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Westernized diets lower arsenic gastrointestinal bioaccessibility but increase microbial arsenic speciation changes in the colon

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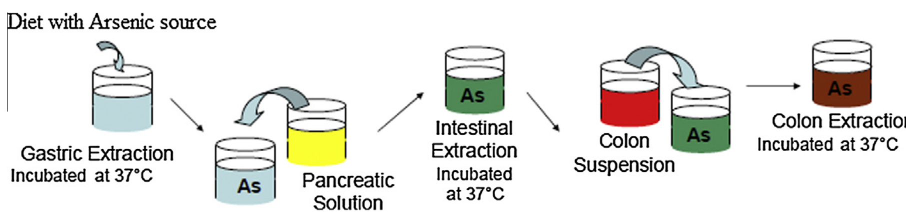
HIGHLIGHTS

- Effect of diet matrix on bioaccessibility of different arsenic species in diet.
- Effect of diet matrix on biotransformation of different arsenic species in diet.
- The difference in formation of new toxic arsenic species basing on diet matrix.
- How difference in diet matrix might be important in case of oral arsenic exposure.

GRAPHICAL ABSTRACT



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ABSTRACT

Arsenic (As) is an important contaminant present in food and water. Several studies have indicated that the occurrence of As based skin lesions is significantly different when root and gourd rich diets are consumed compared to meat rich diets. Additionally, urinary As speciation from orally exposed individuals appears to depend on the composition of the diet. These observations imply that diet composition can affect both the bioavailable As fraction as the As speciation in the body. In this study, we used the *in vitro* gastrointestinal method (IVG) to evaluate how an Asian type diet (fiber rich) and a Western type diet (fat and protein rich), differ in their capability to release inorganic As (iAs^V) and dimethyl arsinate (DMA^V) from a rice matrix following gastrointestinal digestion. Moreover, we used a validated dynamic gut simulator to investigate whether diet background affects As metabolism by gut microbiota in a colon environment. An Asian diet background resulted in a larger As bioaccessibility (81.2%) than a Western diet background (63.4%). On the other hand, incubation of As contaminated rice with human colon microbiota in the presence of a Western type diet resulted in a larger amount of hazardous As species – monomethyl arsonite and monomethylmonothio arsonate – to be formed after 48 h. The permeability of these As species (60.5% and 50.5% resp.) across a Caco-2 cell line was significantly higher compared to iAs^V and DMA^V (46.5% and 28% resp.). We conclude that dietary background is a crucial parameter to incorporate when predicting bioavailability with bioaccessibility measurements and when assessing health risks from As following oral exposure.

1. Introduction

Arsenic (As) is a naturally occurring element in food, soil, air, and water. The major sources of exposure are from food and water. A variety of adverse health effects, e.g., skin and internal cancers, cardiovascular, and neurological effects, have been attributed to As exposure (Chen et al., 1992).

Human health effects from chronic As exposure have also been reported mainly in populations with low socioeconomic status and high levels of malnutrition. Diet composition has a significant

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effect on contaminant bioaccessibility and thereby also oral bioavailability. We previously (Alava et al., 2012a) demonstrated that lipids have a large impact on arsenic bioaccessibility, but also that this depends on the As speciation. Some other studies stated that in protein rich rice (Narukawa and Chiba, 2010) As^{III} is tightly bound to the thiol groups of the proteins and may influence its bioaccessibility. Previous studies demonstrated a relationship between diet composition and levels of MMA^{III} found in urine after ingesting inorganic As (Steinmaus et al., 2005). Additionally, some studies reported that people, who consumed diets relatively high in roots and gourds as opposed to meat or other kinds of vegetables, were less likely to develop As-related skin lesions and also showed different metabolic rate of consumed arsenic (Lammon and Hood, 2004).

Differences in dietary matrix composition may affect release of As from the food source after ingestion. Food contains both organic and inorganic As, whereas drinking water primarily contains inorganic forms of As. In a scenario of dietary As exposure, rice has been demonstrated to be one of the major foodstuffs contributing to human As exposure (Williams et al., 2007) and also widely consumed (Raab et al., 2009). Interestingly, As biotransformations by human gut microbiota are even reported in case of As contaminated rice (Sun et al., 2012). Moreover, rice is an important carbohydrate sources in both Asian and Western style diets. Trenary et al. reported bioaccessibility of arsenic species from different types of rice in USA by using *in vitro* setup. These people extended their results to estimate the exposure level to American people by using probabilistic exposure model. All these above observations and reports lead to an interesting and important question; what is the effect of diet matrix on bioavailability of arsenic from rice?

