from cattle, sheep, chicken and camel produced in Algeria. Annu. Res. Rev. Biol. 4 (8), 1260–1267.

Batista, B.L., Grotto, D., Carneiro, M.F., Barbosa Jr., F., 2012. Evaluation of the concentration of nonessential and essential elements in chicken, pork, and beef samples produced in Brazil. J. Toxicol. Environ. health. Part A 75 (21), 1269–1279.

Bond, A.L., Diamond, A.W., 2009. Total and methyl mercury concentrations in seabird feathers and eggs. Archives Environ. Contam. Toxicol. 56 (2), 286–291.

Bortey-Sam, N., Nakayama, S.M., Ikenaka, Y., Akoto, O., Baidoo, E., Yohannes, Y.B., Mizukawa, H., Ishizuka, M., 2015. Human health risks from metals and metalloid via consumption of food animals near gold mines in Tarkwa, Ghana: estimation of the daily intakes and target hazard quotients (THQs). Ecotoxicol. Environ. Saf. 111, 160–167.

Campbell, L.M., Norstrom, R.J., Hobson, K.A., Muir, D.C., Backus, S., Fisk, A.T., 2005. Mercury and other trace elements in a pelagic Arctic marine food web (Northwater Polynya, Baffin Bay). Sci. Total Environ. 247–263.

DesGranges, J., Rodrigue, J., Tardif, B., Laperle, M., 1998. Mercury accumulation and biomagnification in ospreys (Pandion haliaetus) in the james bay and hudson bay regions of quebec. Archives Environ. Contam. Toxicol. 35 (2), 330–341.

Falandysz, J., Kotecka, W., Kannan, K., 1994. Mercury, lead, cadmium, manganese, copper, iron and zinc concentrations in poultry, rabbit and sheep from the northern part of Poland. Sci. Total Environ. 141, 51–57.

Feng, X., Li, P., Qiu, G., Wang, S., Li, G., Shang, L., Meng, B., Jiang, H., Bai, W., Li, Z., Fu, X., 2008. Human exp osure to methylmercury through rice intake in mercury mining areas, Guizhou province, China. Environ. Sci. Technol. 42 (1), 326–332.

Ghimpeteanu, O.M., Das, K., Melitaru, M., Scippo, M.L., 2012. Assessment of Heavy Metals and Mineral Nutrients in Poultry Liver Using Inductively Coupled Plasma-mass Spectrometer (ICP-MS) and Direct Mercury Analyzer (DMA), vol.69. Bulletin of the University of Agricultural Sciences & Veterinary, pp. 353–359.

Houserova, P., Kuban, V., Kracmar, S., Sitko, J., 2007. Total mercury and mercury species in birds and fish in an aquatic ecosystem in the Czech Republic. Environ. Pollut. 145 (1), 185–194.

Ikemoto, T., Kunito, T., Anan, Y., Tanaka, H., Baba, N., Miyazaki, N., Tanabe, S., 2004. Association of heavy metals with metallothionein and other proteins in hepatic cytosol of marine mammals and seabirds. Environ. Toxicol. Chem. 23 (8), 2008–2016.

JECFA (Joint FAO/WHO Expert Committee on Food Additives), 2003. Summary and Conclusions of the Sixty-first Meeting of the Joint FAO/WHO Expert. Committee on Food Additives, Rome, Italy.

Ji, X., Hu, W., Cheng, J., Yuan, T., Xu, F., Qu, L., Wang, W., 2006. Oxidative stress on domestic ducks (Shaoxing duck) chronically exposed in a Mercury-Selenium coexisting mining area in China. Ecotoxicol. Environ. Saf. 64 (2), 171–177.

Jiang, G., Shi, J., Feng, X., 2006. Mercury pollution in China. Environ. Sci. Technol. 40 (12), 3672–3678.

Kalisinska, E., Gorecki, J., Lanocha, N., Okonska, A., Melgarejo, J.B., Budis, H., Rzad, I., Golas, J., 2014. Total and methylmercury in soft tissues of white-tailed eagle (Haliaeetus albicilla) and Osprey (Pandion haliaetus) collected in Poland. Ambio 43 (7), 858–870.

