

25. Refining the human health risk assessment process in Northern Ireland through the use of oral bioaccessibility data

SHERRY PALMER,¹ ULRICH OFTERDINGER¹ AND JENNIFER MCKINLEY¹

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The Tellus geochemical surveys have identified significantly elevated concentrations of potentially toxic elements (PTEs) in rural soils from both natural sources (geogenic) and human activity (anthropogenic). In some areas PTE concentrations exceed generic soil screening criteria for assessing chronic exposure risks to human health. However, human health risk actually depends on the fraction of a contaminant that can be absorbed by the body rather than its total concentration. We assess the oral bioaccessible concentrations of arsenic, chromium, nickel and lead measured at 145 sites across Northern Ireland. Arsenic total concentrations are high over the mineralised areas of County Tyrone and County Armagh but oral bioaccessibility is highest in the latter. Total concentrations of chromium and nickel are unusually high over the north-east of Northern Ireland but bioaccessibility is lower than for arsenic; enhanced bioaccessibility of chromium and nickel occurs in parts of County Antrim and also for nickel in County Armagh. High total lead occurs throughout the surveyed area, probably with a significant anthropogenic component. Enhanced lead bioaccessibility is mapped in parts of counties Antrim and Armagh and in the vicinity of Belfast.

BACKGROUND

The starting point for assessing human health risks from exposure to land contamination is often generic soil assessment criteria such as the Soil Guideline Values (SGVs) for certain chemical elements and compounds published by the Environment Agency (EA). SGVs are intended for use during the generic quantitative risk assessment process as initial soil screening values. They are not intended for assessing the potential for acute exposure but instead are regarded as the maximum soil contaminant concentrations tolerable over an extended exposure period (EA, 2009a).

¹ Queen's University Belfast

SGVs are calculated using the contaminated land exposure assessment (CLEA) model, which assumes a default contaminant bioavailability of 100% (EA, 2009b), whereby all of an ingested contaminant will be absorbed by the body and therefore available to cause harm. However, contaminant uptake is rarely 100% of intake (Ruby *et al.*, 1999). Assumptions made under such default CLEA settings can undermine the appropriateness of a CLEA-derived SGV. If required during the risk assessment process, simple or detailed adjustments can be made to the CLEA model to suit a variety of site-specific exposure situations. Detailed adjustment of the CLEA model, however, relies on specialist knowledge and the acquisition of site-specific data (EA, 2009a).

Identifying contaminant linkages

Different contaminants pose variable risks depending upon the exposure pathway encountered and, if available, such information is taken into account when SGVs are derived. Exceeding a SGV is not enough to conclude that significant harm will occur and it is not intended as a remediation trigger value (EA, 2009a). Neither does the presence of a high contaminant concentration in soil automatically affirm the presence of risk. Assessing potential human health risks from contaminated sites instead relies upon gathering a body of evidence to determine if risks are present through the identification of contaminant linkages, including consideration of the principal pathways of oral ingestion, dermal exposure or inhalation (DEFRA, 2012). Oral bioaccessibility data are just one piece of the puzzle when assessing exposure risks if oral exposure is a relevant risk pathway. Collation of such data supports the source–pathway–receptor model advocated in the UK CLEA framework (DEFRA and EA, 2004).

Oral bioaccessibility testing *in vitro* assists in the identification of pollutant linkages inside the human body. The oral bioaccessible fraction of a soil contaminant is the portion that is solubilised in the human digestive system and potentially available to cause adverse health effects. Bioaccessibility differs from bioavailability. For a contaminant to be bioavailable when ingested, it must cross the intestinal epithelium, be metabolised by the body, and enter the blood stream after it is rendered bioaccessible (solubilised) in the gastro-intestinal tract. Bioaccessibility is therefore the limiting step to bioavailability, as an ingested contaminant must first be bioaccessible before it can be bioavailable (Ruby *et al.*, 1999). While risk assessors may be accustomed to identifying contaminant linkages in the external environment, such linkages must also exist inside the human body for health risks to be present. An internal pathway (across the intestinal epithelium) exists between the contaminant source (the soil particles) and receptor (human organs and tissues) only if soil contaminants are rendered bioaccessible in the digestive system.

Risk estimation based on the bioaccessible fraction may slightly overestimate health risks compared to the use of bioavailability data because not all soluble contaminants in the digestive system will necessarily be bioavailable. Some solubilised toxins may still be excreted by the body unabsorbed (Fig. 25.1). However, using bioaccessibility data still