As extension to the above question the second part to focus is presystemic metabolism. Upon oral exposure of As contaminated food, gastrointestinal digestion may release a fraction of the matrix-bound As: this is termed the bioaccessible fraction, which is considered a conservative estimator of the oral bioavailable As fraction. The As fraction that is not absorbed across the small intestinal epithelium and the remaining matrix-bound As will reach the colon region. Here, microbial breakdown of the remaining food matrix may further contribute to As release, while the released As in general may get subjected to the diverse metabolic potency of the vast endogenous microbial community. Such conversion of As by colon microbiota is termed presystemic metabolism. Notably, some studies have revealed that microorganisms are important contributors to arsenic speciation changes. A wide range of microbial metalloid biotransformations have been revealed, including oxidation, reduction, methylation and thiolation (Diaz-Bone and Van de Wiele, 2010). Recently, As biotransformations by human gut microbiota have been characterized using As contaminated soils and rice (Van De Wiele et al., 2010; Alava et al., 2012b).

Summarizing the above paragraphs; as rice being one of the major source of arsenic through food and diet components like lipids and fiber affecting the bioaccessibility of ingested arsenic species we aimed to study whether and how bioaccessibility of As is affected by diet composition and further how these diets effect the biotransformations of ingested As. Arsenic through rice is major problem in western countries like America (Trenary et al., 2012) and Eastern countries like Bangladesh, India and Taiwan (Meharg and Rahman, 2003, Chowdhury et al., 2000, Pal et al., 2009), and there is significant difference between diet compositions of these two regions; Asian diet (low in fat, protein and high in carbohydrate) and Western diet (high fat and protein) (Suhana et al., 1999 and Park et al., 2006). We choose these two diet compositions to elucidate whether and how bioaccessibility of As is affected by diet composition, further how these diets effect the biotransformations of As.

2. Experimental

2.1. Arsenic standards and samples

Standards used were: $NaAsO_2$ solution (VWR, Belgium) for Arsenite (As^{III}), $Na_2HAsO_4{\cdot}7H_2O$ (Fluka, Switzerland) for Arsenate (As^V), $(CH_3)_2AsO_2Na{\cdot}3H_2O$ (Fluka, Switzerland) for dimethyl arsinous acid (DMA^V), and $(CH_3)AsNa_2O_3{\cdot}6H_2O$ (Chemservice, Belgium) for mono methyl arsonous acid (MMA^V). Monomethylmonothioarsinic acid (MMMTA) was prepared in the lab. Stability, purity and procedure were already published (Alava et al., 2012). The structure of the product was checked by LC–ESI–MS and MS/MS. MMA^{III} and DMA^{III} were purchased from Argus Chemicals (Italy).

2.2. Certified reference material (CRM) and rice sample

NIST 1568a Rice Flour (National Institute of Standards and Technology, NIST, USA) was used to check the recovery of total arsenic. The certified value of total As in NIST SRM 1568a is $290 \pm 30\ \mu g\ kg^{-1}$, while the As speciation in this reference material is not defined. Basmati rice from Indian origin was purchased from a local supermarket (Colruyt, Gent, Belgium).

2.3. Analysis of rice samples for As content

All rice samples were microwave digested at 80 °C for 30 min using water as extraction solvent. This method has previously been proven to be successful for extracting total As from rice (Alava et al., 2012). NIST 1568a Rice Flour (National Institute of Standards and Technology, NIST, USA) was used to check the recovery of total arsenic. The certified value of total As in NIST SRM 1568a is $290 \pm 0.03\ \mu g\ kg^{-1}$. Digested samples were filtered using a 0.45 μm syringe-type PVDF membrane filter and the filtrate was diluted to 25 mL using double distilled deionized water. This filtrate was analyzed for total arsenic content using ICP–MS (Table 1). The same filtrate was used for speciation analysis using HPLC–ICP–MS (Alava et al., 2011).

2.4. Setup

To better mimic *in vivo* gastrointestinal behavior of As, all regions – stomach, small intestine, and colon – were simulated. The IVG method was previously validated against *in vivo* data for As bioaccessibility (Rodriguez and Basta, 1999) in the upper digestive tract (gastric, small intestine), whereas the SHIME has been validated against *in vivo* data for the colon microbial community composition and metabolic activity toward drugs and phytoestrogens (Molly et al., 1994; Possemiers et al., 2006). To mimic the inter variability of microorganisms in different persons, fecal matter from ten different people representing both Asian and Western countries is collected, mixed well into a homogenous mixture and used as a fecal inoculate in SHIME reactor to develop the microbial source.

Table 1
Optimized instrumental settings for ICP-MS.

Detection DRC mode (As)	
Instrument	Perkin Elmer Elan DRC-e
Plasma RF power	1250 W
Nebulizer flow rate	0.7–1.1 mL min^{-1} (optimized daily)
Lens voltage and autolens	Optimized for AsO daily
Dwell times *m/z*	91 (AsO 500 ms)
Reaction gas	O_2, 0.75 mL min^{-1}
Cell parameters Rpq	0.6