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The UK survey of mosses for metals, nitrogen and microplastics, 2020-2022

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Executive Summary

The data from this study will contribute to the 7th European moss survey, which is coordinated by the ICP Vegetation and reported to the Working Group on Effects (WGE) of the Convention on Long-range Transboundary Air Pollution (LRTAP). The first moss survey at the European scale was conducted in 1990 and has occurred subsequently at 5-year intervals as an indication of atmospheric heavy metal pollution and deposition. The moss data provide a complementary measure of elemental deposition from the atmosphere to terrestrial systems compared to conventional precipitation analysis. The UK has participated in the 1990, 1995, 2000 and 2005 surveys, with the dataset here to be submitted as part of the 2020 survey. Moss samples were collected and analysed for a suite of metals, nitrogen and microplastics. For the metals and nitrogen, methodology followed that of the European Moss Survey Protocol (Frontasyeva and Harmens, 2019), whereas for analysis of microplastics a new technique was developed. The focus for this UK survey was on rural sites in order to assess the input from long-range sources. Data have been mapped and compared to previous surveys where appropriate.

Methods

Moss samples were collected from 124 sites across the UK and analysed for content of metals, nitrogen and microplastics. Moss material was dried at 30°C. Acid-digestion of milled samples was performed in a microwave oven. The metal concentrations were determined by Inductively Coupled Plasma Mass Spectrometry (ICP-MS).

A novel technique was developed for analysis of microplastics from moss samples. Moss samples of 10 g were flushed at high flow rate, with a downstream stainless steel 5 µm filter onto which displaced microplastics were captured. The material captured on the stainless-steel filter was then further cleaned through oxidation by Fenton's reactions before deposition on silver filters for analysis. Analysis of the extracted microplastics used µ-FTIR spectrometry using a pixel size of 25 µm.

Automated spectral matching of the raw data was performed using the Purency Microplastics Finder software to identify 21 common polymers: polypropylene, polyethylene, polyvinylchloride, polyurethane, polyethylene terephthalate, polystyrene, acrylonitrile butadiene styrene, polyamide, polycarbonate, poly(methyl methacrylate), polyoxymethylene, cellulose acetate, ethylene-vinyl-acetate copolymer, ethylene vinyl alcohol, polyacrylonitrile, polybutylene terephthalate, polyether ether ketone, polyphenylsulfone, polysulfone, silicone and polylactic acid.

Results

The focus was on rural sites in order to assess the input from long-range sources. For the metals that will contribute to the 7th European moss survey, hotspots were rare but were most frequently associated with Manchester (an urban site) and Wivenhoe, Essex (which may also have some urban influences). More widespread elevations in concentration in mosses were found for cadmium and zinc, and to a lesser extent, copper.

Although median concentrations of many metals in mosses have declined or stabilised over the UK as a whole since the last UK moss survey in 2005, there are possible

increases in median concentration in mosses for the metals cadmium, zinc and copper. It is possible that these may be associated with vehicle use including lubricants and brake and tyre wear (copper and zinc), in addition to domestic wood burning (cadmium), although this is currently unconfirmed.

There are no signs that nitrogen concentration of mosses has declined since previous surveys, despite a decline in emissions to the air from NO_x and NH_Y.

Microplastic content of moss samples was analysed from 52 sites across the United Kingdom. All except two sites monitored experienced some microplastic (MP) contamination above the limits of detection of the assessment. A diverse range of polymers were detected, with the highest concentrations and diversity concentrated in the more north-westerly regions. The mean total number of microplastics >25 µm in size in moss across the UK was 4.5 MP/g with a maximum of 24.7 MP/g detected across the sampled locations.

The most common polymer detected by particle number per gram of moss was polyurethane. This was detected in 87% of samples, with a mean concentration of 1.7 particles of polyurethane per gram moss. This polymer has very wide-ranging applications, from its use in clothing to its application as a coating and binder, from flexible foams used in construction, to insulation in home furnishings and appliances. Possible sources to the outdoor environment should be explored to know whether this polymer has diverse and numerous local sources. The other major polymers detected were cellulose acetate, polyvinylchloride, ethylene vinyl acetate, polylactic acid, and polyethylene terephthalate. Common polymers associated with packaging and macroplastic litter such as polyethylene and polypropylene were less commonly detected above limits of detection in these samples. The major microplastics found were different to those found in UK rivers e.g. the Thames.

Note that a few samples will be omitted from the submission to the 7th European Moss Survey as they do not meet the rural criteria, however, these samples were used for UK specific analysis.

Further work

Many of the major sources for the metals analysed from moss tissue are associated with vehicles – particularly brake pads. However, as the focus of this study was long-range transport of air pollutants the samples were largely taken from comparatively remote areas. It is possible that concentrations would be significantly higher nearer to major roads and in urban areas.

As the decline in concentration of some metals is slowing in some cases, and may even be reversed for others, it is important to continue monitoring. This is particularly important as large changes in domestic fuel use and in vehicle fleet are forecast over the coming decades. In addition, the change in metal concentration within mosses may not match that seen by changes in emissions, particularly if there is resuspension, or if long-range transport of metals becomes a larger proportion of deposition to UK vegetation.

This study has shown widespread occurrence of microplastics in moss samples in rural areas, which are attributed to airborne deposition. These have been differentiated by polymer, but further work is needed to identify the sources of microplastics, and to model airborne dispersion from these sources.

1 Introduction

Objective

Quantify and map the content of metals, nitrogen and microplastic in moss samples from rural areas across the UK as an indication of airborne deposition. This dataset will contribute to the 7th European moss survey, which is coordinated by the ICP Vegetation and reported to the Working Group on Effects (WGE) of the Convention on Long-range Transboundary Air Pollution (LRTAP).

The first moss survey at the European scale was conducted in 1990 and has been repeated every five years since then. The aim of the survey is to identify the main polluted areas for various metals and to further develop the understanding of long-range transboundary air pollution of heavy metals and nitrogen. Apart from spatial patterns, the repeated surveys also provide an indication of temporal trends of heavy metal and nitrogen deposition. The UK has participated in the survey intermittently, including in 1990, 1995, 2000 and 2005. The data from this study will contribute to the 7th European Moss Survey (2020).

The European Moss Survey forms part of the workplan of the UNECE Convention on Long-range transboundary Air Pollution (LRTAP) and is coordinated by the ICP Vegetation. The ICP Vegetation provides information for the review and possible revision of the Protocols of the LRTAP Convention. In 1998, the first Protocol for the control of emissions of heavy metals was adopted in Aarhus. The Protocol states that "an effects-based approach should integrate information for formulating future optimized control strategies taking account of economics and technological factors". Cadmium, lead and mercury emissions were targeted as they are the most toxic of metals. The Joint World Health Organization/Convention Task Force on the Health Aspects of Air Pollution (Task Force on Health) has evaluated the potential health risks of the priority metals cadmium, lead and mercury in Europe in more detail (Task Force on Health, 2007).

In recent decades, mosses have been applied successfully as biomonitors of heavy metal deposition across Europe (e.g. Harmens et al., 2007, 2008). Carpet forming, ectohydric mosses obtain most trace elements and nutrients directly from precipitation and dry deposition; there is little uptake of metals from the substrate (Tyler, 1970). At the European scale *Pleurozium schreberi* is the most frequently sampled moss species, followed by *Hypnum cupressiforme*, *Hylocomium splendens* and *Pseudoscleropodium purum*.

From the start, the European moss survey has provided data on concentrations of ten heavy metals (arsenic, cadmium, chromium, copper, iron, lead, mercury, nickel, vanadium and zinc) in naturally-growing mosses. Since 2005, the concentration of aluminium (a good indicator of wind-blown dust as it is present in high concentrations in the earth's crust), antimony (a good indicator of anthropogenic pollution as it is present in very low concentrations in the earth's crust) and nitrogen were also determined. The moss data provide a complementary measure of elemental deposition from the atmosphere to terrestrial systems, it is easier and cheaper than conventional precipitation analysis, and therefore enables a high sampling density to be achieved.

Across Europe the lowest concentrations of heavy metals in mosses were generally found in northern and western Europe and the highest concentrations in (south-)eastern Europe, resulting in a north-west to south-east gradient (Frontasyeva et al., 2020). Some metals e.g. lead have shown a clear decline in concentration within moss over the past 20 years, in line with reductions in emissions within the European region. However, other metals e.g. mercury, have concentrations in mosses that have remained stable despite a reduction in emissions (Frontasyeva et al., 2020).

Previous studies have shown that there is a good linear relationship between the total nitrogen concentration in mosses and atmospheric nitrogen deposition rates for areas with bulk atmospheric nitrogen deposition rates up to about 20 kg ha^{-1} y^{-1} (Harmens et al., 2011). Nitrogen was included in the moss survey for the first time in 2005. Synthetic fertilizer production together with industrialization, population growth and associated demand for food has resulted in a five-fold increase in emission of reactive nitrogen compounds. Nitrogen tends to stimulate plant growth up to a certain level, above which detrimental effects occur. However, enhanced nitrogen deposition is known to reduce plant diversity in areas and habitats where plants are adapted to low atmospheric nitrogen input. The total nitrogen concentration in mosses can be used to identify areas at risk from nitrogen pollution at a high spatial resolution. Potentially it can also be used as a complementary method to estimate total nitrogen deposition, particularly in lower nitrogen deposition areas (Harmens et al., 2011), although due to the high local variation in nitrogen deposition, the relationship between total nitrogen deposition and the nitrogen concentration in mosses is most robust when deposition rates are measured at the moss sampling sites rather than modelled over a larger area.

More recently mosses have been proposed as a passive biomonitor for the deposition of airborne microplastics. Presence of microplastics within water samples is comparatively well established (e.g. McCormick et al., 2016). A large amount of litter and plastic waste is known to enter water courses and break down into smaller and smaller pieces. A small number of studies have shown the presence of microplastics in rural moss samples (e.g. in Ireland, Roblin and Aherne, 2020) and these are thought to be airborne, but the extent of this and the major sources are not known.

In this study we aim to quantify the metal, nitrogen and microplastic content of mosses sampled from rural areas around the UK. This will give an indication of the spatial patterns of occurrence and a comparison with concentrations within other countries participating in the European Moss Survey. In addition to the UK analysis presented here, the data will also be included within the 7th European Moss Survey.

2 Methods

2.1 Sampling

As in previous surveys, and consistently with the 2020-2022 European Moss Survey, moss samples were collected according to a standardised protocol (Frontasyeva and Harmens, 2019). In line with the 2020-2022 European Moss Survey, samples were collected during the period 2020-2022. Each sampling site was located at least 300 m from main roads and populated areas and at least 100 m from any road or single house. In forests or plantations, samples were collected as far as possible in small open spaces to preclude any significant effect of canopy drip. A few additional samples were collected within conurbations as a contrast, including some from sub-urban country parks, although for all but two samples (Manchester crematorium and Manchester-Withington) the criteria for road and house distance from the sampling site were met.

Due to the travel and site access restrictions due to the Covid-19 pandemic, many samples were collected from volunteers and site managers. Each sample was a composite of about ten sub-samples. Samples were collected into paper bags from England (56 samples), Wales (22 samples), Scotland (22 samples) and Northern Ireland (7 samples). An additional 10 samples were collected but subsequently discarded due to excessive sample contamination with other material (usually soil).

Dead material and litter were removed from the samples in a ventilated laboratory, and only the last two to three years' growth segments were used for the analyses. Samples were dried at room temperature, and stored until chemical analysis.

The most frequently sampled moss species was *Pleurozium schreberi* (31%), followed by *Hylocomium splendens* (23%), *Pseudoscleropodium purum* (10%), *Hypnum cupressiforme* (7%) and *Rhytidiadelphus squarrosus* (2%), with other species and unidentified species (which may have included the target moss species) accounting for the remainder of samples.

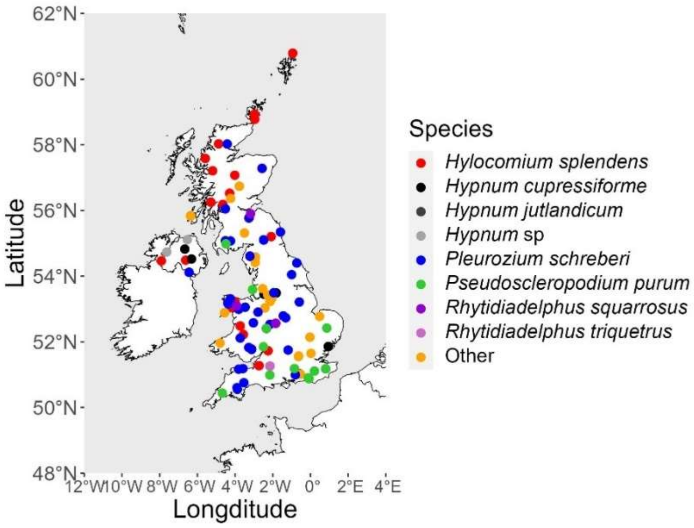

Figure 1: Distribution of moss species collected in the UK, 2020.

