Science of the Total Environment 536 (2015) 368–381

Contents lists available at ScienceDirect

Science of the Total Environment

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

Review

# Arsenic relative bioavailability from diet and airborne exposures: Implications for risk assessment

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## HIGHLIGHTS

• Focuses on relative bioavailability of inorganic arsenic (iAs) in foods and fly ash
• Identifies iAs bioavailability related to coal fly ash inhalation and ingestion
• Explores current database strengths, limitations and needs for dietary iAs
• Discusses implications of iAs bioavailability for human health risk assessment

## ARTICLE INFO

*Article history:*
Received 19 December 2014
Received in revised form 18 May 2015
Accepted 19 May 2015
Available online 28 July 2015

Editor: D. Barcelo

*Keywords:*
Bioavailability
Bioaccessibility
Arsenic
Coal ash
Rice

## ABSTRACT

Major human environmental health concern has been associated with inorganic arsenic (iAs) in drinking water in which dissolved iAs is highly bioavailable. More recently health concerns have been raised regarding the extent of iAs exposure via food and other potential sources. Arsenic relative bioavailability (RBA) in soil is known to be variable; the extent and role of iAs bioavailability in food are not well characterized. iAs in coal fly ash and bottom ash are other potential exposure media for which RBA has not been well characterized. A comprehensive literature search was conducted to support evaluation of the contribution of food and coal fly ash to iAs exposure. Few studies were found that investigated bioavailability associated with As-containing coal ash or airborne As-containing particles; estimated bioavailability in these studies ranged from 11% to 50%. The implications and potential usefulness of iAs bioavailability associated with inhalation exposure to human health risk assessment remain unknown at this time. Main sources of dietary iAs intake in the U.S. include rice and other grains, vegetables, and fruits. Due to low concentrations of iAs, seafood is not a primary contributor to dietary iAs intake. Three general kinds of food studies were identified: studies of As bioaccessibility in composites, As bioavailability and bioaccessibility in specific foods, and As consumption and urinary excretion in human volunteers. One in vivo study was identified that examined As bioavailability in food. A variety of experimental in vitro gastro-intestinal protocols have been used, however, few studies have included As speciation before and after the in vitro extraction. Current data suggest that the bioaccessibility of iAs in rice is quite high, typically 70% or more indicating that iAs in rice is highly bioavailable. Adjusting for RBA may not have a meaningful impact on iAs exposure estimates for rice-based foods.



**Contents**



*Abbreviations:* As, arsenic; iAs, inorganic arsenic; BA, bioavailability; RBA, relative bioavailability; HHRA, human health risk assessment.

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http://dx.doi.org/10.1016/j.scitotenv.2015.05.141
0048-9697/

## 1. Introduction

Arsenic (As) is a commonly occurring trace element in the earth's crust typically occurring in +3 and +5 oxidation states in inorganic forms in air, water, and soil. Plant and animal foods may contain both inorganic and organic compounds. Human exposure is primarily by ingestion. Inhalation of airborne As and dermal contact with As in water or soil may also occur. While exposure doses (intake) may be estimated by measurement of total or speciated As concentrations in the immediate matrix of interest (e.g., air, water, food, soil), the fraction of an As dose that is taken up and reaches the systemic circulation or its bioavailable fraction represents a more exact measure of internal exposure, potential toxicity and possible subsequent health risk.

Absolute oral bioavailability (ABA) is defined as the ratio of the amount of arsenic absorbed as seen in the systemic circulation to the amount ingested (U.S. EPA, 2012a). Relative bioavailability (RBA) is defined as the ratio of the absolute bioavailability of As in the matrix of interest to the absolute bioavailability of As in an appropriate reference material. For As, the appropriate reference point is often the oral toxicity values in EPA's Integrated Risk Information System (IRIS) which are based upon exposure to As in water (U.S. EPA, 2012b). Thus the default assumption for assessing risk from As in exposure matrices other than water is that the bioavailability in that matrix is the same as the bioavailability of As dissolved in water. This assumption will result in an overestimation of the true risk if the bioavailability of As in the exposure matrix of interest is less than that of As in water.