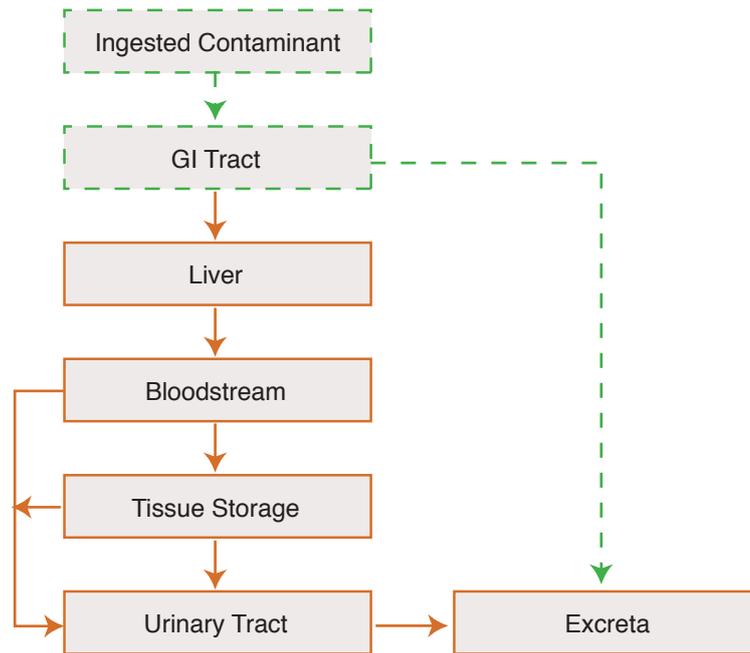


Figure 25.1. Simplified illustration of pathways an ingested contaminant could follow in the human body. Toxic health effects may occur when a toxin follows the metabolic pathway, indicated by solid lines in the diagram. Dashed lines illustrate the measured bioaccessible fraction.

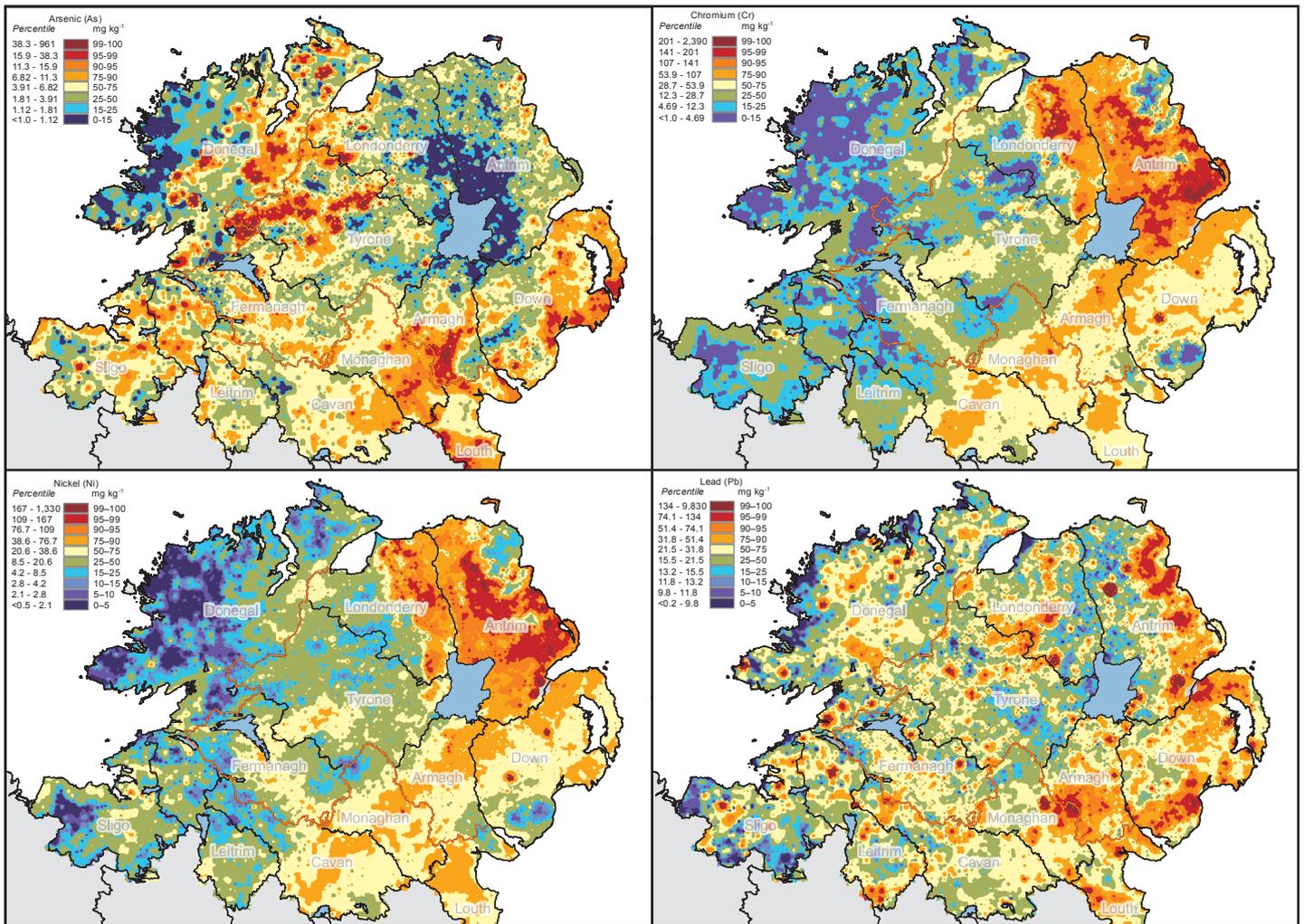
provides a more accurate basis for risk assessment than the use of total soil concentrations and also supports the application of a precautionary principle to conducting a CLRA.

The benefits of oral bioaccessibility testing

Informed contaminated land management practices can save stakeholders substantial sums in remediation costs while also preserving soil resources. Oral bioaccessibility data supplement a lines-of-evidence approach to more accurately characterise human health risk from oral contaminant exposure. Oral bioaccessibility data are not intended for use in isolation to assess risk but instead must be interpreted with all available site-specific information, including land-use and receptor sensitivity.

