

# **Arsenic—Trials, Tribulations and Triumph in Site Remediation**

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## **ABSTRACT**

For better or worse, arsenic possesses a notoriety when it comes to setting acceptable limits for soil remediation. It is unquestionably a human carcinogen and the subject of considerable continuing research regarding its mode of action. There is on-going debate as to the applicability of the cancer slope factor (determined by the US EPA, and independently corroborated by the US National Academy of Sciences, from studies of Taiwanese communities exposed to high drinking-water levels of arsenic) in extrapolating to low level exposures.

In many parts of the world, particularly in jurisdictions that have not set their own soil clean-up guidelines, the approach adopted by the US EPA is taken as the “default”. However, uncritical application of this approach can lead to difficulties, for regulatory authorities and the entities whom they regulate, in arriving at practicable solutions for many “real-life” remediation projects. The soil clean-up target level based on the cancer slope factor can lead to remediation targets that are lower than natural levels in the surrounding soils.

In this paper, we examine general approaches taken by Australia, Canada, UK (in addition to the US EPA) in handling soil-remediation guideline-setting for arsenic. We look at the cancer slope factor question, and discuss the role of bioavailability of arsenic from soil matrices and of house dusts on the human exposure and realistic quantification of exposure through such pathways as house dust and remediated soil in a suburban residential development.

We conclude by describing a successful outcome of the approach in the cost effective remediation of a substantial industrial site located near Cape Town, South Africa.

## **INTRODUCTION**

In *Risk and Reason: Safety, Law and the Environment*,<sup>1</sup> Sunstein uses cost-benefit analysis as a tool to examine society’s perception of environmental risks and how government may (or does) regulate those risks. That Sunstein devotes a whole chapter of his book to the ramifications of arsenic risk assessment is testament not only to arsenic’s high “political” profile, but also to the considerable data available on arsenic’s carcinogenic dose-response and on cost-benefits of arsenic regulation. In this chapter,

Sunstein discusses the considerable uncertainties associated with cancer dose-response relationships derived from Taiwanese communities exposed to high levels of arsenic in their drinking water—of all the community data sets that are available, the U.S. National Academy of Sciences considers the Taiwanese data to be the most appropriate for developing dose-response curves.<sup>2</sup> Sunstein also observes that “very few people are expert on the risks posed by exposure to low levels of arsenic.” We do not count ourselves among these few but, along with very many others, we do have an interest in seeing soil clean-up targets for arsenic that are, at the same time, feasible economically and fully protective of a community who will reside on the remediated land.

In this paper, we first look at clean up of contaminated soil in brownfield sites in general terms, with arsenic as the contaminant. We follow with a description of how site-specific risk assessment aided the cost effective clean up of a decommissioned industrial site, AECI’s Agrochemicals site at Somerset West, southeast of Cape Town, South Africa.

## SOIL CLEAN-UP

Many jurisdictions give guidance to those engaging in brownfield redevelopment by providing soil screening levels (SSLs) for pollutants. These levels represent soil-pollution concentrations that the jurisdiction considers pose no unacceptable risk to exposed populations. The SSLs take into account all possible routes of exposure and are, by their nature, set conservatively. Jurisdictions that do not set their own SSLs commonly use, either formally (i.e., stated in regulations) or informally, SSLs developed by the U.S. Environmental Protection Agency.

In the following table, we list SSLs that have been published in the last few years. We do this because we wanted to be certain that they represent values developed at a time when full recognition of the carcinogenic properties of arsenic would have been made (explicitly or implicitly) by the setting jurisdictions:

TABLE 1 *Examples of Recent Soil Screening Levels (SSLs) for Arsenic*

Country	Year Published	mg/kg (dry weight)	
		Residential	Industrial
U.S.A. <sup>3</sup>	2006	0.43	1.9
Canada <sup>4</sup>	2006	12	12
United Kingdom <sup>5</sup>	2002	20	500
Australia <sup>6</sup>	2001	100	500

As may be seen, there is quite a range in the values for arsenic SSLs developed by the various jurisdictions. This variation is partly a reflection of the individual jurisdictions’ differing approaches to handling arsenic carcinogenicity in their guideline-setting processes, as well as different weightings given to apportionment of various exposure routes. But, in the final analysis, it must be recognized that each jurisdiction considers that the SSLs it sets are fully protective of the citizens it represents.