As oral bioavailability has been studied in various in vivo models, including primates, juvenile swine, and the mouse (U.S. EPA, 2012a; Ng et al., 2013). Soluble As compounds are readily absorbed from the gastro-intestinal tract of laboratory animals. Determination of As concentration in blood and organs, or the amount excreted in urine or feces following exposure is used to estimate the fraction of the dose reaching the systemic circulation.

In vitro systems designed to simulate key processes in the milieu of the human gastro-intestinal tract are also used to estimate As RBA (Rodriguez et al., 1999; Ruby et al., 1996, 1999). The portion of a compound soluble in an in vitro system provides a measure of bioaccessibility, which is assumed to represent the maximum fraction of a dose available for intestinal absorption. Multiple versions of the in vitro test system have been developed; gastric and intestinal phases are included in most protocols with varying compartment constituents among methods (Ng et al., 2013). Digestive enzymes (e.g., pepsin) mucins, proteins, lipids, and selected intestinal microbes may be included in the gastric phase, whereas bile and pancreatin are usual constituents of the gastric phase (Charman et al., 1997; Dean and Ma, 2007; Molly et al., 1993; Oomen et al., 2003, 2004; Ruby et al., 2002; Tang et al., 2006). Incorporation of a microbial component has not been found to affect As bioaccessibility estimates consistently compared with methods that do not include microbial activity (Laird et al., 2009, 2013). The human gastro-intestinal system may attain varying pH levels depending on fasting conditions or the presence of food. In vitro methods have been designed to simulate the pH of the fasting state in a young child. For many metals, these conditions have been ascertained as likely to result in higher bioaccessibility values and thus are considered a more conservative approach (Pelfrene et al., 2011; Ruby et al., 1996, 1999; Wragg et al., 2011). The gastric phase is generally adjusted to a pH of around 1–2 and the intestinal phase near neutral at 6.5–7.5 although pH values utilized may vary slightly outside of those parameters (Ruby et al., 1996; Oomen et al., 2002).

Methods and the majority of available ABA and RBA As data have been developed to assess As in soil. Development of this large database stems from intensive work during the last decade or more on elucidating the RBA of As in soil (ENVIRON, 2011; U.S. EPA, 2012a; Ng et al., 2013). Estimation of an As RBA in soil is critical in risk assessment procedures used to establish clean-up levels at U.S. EPA Superfund sites as well as at other locations where potential soil ingestion is of health risk concern (Schoof, 2004). Based on extensive compilation and review of data on ABA of As in soil, U.S. EPA recommended a default RBA value of 60% for As in soil (U.S. EPA, 2012a). However, it is recommended that, when available, site specific data for an As RBA in soil be generated and utilized since results are highly variable and affected by the choice of method used to measure RBA values, the compound characteristics of As in the soil and its residence time at that site and the properties (e.g., iron content) and pH of the soil matrix itself, among others.

The purpose of this review is to critically evaluate the current state of knowledge on bioavailability of As in potential human exposure media other than soil and water, such as the diet, and potential environmental sources such as coal ash and to consider the possible impacts of this knowledge on human health risk assessment (HHRA). As the current drinking water U.S. EPA Maximum Contaminant Level (MCL) of 10 ppb in municipal drinking water has been achieved in most of the U.S. along with successful efforts to reduce drinking water As levels in many parts of the world, the contribution of As from food and other exposure sources has become increasingly evident. Overall, dietary As accounts for approximately 70% of iAs intake with a range of 1–20 μg/day. Numerous studies assessing U.S. population dietary and multi-pathway As exposures have demonstrated that diet is the primary contributor to total and inorganic As exposure when drinking water As levels are not elevated (Meacher et al., 2002; Tsuji et al., 2007; Georgopoulos et al., 2008; Xue et al., 2010; Kurzius-Spencer et al., 2014). In these studies potential variation in RBA of As among foods was not considered contributing to uncertainty in health risk model outcomes. This paper seeks to extend observations on As RBA in soil to include other exposure media including coal fly ash and food, and provides an assessment of best practices and the relevance of RBA in those media for HHRA.

## 2. Materials and methods

A comprehensive literature search was performed originally spanning the last five years utilizing the U.S. National Library of Medicine's