Bioaccessibility data are particularly useful if the cost of *in vitro* testing will be less than the cost of site remediation. For example, oral bioaccessibility testing may have the greatest merits when applied to sites where an area of land that is too large to be remediated is classified as contaminated, particularly if there is widespread natural or diffuse anthropogenic contamination. Oral bioaccessibility data can also supplement cost-effective *in situ* remedial strategies on smaller sites and can provide the justification for the reuse of excavated material off-site.

In a case study commissioned by the Natural Environment Research Council (NERC, 2012) it was reported that an estimated £3.75 million in remediation costs were avoided during the redevelopment of a contaminated former coal mining site by employing *in vitro*



oral bioaccessibility testing during the risk assessment process. *In vitro* soil tests showed that arsenic and hydrocarbons present were not highly bioaccessible, supporting the conclusion that significant risk was not present and eliminating the need for complex remediation. On a housing development project profiled in the same NERC case study, oral bioaccessibility testing supported the conclusion that future residents would not encounter significant health risks from exposure to soil contaminants. This resulted in estimated savings of at least £7 million in remediation costs and further savings in terms of avoiding delays in development.

Soil contamination in the north of Ireland

Figure 25.2 shows the concentrations in topsoil of four heavy metals – arsenic (As), chromium (Cr), nickel (Ni) and lead (Pb) – over the Tellus and Tellus Border survey areas,

Figure 25.2. Total concentrations of As, Cr, Ni and Pb in topsoil across the Tellus and Tellus Border survey areas.

analysed by inductively coupled plasma mass spectrometry (ICP-MS). High total arsenic occurs in the mineralised areas of County Tyrone, extending into County Donegal, and of County Armagh, extending into counties Monaghan and Louth. High concentrations of chromium and nickel are mapped widely across the basalts of the Antrim Plateau, and from south of Lough Neagh, through County Armagh into counties Monaghan, Cavan and Louth. Total lead in topsoils displays a varied distribution throughout the Tellus and Tellus Border areas, probably reflecting its partly anthropogenic origin; like the other PTEs high lead concentrations are found in counties Armagh, Monaghan and Louth. Details of the geological provenance and geochemical characteristics of these elements are given in Young and Donald (2013).

This study examines the concentrations in Northern Ireland soils of As, Cr, Ni and Pb, which in places exceed average UK concentrations as well as generic soil screening values (Table 25.1). In an Environment Agency survey (2006) of local authorities in England and Wales, over 90% of respondents indicated that As, Ni and Pb were present at elevated levels within their jurisdictions. Such elevated concentrations resulted both from anthropogenic activity and from natural background sources (EA, 2006). Numerous sites in Northern Ireland are known to be affected by industrial contamination, although the area that could be designated as contaminated is unknown due in part to the widespread distribution in soil of naturally occurring enhanced levels of certain metals and metalloids.

TABLE 25.1. CONCENTRATIONS OF SELECTED PTEs (MG KG⁻¹): CRUSTAL AVERAGE; UK SOILS; NORTHERN IRELAND SOILS; LOWEST PUBLISHED ORAL EXPOSURE SGV OR GENERIC ASSESSMENT CRITERIA (GAC)

Element	Crustal average ¹	UK average ²	NI average ³	NI max.	SGV or GAC ⁴
As	2–3.4	10.9	10.5	271.2	32 (inorganic)
Cr	100–110	34.4	131.0	1229	Cr(III): 3000 Cr(VI): 2.1
Ni	75–89	21.1	46.2	333.6	230
Pb	12–13	52.6	41.7	18,757	(450)

¹ ATSDR (2007), Garrett (2013), Taylor (1964), Lee & Yao (1970), Alloway (2013).

² UK Soil and Herbage Pollutant Survey (EA, 2007).

³ Tellus Survey total XRFs concentrations in shallow soils (speciation not identified during analysis: toxic effects are species-dependent for some elements).

⁴ Lowest available UK criteria reported where ingestion pathway considered relevant, assuming chronic exposure. The Pb and Ni SGVs have been withdrawn and are provided for reference only. DEFRA and EA (2002), EA (2009c, 2009d), Nathanail *et al.* (2009).