The United States SSLs are based on a cancer slope factor for oral exposure obtained from analysis of skin-cancer morbidity data from communities in southwestern Taiwan exposed to high levels of inorganic arsenic in their drinking water—the US EPA, in developing the cancer slope-factor for ingestion used in the SSL, felt that data (from the same communities) were inadequate, at the time (1998), to develop dose responses for internal cancers.<sup>7</sup> The United States SSLs reflect a *de minimis* lifetime cancer risk of  $10^{-6}$ . (If a jurisdiction using the US EPA’s approach to deriving SSLs decided, as public policy, to use a different *de minimis* cancer risk, the resulting SSLs would be modified correspondingly—e.g., if a cancer risk of  $10^{-4}$  were chosen, the SSLs would be 43 mg/kg and 190 mg/kg for residential and industrial uses, respectively.)

Canada is more “flexible” about the choice of *de minimis* lifetime cancer risk in setting soil clean-up levels for substances considered to be carcinogenic (or probably carcinogenic) to humans (i.e., non-threshold toxicants). The Canadian Council of Ministers of the Environment (CCME),<sup>8</sup> while agreeing “in principle with the philosophy ... that human exposure to non-threshold toxicants should be reduced to the lowest levels deemed reasonably feasible”, adopt the position that “contaminated site related risks arising from human exposure to non-threshold agents be ... remediated to levels within the range of  $10^{-4}$  to  $10^{-7}$ .” It is, however, unclear to us whether CCME’s 12 mg/kg SSLs for all land uses, presented in Table 1, are derived from lifetime cancer risk estimates, from an assessment of “the lowest levels deemed reasonably feasible” (which might explain why the SSL is the same for all land uses), or whether NOEL and LOEL arguments (implying that arsenic carcinogenicity is a threshold phenomenon) were used to derive the levels.

The United Kingdom takes a somewhat different approach to deriving SSLs.<sup>5,9</sup> This jurisdiction first establishes “Index Doses” (for both oral and inhalation exposure) which “represent doses that pose minimal risk levels from possible exposure to contaminants in soil”, with a rider “that exposure needs to be reduced to as low a level as reasonably practicable”.<sup>\*</sup> The SSLs (called “Soil Guidance Values” in the UK) are derived from the Index Doses, taking into account possible exposure to soil contamination for the different land uses. For residential uses, the Soil Guidance Value is set to be protective of young children because, in general, they are more likely to have higher exposure to soil contaminants. For industrial land use, an adult is assumed to be the critical receptor, with exposure considered over the working lifetime. In the case of arsenic, only oral exposure is considered as it is felt that inhalation and dermal exposure contributes “much less than 1%” to overall exposure.

Australia<sup>11</sup> develops its arsenic SSLs—called Health-based Soil Investigation Levels (HILs) in that country—from the Provisional Tolerable Weekly Intake (PTWI) of

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\* The Index Doses for arsenic are  $0.3 \mu\text{g kg}^{-1} \text{ bw day}^{-1}$  for oral exposure and  $0.002 \mu\text{g kg}^{-1} \text{ bw day}^{-1}$  for inhalation exposure. The oral Index Dose is derived from the WHO recommended provisional drinking-water guideline value of  $10 \mu\text{g/L}$ ,<sup>10</sup> which is based on drinking-water studies in which the incidence of skin effects was observed to be related to arsenic intake. By assuming that 2 L/day of water is consumed by a 70 kg adult, this would correspond to approximately  $0.3 \mu\text{g kg}^{-1} \text{ bw day}^{-1}$ .<sup>5</sup>

[Although it was not explicitly stated, we assume that in the case of arsenic the UK authorities consider the WHO drinking water guideline to be a measure of a dose that poses a minimal risk. There is no consideration given to “background sources” of arsenic (e.g., arsenic from food) in the development of the UK Soil Guidance Value.]