2.2 Analysis for metals and N

Moss material was dried at 30 °C. Aqua regia acid-digestion of milled samples was performed in a microwave oven. The metal concentrations were determined by ICP Mass Spectrometry (ICP-OES for aluminium, iron and phosphorus).

Metals analysed were aluminium, antimony, arsenic, barium, cadmium, chromium, cobalt, copper, iron, lead, manganese, mercury, molybdenum, nickel, phosphorus, strontium, titanium and zinc. Nitrogen content was analysed separately by organic elemental analyser (VARIO).

All metal concentrations (including mercury) are expressed as mg kg^{-1} dry weight at 40 °C.

2.3 Analysis for microplastics

A novel technique was developed for analysis of microplastics from moss samples. Analysis of microplastics within organic samples is a relatively new technique. Most have used a conventional organic material digestion and density separation method, established and optimised for other environmental media. This involves wet peroxidation of organic matter using Fenton's reactions and enzymatic treatment of samples (e.g. Horton et al., 2021). The objective when processing samples in this way is to remove infra-red (IR)-interfering material without eliminating the plastic particles themselves. However, this was not found to be successful for the moss samples, as whilst it broke the tangled web of recalcitrant material into smaller fragments, these could not be filtered out from the microplastics, resulting in interference that obstructed Fourier transformed infrared (FTIR) analysis. The organic matter digestion and density separation proved ineffectual in extracting microplastics from larger moss samples, limiting its suitability as a method to provide adequate sample sizes to be representative of the moss as a whole.

It is likely that physiological constraints to the maximum size microplastic that may be internalised into moss tissue is due to stomata, and in the moss *Physcomitrium patens* these are ~10 µm in diameter (Caine et al., 2020). The lower limit to the size of microplastic particle that may be detected by µ-FTIR typically is ~25 µm so rather than extracting microplastics from within the moss tissue the technique developed was to displace and flush microplastics from the superstructure of the moss.

Briefly, moss samples of 10 g were flushed through a prototype chamber at high flow rate using pre-filtered water, with a downstream stainless steel 5 µm filter onto which displaced microplastics were captured. The material captured on the stainless-steel filter was then further cleaned through oxidation by Fenton's reactions for removal of any remaining moss fragments, before deposition on silver filters for analysis.

The technique was refined with comprehensive laboratory testing using the recovery rate of artificially spiked samples of *Pleurozium schreberi.* QA/QC assessment of the method demonstrated 100% recovery of a commercial standard of 45 µm polystyrene spheres. Full procedural blanks demonstrated that this high-pressure flow displacement method contributes very low contamination of plastic to samples, with good limits of detection. Note, this selected method is suitable for analysis of microplastics using µ-FTIR spectrometry, for which the lower size limit of detection is 25 µm. This lower particle size limit is above the physical limits of internalisation into the moss tissues themselves, which is dictated by the size of stomata. Therefore, full digestion of the moss tissue is not required, only displacement of MPs captured in the structure of the moss.

Analysis of the extracted microplastics used μ-FTIR spectrometry. A sub-sample was deposited onto a 25 mm diameter 3 μm pore size silver membrane filter (Sterlitech, Washington USA) using a glass pipette. All microplastics within the deposition area (approximately 11 x 11 mm) were identified and quantified with an imaging μFTIR spectrometer (PerkinElmer Spotlight 400) set to collect spectra in the range between 4000 and 700 cm^{-1} wave numbers. A background spectrum of the silver filter was collected and removed from resulting data. The pixel size selected was 25 μm to give a reasonable compromise between resolution, processing time and resulting file size, this, therefore, limiting the minimum particle size that could be quantified. Mapping was carried out at a resolution of 8 cm^{-1}, with two scans per pixel, and an interferometer speed of 2.2 cm/s.

Automated spectral matching of the raw data was performed using the Purency Microplastics Finder software (pMPf, https://www.purency.ai/product1/microplastics-finder). This software uses machine learning algorithms to automate the data analysis of microplastics measurements. The automated particle finding and analysis prevents operator bias which can occur with manual methods such as with some Raman analysis. The output generates particle counts by polymer type and provides information on the two-dimensional aspects of each particle. A total of 21 common polymers were searched for in the library: polypropylene (PP), polyethylene (PE), polyvinylchloride (PVC), polyurethane (PU), polyethylene terephthalate (PET), polystyrene (PS), acrylonitrile butadiene styrene (ABS), polyamide (PA), polycarbonate (PC), poly(methyl methacrylate) (PMMA), polyoxymethylene (POM), cellulose acetate (CA), ethylene-vinyl-acetate copolymer (EVAc), ethylene vinyl alcohol (EVOH), polyacrylonitrile (PAN), polybutylene terephthalate (PBT), polyether ether ketone (PEEK), polyphenylsulfone (PPSU), polysulfone (PSU), silicone and polylactic acid (PLA).

2.3.1 Quality Assurance

The analytical chemistry group at UKCEH holds UKAS accreditation to ISO 1705:2017. Laboratory standards were used for all metals analysed, and for N. Standards were also used to calculate recovery rates and in cases where the % recovery of the standard metal was below thresholds, then the samples were re-extracted and re-analysed. This re-analysis occurred for arsenic, mercury and antimony. In addition to certified laboratory standards, moss reference material ‘M3’ was obtained from Juha Piispanen (National Resources Institute Finland (Luke), Oulu) for quality assurance purposes and to allow comparison with the wider European moss survey.

For microplastics, a specialist laboratory within UKCEH was used with equipment and ventilation designed to minimise potential contamination of samples. QA/QC was an integral part of developing the successful extraction technique described above. Some moss samples were spiked with a known amount of a polystyrene microplastic standard in order to test recovery. In addition, laboratory standards were used to allow identification of the specific microplastics. As the large proportion of polyurethane was unexpected, identification was checked and confirmed through use of another infra-red spectrometry technique, Agilent's 8700 Laser Direct Infrared (LDIR) imaging system. A particle that was clearly identifiable both in the Purency MP map and the visible image from the FTIR was selected for corroboration by LDIR. A full infrared spectrum in the range of 975 cm^{-1} to 1800 cm^{-1} was taken of this particle in the LDIR and compared against the library on the Agilent Clarity analysis software, confirming identification of polyurethane (version 1.4.1, Agilent Technologies, USA).

For all samples, data were checked for outliers. Geographical data were used to verify that samples had been taken from sites that met the rural criteria – with the exception of the few sites that had been specifically selected to allow an urban comparison.

2.4 Mapping the data

UK metal and nitrogen data were plotted using the statistical software R (R Core Team, 2022), using the ggplot2 package (version 3.4.1). To allow potentially overlapping data points to be viewed more easily, points were ‘jittered,’ which is the addition of a small amount of random variation to the location of each point. This means that the appearance of locations of samples varies slightly between Figures, even though the sample sites were the same.

3 Results

Note: All maps and figures (Figures 2 - 28) are presented at the end of each subsection within the results to avoid breaking up the text.

3.1 Metals

Analysis of metal content occurred for moss samples from 124 sites. Mosses were analysed for the metals aluminium, antimony, arsenic, barium, cadmium, chromium, cobalt, copper, iron, lead, manganese, mercury, molybdenum, nickel, phosphorus, strontium, titanium and zinc. Some of these metals were included in the analysis in previous UK moss surveys. Barium, cobalt, manganese, mercury, molybdenum, phosphorus, strontium and titanium were included for the first time in this survey. The colour scheme according to metal concentration that was used for presentation of the results is as used in the Frontasyeva et al. (2020) for the 2015/16 European survey for ease of comparison. However, barium, cobalt, manganese, molybdenum, phosphorus, strontium and titanium were not routinely included in the 2015/16 survey, meaning that there is no comparable colour scale to indicate low vs high concentrations for these.

Aluminium (Figure 2) concentrations across the UK were generally low, with the exception of small hotspots near to Manchester, Birmingham and Essex. Aluminium can be re-suspended within soil particles, and thus can sometimes reflect historical deposition. Sources of aluminium include from aluminium production, coal combustion, waste incineration and vehicle exhaust emissions.

Antimony (Figure 3) is an indicator of anthropogenic emissions. Several hotspots of concentrations in mosses were found and these included near Manchester, Birmingham, South Wales, London and Essex. Sources of antimony include fuel combustion, brake wear and waste incineration. Although concentrations of antimony in mosses appear slightly higher in the UK than in neighbouring European countries, the median concentration of antimony in moss samples has declined slightly since the previous (2005) survey.

Although concentrations of **arsenic** (Figure 4) in mosses are low compared to eastern and southern Europe, hotspots of arsenic concentrations in mosses were found in samples from near to Manchester, Essex, and some sites in central England. Historically the largest source of arsenic emissions was coal combustion, and UK emissions have declined dramatically since 1990 as coal use has declined. The largest source of arsenic emissions in 2020 was from open burning of treated wood (63% of 2020 emissions, National Atmospheric Emissions Inventory, NAEI, https://naei.beis.gov.uk/overview/ap-overview). Iron and steel production was the next most significant source, contributing 19% of emissions in 2020.

The highest concentrations of **barium** (Figure 5) in moss samples were found near to Manchester, Birmingham and Essex. Barium is not routinely measured in the European moss survey, so it is not possible to identify whether or not these are hotspots compared to whether they still represent a low background. Anthropogenic sources of barium are mainly associated with industrial processes.

Cadmium (Figure 6) concentrations in moss samples were highest in central Manchester and also in south Wales and the south-west of England. The main sources of cadmium emissions in the UK are from the residential sector and industrial use of wood and other biomass fuels. These contributed 39% of emissions in 2020 (NAEI). Industrial emissions of cadmium in the UK and in Europe have reduced since 1990 due to improved abatement of industrial sources.

Chromium (Figure 7) was one of several metals with concentrations in mosses having a relatively uniform distribution across the UK. Vehicle use contributed to emissions, including from tyre and brake wear, which accounts for 28% of UK chromium emissions (NAEI). The moss survey focussed on sampling from rural areas, therefore, chromium is one of several metals that might have higher concentrations nearer to major roads and in urban areas.

Cobalt (Figure 8) has not been measured previously in UK surveys and is not routinely measured as part of the European survey. The highest concentrations in mosses were found in northern, central and south-East England. Cobalt is a by-product of copper and nickel mining, but this is unlikely to be a local source in the UK as nickel has only been mined at small quantities and very few locations in the UK, and historic copper mines were located in Wales, Cheshire and the south-west of England. Cobalt is primarily used in magnetic and hard alloys, and in lithium batteries but it does have a variety of other uses and potential sources.

Copper (Figure 9) has a fairly uniform distribution across the UK, and at low concentrations consistent with neighbouring European countries. Automobile tyre and brake wear contributed 51% of UK copper emissions in 2020 (NAEI). An additional 45% of copper emissions were from lubricants used in road vehicle engines. Since the distribution of vehicle use is concentrated in central and south-east England, this suggests that copper deposition in the UK in rural areas is a result of medium to long-range transport rather than local sources.

Iron (Figure 10) concentration in mosses in the UK was mostly low, with a few sites in England showing elevated concentrations in comparison to these. Major sources of iron to the atmosphere include from windblown desert dust, including from the Sahara. Europe received a large deposition of Saharan dust in February 2021, but in the UK this was largely confined to the south-east, and iron-containing particles were generally concentrated closer to the source (Dumont et al., 2023). Possible anthropogenic sources include from combustion of fossil fuels.

Concentrations of **lead** (Figure 11) in the moss samples were mostly low, with isolated hotspots in Manchester and Essex. Within Manchester the elevated concentrations are likely due to localised emissions within the city, rather than atmospheric deposition. Re-suspension of existing road dust and topsoils may also contribute to this. Following the phasing out of leaded petrol since 1999, emissions from petrol decreased to just 1% of UK emissions. Lead emissions from tyre and brake wear were estimated for the first time in 2019, using emission factors provided in the 2016 EMEP/EEA Guidebook (EEA, 2016). In 2020, the major sources of lead were tyre and brake wear, which now accounts for 36% of the national total.

Manganese (Figure 12) has not been measured previously in UK surveys and is not routinely measured as part of the European survey. There was no clear pattern of manganese concentrations in mosses across the UK. Manganese occurs naturally in some rocks. Emissions to the air increase due to industrial combustion of biomass and wood and this was estimated to account for 78% of emissions in the UK in 2020 (NAEI).

Other sources include emissions from alloy, steel, and iron production and combustion of fossil fuels.

The concentration of **mercury** (Figure 13) within the mosses was generally low and also had a uniform distribution across the UK, with the exception of two sites in Manchester and Essex. Main sources of mercury emissions in the UK in 2020 were coal use in electricity generation and industrial combustion including for iron and steel production. Other sources include from cremations, and disposal of products containing mercury (NAEI).