- 2009 'A' UBM Soils
- 2013 'A' UBM Soils

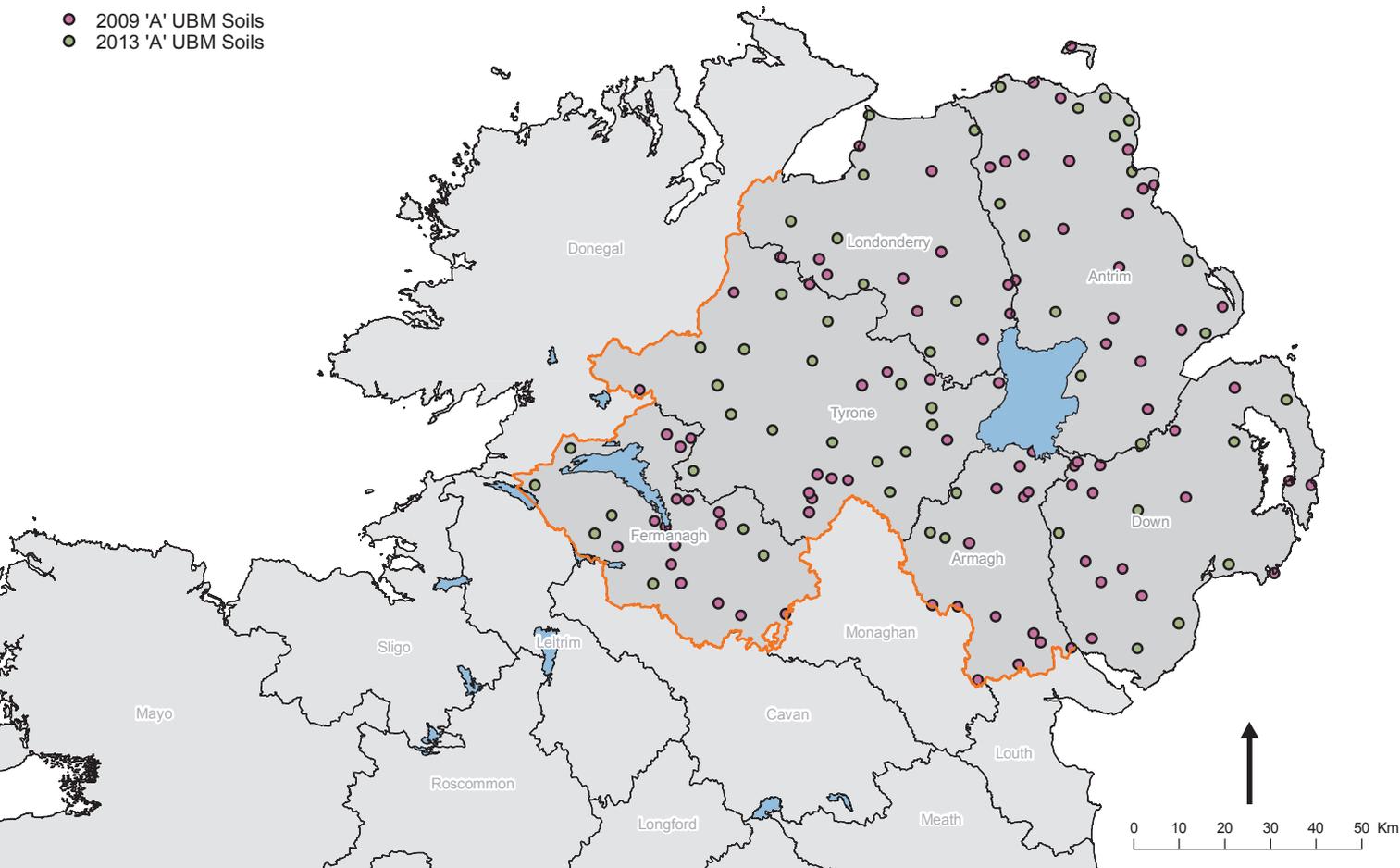


Figure 25.3. Locations of soil samples for UBM analysis.

METHODS

Laboratory procedures

In order to determine whether oral exposure to high levels of PTEs in Northern Irish soils poses a possible human health risk, *in vitro* oral bioaccessibility testing was carried out on a subset of archived Tellus Survey shallow ('A' profile) soils in 2013 using the Unified BARGE (Bioaccessibility Research Group of Europe) Method (UBM). The UBM is an extraction technique that measures the bioaccessible portion of a toxic substance by simulating the conditions of the human stomach (gastric, G) and upper intestine (gastrointestinal, G-I) (Wragg *et al.*, 2011). The full UBM procedure is available online through the BARGE website (BARGE/INERIS, 2011).

UBM extracts were analysed by the British Geological Survey (BGS) Analytical Geochemistry Facility in Nottingham using ICP-MS to provide bioaccessible concentration data (mg kg^{-1}). PTE bioaccessible fractions (BAFs, %) were calculated as percentages of total and partial soil concentrations as measured by X-ray fluorescence spectrometry (XRFS) and ICP-MS respectively (Smyth, 2007). ICP-MS measures only the fraction of trace elements solubilised by *aqua regia* digestion and such analytical results are therefore often regarded as partial rather than total concentrations.

Spatial analysis

The UBM extraction data obtained in 2013 were amalgamated with a pre-existing UBM data set from previous analyses of Tellus soil samples in 2009 (Barsby *et al.*, 2012). A total of 145 'A' profile soil sample locations was thus considered (Fig. 25.3), covering a comprehensive geographical range of soil and underlying rock types across the study area. By joining these two UBM data sets, it was possible to develop geographical models of PTE bioaccessibility covering all of Northern Ireland.

Regional maps of As, Cr, Ni and Pb bioaccessibility were generated using inverse distance weighting (IDW) to illustrate trends in PTE bioaccessibility. IDW is an exact interpolator that estimates mapped values between sample locations by giving the greatest weight to the nearest neighbouring sample locations. For comparison against bioaccessible concentrations, total PTE concentration maps were generated from the shallow (top)soils XRFS Tellus data ($n = 6862$) using ordinary kriging (OK) with 10–12 nearest neighbours. IDW outputs favoured the smaller sample size represented by the bioaccessibility data set while OK was well suited to providing outputs of total soil PTE concentrations for which a larger number of sample locations was available.

The extraction phase yielding the highest average bioaccessible PTE concentration was chosen for illustrative purposes in line with a precautionary principle to ensure that health risks from oral contaminant exposure were not underestimated. G concentrations are presented for Pb, Ni and Cr. In line with available validation criteria (Wragg, 2009), G-I concentrations are presented for As.