0.015 mg/kg-wk for arsenic that the WHO used to develop the provisional drinking-water guideline level in the 2<sup>nd</sup> Edition of its *Guidelines for Drinking Water Quality*.<sup>12</sup> (This PTWI, in turn, was taken from earlier deliberations, in 1989, of the Joint FAO/WHO Expert Committee on Food Additives.<sup>13</sup>) The HIL for residential areas is based on exposure of young children, who are assumed to ingest 100 mg soil per day (an amount equivalent to 40% of the PTWI). The HIL for industrial areas is, like the UK Soil Guidance Value, based on adult exposure and also uses the PTWI concept.

### **Cancer Slope-factors<sup>†</sup>**

Of the SSLs presented in Table 1, only those produced by the US EPA are explicitly derived from a quantified cancer dose response—incidence of skin cancer in communities in southwestern Taiwan exposed to high levels of inorganic arsenic in their drinking water. An essentially linear extrapolation of the dose-response data to low exposure levels was used (described by the US EPA as a “time- and dose-related formulation of the multistage model”). However, at the time the slope factor was developed (circa 1998), it was acknowledged that “the dose-response for arsenic at low doses would likely be truly nonlinear (i.e., with a decreasing slope as the dose decreased) ... [but that] at very low doses such a curve might be linear but with a very shallow slope, probably indistinguishable from a threshold”. The EPA also acknowledged that there were weaknesses and uncertainties, occasioned by the status and situation of the group in Taiwan, in the derived dose response that limit the usefulness in risk estimation. These problems included poor nutritional status of the exposed populations, their genetic susceptibility, and exposure to inorganic arsenic from non-water sources; dietary inorganic arsenic was not considered, nor was the potential confounding of contaminants other than arsenic in their drinking water.<sup>‡</sup>

Sunstein<sup>1</sup> examines the use of (the potentially conservative) linear extrapolation of observed dose-response to arsenic (and to carcinogenic pollutants in general) at high exposures to low exposure scenarios. He observes that the choice of extrapolation model—supralinear, linear, sublinear, threshold below which exposure produces no adverse effects (essentially an extreme case of the sublinear model), or “U-shaped” response (a situation where exposure at high doses produces adverse effects, but exposure at low doses can have a beneficial effect)—can have a profound effect (often orders of magnitude) on the low-dose risk estimation; an observation with which toxicologists and risk assessors would readily agree.

It seems to us, at times, that the debate concerning extrapolation of dose response data to low exposure levels, in the absence of a credible understanding of the mechanism(s) that

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<sup>†</sup> Unless otherwise stated, information concerning the US EPA slope factors in this section was taken from the Integrated Risk Information System (IRIS).<sup>7</sup>

<sup>‡</sup> Since the US EPA first proposed reducing the Maximum Contaminant Level (MCL) for arsenic in drinking water from 50µg/L to 10 µg/L, there has been an extensive literature criticizing (both positively and negatively) the Taiwanese data, the dose-response relationships obtained, and the risk estimations of the cancer endpoints (principally skin, bladder and lung cancers). A good selection of such publications are described and analyzed in the National Academy of Sciences re-evaluation of their original investigation of the US EPA’s risk assessment for the establishment of the arsenic drinking-water standard,<sup>2</sup> the US EPA’s Final Rule for arsenic in drinking water,<sup>14</sup> and in a more recent US EPA issue paper on the arsenic slope factor.<sup>15</sup>

may cause the response, can seem like an exercise akin to the interminable medieval theological debates about how many of God's angels are able to dance on the head of a pin. But we are not saying that using cancer slope factors to develop SSLs is imprudent—indeed, quite the contrary, given that the substance is an acknowledged human carcinogen and that there are human epidemiological data available, it is probably the most “scientific” way to initially proceed. However, considering the uncertainties inherent in slope-factor extrapolations and in the risk conclusions drawn from them, it may be worthwhile to consider other means to set soil-clean-up levels. And perhaps this was the thinking behind the other jurisdictions' use of such concepts as “tolerable” daily intake, and NOEL and LOEL arguments (specifically regarding dermal effects).