Molybdenum (Figure 14) showed no clear pattern in concentrations in mosses. It has not been measured previously in UK surveys and is not routinely measured as part of the European survey. Natural sources include sea-spray and desert dust, whereas potential anthropogenic sources include from combustion of fossil fuels.

Nickel (Figure 15) concentrations in moss were low and uniform across the UK including in Manchester and the south-East of England. Nickel emissions in 2020 were dominated by combustion of fuels containing petroleum coke and heavy fuel oil predominantly by the residential sector, but also by industry (NAEI).

Concentrations of **phosphorus** (Figure 16) in mosses showed no clear pattern across the UK. Phosphorus has not been measured previously in UK surveys and is not routinely measured as part of the European survey. Potential sources to the atmosphere include dust from soils, biomass burning, and combustion of oil and coal.

Strontium (Figure 17) occurs naturally, particularly in igneous rocks. Strontium levels in the air can be increased by coal and oil combustion, and are released from mining activities. Strontium is also a component of carbonate in television screens. The strontium concentration in mosses showed no clear pattern across the UK, with concentrations varying even between fairly nearby sites. Strontium has not been measured previously in UK surveys and is not routinely measured as part of the European survey.

Titanium (Figure 18) also showed no clear pattern for concentrations in mosses across the UK, and also has not been measured previously in UK surveys and is not routinely measured as part of the European survey. Titanium dioxide engineered particles are widely used in the urban environment both as pigments in paint and nanosized particles in self-cleaning and photocatalytic surfaces.

Slightly elevated concentrations of **Zinc** (Figure 19) in mosses were found near to Manchester, Birmingham and Essex. According to the UK National Atmospheric Emissions Inventory, the main sources in 2020 were use of recovered waste lubricants as fuel (43% of UK emissions), road transport (22%), and iron and steel production (13%). Emissions arising from road transport include those from brake and tyre wear which contributed 22% of total zinc emissions in 2020. The remainder of the road transport emission is from use of both diesel and petrol.

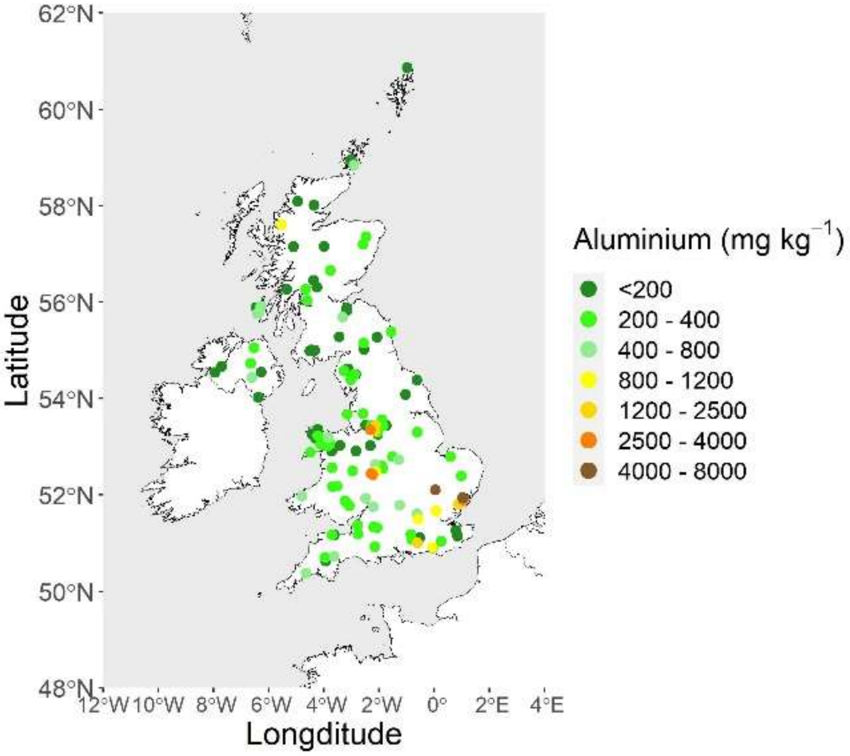

Figure 2: Aluminium concentration in mosses in 2020/2021.

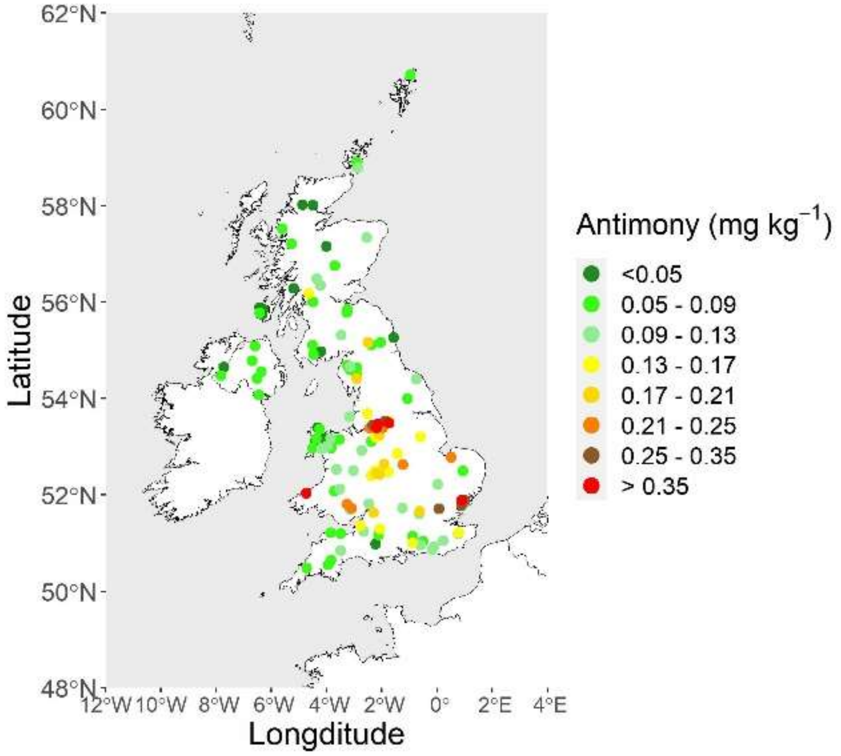

Figure 3: Antimony concentration in mosses in 2020/2021.

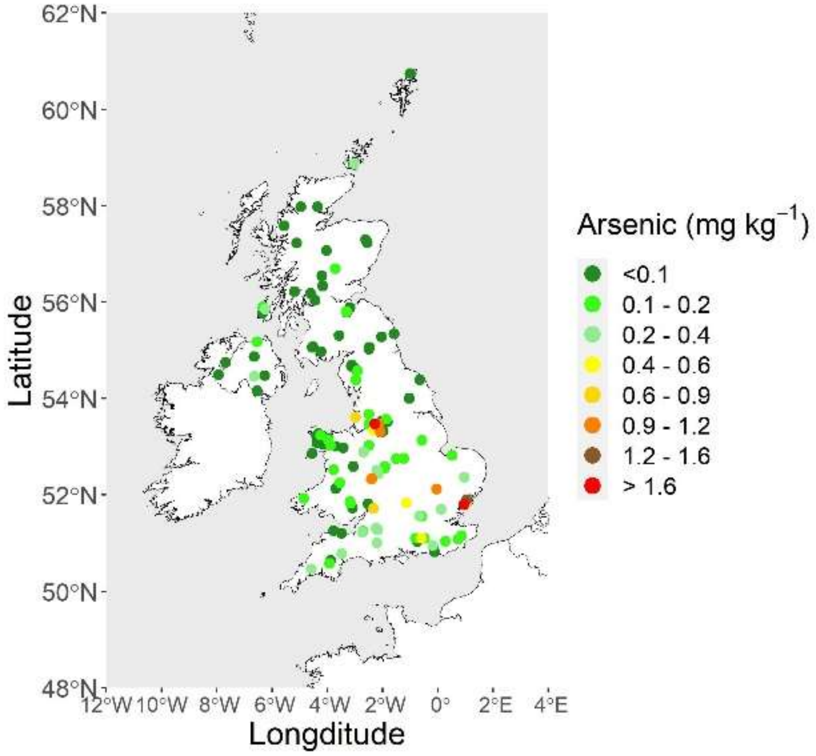

Figure 4: Arsenic concentration in mosses in 2020/2021.

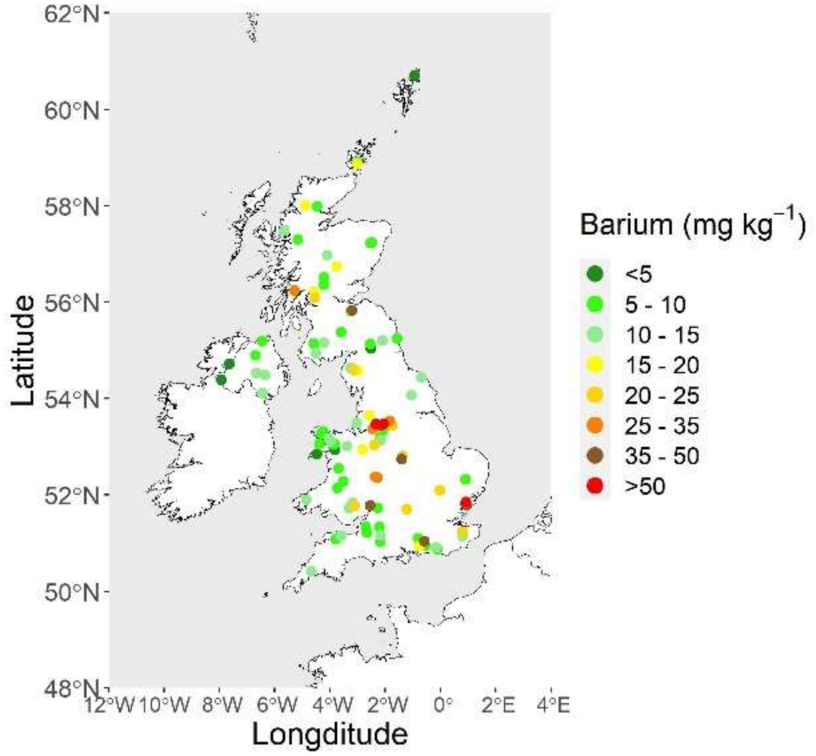

Figure 5: Barium concentration in mosses in 2020/2021.

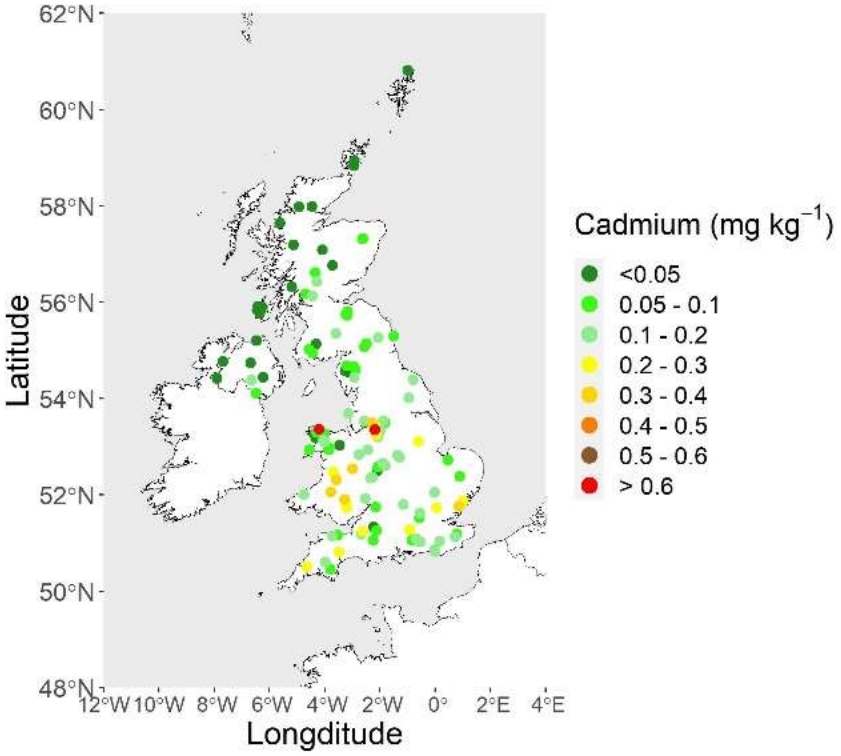

Figure 6: Cadmium concentration in mosses in 2020/2021.