KEY FINDINGS

Table 25.2 provides summary statistics for total, partial and bioaccessible concentrations and fractions of As, Cr, Ni and Pb. For these elements, Figs 25.4–25.7 compare the spatial distributions of their total concentrations with their bioaccessibility.

TABLE 25.2. SUMMARY STATISTICS FOR As, Cr, Ni AND Pb SOIL AND BIOACCESSIBLE CONCENTRATIONS (MG KG⁻¹) AND FRACTIONS (BAF, %) OF TOTAL XRFS AND PARTIAL ICP-MS SOIL CONCENTRATIONS (PALMER *et al.*, 2014, 2015; BARSBY *et al.*, 2012). WHERE LARGE DIFFERENCES ARE OBSERVED BETWEEN ICP-MS AND XRFS CONCENTRATIONS AND FRACTIONS, LOW PTE SOLUBILITY IS INFERRED, WHICH IS COMMON FOR GEOGENIC ELEMENTS. SD = STANDARD DEVIATION.

As	Total XRFS	Partial ICP	G	G-I	XRFS		ICP-MS	
					G BAF	G-I BAF	G BAF	G-I BAF
Min.	2.30	0.05	0.18	0.20	2.58	3.35	3.31	5.26
Mean	10.58	6.49	1.46	1.39	12.01	11.83	24.31	24.15
Med.	8.40	4.50	0.96	0.94	11.15	11.67	22.20	22.49
Max.	135.6	108.0	45.15	37.17	58.61	49.35	90.81	91.95
SD	12.09	10.10	3.83	3.16	6.73	5.62	14.20	13.36
Cr								
Min.	4.60	0.50	0.00	0.00	0.04	0.00	0.20	0.02
Mean	117.4	39.21	1.50	1.11	1.20	1.01	3.67	3.31
Med.	85.10	31.00	1.00	0.82	1.09	0.89	3.32	2.81
Max.	510.2	180.0	9.96	7.39	5.36	4.44	13.04	19.84
SD	100.3	33.51	1.57	0.98	0.65	0.55	1.69	2.31
Ni								
Min.	2.20	1.20	0.33	0.07	1.42	0.60	1.64	0.70
Mean	41.12	34.83	3.84	1.89	12.43	5.69	15.24	6.89
Med.	26.20	23.00	2.45	1.31	9.05	5.23	10.12	6.13
Max.	235.1	194.0	30.70	11.10	46.30	16.02	53.64	18.11
SD	42.40	34.47	4.39	1.86	9.17	2.84	11.66	3.57
Pb								
Min.	6.00	5.70	1.49	0.03	8.22	0.17	9.69	0.29
Mean	40.62	36.04	15.34	5.96	32.75	12.46	38.02	14.43
Med.	29.20	24.40	8.38	3.55	32.51	12.40	38.48	14.32
Max.	291.2	268.0	199.8	85.90	68.63	35.06	74.57	38.08
SD	38.18	36.34	21.78	9.52	12.41	6.56	13.60	7.46

