

INVESTIGATIONS NEAR DEVON GREAT CONSOLS MINE INTO AIRBORNE DUST POLLUTION

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An investigation was carried out into whether toxic elements at Devon Great Consols mine and mineral processing site are transferred into airborne dust particles. Deposited dust samples were collected across an area of approximately 15 hectares in the summer of 2011. Soil samples were also collected from within and near the former working areas. Dust sampling took place at locations up to 550 m from the former working areas. Samples were prepared for ICP-MS analysis by strong acid digestion. Crustal enrichment factors (EFs) were determined to compare concentrations in the samples with typical crustal values. EFs for arsenic and copper were very high and showed little significant difference between the soils and the dusts in the source area. EFs appear to reduce beyond the former mine workings. The soil appears to be a principal source for arsenic and copper in the dust particles. On the basis of comparison with findings elsewhere, it is possible that the UK target value for airborne arsenic could have been exceeded significantly. Given that the site is a current public open space, and that the bioavailability of these elements at this site has been recognised by others, there may be grounds for health risk concerns.

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INTRODUCTION

This paper describes an investigation into whether potentially-toxic elements at the Devon Great Consols mine and mineral processing site have been transferred into airborne dust particles which might have left the site. Devon Great Consols, an old mining site in South-West England, has had a plethora of scientific studies analysing the mine spoils resulting from its extensive extraction in the late 19th Century. Studies have shown very high levels of elements such as arsenic and copper present in the soil and water column (Hamilton, 2000), but no data have been published on the pathway of dust propagation at and around the site. This study therefore aims to use modern dust collection techniques, supplemented by quantitative elemental analysis, to determine if dust in the area can be considered a potential risk to local receptors.

There were three main objectives in the study:

- 1) To collect a set of dust samples in order to investigate levels of toxic elements present in dust at and around mine spoil sites, assessing the potential link between distance from the contaminated land and toxic element concentrations.
- 2) To collect soil samples in order to investigate whether elemental levels found in the dust samples can be directly attributed to the former mining activity and the adjacent soils.
- 3) To compare concentrations of toxic elements found in the dust samples to background levels, and national limit values, in order to highlight potential impacts on local receptors.

BACKGROUND

Devon Great Consols

Devon Great Consols [SX 431 735] is a highly mineralised ex-mining area of 68 hectares (Kavanagh *et al.*, 1997) which lies in the parish of Gulworthy, in the county of Devon, approximately 10 miles north of Plymouth. It is immediately west of Dartmoor National Park, and adjacent to the River Tamar, which forms the western boundary of the site. The River Tamar was crucial to the industry that took place, with boats used to transport excavated minerals (Otter, 1994).

The Devon Great Consols site is set within the Tamar Valley, an area underlain by Late Devonian Tavy Formation sediments (Leveridge and Hartley, 2006). The Dartmoor granite pluton was intruded into the surrounding sedimentary rocks in the Early Permian Period, as part of a group of intrusions produced from the Cornubian Batholith (e.g. Watson *et al.*, 1984). Hydrothermal processes related to and following the plutonism caused a wide range of minerals to be formed, with fluids depositing their metal ores within and around the edge of the pluton, in locations such as Devon Great Consols (Natural England, 2012).

The mining site that gave its name to Devon Great Consols began extraction in 1844 and quickly established itself as the world's richest copper and arsenic mine (Hamilton, 2000). The underground mine workings were 4 km in length along the main ore vein; up to 1,300 people were employed, and a railway link was established nearby to transport the ore for shipment and handling. The copper mine was slowly depleted and a shift to arsenic and tin production began in 1880. In the

20th Century output continued to decrease, until the site's final abandonment in 1925 (Devon County Council, 2012). Extensive restoration of the site took place more recently and it was opened for public access in 2009 as part of the Mining Heritage Project: that project aims to recount the area's history. The Tamar Trails were subsequently set up and comprise a collection of trails and footpaths which run through the mining area (Tamar Valley Area of Outstanding Natural Beauty, 2010). The location is a Site of Special Scientific Interest (SSSI) under the Wildlife and Countryside Act of 1991 since it provides excellent examples of primary mineralisation in the area, in particular of weather-resistant arsenopyrite, and crystalline carbonates such as siderite (Sites of Special Scientific Interest, 2012).

