Available online at www.sciencedirect.com
ScienceDirect
Science of the Total Environment 371 (2006) 89–98
Science of the Total Environment
An International Journal for Scientific Research into the Environment and its Relationship with Humankind
www.elsevier.com/locate/scitotenv

Bioaccessible arsenic in the home environment in southwest England

J.S. Rieuwerts *, P. Searle, R. Buck

School of Earth, Ocean and Environmental Sciences, University of Plymouth, Portland Square, North Hill, Plymouth, PL4 8AA, United Kingdom

Received 7 July 2006; received in revised form 26 August 2006; accepted 29 August 2006
Available online 4 October 2006

Abstract

Samples of household dust and garden soil were collected from twenty households in the vicinity of an ex-mining site in southwest England and from nine households in a control village. All samples were analysed by ICP-MS for pseudo-total arsenic (As) concentrations and the results show clearly elevated levels, with maximum As concentrations of 486 μg g^{-1} in housedusts and 471 μg g^{-1} in garden soils (and mean concentrations of 149 μg g^{-1} and 262 μg g^{-1}, respectively). Arsenic concentrations in all samples from the mining area exceeded the UK Soil Guideline Value (SGV) of 20 μg g^{-1}. No significant correlation was observed between garden soil and housedust As concentrations. Bioaccessible As concentrations were determined in a small subset of samples using the Physiologically Based Extraction Test (PBET). For the stomach phase of the PBET, bioaccessibility percentages of 10–20% were generally recorded. Higher percentages (generally 30–45%) were recorded in the intestine phases with a maximum value (for one of the housedusts) of 59%. Data from the mining area were used, together with default values for soil ingestion rates and infant body weights from the Contaminated Land Exposure Assessment (CLEA) model, to derive estimates of As intake for infants and small children (0–6 years old). Dose estimates of up to 3.53 μg kg^{-1} bw day^{-1} for housedusts and 2.43 μg kg^{-1} bw day^{-1} for garden soils were calculated, compared to the index dose used for the derivation of the SGV of 0.3 μg kg^{-1} bw day^{-1} (based on health risk assessments). The index dose was exceeded by 75% (18 out of 24) of the estimated As doses that were calculated for children aged 0–6 years, a group which is particularly at risk from exposure via soil and dust ingestion. The results of the present study support the concerns expressed by previous authors about the significant As contamination in southwest England and the potential implications for human health.

Keywords: Bioaccessibility; Southwest England; Arsenic; Housedust; Soil; PBET

1. Introduction

The toxic properties of As have been known for many centuries. It is generally accepted that exposure to elevated levels of inorganic forms of As is associated with increased risk of cancer of the skin and other organs (World Health Organisation, 2000). Human exposure to elevated levels of As in the environment may result from: (i) ingestion of dust, soil, foodstuffs and water (ii); dermal contact and; (iii) inhalation of dust. Young children are particularly vulnerable to exposure to As in soil and dust for physiological and behavioural reasons. Vigilance is therefore required in areas of environmental As contamination to ensure that young children and other vulnerable sub-groups of the population are not subject to intakes which exceed acceptable levels.

In the UK, a Soil Guideline Value (SGV) for As has been derived (DEFRA and Environment Agency, 2002a) from the Contaminated Land Exposure Assessment

* Corresponding author.
E-mail address: jrieuwerts@plymouth.ac.uk (J.S. Rieuwerts).

0048-9697/$ - see front matter © 2006 Elsevier B.V. All rights reserved.
doi:10.1016/j.scitotenv.2006.08.039

(CLEA) model, using accepted risk assessment criteria and index doses. The SGV is set to be protective of young children who are at particular risk of direct exposure to As in soil and dust via ingestion. The SGV of 20 mg As per kg of soil is based on an oral index dose (health criteria value) of 0.3 μg kg^{-1} bw day^{-1}. The oral index dose is, in turn, based on studies relating the incidence of skin lesions to As intake from drinking water. Index doses (rather than 'tolerable daily intakes') are allocated to substances, like As, for which a threshold of adverse health effects cannot be presumed (i.e. As carries some risk at any level of exposure). DEFRA and EA (2002a) state that exceedance of the SGV (and, by implication, the index dose) "can indicate a potentially significant risk to human health". They add, however, that "this does not necessarily imply that there is an actual risk to health."