## **Mitigating Considerations**

As we have said, SSLs are set conservatively. They consider all possible routes of exposure and, usually, assume 100% absorption. When one turns to developing soil-cleanup-target levels (SCTLs) for a specific site, there are a number of mitigating factors that have a bearing on the level chosen. We deal with two factors as examples—one of them, the question of bioavailability from soil matrices is of general application to all arsenic soil remediations; the other, the contribution of soil to levels of arsenic in house dust, although specific for the site we describe in the next section, may also be considered to have more general applicability.

### ***Bioavailability of Arsenic from Soil***

For arsenic, the observed effects in the Taiwanese communities came from exposure to arsenic in drinking water. Virtually all the soluble arsenic in water (both As<sup>III</sup> and As<sup>V</sup>) can be absorbed from the gut. The same cannot be said for arsenic in a soil matrix.

Discrete arsenic mineral phases commonly include such less soluble forms as sulfide minerals, complex oxides, and arsenic present in iron, manganese, and phosphate mineral species; and, typically, arsenic in soil is only one-half to one-tenth as bioavailable as arsenic in solution.<sup>16</sup> A recent study by Roberts *et al.*<sup>17</sup> measured oral bioavailability of arsenic in soils using five male *Cebus apella* monkeys. The soils were obtained from five waste sites in Florida and the relative bioavailability ranged from 10.7±4.9% to 24.7±3.2%. So, clearly, this lessened availability can have a considerable bearing on SCTLs.

### ***House Dust in Exposure Calculations***

Site-specific development features have a major influence on exposure scenarios. For example, approximately 80 per cent of house dust is normally considered to be derived from local soil and would therefore represent outdoor soil contamination.<sup>18</sup> A child, representing the most sensitive subpopulation, would therefore be exposed whenever on-site, and not only just when outdoors. That is why exposure to site contaminants is normally assessed for 350 days per year. (It is assumed that 15 days per year would be spent at a non-contaminated area away from the residential location.) The nature of the house dust (and its attendant arsenic levels) would very much depend on the remediation program adopted and would depend on such things as the nature of replacement topsoil, paved surface areas, and so on.

## Postscript

The broad variations in approaches, interpretation of cancer risks, and development of SSLs make it difficult to choose one approach over another. In the site-specific example presented in the next section, we rely upon “common sense” arguments, rather than on developing absolute (and rather abstract) estimates of risks.

## SOMERSET WEST

### Site History and Proposed Redevelopment

The AECI Somerset West site is located on False Bay, some 40 minutes drive southeast of downtown Cape Town. The site had been used for chemical manufacturing operations for the past 100 years. Operations included the manufacture of explosives, fertilizers, acids, agrochemicals, and a host of other products.

FIGURE 1 *The Somerset West Site*



**The site in production years**



**The site as it is now**

The area of interest for this paper is the Agrochemicals area, which covers approximately 20 hectares (50 acres). Operations began in 1920 with the manufacture of sulfur-based fungicides followed, later on, by the manufacture and formulation of both inorganic and organic pesticides. Operations ceased in 1999 and the plant was decommissioned in 2001.

Key to the arsenic discussion in this paper was the manufacture, at the Agrochemicals production area, of such products as arsenic acid, lead arsenate, copper arsenate and calcium arsenate. The effluent streams from the manufacturing processes had been directed into what were euphemistically known as *soak away areas*. As Eh and pH varied considerably, these products were mobilized to a greater or lesser extent. The mobilization, however, took place through a matrix that is highly calcareous and also contained elevated levels of iron. The resultant chemical complexing and mobilization of arsenic, lead and copper were therefore difficult issues to deal with. What made the situation even more problematic was the presence of a wide range of pesticides.

FIGURE 2 *A Euphemistically-termed Soak-away Area*



AECI is now in the process of releasing this landholding for re-development. A conceptual development framework (CDF) for the site was developed with a mixed land end use of residential, light commercial and public open space. The Agrochemicals Area, where arsenic levels were elevated in soil and were present in ground water, is an area that will incorporate areas used as public open spaces, low density residential development and, potentially, tourism facilities.