Although public access is now available, there are still many safety issues at the site including evidence of both arsenic and copper mineralisation surrounding the Wheal Anna Maria site, with both elements demonstrating high toxicity (Hamilton, 2000). Dangers also remain from exposed mine shafts (Devon County Council, 2012). Hazardous dusts created from the site were found to have been previously limited to heights of 1.2 m above ground level and were therefore determined not to pose significant problems to surrounding farmlands (Hamilton, 2000). In 1990, however, West Devon Borough Council (1990) approved the site for use as a rally driving circuit for 28 days a year and for various other off-road events each year. The council was of the opinion that this did not cause statutory nuisance, but dust levels were observed to have risen to over 100 m in height throughout the periods of use (Hamilton, 2000), and rallies were stopped.

Previous work

A review of contaminated land in West Devon was conducted by Hamilton (2000), with arsenic chosen as the prime focus of the investigation. Water, soil and stream sediment concentrations were evaluated at mine sites such as Devon Great Consols and found to be consistently high, with many areas of soil containing at least 1000 mg kg⁻¹ of arsenic. Potential levels of exposure therefore appear to be very high but there is no body of evidence that has yet indicated a link to impacts on human health. This might be due to a lack of appropriate testing, and it is considered that pathways, such as the potential exposure to dusts, need further evaluation.

Many studies of Devon Great Consols focus on the bioavailability of the high levels of arsenic in the soil (Kavanagh *et al.*, 1997; Palumbo-Roe and Klinck, 2007) and often on how it has affected local populations of earthworms (Button *et al.*, 2010, 2011; Langdon *et al.*, 2001; Pearce *et al.*, 2002). Earthworms are frequently used to evaluate soil contamination (Pearce *et al.*, 2002), and are known to play a major role in the breaking down of soil, thereby accumulating any contaminants present (Langdon *et al.*, 2001). Earthworms in the area were found to have acquired very high tolerances to both Cu and As toxicity in contrast to worms from uncontaminated sites, and it was therefore demonstrated that the soils at Devon Great Consols are genotoxic to non-native fauna (Button *et al.*, 2010; Langdon *et al.*, 2001).

Many toxic elements can be harmful even in small doses, whilst others can be essential for the survival and good health of both plants and animals. These two properties are not mutually exclusive however, and many metals can demonstrate a fine line between deficiency, homeostasis and toxicity (Nordberg *et al.*, 2002).

Source-pathway-receptor

Contaminated land risks can be assessed using the source-pathway-receptor model. Firstly, the source in which the elements are present is identified, and pathways are recognised that could transport the elements into sensitive receptors (Stewart, 2004). For dust particles this pathway might involve

oral bioaccessibility, with the receptor being either the human stomach or lungs. A study by Okorie *et al.* (2012) on the bioaccessibility of six potentially toxic elements (PTEs) from urban street dust found widely contrasting levels for each PTE. Using the average concentrations of street dust samples, it was found that for all cases except lead, the bioaccessible concentrations were below guideline values on unintentional consumption (estimated at 100 mg day⁻¹ by the US EPA (2008)), and therefore not harmful to human health. However, a study by Turner (2011) reviewed the oral bioaccessibility of trace elements in household dusts, and found that different attempts to mimic the intestine can have considerably different outcomes. Measurements of Pb bioaccessibility in the stomach ranged widely from 25 to 80%, demonstrating that the full effects of trace elements in dusts are still difficult to quantify.

Toxic elements

Arsenic and copper have been chosen as the focus of this study due to their toxicity and observed abundance in previous local studies. Nickel, vanadium, chromium, zinc and iron were also chosen for comparison because of their observed variability and concentrations in the data.