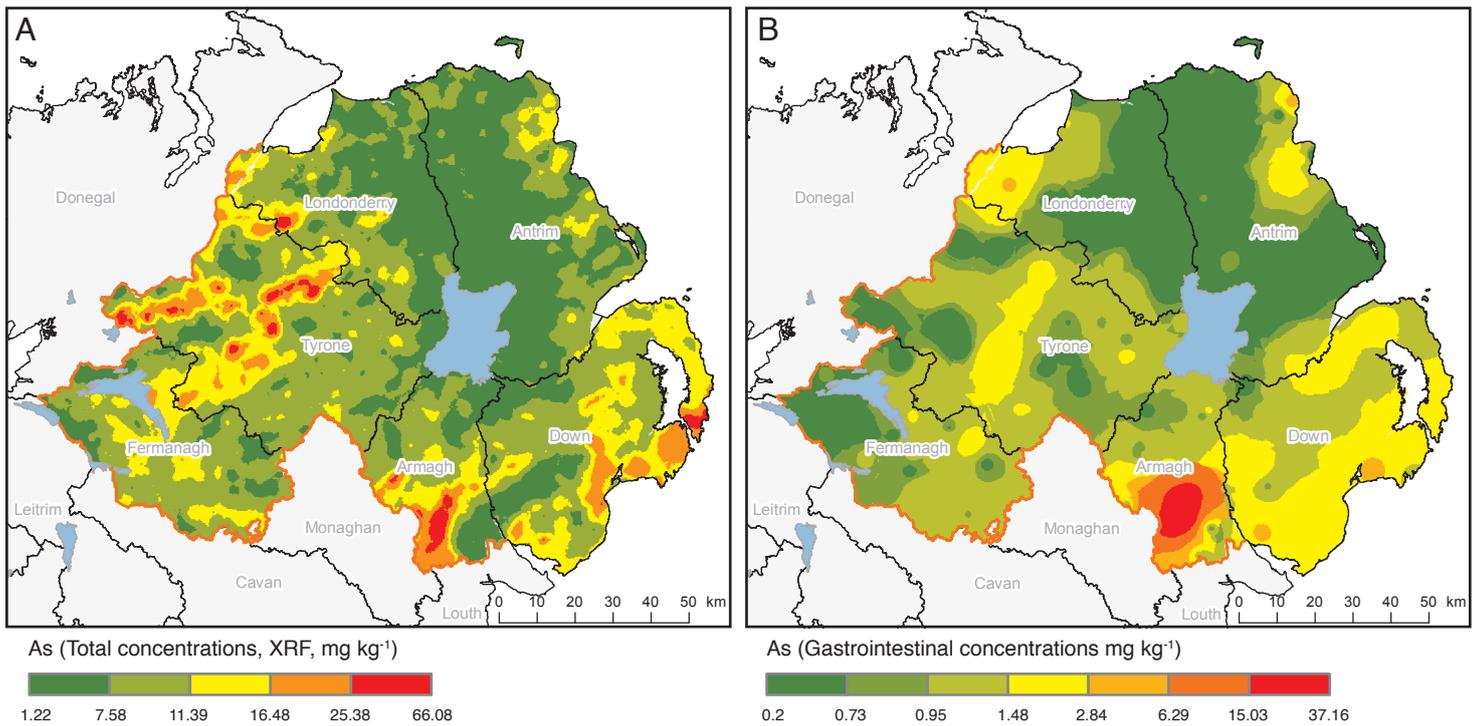


Figure 25.4. Arsenic (As) (mg kg^{-1}): (A) total XRF concentrations (ordinary kriging); (B) G-I As concentrations (inverse distance weighting), showing that higher measured oral bioaccessibility spatially aligns with higher total As concentrations.

Arsenic

On average across the study area of Northern Ireland, less than one quarter of soluble As was bioaccessible in soil (median G-I BAF = 22%). A maximum of 37 mg kg^{-1} of As was solubilised by G-I UBM extraction and this maximum occurred in the central southern portion of the study area in the County Armagh mining district (Fig. 25.4b).

Chromium

Despite high total Cr concentrations in the north-east of the study area (Fig. 25.5a), Cr displayed the lowest measured oral bioaccessibility compared with other PTEs (Table 25.2). This is likely to be due to Cr residing in insoluble forms in soil (Palmer *et al.*, 2014; Cox *et al.*, 2013). However, all exposure pathways should be investigated for this PTE as dust inhalation could also present toxic health risks to exposed populations. Furthermore, Cr speciation has not yet been analysed in Northern Ireland and toxic effects are species-dependent (Alloway, 2013). A maximum Cr G concentration approaching 10 mg kg^{-1} was recorded in the north-east of Northern Ireland (Table 25.2, Fig. 25.5b).

Nickel

Although Ni shares geogenic origins with Cr in soils of the study area, different trends in Ni oral bioaccessibility were observed. Overall, higher measured G and G-I bioaccessibility was recorded compared with Cr, although the average G fraction of total Ni concentrations reached only 12.4% (Table 25.2). Higher G Ni concentrations were found south of