## **Site Remediation Approach and Resolution**

### *Site Characterization*

The Agrochemicals site was part of an extensive evaluation of the whole AECI Somerset West property. Site characterization followed a classical, and reiterative, approach: historical review of the site's manufacturing activities to determine the field work to be undertaken; systematic collection and analysis of soil and groundwater samples; hydrogeologic modeling; and then, based on the results, additional sampling, analysis and modeling to better define and understand the features.

The main feature of arsenic contamination in the Agrochemicals site, as would be expected from the “soak away” practices, were distinct localized areas with very high arsenic levels (up to the tens of thousands of ppm). These “hot spots” did not really merit much additional consideration—they would obviously have to be removed. For the remaining areas—the bulk of the site—a human-health hazard analysis was undertaken.

### *Determination of Cleanup Goals*

In the assessment of human health risks associated with environmental contaminants and the development of goals for corrective action, all relevant pathways and routes of exposure have to be considered—for example:

- ingestion from drinking water derived from ground water underlying the soils;
- ingestion from vegetables irrigated by ground water underlying the soils;
- inhalation of dust and vapors;
- ingestion of soil; and
- dermal contact with soil.

This *multi-route approach* is based on work published by the Center for Environmental and Human Toxicology in the University of Florida in the USA.<sup>19</sup>

The groundwater underlying the Agrochemicals Area is in a secondary aquifer which has high salinity (the site is comparatively low lying and within 500 m of the ocean, and the underlying rock is porous and fractured). This groundwater is *totally unsuitable* both as a drinking-water source and for irrigation. Accordingly no consideration is given to the potential for leaching of arsenic from the remediated soil into groundwater as a human exposure pathway. Because the inorganic arsenic is not volatile, and because the remediated soil will be covered with uncontaminated topsoil (see below), the probability of notable levels of arsenic-contaminated windblown dust generated in the proposed developed residential area at AECI Somerset West will also be low. Thus the potential for exposure to soil contaminants from inhalation is of minimal significance. The assessment, therefore, focused on the potential for exposure to arsenic through ingestion of soil and, to a lesser extent, dermal contact with soil.

The equation for the calculation of SCTLs includes the standard parameters for exposure quantification, adjusted for site-specific conditions, at a chosen target risk level (normally one in a million for cancer). We will show in the following paragraphs how careful assessment of site-specific conditions may have major effects on the calculated SCTLs.

As has already been mentioned, the US EPA cancer slope factor for arsenic of 1.5 per (mg/kg-day) has been the subject of much debate. Uncritical application of it often leads to soil cleanup levels that are below the natural uncontaminated background of the area being considered. The New Zealand government has suggested that a cancer slope factor ten times lower would be more realistic and is, any way, in accordance with interpretations of epidemiological data by many scientists.<sup>18</sup> A cancer slope factor of 0.15 per (mg/kg-day) was therefore applied in this project.

We have already discussed the bioavailability of arsenic from soil matrices in the section above. For the purposes of this project, bioavailability of 15 per cent was assumed for oral intake from soil—essentially the mid-point of the range of bioavailabilities experimentally determined in monkeys by Roberts *et al.*<sup>17</sup>

With regard to the potential house-dust exposure route, the scenario at the proposed residential development in Somerset West would be much different from a scenario where the entire area (all outdoor soil) is remediated to the SCTL. During development of the Somerset West site, establishment of open parks and garden areas would require extensive landscaping. Because of the natural poor soil quality for gardening purposes, large quantities of clean topsoil and compost will have to be imported for the development. Together with building of roads and paving of many areas, as well as building of residential and commercial facilities, a large proportion of the current surface and shallow soil is likely to be either covered or mixed with clean imported soil and compost. Where outdoor surface soil is considered to be largely uncontaminated (contaminants at background levels in soil), as would be the case in the Somerset West development, house dust would primarily derive from this soil and exposure to house dust should not affect the calculation of SCTLs. Field studies at the Agrochemicals Production Areas identified specific locations where unacceptable levels of contamination were indicated for removal and disposal. Exposure of residents (including children) to residual contaminants, for which SCTLs are determined, would be infrequent or intermittent at the locations where “hot spots” were removed. Although house dust

would not form a significant component in the derivation of SCTLs across the entire study area under these conditions, we considered it precautionary to allow at least a component of 10 per cent of house dust to be associated with residual contaminants at and below the SCTLs. This would also allow for intake of substances that may be present in soil at background levels. This was taken into account in the derivation of SCTLs. For the current assessment, exposure to house dust was calculated for 300 days per year, allowing 50 days per year for outdoors exposure to soil in areas where contaminant sources were removed and soil was remediated to the SCTLs.