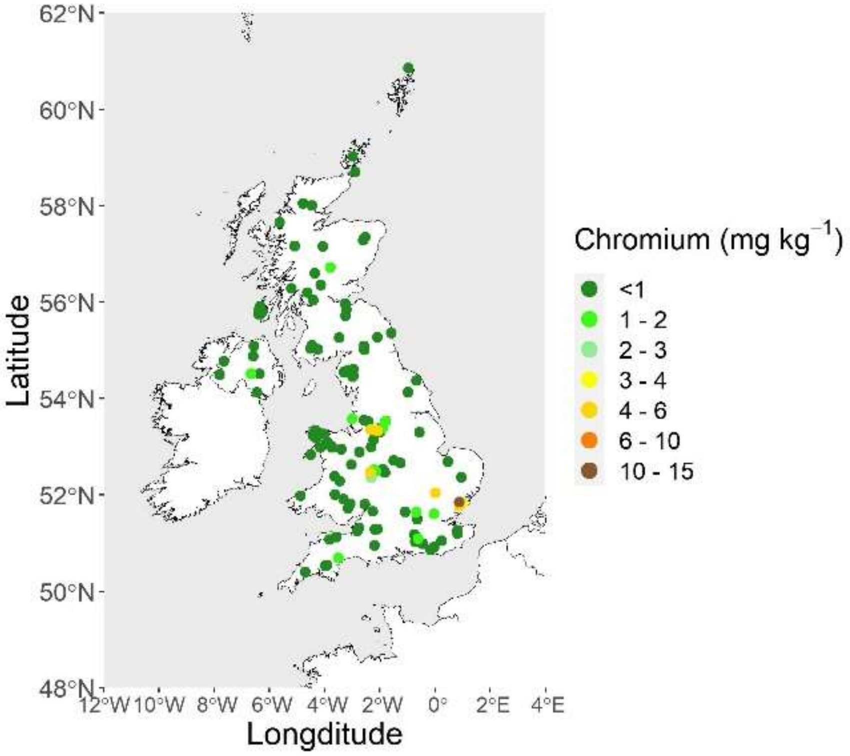

Figure 7: Chromium concentration in mosses in 2020/2021.

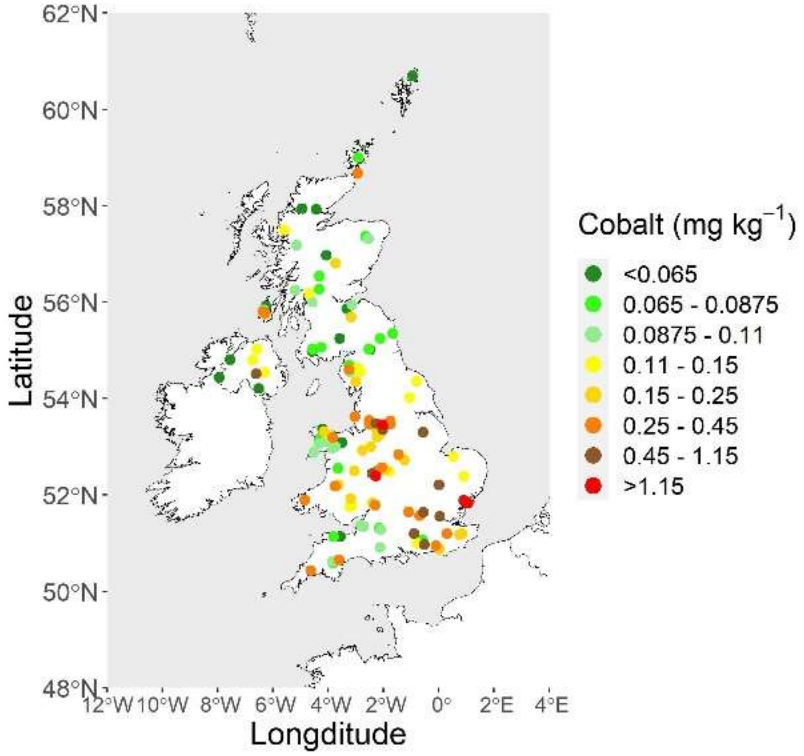

Figure 8: Cobalt concentration in mosses in 2020/2021.

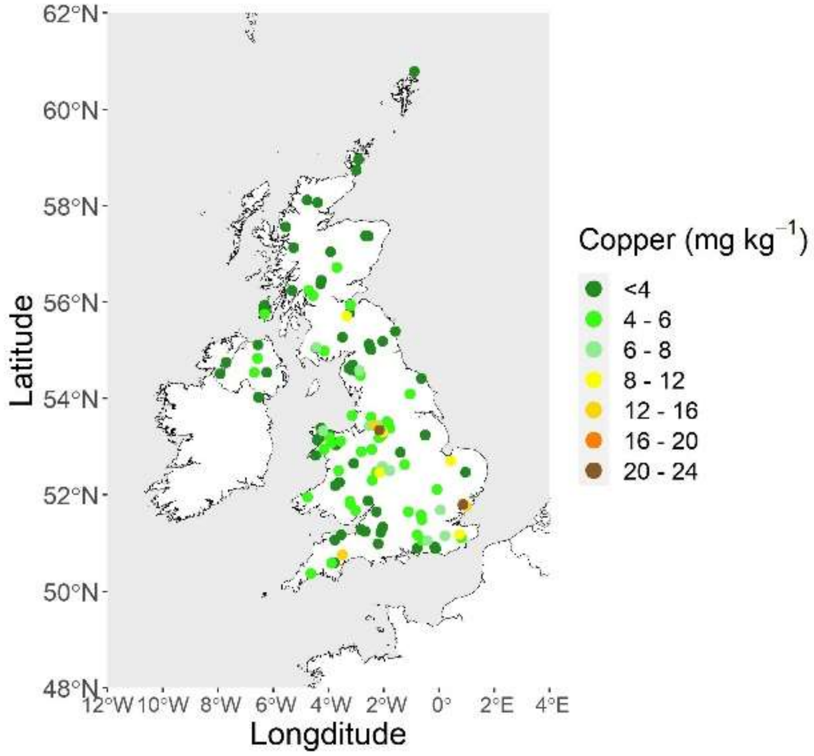

Figure 9: Copper concentration in mosses in 2020/2021.

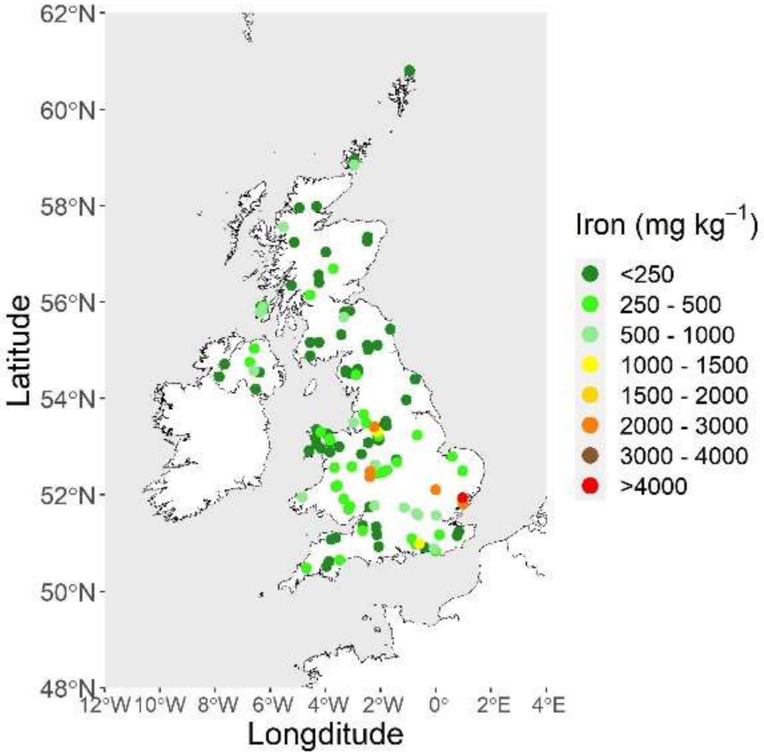

Figure 10: Iron concentration in mosses in 2020/2021.

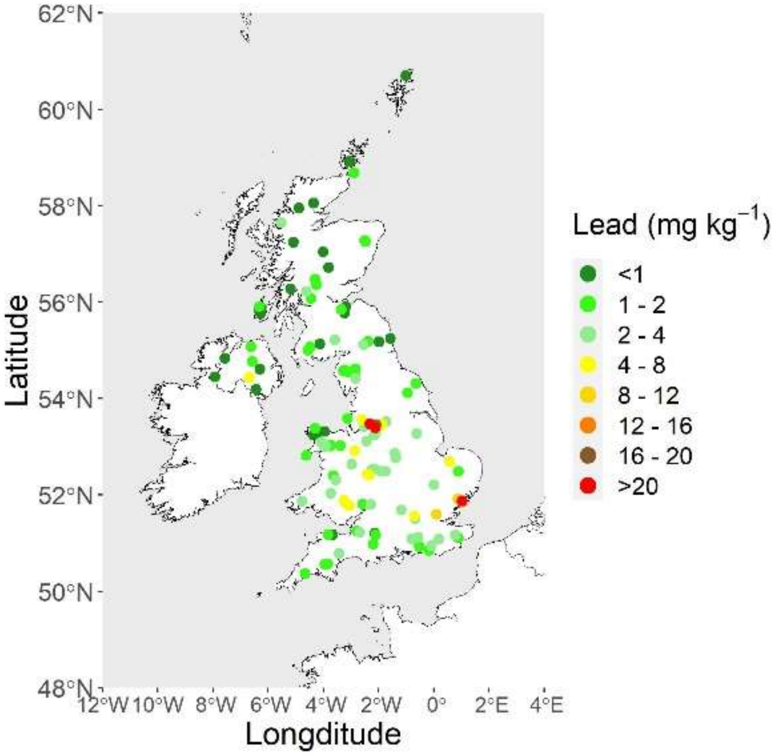

Figure 11: Lead concentration in mosses in 2020/2021.

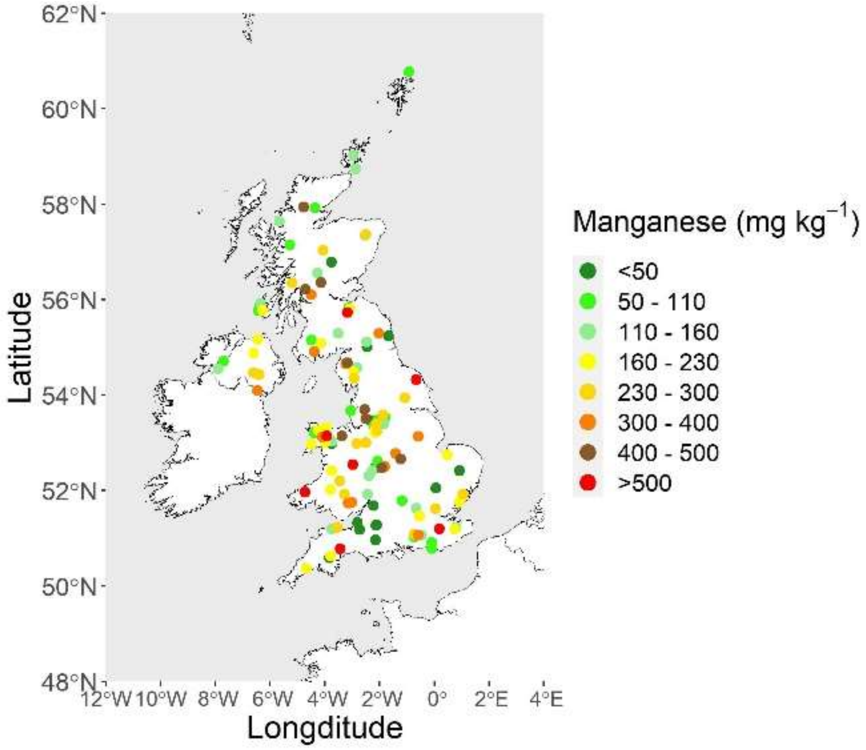

Figure 12: Manganese concentration in mosses in 2020/2021.