Arsenic: has an average crustal abundance of approximately 1.5-3 mg kg⁻¹ (Hindmarsh *et al.*, 2002). It is present in soils throughout the world at background levels of 1-40 mg kg⁻¹, with concentrations often determined by the parent rock (Button *et al.*, 2010). In the United Kingdom, half of all soils contain less than 25 mg kg⁻¹ of arsenic, with much higher concentrations present in parts of Cornwall, Devon and South Wales (Rawlins *et al.*, 2012). These areas can have considerably higher concentrations for a wide variety of reasons: anthropological sources of contamination can include industrial processes such as mining and wood preservation (Bhattacharya *et al.*, 2002; Button *et al.*, 2010). Arsenic is widely known to be carcinogenic (IARC, 1980). Schedule 3 of The Air Quality Standards Regulations 2010 (see <http://www.legislation.gov.uk/ukxi/2010/1001/contents/made>) has therefore set a target ratio for dust particulate matter, to be met by the end of 2012, of 6 ng m⁻³ of arsenic in PM₁₀ form (see below). Recommended garden soil levels are additionally set by the Environment Agency at a maximum of 20 mg kg⁻¹ (Ravenscroft, 2009).

The arsenic present in soils in Devon Great Consols mostly comprises arsenopyrite and the products of its weathering, and can reach concentrations of 34,000 mg kg⁻¹ (Langdon *et al.*, 2001). Another study found concentrations were dependant on proximity to spoil areas and ranged from 204 to 9,025 mg kg⁻¹ (Button *et al.*, 2010), and overall bioaccessibility levels for As were found to be approximately 15% of the total arsenic (Palumbo-Roe and Klinck, 2007).

Copper: is a potentially toxic metal that has an average soil concentration of the order of 50 mg kg⁻¹, and is a necessary component of enzymes used in human metabolic functions (New Hampshire Department of Environmental Science, 2005). It has a recommended daily allowance (RDA) of 0.9 mg, with safe levels over an extended period of time defined as <10 mg day⁻¹ by the US EPA (US EPA, 2011). Exposure to high levels of copper may be toxic, with long-term effects including kidney and liver damage (New Hampshire Department of Environmental Science, 2005). Its bioavailability is limited however, because it strongly adsorbs onto organic matter, so Cu poisoning is relatively rare (US EPA, 2011). Copper, largely present as chalcopyrite and weathered products, can be found in concentrations up to 12,000 mg kg⁻¹ at Devon Great Consols (Langdon *et al.*, 2001).

Dust definitions

In the atmosphere, particulate matter is the collective name to describe small liquid and solid particles that exist over short (seconds to minutes) to prolonged (days to weeks) periods of

time. Particles can vary widely in mass, size, geometry, concentration, physical properties and chemical composition and can be natural or result from human activities. Dust particles can essentially be categorised by size in accordance with National Air Quality Standards (NAQS), with PM₁₀ measured and reported as mass concentration over time and defined as “those particles passing an inlet of defined characteristics with a 50% sampling efficiency at 10 μm aerodynamic diameter” (The Quality of Urban Air Review Group, 1996). Size strongly influences the atmospheric lifetime of particles and deposition patterns in human lungs (Godish, 1997). The European Directive 1999/30/EC has set daily average limits of 50 $\mu\text{g m}^{-3}$ for PM₁₀ with a yearly average limit required to be $\leq 20 \mu\text{g m}^{-3}$.

METHODOLOGY

The methodology for this study focuses on the collection of deposited dust samples around the Devon Great Consols site. DustDiscs, a sticky-pad dust collection sampler provided by DustScan Ltd, were used to sample dust deposition (Figure 1). Soil samples from mine spoil areas of 50 g were taken for reference and consideration as potential source material.