Taking all these factors into account, a soil cleanup target level for arsenic in the range 1 900 mg/kg to 2 900 mg/kg was derived for the site-specific scenario.<sup>§</sup> The arsenic levels in the bulk of the site area, other than the “hot spots” (which were dug up and removed), are 10 to 20 times lower than these SCTLs.

### ***Remediation Plan and Approval***

Although a number of technologies for the removal of these compounds were considered and tried, the bulk of the materials (~350 000 tons) needed to be dug up and transported to, and disposed of at, a registered hazardous waste site roughly 55 km (34 miles) from Somerset West. The cost was approximately R90 m (ca. \$12m USD). The disposal was into a “monocell” developed, and approved by the authorities, specifically for the waste from the Somerset West site. This proved to be the most cost-effective method of reducing levels of contamination to the site-specific soil cleanup target levels (SCTLs).

FIGURE 3 *Remediation and Disposal*



**“Stockpile” of contaminated soil**



**“Monocell” at hazardous waste site**

In February of last year, the Department of Water Affairs gave its approval to the proposed remediation plan and removal of contaminated soil is now essentially complete.

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<sup>§</sup> In deriving these SCTLs, we used a lifetime cancer risk factor of  $1 \times 10^{-5}$ —a risk value used in New Zealand’s derivation of SSLs.<sup>18</sup> A risk factor of  $1 \times 10^{-6}$  would, of course, yield SCTLs of 190 mg/kg to 290 mg/kg—values that still would have been greater than (or similar to) the arsenic levels in the bulk of the site area.

## **Economic Factors\*\***

The costs of the remediation of the approximately 20 ha (50 acres) area following the plan described in the section above, has amounted to approximately R90m. If one had attempted, *a priori*, to clean up the site to a reasonably strict SSL for arsenic (say the Canadian value of 12 mg/kg), without conducting the site-specific health-risk analysis another approximately 400 000 tons of material at a cost of approximately R105m would have had to be disposed of. In addition, this amount would have had to be backfilled. As the site is “soil deficient”, the back fill material would have had to be imported at a cost of R40/m<sup>3</sup>, and compacted. This would have cost an additional R20m.

As a point of comparison, if one had chosen to only remediate to brownfield site clean up levels (at the same cost that was actually spent—i.e. the R90 m), the site would been judged only suitable for commercial/industrial usage. And this would have potentially realized some R400m-R500m less in real estate sales than one would be getting for the approved residential/parkland use.

In summary, some R500-R600 m in revenue would have been lost using a “broad-brush” non-site-specific approach. These are very conservative figures as this particular area of the site is positioned along a pristine and much sought after beach front area, some 1.5 km in length. Property prices are unpredictable, but the development has attracted a lot of foreign capital.

## **CONCLUSION**

If one had to uncritically apply the US EPA approach to the arsenic soil clean-up levels adopted for Somerset West, re-development of the site would not occur—clean-up to levels at (or, even, below) those of the uncontaminated surroundings, that would be demanded by such an approach, would be economically infeasible. Instead, as we hope the discussions in this paper show, a solution has been reached that is economically feasible, with little (and, realistically, non-measurable) increase in potential risk from the ideal. By approving the plan, government (which, after all in a democratic society, is ultimately answerable to the citizenry) obviously agrees.

The end result is, using Sunstein’s cost-benefit paradigm, a net benefit to society. Land, rendered unproductive for its original industrial usage, has the potential to again produce a benefit to the population by providing much needed housing and commercial space to meet the demands of an expanding Cape Town area economy.

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\*\* At the time of writing, \$1US is worth close to R7.5.

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