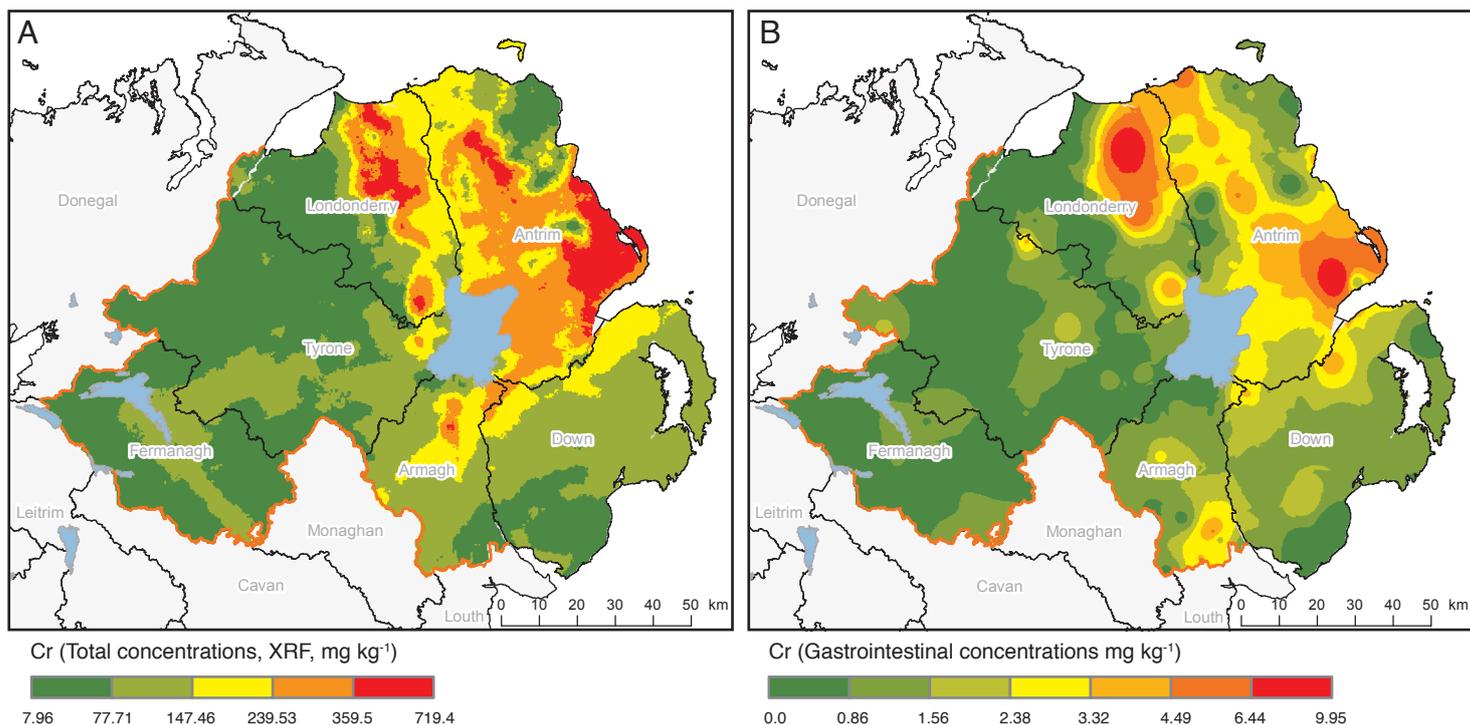


Figure 25.5. Chromium (Cr) (mg kg⁻¹): (A) total XRF concentrations (ordinary kriging); (B) G Cr concentrations (inverse distance weighting), showing higher total and bioaccessible Cr concentrations in the north-east of the study area (Palmer *et al.*, 2014).

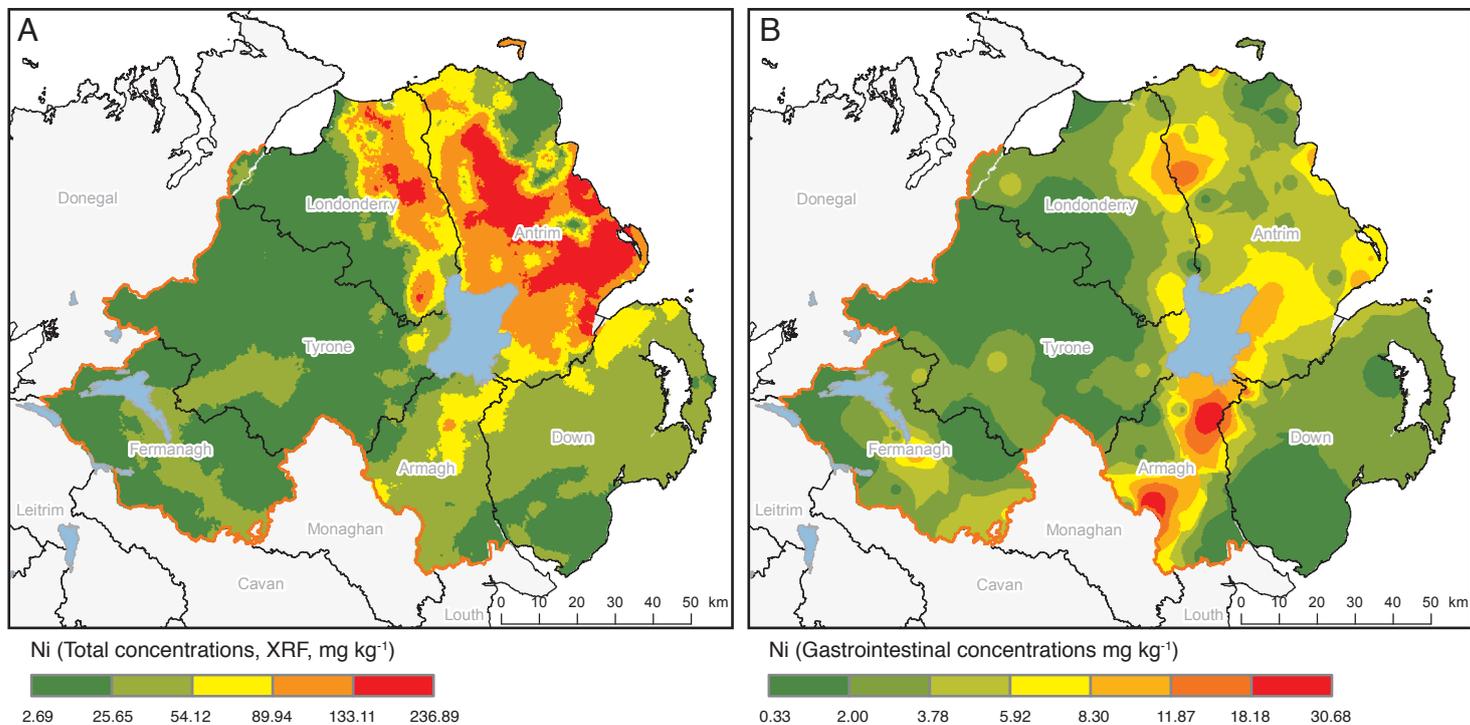


Figure 25.6. Nickel (Ni) (mg kg⁻¹): (A) total XRF concentrations (ordinary kriging); (B) G Ni concentrations (inverse distance weighting), showing higher measured Ni bioaccessibility south of Lough Neagh despite the highest total Ni concentrations occurring in the north-east (Palmer *et al.*, 2014).

