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Antimony exposure and speciation in human biomarkers near an active mining area in Hunan, China

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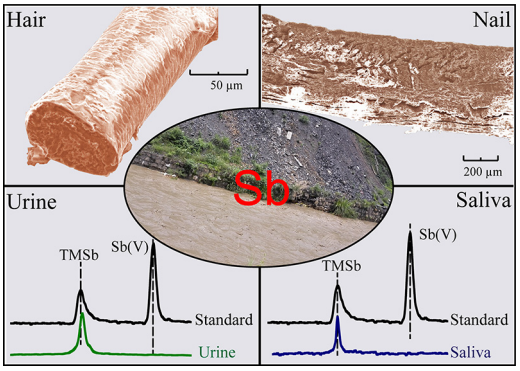
HIGHLIGHTS

- Sb content in water and food near the mining area is higher than the reference level.
- Hair can be used as a non-invasive biomarker for Sb exposure.
- Interaction effects explained most of the Sb concentration in biomarkers.
- Methylated Sb was the predominant species in urine and saliva.

GRAPHICAL ABSTRACT



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ABSTRACT

Antimony (Sb) exposure threatens human health. To identify human biomarkers for Sb exposure, we analyzed 480 environmental samples from an active Sb mining area in Hunan, China. Elevated Sb concentrations exceeding the reference level were detected in drinking water (70% of $n = 83$ total samples), foods (80%, $n = 188$), urine (95%, $n = 63$), saliva (44%, $n = 48$), hair (80%, $n = 51$) and nails (83%, $n = 47$). Drinking water contributed 85%–100% of the average daily dose (ADD) of Sb, and the total ADD (11.7 μg/kg bodyweight/day) was up to thirty times higher than the oral reference dose (0.4 μg/kg bodyweight/day) as recommended by USEPA. A positive correlation was found between ADD and Sb content in hair ($p = 0.02$), but not in urine ($p = 0.051$), saliva ($p = 0.52$) or nails ($p = 0.85$), suggesting that hair is the best non-invasive biomarker. Micro X-ray fluorescence analysis indicated that Sb is distributed in discrete spots in hair and nails, and Sb distribution is correlated with other metals. Methylated Sb species were predominant in urine (46%–100%) and saliva (74%–100%) in collected samples, implying that the human metabolic system adopts methylation as an effective pathway to detoxify and excrete Sb.
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1. Introduction

Exposure to antimony (Sb) is a great threat to human health (El Shanawany et al., 2017; Guo et al., 2016) through air, drinking water and food (Fort et al., 2016; Nelson et al., 2011; Wu et al., 2011). Drinking water with high Sb content is an important source of human Sb intake. Moreover, Sb-laden water and soil unavoidably lead to Sb accumulation

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in cereals and vegetables in polluted areas (He, 2007; Zhang et al., 2014). A previous study found that the dietary Sb intake by residents near Sb mining areas is 1.5 times higher than the tolerable daily intake (360 μg/day) (Wu et al., 2011).

Sb dietary intake may lead to elevated Sb levels in noninvasive human biomarkers such as urine (Filella et al., 2013; Shao et al., 2017), hair (Huang et al., 2015; Rapant et al., 2006), nails (Rapant et al., 2006) and saliva (Chicharro et al., 1999; Tu and He, 2015). Among the four biomarkers, urine and saliva are used to reveal short-term Sb exposure (Lidia et al., 2011; Sun et al., 2007), while hair and nails indicate long-term Sb exposure (Huang et al., 2015). In fact, the Sb content in the urine, hair and nails of residents in polluted areas can be over 100 times higher than that of unexposed people (Rapant et al., 2006; Wu et al., 2011). The mean Sb value in saliva is 1.8 μg/L in unpolluted areas (Zaichick et al., 1995), but little is known about saliva Sb concentrations in polluted areas (Filella et al., 2013). More importantly, the correlation between Sb levels in biomarkers and dietary Sb intake is still unknown. This correlation, however, is central to identifying the best noninvasive human biomarker for Sb exposure.

The toxicity of Sb depends mainly on its speciation and is assessed from species levels in human biomarkers (Cui et al., 2013; Ji et al., 2017). However, only limited articles have reported on Sb speciation in urine and saliva, and their focus has been on developing analytical methods (Filella et al., 2013; Hansen and Pergantis, 2008). For example, a previous study measured urinary Sb species from two occupationally exposed people working in a battery plant, and found that Sb(V) was the predominant Sb species, followed by trimethylantimony (TMSb) (Krachler and Emons, 2000). Until now, no knowledge has been available about Sb species in hair, nails (Ponomarenko et al., 2014; Xing et al., 2013), and saliva (Filella et al., 2013), although these biomarkers are usually used for metal exposure assessment (Bhowmick et al., 2014; Israelsson et al., 2015; Katsikini et al., 2010; Lew et al., 2010).