Field sampling

On the basis of a site reconnaissance visit in July 2011, three soil sample sites were selected, all within close proximity to a relevant source of contamination. Soil sample 1 was located immediately next to a mineral processing tip. Soil sample 2 was situated adjacent to a spoil site with discoloured soil, approximately 100 m north of soil site 1. Soil sample 3 was located next to a former arsenic calciner.

A sample grid was constructed for the placement of the DustDiscs. Four DustDiscs were deployed in a rectangular array surrounding the soil sample sites (and therefore spoil sites), with a further six laid out downwind of the spoil sites, in varying distances to the east. The data collected thus facilitated assessment of dust propagation away from the source.

A second site visit took place in August 2011 and DustDisc locations were selected to ensure that each disc was placed in a location which fits the following criteria:

- 1) It must be at least partly hidden from general view (in order to avoid tampering and theft).
- 2) It must not be in an area sheltered from wind (in order to achieve maximum dust exposure).
- 3) It should be at an optimal height (in order to avoid contamination or interference from local wildlife or soil splash).

Each DustDisc placement was recorded using GPS coordinates (Figure 2).

The dust sampling locations were categorised by distance from former working areas of the mining site into three DustDisc groups (Figure 2). The distance from the closest working area was calculated for each DustDisc point. Group 1 included those closest to the former working areas (DustDiscs 1-4), Group 2 included those within 300 m (DustDiscs 5-7) and Group 3 included those up to 550 m away (DustDiscs 8-10).

Laboratory analyses

Soil samples were dried, disaggregated and homogenised by gentle crushing with an agate pestle and mortar. Dust samples were prepared for analysis by removing the acetate sealing sheet to expose the accumulated dust. Both were subjected to a hot acid (aqua-regia) digestion, made to a final volume of 25 ml at a final acid strength of 8%.

All samples were analysed on an Agilent 7500ce Inductively Coupled Plasma-Mass Spectrometer (ICP-MS) at the University



Figure 1. DustDisc (DustScan, 2013).

of Portsmouth, using an octopole collision cell in He mode. Calibrations were defined using dilutions of a commercially-available multi-element standard solution, to encompass the expected range of concentrations. Each analytical batch included suitable certified reference materials, blanks and replicates. Accuracy and precision are estimated to be better than *c.* 10% for most elements, except when approaching detection limit (generally in the ppb range, in the analysed solution).

RESULTS

Field sampling data

Table 1 shows concentrations for all elements as analysed by ICP-MS compared to UK soil averages (Rawlins *et al.*, 2012). Arsenic and copper concentrations are very high for soil sites 1 and 3, which were located next to the arsenic calciner and mineral processing tip respectively. Table 2 shows crustal enrichment factors (EFs) (Datson and Fowler, 2007) for selected elements from the soil samples. Soil EFs were determined to compare concentrations in the samples with typical crustal values, and for comparison with dust sample EFs. Aluminium is often chosen as a reference element because its high abundance is relatively uniform throughout the Earth's crust and soils (Alloway, 1994). Table 3 shows average EFs for each dust sampling point. It is apparent that arsenic is extremely high in dust sample sites 1-7 and that copper, zinc and chromium appear to be consistently enriched.

Comparisons of data in Table 4 and Figure 3 indicate that the EFs for soil and dust were not significantly different for As and Cu from soil and dust samples. This was confirmed with a suite of appropriate statistical tests. An independent t-test was used for arsenic ($p = 0.198$) and Welch's t-test was used for copper ($p = 0.47$). This is therefore consistent with the soil being the source of the dust, at least for these elements. Many other elements are enriched in the dust, although EFs for elements such as chromium can be highly variable between dust and soil, for unknown reasons (Figure 3).

Toxic element EFs by group

Figures 4 and 5 show a clear decrease in arsenic and copper levels in relation to DustDisc location. This indicates that arsenic and copper concentrations in dusts decrease with distance from mine spoil areas, which strongly suggests that the mine spoils and soils in the Group 1 area could be the source of the dust in both the source area and downwind.