For many centuries, ores of tin, copper, arsenic and other valuable minerals were mined and processed in Cornwall and West Devon. These historic industries left extensive areas of mine-related remains in the region, including spoil heaps with elevated levels of As. Natural mineralization and the dispersal of ore materials by mining activities have resulted in large scale contamination of the region by As and other potentially toxic elements. For example, Abrahams and Thornton (1987) estimate that 722 km^2 of the land area of southwest England are contaminated by As. Elevated concentrations of As have been reported in the region in soils, stream sediments, freshwaters, homes (housedusts and garden soils) and in human biomarkers such as hair and urine (Colbourne et al., 1975; Xu and Thornton, 1985; Johnson and Farmer, 1989; Li and Thornton, 1993; Kavanagh et al., 1998; Peach and Lane, 1998; Farago and Kavanagh, 1999; Hamilton, 2000).

In the UK, As concentrations in foodstuffs and public water supplies generally fall within safety guidelines (e.g. Xu and Thornton, 1985). Arsenic ingestion via these sources is unlikely to be important in southwest England, although exposure to unregulated sources such as home-grown food and private water supplies may be higher than desirable in some cases. For example, Thornton and Farago (1997) state that there are up to 30 000 private wells in Cornwall for which little data is available. The dermal pathway may be of significance for young children, who are more likely than adults to come into bodily contact with spoil heaps and contaminated soils during normal play activities. However, in southwest England, direct ingestion of soil and dust is likely to be the main exposure source of As, particularly for young children (see next paragraph). Inhalation is thought to be a negligible exposure pathway for As in contaminated areas, compared with intake from soil and dust ingestion (DEFRA and EA, 2002b). It is worth noting that much of the particulate matter inhaled by humans is subsequently ingested (Wagner, 1980).

Ingestion of dust and soil is of particular importance, as an exposure route, for young children, a sub-group of the population which is known to ingest relatively large amounts of soil and dust. This is partly because of their tendency to play on the floor, thereby increasing the likelihood of coming into contact with soil and dust, and partly because of their frequent hand (or object) to mouth behaviour. For example, in a large scale survey in the United States, the average hand-to-mouth contact rate of children was observed as 9.5 times/h (USEPA, 2006). Studies of soil/dust ingestion in children, using soil and dust tracer elements, have yielded estimates of 39 mg day^{-1} (median value), 83 mg day^{-1} (mean value), 273 mg day^{-1} (90th percentile) and 515 mg day^{-1} (maximum value) (Calabrese and Stanek, 1993). Some individuals, including young children, can also indulge in geophagy, the deliberate ingestion of relatively large masses of soil.

In southwest England, the risks of exposure to As are clear but a relatively small amount of data has been published on levels of As in the home environment in the region. One objective of the present study was to update and add to this limited database by collecting and analysing garden soils and household dusts from an ex-mining area in southwest England. The few published studies that have reported concentrations of As in house-dusts and garden soils in southwest England, (summarised in Tables 1 and 2, together with unpublished

Table 1
Literature values of As in household dust in southwest England

Location	*n*	Mean conc. (μg g^{-1})	Range (μg g^{-1})	Reference
Camborne/ Hayle, Cornwall	74	81	9–330	Johnson (1983)
Mining villages, Cornwall	15	68	14–147	Elghali (1994) (unpublished data)
Devon Great Consols, Devon	13	1167	24–3740	Farago and Kavanagh (1999)
Gunnislake, Cornwall	9	217	33–1160	Farago and Kavanagh (1999)
Mining villages, Cornwall	[a]	58[b]	1–330	Culbard and Johnson (1984)
West Cornwall	[a]	[a]	460 (max.)	Harding (1993) (unpublished data)
Coombe Martin, Devon[c]	76	14	2–66	Johnson (1983)

[a] unknown.
[b] median — all other mean concentrations are arithmetic means.
[c] used by Johnson as a control area.