The purpose of this study was to identify human biomarkers for Sb exposure. The study region was an active Sb mining area in Hunan, China. The dietary intake of Sb and its correlation with Sb accumulation and speciation in biomarkers were analyzed. High-performance liquid chromatography (HPLC) coupled with atomic fluorescence spectrometry (AFS) was used to analyze Sb speciation in urine and saliva samples. The Sb distribution in hair and nails was explored by micro X-ray fluorescence (μ-XRF). This comprehensive study will provide insights into Sb exposure and its health risks.

2. Materials and methods

2.1. Sample collection

The study area lies in an active mining area in Hunan, China (Fig. 1). This anthropogenic Sb pollution region covers an area of about 70 km^2 containing a large Sb deposit (Wang et al., 2011; Wei et al., 2015). Samples of drinking water (n = 83), garden vegetables (n = 168), eggs (n = 11), rice (n = 9), urine (n = 63), saliva (n = 48), nails (n = 47) and hair (n = 51) were collected as detailed in the supporting material (SM). Local residents, including 54 males and 46 females in 20 villages, were asked to complete a questionnaire with information such as sex, age, smoking habits, years working in mine, and residence time. Ten families were randomly selected to investigate dietary habits in the study area, as detailed in Table S1.

2.2. Sample preparation and analysis

Vegetables and rice samples were washed with deionized (DI) water to remove dust particles and soil, then crushed and dried at 60 °C in a drying oven for microwave digestion analysis. Hair and nail samples were washed successively by DI water-methanol-DI water, and then dried at 60 °C for microwave digestion (Cui et al., 2013). The digestion of solid samples was completed with a microwave digestion system (MARS, CEM Corporation, U.S.) as shown in the SM.

For Sb speciation analysis in liquid samples, the urine was centrifuged at 12,000 ×g for 10 min, and the supernatant was filtered through a 0.22 μm membrane. Saliva was vortexed for 1 min and diluted three times with DI water. The diluted saliva samples were ultrasonicated for 5 min and then centrifuged at 6000 ×g for 15 min, and finally the supernatant was filtered as described above (Lew et al., 2010).

The total dissolved Sb was detected using an atomic fluorescence spectrometer (AFS-8800 spectrometer, Jitian, China). Online hydride generation was achieved by reacting with 2% wt. KBH_4 and 7% wt. HCl. The hydride was atomized in a hydrogen flame and the fluorescence signal was recorded. For Sb speciation analysis, Sb(III), Sb(V) and TMSb in urine and saliva samples were determined using HPLC-AFS (Vinas et al., 2006; Wang et al., 2018). An anion exchange column was used (PRP-X100, 4.1 × 250 mm, 10 μm; Hamilton) to separate different Sb species. The details of Sb analysis are shown in the SM.

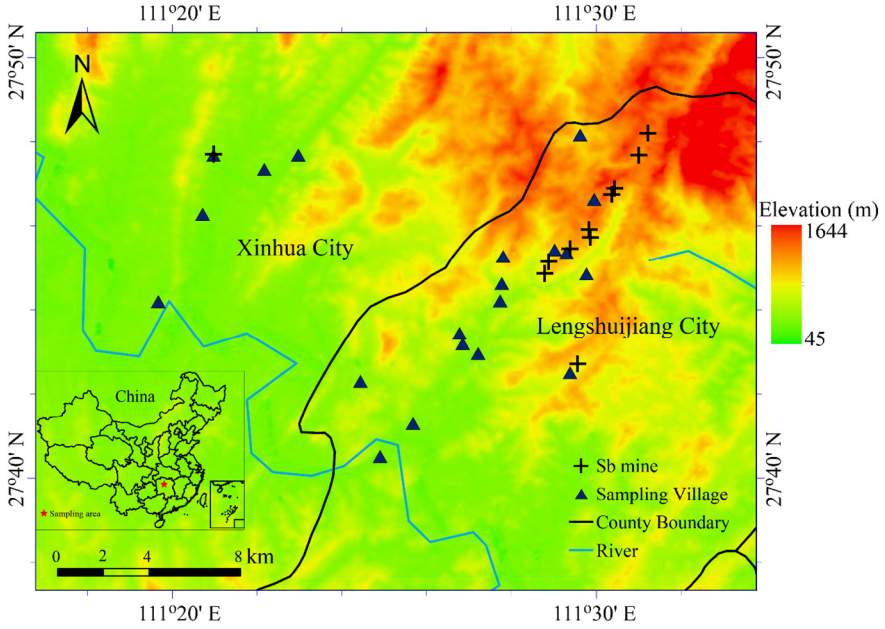

Fig. 1. Map of the study area.

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