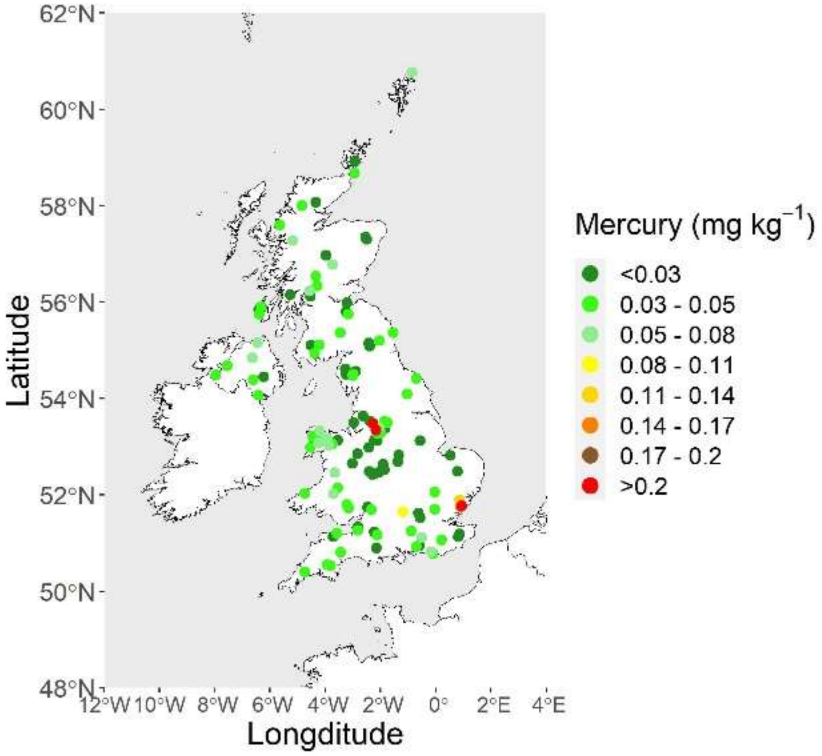

Figure 13: Mercury concentration in mosses in 2020/2021.

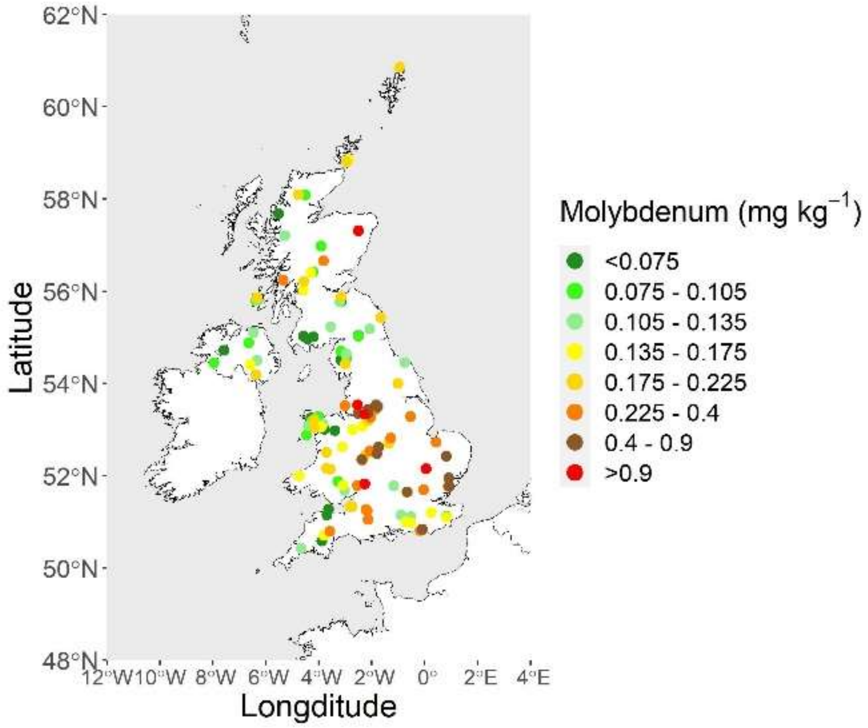

Figure 14: Molybdenum concentration in mosses in 2020/2021.

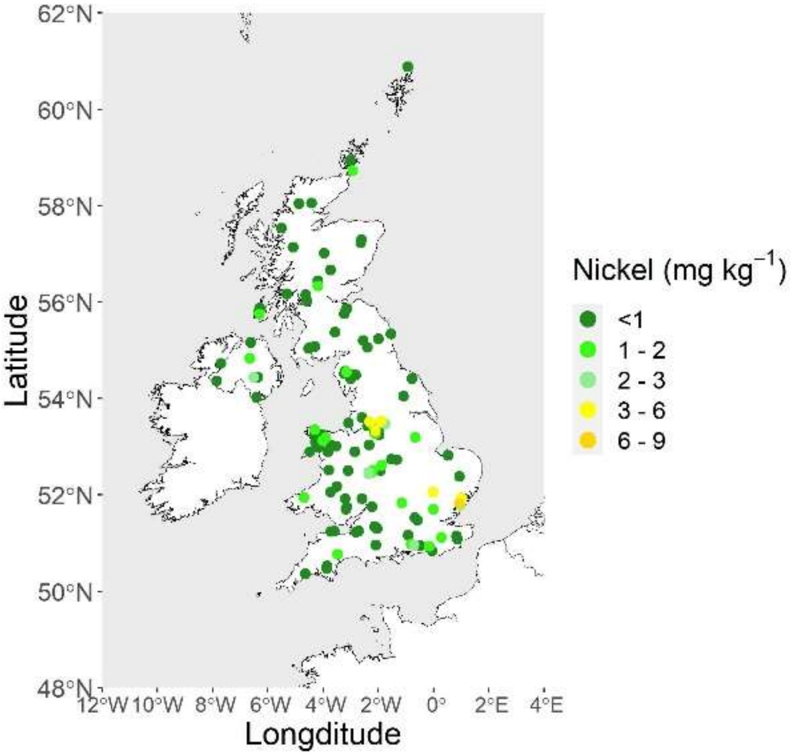

Figure 15: Nickel concentration in mosses in 2020/2021.

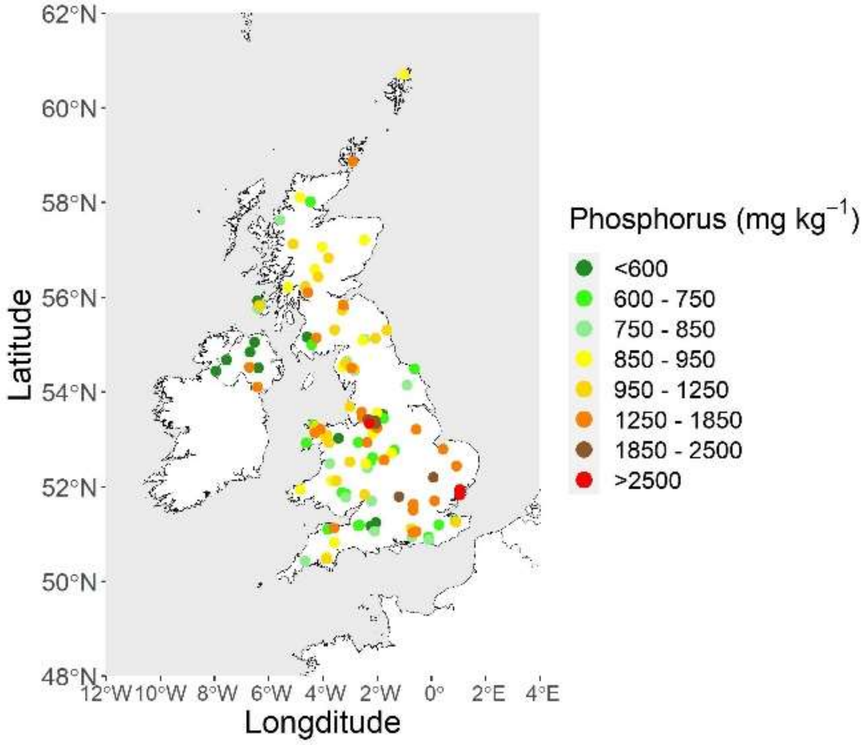

Figure 16: Phosphorus concentration in mosses in 2020/2021.

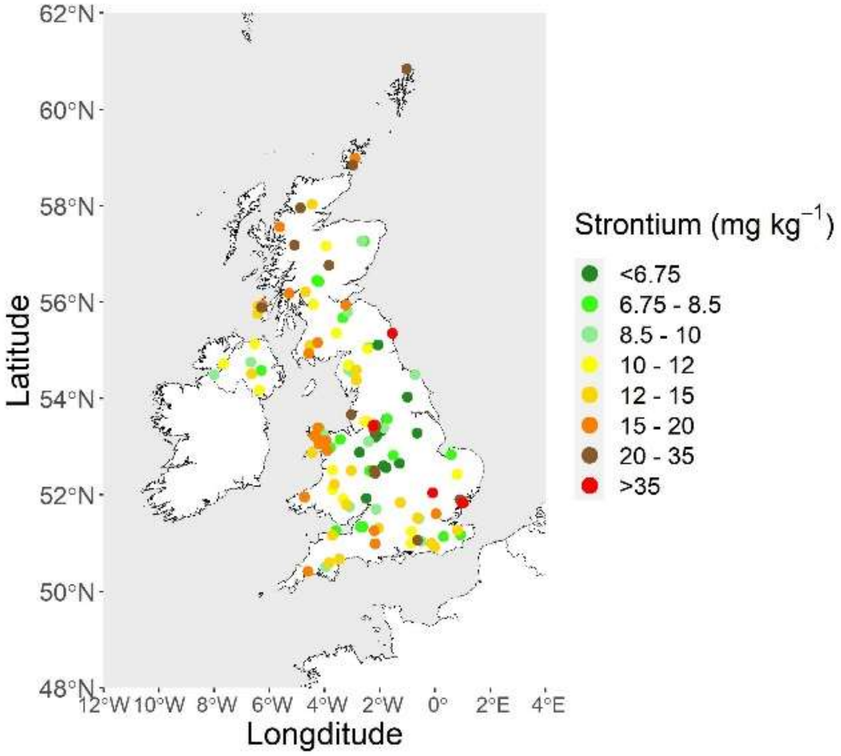

Figure 17: Strontium concentration in mosses in 2020/2021.

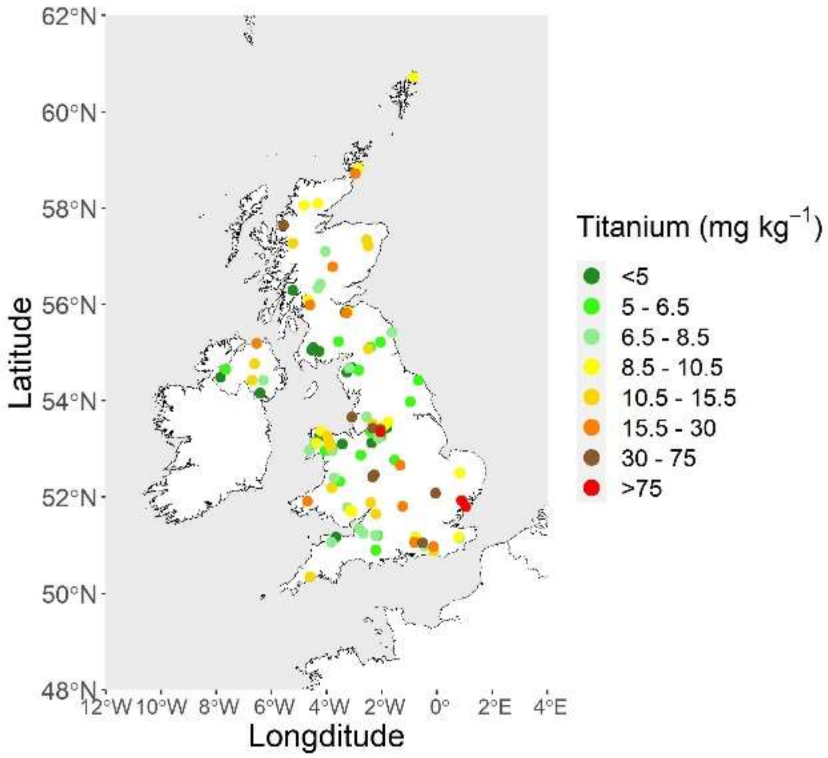

Figure 18: Titanium concentration in mosses in 2020/2021.

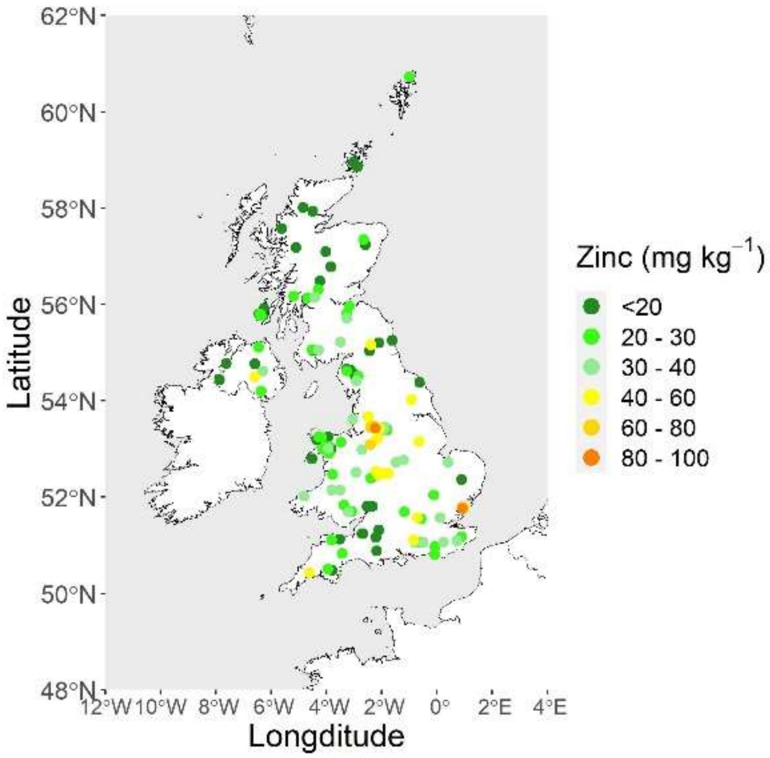

Figure 19: Zinc concentration in mosses in 2020/2021.