Kambamanoli-Dimou, A., Kilikidis, S., Kamarianos, A., 1989. Methylmercury concentrations in broiler's meat and hen's meat and eggs. Bull. Environ. Contam. Toxicol. 42 (5), 728–734.

Kim, E.Y., Murakami, T., Saeki, K., Tatsukawa, R., 1996. Mercury levels and its chemical form in tissues and organs of seabirds. Archives Environ. Contam. Toxicol. 30 (2), 259–266.

Li, P., Feng, X., Qiu, G., Shang, L., Wang, S., Meng, B., 2009. Atmospheric mercury emission from artisanal mercury mining in Guizhou Province, Southwestern China. Atmos. Environ. 43 (14), 2247–2251.

Li, P., Feng, X., Yuan, X., Chan, H.M., Qiu, G., Sun, G.X., Zhu, Y.G., 2012. Rice consumption contributes to low level methylmercury exposure in southern China. Environ. Int. 49, 18–23.

Mori, N., Yamamoto, M., Tsukada, E., Yokooji, T., Matsumura, N., Sasaki, M., Murakami, T., 2012. Comparison of in vivo with in vitro pharmacokinetics of mercury between methylmercury chloride and methylmercury cysteine using rats and caco2 cells. Archives Environ. Contam. Toxicol. 63, 628–636.

Peterson, S.H., Ackerman, J.T., Eagles-Smith, C.A., 2017. Mercury contamination and stable isotopes reveal variability in foraging ecology of generalist California gulls. Ecol. Indic. 74, 205–215.

Pirrone, N., Cinnirella, S., Feng, X., Finkelman, R.B., Friedli, H.R., Leaner, J., Mason, R., Mukherjee, A.B., Stracher, G.B., Streets, D.G., Telmer, K., 2010. Global mercury emissions to the atmosphere from anthropogenic and natural sources. Atmos. Chem. Phys. 10 (13), 5951–5964.

Pirrone, N., Mahaffey, K.R., 2005. Dynamics of Mercury Pollution on Regional and Global Scales: Atmospheric Processes and Human Exposures Around the World. Springer Science & Business Media.

Qiu, G., Feng, X., Li, P., Wang, S., Li, G., Shang, L., Fu, X., 2008. Methylmercury accumulation in rice (Oryza sativa L.) grown at abandoned mercury mines in Guizhou, China. J. Agric. Food Chem. 56 (7), 2465–2468.

Roos, D.H., 2011. Modulation of methylmercury uptake by methionine: prevention of mitochondrial dysfunction in rat liver slices by a mimicry mechanism. Toxicol. Appl. Pharmacol. 252 (1), 28–35.

Rothenberg, S.E., Feng, X., Dong, B., Shang, L., Yin, R., Yuan, X., 2011. Characterization of mercury species in brown and white rice (Oryza sativa L.) grown in water-saving paddies. Environ. Pollut. 159 (5), 1283–1289.

Ruelas-Inzunza, J., Hernandez-Osuna, J., Paez-Osuna, F., 2009. Organic and total mercury in muscle tissue of five aquatic birds with different feeding habits from the SE gulf of California, Mexico. Chemosphere 76 (3), 415–418.

Scheuhammer, A.M., Meyer, M.W., Sandheinrich, M.B., Murray, M.W., 2007. Effects of environmental methylmercury on the health of wild birds, mammals, and fish. Ambio 36 (1), 12–18.

Shah, A.Q., Kazi, T.G., Baig, J.A., Afridi, H.I., Kandhro, G.A., Arain, M.B., Kolachi, N.F., Wadhwa, S.K., 2010a. Total mercury determination in different tissues of broiler chicken by using cloud point extraction and cold vapor atomic absorption spectrometry. Food Chem. Toxicol. 48 (1), 65–69.