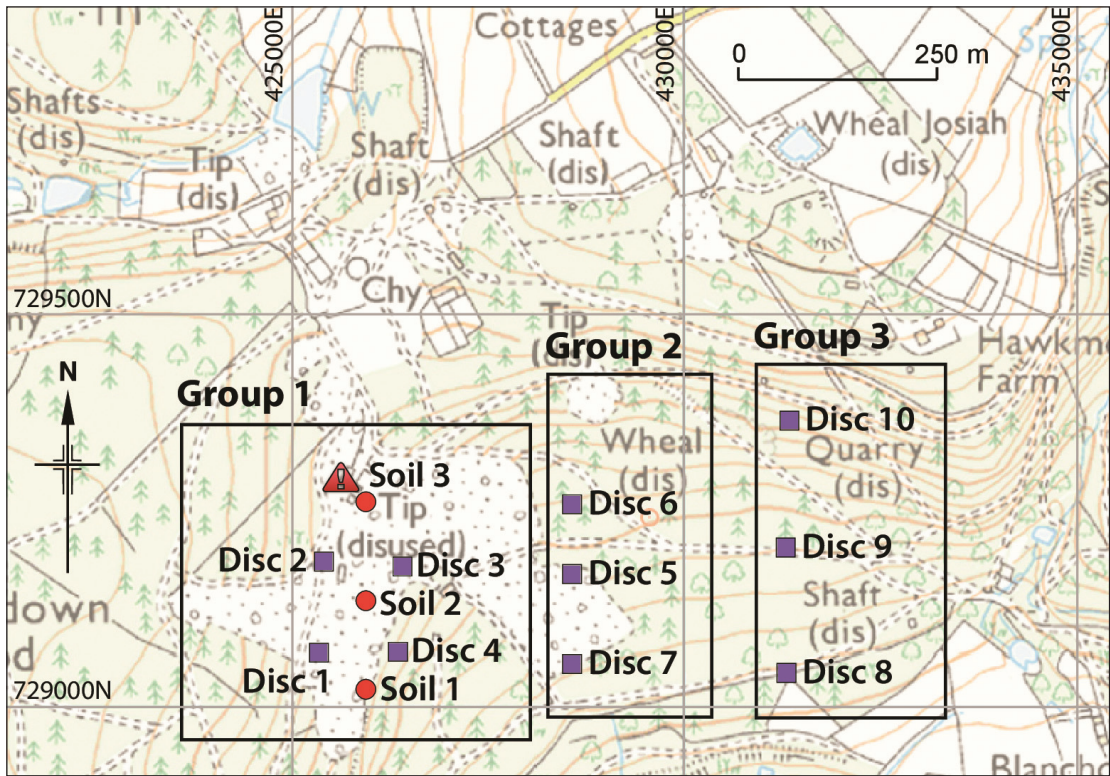


Figure 2. DustDisc locations. The red hazard symbol refers to the arsenic calciner (Ordnance Survey, 2012). Reproduced by permission of Ordnance Survey on behalf of HMSO. © Crown Copyright. All rights reserved.

Sample	Vanadium	Chromium	Iron	Nickel	Copper	Zinc	Arsenic
Soil 1	26	17	106,404	30	10,332	489	25,865
Soil 2	14	4.8	44,471	1.8	92	12	806
Soil 3	4.1	4.7	40,899	6.9	883	71	32,363
UK Average	79	68	29,000	23	24	91	20

Table 1. Element concentrations in soil samples compared to UK average (mg kg⁻¹).

Sample	Vanadium	Chromium	Iron	Nickel	Copper	Zinc	Arsenic
Soil 1	0.9	0.8	1.0	1.8	861	32	65,874
Soil 2	6.3	2.7	5.1	1.3	96	7.8	25,797
Soil 3	0.4	0.6	1.1	1.2	216	13	242,448
Average	2.5	1.4	2.4	1.5	391	18	111,373

Table 2. Crustal enrichment factors for soil with reference to aluminium.