3.1.1 Trends for concentrations of selected metals

For metals that have been measured in the UK since 1990 it is possible to identify some trends in concentrations within mosses, although note that the sites sampled are different in the different years, which may affect the analysis. Due to the different sites and different number of sites in each sampling year, analysis is based on median values, which would reduce the influence of any hotspots of deposition.

For chromium, the concentration in moss has declined since the previous survey in 2005, following a dramatic rise between 1990 and 1995 to reach the peak concentration in 2000 (Figure 20). Sources of chromium include emissions from combustion processes including incineration facilities, metal industries, and catalytic converter erosion and wear of brakes from vehicles. The median concentration in moss in 2020 is less than that found for the 1990 survey (the first survey that the UK participated in).

Concentrations of nickel, arsenic and lead (Figure 20) have continued to decline slightly since 2005, following the larger declines found since peak concentrations were observed in moss in 1990 (Ni) or 1995 (As and Pb). Nickel sources include combustion of coal, fuel oil and diesel oil, and incineration of waste and sewage. Arsenic sources include coal burning, smelting and mining. Lead sources were previously dominated by leaded petrol and currently include smelting, battery manufacturing, industrial emissions and leaded paint.

Concentrations of cadmium, copper and zinc in moss may have increased slightly since the last survey in 2005, despite decreases that had occurred prior to this (Figure 21). In addition to emissions from mining and smelting, cadmium has various industrial uses including NiCd batteries, metal plating, pigments, and as stabilisers in some plastics. In addition to mining and smelting sources, zinc is found in some fertilisers and some wood preservatives. Sources of copper include mining and smelting, burning of fossil fuels, wood preservatives, and brake pads of vehicles.

Antimony, aluminium, barium, cobalt, manganese, mercury, molybdenum, phosphorus, strontium and titanium have not been analysed in a sufficient number of surveys to determine trends.

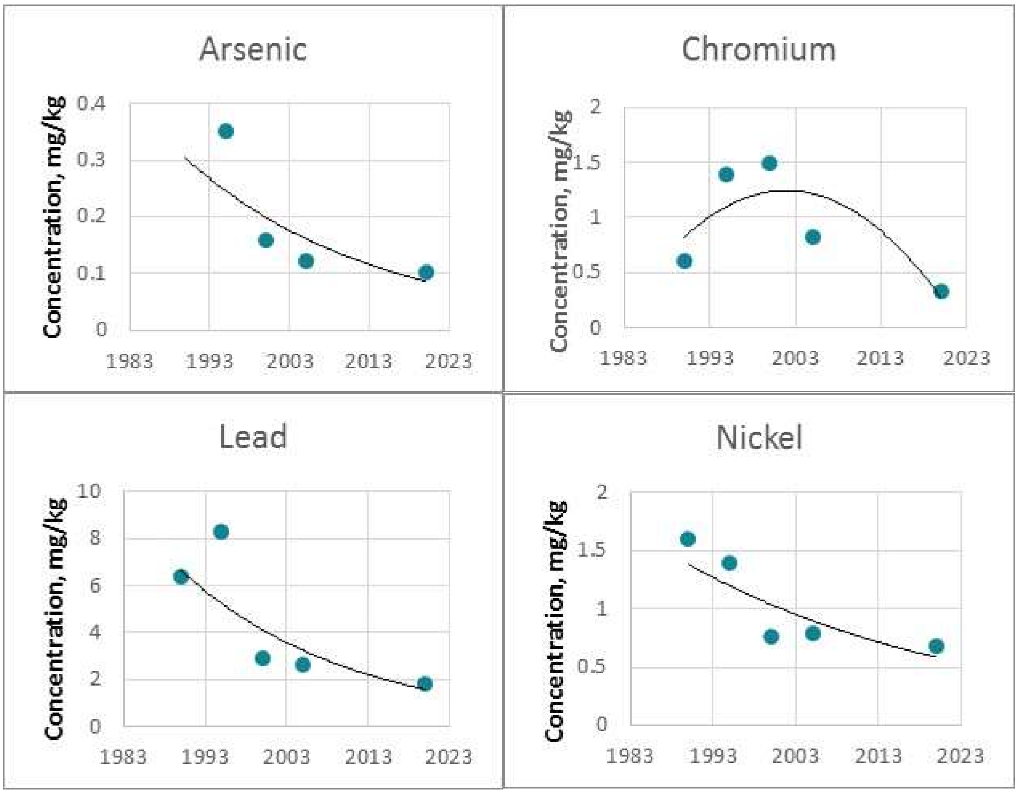

Figure 20: Trends in concentration of metals in mosses for a) arsenic, b) chromium, c) lead and d) nickel.

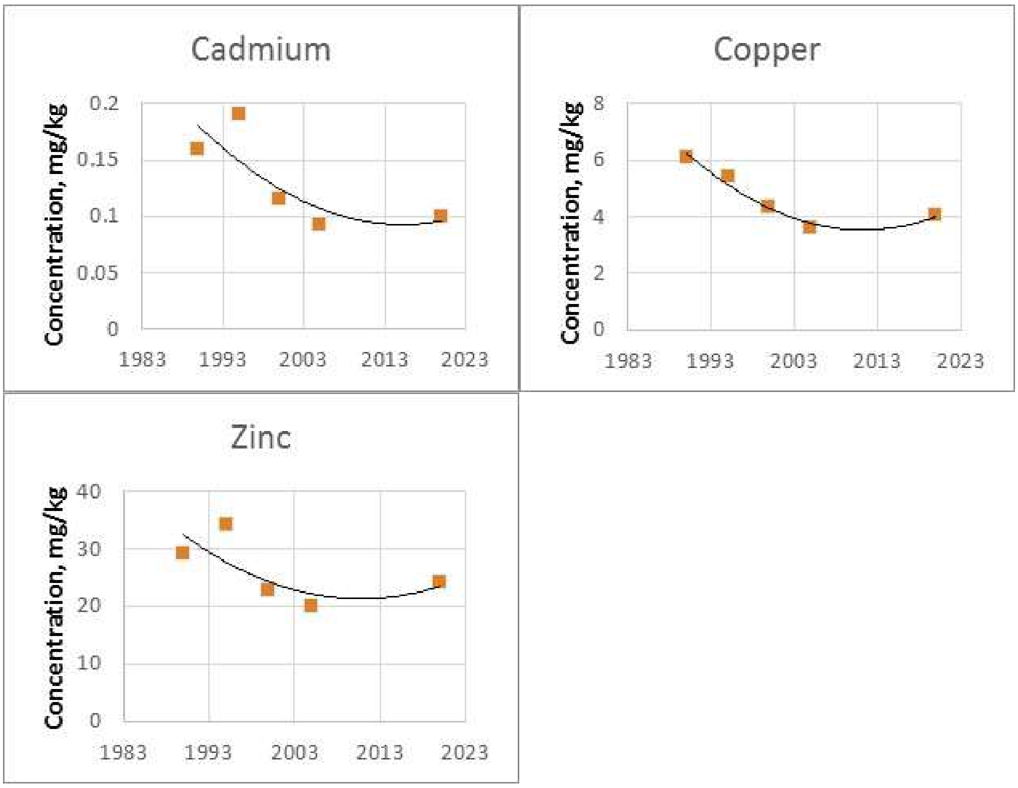

Figure 21: Trends in concentration of metals in mosses for a) cadmium, b) copper, and c) zinc.

3.1.2 European context for selected metals

The UK data from this survey will contribute to the wider (predominantly European) dataset for analysis, due for publication 2024. However, to provide immediate context for some of the metals, results can be compared to the previous European survey (2015 – to which the UK did not participate). A comparison is shown for cadmium, copper and zinc, as these may have increased in the UK slightly compared to previous surveys. A comparison is also shown for mercury, as the UK did not include mercury in previous surveys. Note that the UK maps are repeated from those above, for ease of comparison.

For mercury, concentrations in UK moss in 2020 were similar to those in Europe in the 2015 survey (Figure 22). The concentration of mercury in moss across Europe was fairly homogenous and low (with the exception of France), and with the occasional slightly elevated point. The same pattern and similar concentrations are shown for the UK – note that the ‘hotspot’ in Manchester is from a site within the grounds of a crematorium and does not fulfil the rural site criteria used for the European survey. This suggests that mercury pollution in the UK is dominated by long-range sources, possible outside of the UNECE region, as is the case for Europe.

Concentration of both cadmium (Figure 23) and zinc (Figure 25) in moss has a less homogenous distribution than for mercury, both in Europe and in the UK. However, despite a small increase in median concentrations since the previous surveys, the concentrations in moss in the UK appears similar to those of Europe. The uneven spatial pattern of concentrations is also similar to that shown in Europe.

For copper, which also shows a slight increase in concentration in moss compared to previous UK surveys, the pattern is slightly different (Figure 24). There are a number of ‘hotspots’ around the UK – albeit these are not particularly high. Generally the pattern in Europe is more homogenous, either with low concentrations (such as in France, Germany and Scandinavia), or with higher concentrations as seen in South-Eastern Europe. Possible (non-industrial) sources include domestic wood burning, which may explain hotspots being co-located with some residential areas, but this has not been confirmed.

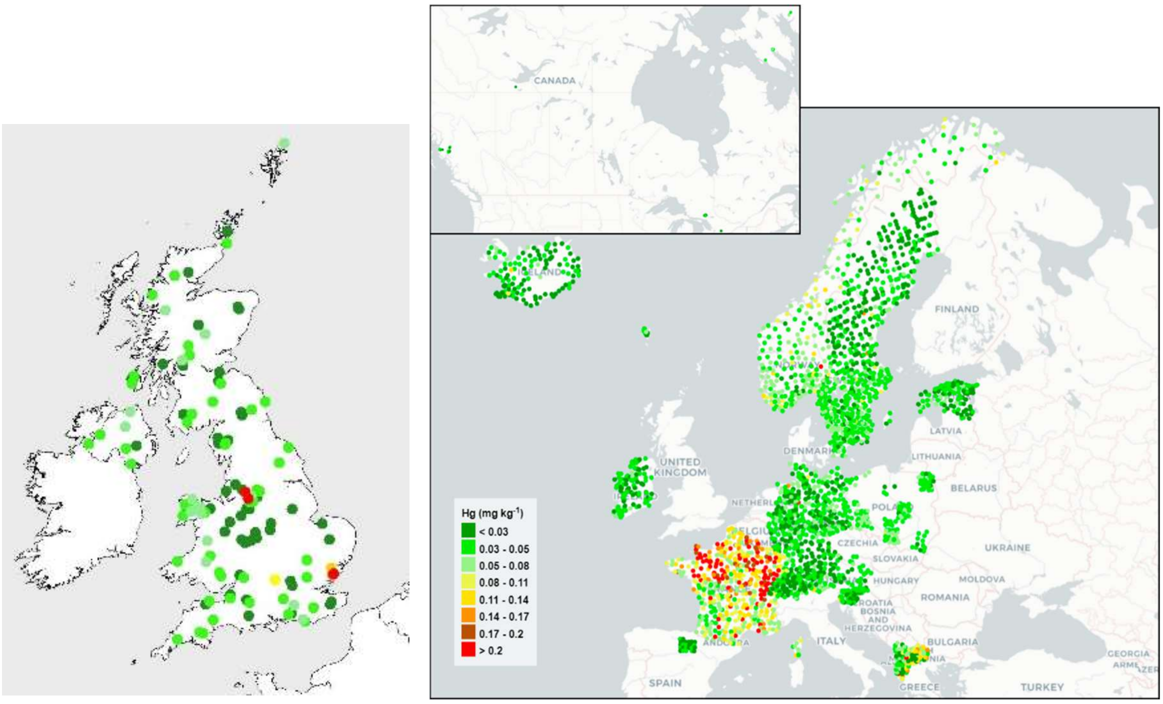

Figure 22: Mercury concentration in mosses in 2015/16, compared to concentrations in mosses in the current UK survey.