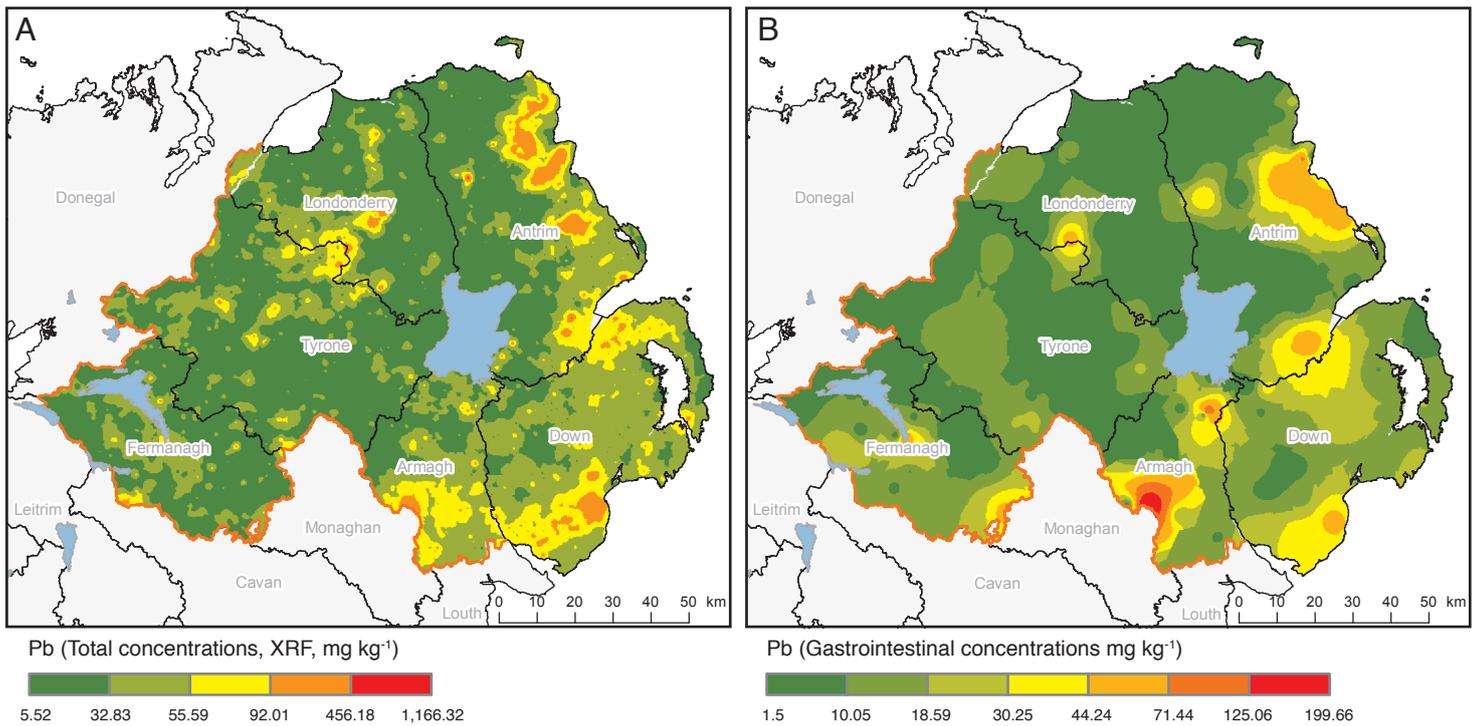


Figure 25.7. Lead (Pb) (mg kg⁻¹): (A) total XRFs concentrations (ordinary kriging); (B) G Pb concentrations (inverse distance weighting), showing higher total and bioaccessible Pb concentrations near urban, mined and mineralised areas (Palmer *et al.*, 2015).

Lough Neagh and in the Armagh mining district, reaching a maximum of 31 mg kg⁻¹, despite higher total Ni concentrations occurring in the north-east (Fig. 25.6).

Lead

On average, approximately one third of total Pb soil concentrations were measured as bioaccessible in the stomach *in vitro*. This average decreased substantially in the intestinal phase, however, with 12.5% of total Pb potentially available for intestinal absorption.

Similarities in total and partial Pb concentrations and BAF suggest that a large proportion of Pb in the study area is highly soluble and therefore potentially bioaccessible (Table 25.2; Palmer *et al.*, 2015). Geographically, the highest concentrations of bioaccessible Pb occur along the north coast in an area of historical mining activity, around the Belfast Metropolitan Area and also in the Armagh mining district (Fig. 25.7).

CONCLUSIONS

Conducting a thorough contaminated land risk assessment relies on the collation of data about all possible sources of and exposure pathways of toxic substances and proposed site-specific land-use activities. Here only the oral pathway has been considered. However, these results demonstrate how total concentrations of PTEs are not the sole determinant for possible human health risks from soil contaminant exposure. It is equally important to determine the fractions of PTEs in soils that are both bioaccessible and bioavailable for human absorption. These are demonstrated to be relatively low in Northern Ireland

for naturally occurring nickel and chromium, which both show unusually high total concentrations. Not all geogenic PTEs are devoid of potential health risks, however. For example, because of the documented spatial correlation between oral arsenic exposure and an increase in human disease burdens (McKinley *et al.*, 2013), further risk evaluation of this largely geogenic element may be warranted in Northern Ireland.

Elevated and bioaccessible lead concentrations appear to arise in part from anthropogenic sources, notably in the historical mining area of County Armagh, which extends into County Monaghan. Despite the low measured G-I lead fractions relative to total and partial lead concentrations measured in soils, it is arguable that any measured bioaccessible lead should be investigated, in view of the current lack of a published threshold (Palmer *et al.*, 2015).

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