DustDisc Sample	Vanadium	Chromium	Iron	Nickel	Copper	Zinc	Arsenic
1	5.3	32	2.3	6.4	194	269	214,824
2	4.3	25	1.5	9.7	92	218	35,968
3	6.1	58	8.2	47	369	414	34,240
4	12	59	2.8	36	149	633	31,359
5	7.4	56	3.2	20	149	510	41,232
6	1.6	10	2.4	3.8	100	35	51,154
7	1.6	4.0	5.0	3.7	268	56	81,753
8	23	288	1.9	59	18	1,231	4,013
9	15	65	0.8	31	243	1,055	BDL
10	16	184	1.9	42	BDL	1,057	4,561

BDL = below detection limit

Table 3. Crustal enrichment factors for dust samples with reference to aluminium (averages of monitoring periods 1 and 2).

Samples	Vanadium	Chromium	Iron	Nickel	Copper	Zinc	Arsenic
DustDiscs 1-7 Period 1	3.6	26	3.5	9.4	218	217	82,357
DustDiscs 1-7 Period 2	7.4	43	3.8	27	159	394	57,794
Soil average	2.5	1.4	2.4	1.5	391	18	111,373

Table 4. Crustal enrichment factors for averages of dust samples 1-7 with reference to aluminium for sample monitoring periods 1 and 2 in relation to soil averages.

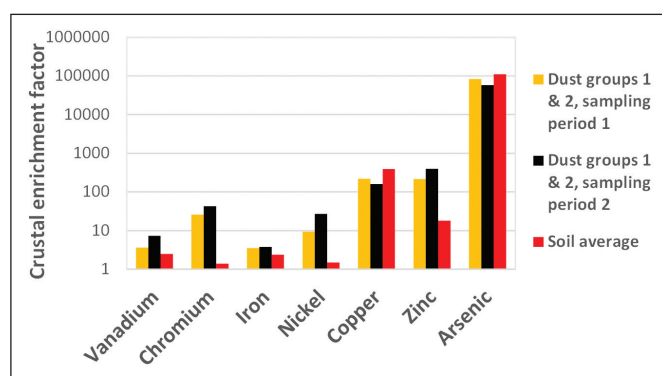


Figure 3. Average crustal enrichment factors with reference to aluminium for soil and dust samples 1-7 for sample monitoring periods 1 and 2.

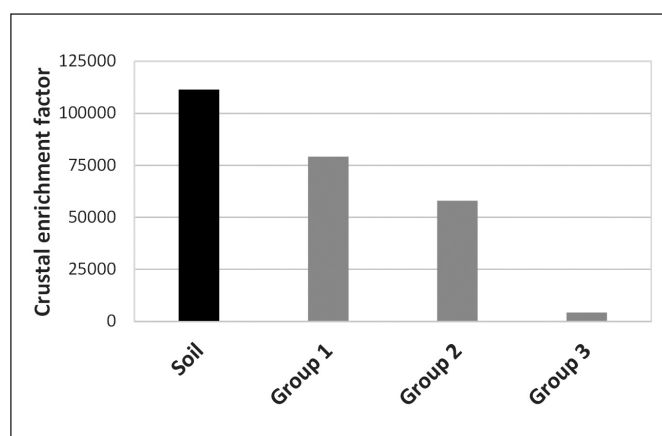


Figure 4. Average crustal enrichment factors of DustDisc groups for arsenic.

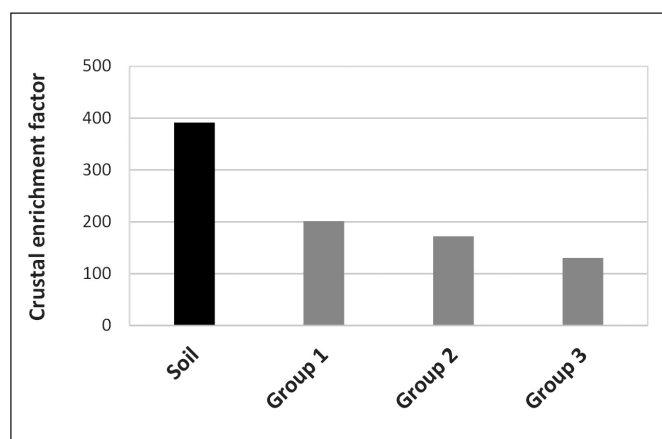


Figure 5. Average crustal enrichment factors of DustDisc groups for copper.