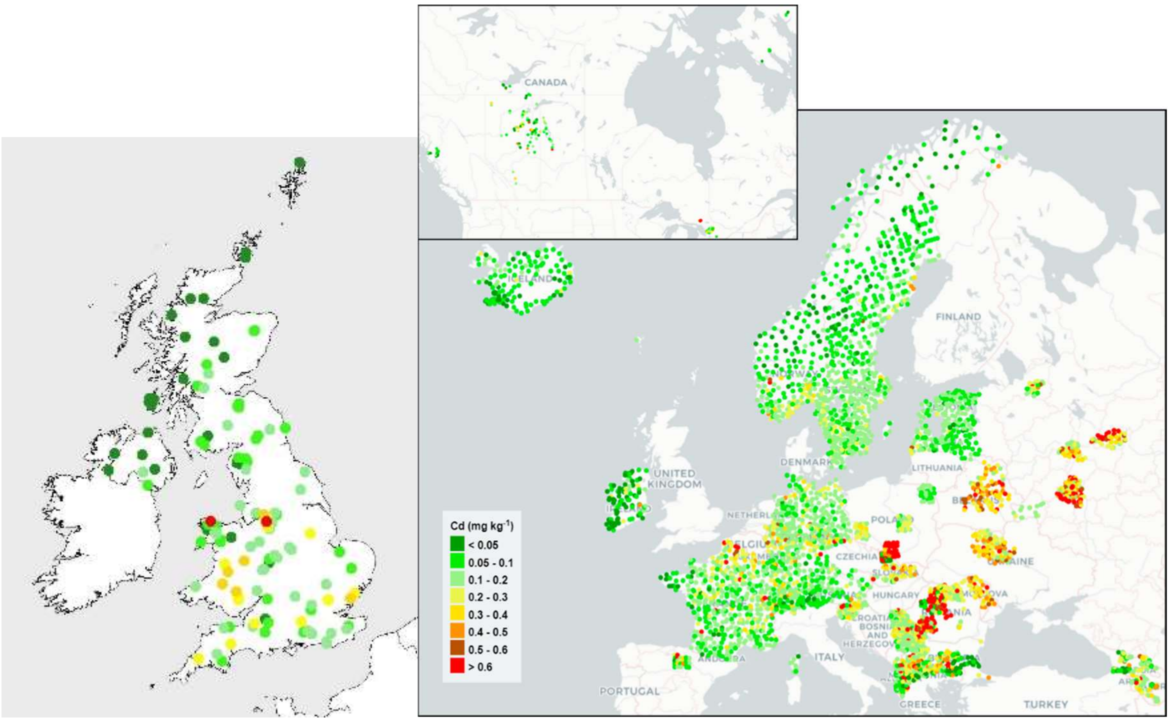

Figure 23: Cadmium concentration in mosses in 2015/16, compared to concentrations in mosses in the current UK survey.

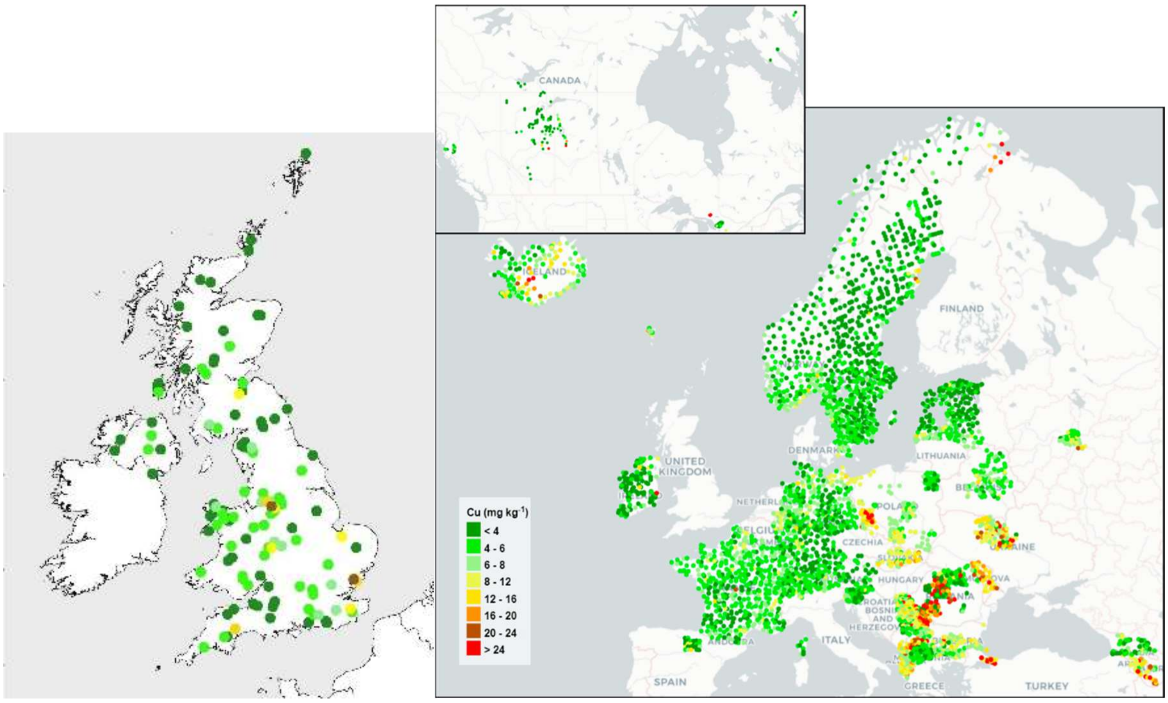

Figure 24: Copper concentration in mosses in 2015/16, compared to concentrations in mosses in the current UK survey.

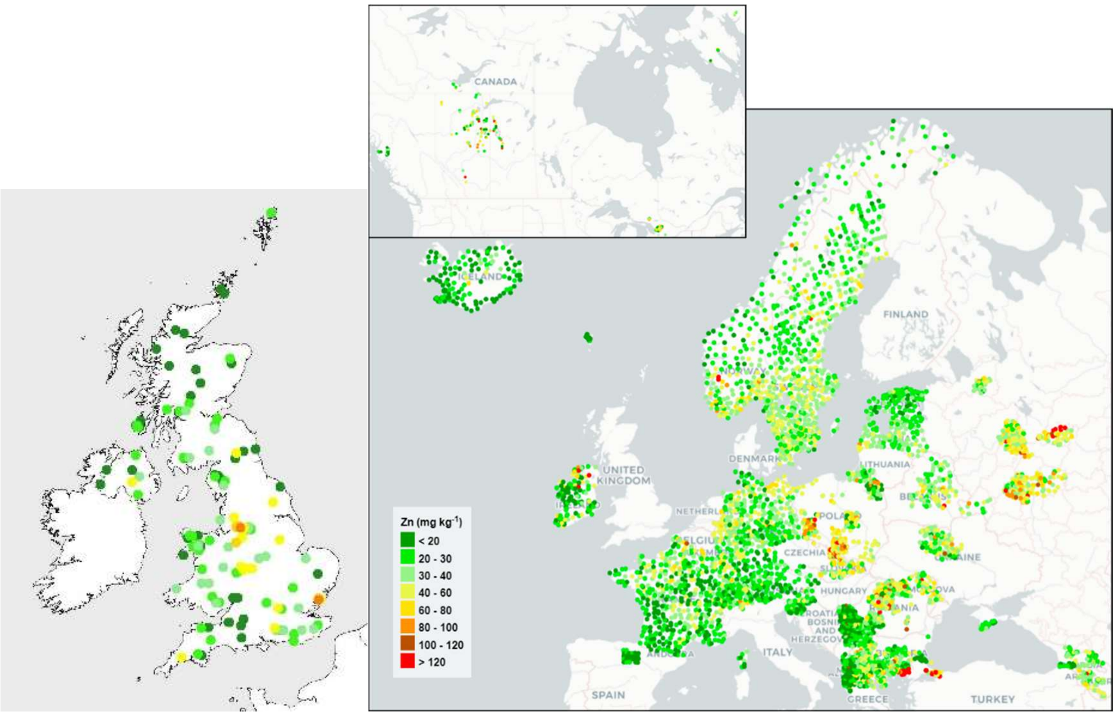

Figure 25: Zinc concentration in mosses in 2015/16, compared to concentrations in mosses in the current UK survey.

3.2 Nitrogen

Analysis of nitrogen content occurred for moss samples from 124 sites.

The concentration of N in moss samples was quite variable across the UK (Figure 26). Generally, concentrations were higher in England, Wales and Northern Ireland than in Scotland. Some local hotspots of nitrogen concentration were found, including in Wales, NW England and SW England. However, these often had neighbouring sites with much lower nitrogen concentrations, indicating a large influence of local factors and emission sources at many sites. It is not clear whether these could relate to local emission sources such as agriculture, or whether some sites were influenced by local deposition sources e.g. livestock and rabbits. The median nitrogen concentration of the moss samples collected in the current survey was 0.97%. This compares to a median concentration within the moss of 0.79% from samples collected during the 2005 UK survey. Although total UK nitrogen deposition has decreased since the peak in 1990, not all forms of nitrogen have decreased. Modelled data for the UK at 1 km resolution suggests that deposition of NO_x has declined markedly, whereas NH_Y has not declined (Tomlinson et al., 2021). It has been calculated that much of the UK and the native habitats are at risk of adverse impacts of nitrogen deposition based on current deposition rates (Rowe et al., 2022).

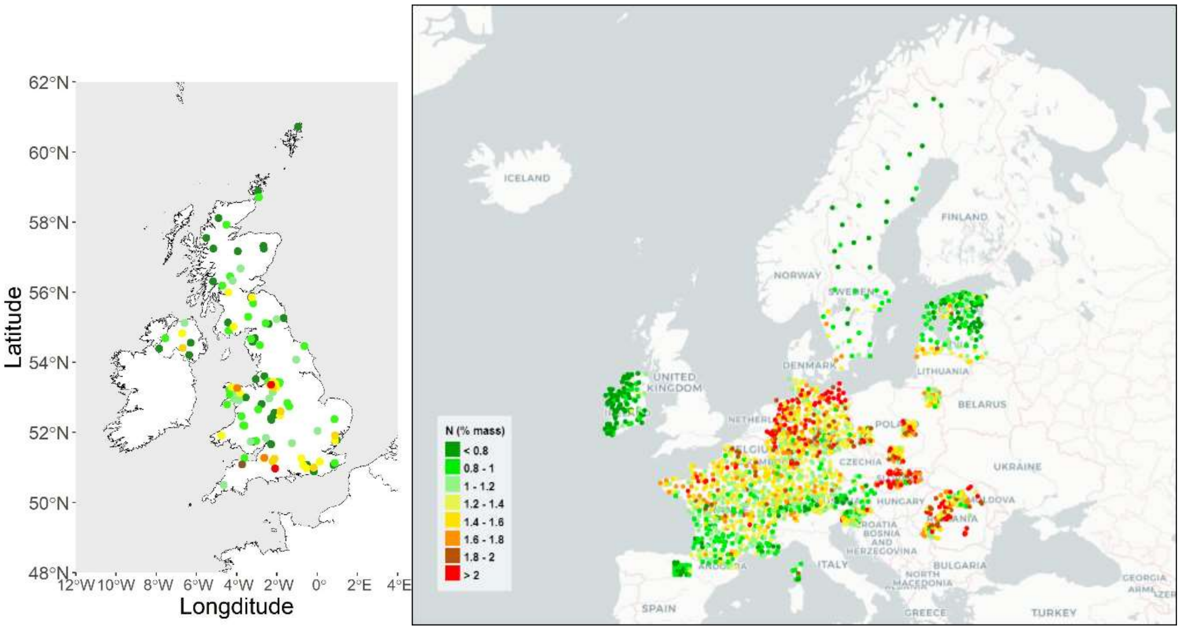

Figure 26: Nitrogen concentration in mosses in 2020/2021, compared to those of the 2015 European survey.

3.3 Microplastics

Moss samples were analysed from 52 sites across the United Kingdom. All sites except two of those sampled showed some contamination above the limits of detection. The mean total number of microplastics >25 µm in size in moss across the UK was 4.5 MP/g with a maximum of 24.7 MP/g detected across the sampled locations. This stresses the importance of having a method which can quantify microplastics in an adequate mass of sample to ensure that the numbers detected are above limits of detection. In this study we were able to process and analyse around 10 g of moss per sample. It is difficult to put these concentrations in context due to no equivalent published data for other regions that is suitable for direct comparison, i.e. equivalent assessment using µ-FTIR and of MPs in the same size region. Roblin and Aherne (2020) did report on microfibre concentrations in remote moss on the island of Ireland, finding an average of 24 fibres/ g moss, with an estimated 25% potentially plastic, indicating good agreement with our findings across the UK. Their study, however, focused only on microfibres and did not confirm polymer identity through chemical analysis as we report here, rather relied on visual criteria and analyst's judgment.