Shah, A.Q., Kazi, T.G., Baig, J.A., Afridi, H.I., Kandhro, G.A., Khan, S., Kolachi, N.F., Wadhwa, S.K., 2010b. Determination of total mercury in chicken feed, its translocation to different tissues of chicken and their manure using cold vapour atomic absorption spectrometer. Food Chem. Toxicol. 48 (6), 1550–1554.

Sharkawy, A.A., Ahmed, E.K., 2002. Determination of lead, cadmium, mercury and copper concentrations in hen's eggs at Assiut Governorate. Assiut Veterinary Med. J. 48 (95), 45–59.

Surtipanti, S., Suwirma, S., Yumiarti, S., Mellawati, Y., 1995. Determination of heavy metals in meat, intestine, liver, eggs, and chicken using neutron activation analysis and atomic absorption spectrometry. Atom Indonesia. Life Earth Health Sci. 21 (1), 1–8.

Teraoka, H., Tagami, Y., Kudo, M., Miura, Y., Okamoto, E., Matsumoto, F., Koga, K., Uebayashi, A., Shimura, R., Inoue, M., Momose, K., Masatomi, H., Kitazawa, T., Hiraga, T., Subramanian, A., 2012. Changes of mercury contamination in red-crowned cranes, grus japonensis, in east Hhokkaido, Japan. Archives Environ. Contam. Toxicol. 63 (1), 153–160.

Thompson, D.R., Furness, R.W., 1989. The chemical form of mercury stored in South Atlantic seabirds. Environ. Pollut. 60, 305–317.

USEPA, 1997. Mercury Study Report to Congress. Washington, USA.

USEPA, 2001a. Method 1630: Methyl Mercury in Water by Distillation, Aqueous Ethylation, Purge and Trap, and Cold Capor Atomic Fluorescence Spectrometry. U.S. Environmental Protection Agency, Washington, DC, pp. 1–41. EPA-821-R-01-020.

USEPA, 2001b. Water Quality Criterion for the Protection of Human Health: Methylmercury. U.S. EPA, Washington, DC. EPA-823-R-01–001.

USEPA, 2002. Method 1631: mercury in water by oxidation, purge and trap, and cold vapor atomic fluorescence spectrometry. U. S. Environmental Protection Agency, Washington, DC, pp. 1–33. EPA-821-R-02-019.

Ullrich, S.M., Tanton, T.W., Abdrashitova, S.A., 2001. Mercury in the aquatic environment: a review of factors affecting methylation. Crit. Rev. Environ. Sci. Technol. 31 (3), 241–293.

WHO, 1993. Environmental Health Criteria 101: Methylmercury. World Health Organization, Geneva, Switzerland.

Yasutake, A., Hirayama, K., 2001. Evaluation of methylmercury biotransformation using rat liver slices. Archives Toxicol. 75 (7), 400–406.

Yin, R., Feng, X., Wang, J., Li, P., Liu, J., Zhang, Y., Chen, J., Zheng, L., Hu, T., 2013a. Mercury speciation and mercury isotope fractionation during ore roasting process and their implication to source identification of downstream sediment in the Wanshan mercury mining area, SW China. Chem. Geol. 336, 72–79.

Yin, R., Feng, X., Meng, B., 2013b. Stable mercury isotope variation in rice plants (Oryza sativa L.) from the Wanshan mercury mining district, SW China. Environ. Sci. Technol. 47 (5), 2238–2245.

Yin, R., Gu, C., Feng, X., et al., 2016a. Transportation and transformation of mercury in a calcine profile in the Wanshan Mercury Mine, SW China. Environ. Pollut. 219, 976–981.

Yin, R., Gu, C., Feng, X., Hurley, J.P., Krabbenhoft, D.P., Lepak, R.F., Zhu, W., Zheng, L., Hu, T., 2016b. Distribution and geochemical speciation of soil mercury in Wanshan Hg mine: effects of cultivation. Geoderma 272, 32–38.

Zhang, H., Feng, X., Larssen, T., Qiu, G., Vogt, R.D., 2010. In inland China, rice, rather than fish, is the major pathway for methylmercury exposure. Environ. Health Perspect. 118 (9), 1183–1186.