DISCUSSION

The first objective was to investigate the levels of toxic elements present in the dust downwind of the mine spoil sites and assess whether a link existed between distance from the contaminated land and toxic element concentrations. Many of the elements did not show any particular trends or patterns. It is apparent that arsenic and copper, the two main elements mined on site, demonstrated a decrease in EF with distance away from mine spoil sites. It is suggested that levels of arsenic present in dust particles are higher around the mine spoil sites, and may pose contamination risks to surrounding areas as they travel downwind.

The second objective was to investigate whether elemental levels found in the dust samples could be attributed to the soil and surrounding mine spoils. The data for many elements again did not show significantly high levels, but for arsenic and copper, the two data sets were linked. This shows that the high levels of soil contamination, in the form of toxic elements, are also present in the dust particles. This suggests that the area might be considered hazardous, not solely for direct dermal contact with the surrounding mine spoils, but for potential ingestion and inhalation of dusts by visitors to other places in the area.

The last objective was to investigate whether the toxic elements found in the dust samples were comparable to background levels and national air quality standards, and to highlight any potential impacts on receptors. A study by Wang and Mulligan (2006) found that average atmospheric levels of arsenic throughout Canada were less than $0.3 \mu\text{g m}^{-3}$, arising from uncontaminated soils with 4 to 150 mg kg^{-1} arsenic. In areas of soils with similar arsenic concentrations to Devon Great Consols ($25,000 \text{ mg kg}^{-1}$) however, arsenic levels were found in the air to be up to $6.5 \mu\text{g m}^{-3}$. Given that soil sample concentrations at the site were found to be significantly above background levels, and were linked to levels of arsenic in dust particles, it could be speculated that it provides a pathway for enhanced levels of arsenic to sensitive receptors such as the human body. The Air Quality Standards guideline for arsenic currently stands at 6 ng m^{-3} , and given the data reported above, it is feasible that concentrations may be many times that around the source area and up to 300 m downwind in the Group 2 area. However, the results also showed that dust abundance at Group 3 samples 500 m downwind was significantly lower. It is therefore reasonable to speculate that sensitive receptors might be affected by high levels of arsenic in the dust over several hectares east of the dust source area, but that further study is needed to define the limits of this pollution.

CONCLUSIONS

With average UK levels of arsenic in soil at 20 mg kg^{-1} , and soil levels on site measured at over $30,000 \text{ mg kg}^{-1}$, it is clear that some of the soils present at Devon Great Consols represent an increase of over 1,500 times the UK average. Given high levels of bioaccessibility reported for similar soils, such values are potentially toxic. Soil and dust EFs for arsenic and copper are shown not to be significantly different from each other, and

therefore the high levels of these elements present in soils can be linked to high levels in fugitive dust from the site.

The current Air Quality Standards Regulations 2010 for arsenic in PM₁₀ form indicate that levels at Devon Great Consols could potentially be in breach of these standards. High EFs for As and Cu were found in the source area of Group 1 in addition to those downwind at Group 2, collected from the pathway between source and potential receptor. This indicates that toxicants are present in areas up to 300 m from their respective source. Dust particles of the correct size (i.e. PM₁₀ and below) can be transmitted to the lungs or stomach of human receptors by inhalation or ingestion, and with the known toxicities of these elements, this could potentially be a cause for public concern. It is consequently recommended that further study takes place over an extended period of time to observe dust flux and deposition in the surrounding areas.

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