A diverse range of polymers were detected, with the most common polymer detected by particle number per gram of moss being polyurethane (PU) (Figure 27). This was detected in 87% of samples, with a mean concentration of 1.7 particles of PU per gram moss. This polymer has very wide-ranging applications, from its use in clothing to its application as a coating and binder, from flexible foams used in construction, to insulation in home furnishings and appliances. Possible sources to the outdoor environment should be explored to know whether this polymer has diverse and numerous local sources. The other major polymers detected were cellulose acetate (CA), polyvinylchloride (PVC), ethylene vinyl acetate co-polymer (EVAc), polylactic acid (PLA), and polyethylene terephthalate (PBT). Interestingly it is PU, CA, PVC and EVAc which dominate the plastic fragments found >25 µm in size in moss, rather than the most commonly produced packaging plastics such as polyethylene (PE) and polypropylene (PP) which might be expected to contribute significantly to ambient microplastics transported through the air, arising from the fragmentation of exposed litter. The most commonly found microplastics found in the mosses were also different to those found in UK waters, for example, in the river Thames the most commonly found types of microplastic were polyvinyl chloride, polystyrene, polychloroprene, polyethylene chlorinated and polypropylene (Devereux et al., 2023). These microplastics found in water bodies may originate from plastic water bottles and other fragmented packaging materials.

Sampling sites covered a range of land cover class, species and a good distribution from across the United Kingdom representing more urbanised as well as more rural locations. On only two occasions were microplastics not quantified above the limits of detection, in Thetford and Warkworth. This demonstrates ubiquitous contamination of mosses with microplastics irrespective of their location across the United Kingdom, and is an indication that a diffuse atmospheric source may play a role in this widespread contamination of moss. In general, there was an increase in total microplastic abundance in mosses in the northwest of the UK compared to the southeast, with the highest concentrations per gram moss in locations including Ward Hill in Scotland and in rural northwest Wales (Figure 28a).

The diversity of polymers found in mosses was also not consistent across the UK (Figure 28b). There are tentative indications that the diversity of polymers is positively correlated with the total abundance of microplastics, with regression analysis of microplastic abundance and polymer diversity showing a slight positive correlation ($R^2 = 0.19$, slope 0.22). Again, this is due to the greater diversity of microplastic polymers in the northwest of the UK compared to the southeast.

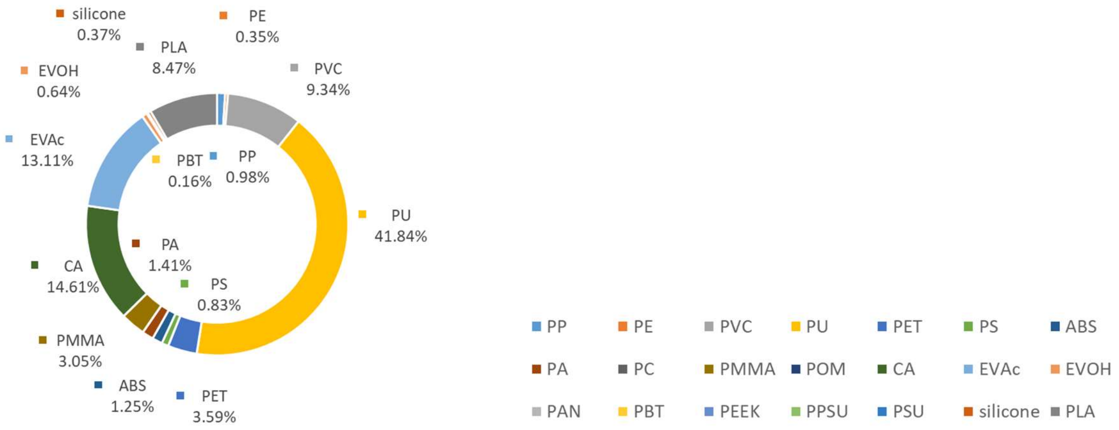

Figure 27: Relative abundance of different microplastics across all UK moss samples.

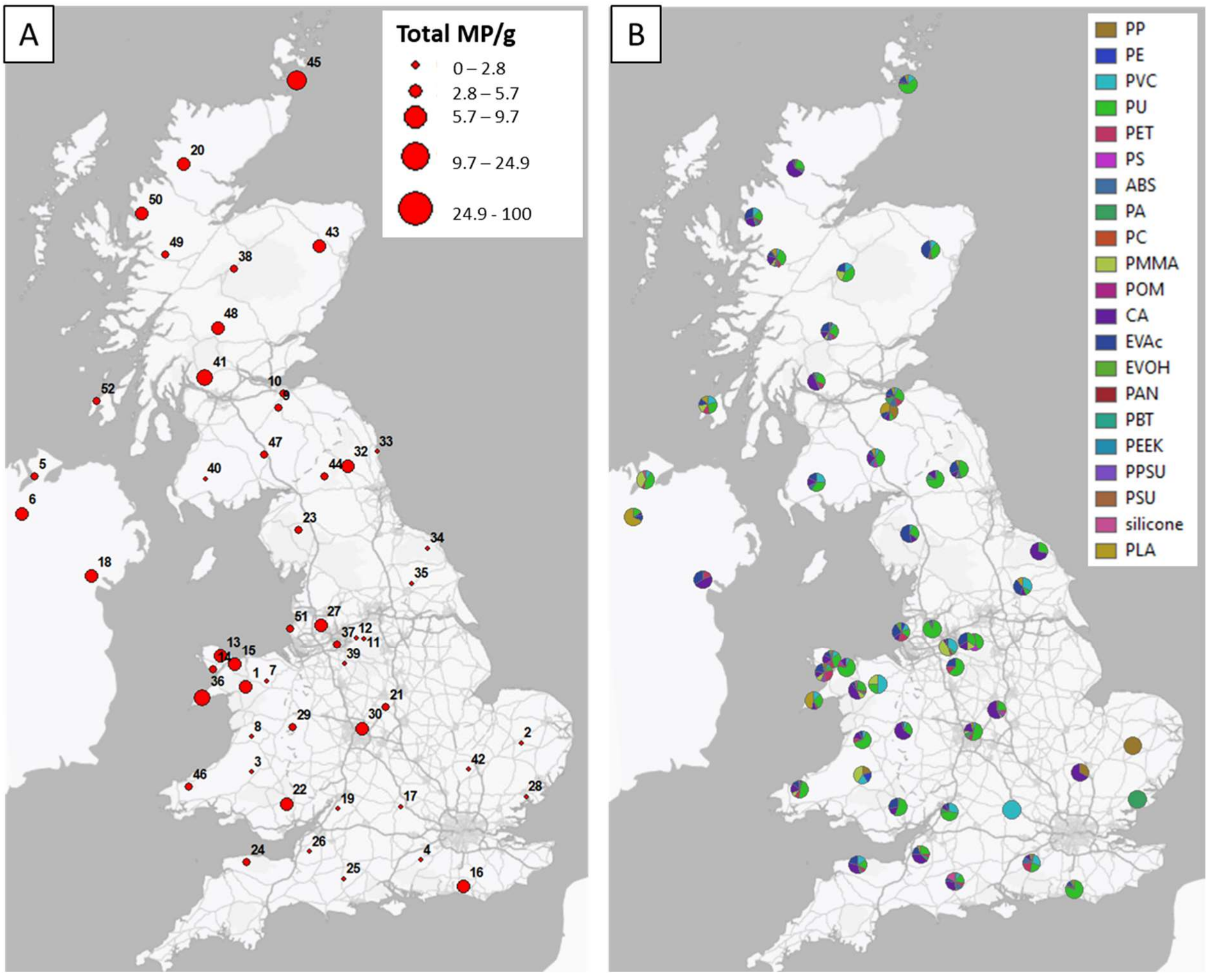

Figure 28: Microplastics in moss samples from the UK showing a) total microplastic (MP) abundance in mosses across the United Kingdom. Each red datapoint on the map represents moss from a single sampling location. The size of each data point indicates the total abundance of microplastics per gram of moss from 0 – 100 MP/g. b) Pie charts of the proportional contribution of different polymers to the overall microplastic contamination in moss at each sampling location. Further detail on numbers of microplastic particles and polymer diversity are given in the Annex.

4 Conclusions

This study has found that although concentrations of many metals in mosses have declined or stabilised since the last UK moss survey in 2005, there are possible increases in concentration in mosses for the metals cadmium, zinc and copper. These may be associated with vehicle use including lubricants and brake and tyre wear (copper and zinc), in addition to domestic wood burning (cadmium), although this is currently unconfirmed.

Microplastic content of moss samples was analysed from 52 sites across the United Kingdom. All except two sites monitored experienced some microplastic contamination above the limits of detection of the assessment. A diverse range of polymers were detected across the United Kingdom, with the highest concentrations and diversity concentrated in the more north westerly regions. The mean total number of microplastics >25 µm in size in moss across the UK was 4.5 MP/g with a maximum of 24.7 MP/g detected across the sampled locations. The most common polymer detected by particle number per gram of moss was polyurethane. This was detected in 87% of samples, with a mean concentration of 1.7 particles of PU per gram moss. The other major polymers detected were cellulose acetate, polyvinylchloride, ethylene vinyl acetate, polylactic acid, and polyethylene terephthalate. Common polymers associated with packaging and perhaps macroplastic litter such as polyethylene and polypropylene were less commonly detected above limits of detection in these samples.

The results from this study will be submitted to the 7th European Moss Survey, with the exception of a few locations that do not meet the rural criteria, but were useful for this UK specific analysis.

5 Further work

Many of the major sources for the metals analysed from moss tissue are associated with vehicles – particularly brake pads. However, as the focus of this study was long-range transport of air pollutants the samples were largely taken from comparatively remote areas. It is possible that concentrations would be significantly higher nearer to major roads and in urban areas.

As the decline in concentration of some metals is slowing in some cases, and may even be reversed for others, it is important to continue monitoring. This is particularly important as large changes in domestic fuel use and in vehicle fleet are forecast over the coming decades. In addition, the change in metal concentration within mosses may not match that seen by changes in emissions, particularly if there is resuspension, or if long-range transport of metals becomes a larger proportion of deposition to UK vegetation.

This study has shown widespread occurrence of microplastics in moss samples in rural areas, which are attributed to airborne deposition. These have been differentiated by polymer, but further work is needed to identify the sources of microplastics, and to model airborne dispersion from these sources.

6 Acknowledgements

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8 Annex

Table 1: Summary of key descriptors of the sampling sites and both the total microplastic contamination (MP/g) and the diversity of polymers in each moss sample (the total number of distinct polymers detected >LOD). Note the sample location number matches those of the maps in Figure 28.

Location	Date sampled	Land cover	Species	Total (MP/g)	Polymer diversity
1	16/03/2021	Moors & heathland	*Pleurozium schreberi*	6.52	8
2	02/03/2021	Mixed forest	*Pseudoscleropdium purum*	0.00	0
3	02/03/2021	Mixed forest	*Pleurozium schreberi*	0.37	2
4		Mixed forest	*Pseudoscleropdium purum*	1.76	5
5	23/03/2021	Moors & heathland	*Hypnum cupressiforme*	3.41	4
6	22/03/2021	Moors & heathland	*Hypnum sp*	8.25	4
7	01/04/2021	Moors & heathland	*Pleurozium schreberi*	1.20	3
8	06/04/2021	Moors & heathland	*Hylocomium splendens*	2.16	4
9	18/03/2021	Moors & heathland	*Hypnum jutlandicum*	3.76	5
10	27/03/2021	Grassland	*Rhytidiadelphus squarrosus*	3.07	8
11	31/03/2021	Moors & heathland	*Hylocomium splendens*	2.17	4
12	01/04/2021	Moors & heathland	*Pleurozium schreberi*	1.48	4
13	07/04/2021	Grassland	*Pleurozium schreberi*	5.91	8
14	09/04/2021	Grassland	*Pleurozium schreberi*	4.33	8
15	13/04/2021	Grassland	*Hylocomium splendens*	6.84	5
16	08/04/2021	Grassland	*Pseudoscleropdium purum*	6.40	5
17	14/04/2021	Grassland	*Pleurozium schreberi*	0.86	1
18	14/04/2021	Moors & heathland	*Pleurozium schreberi*	6.06	4
19	11/04/2021	Grassland	*Hylocomium splendens*	1.05	2
20	20/04/2021	Moors & heathland	*Hylocomium splendens*	5.71	5
21	18/04/2021	Grassland	*Pleurozium schreberi*	3.85	3
22	16/03/2021	Moors & heathland	*Pleurozium schreberi*	7.51	4
23	23/04/2021	Mixed forest	*Hylocomium splendens*	4.39	4
24	14/04/2021	Moors & heathland	*Pleurozium schreberi*	5.11	5
25	25/04/2021	Grassland	*Pseudoscleropdium purum*	2.13	6
26	27/04/2021	Grassland	*Hylocomium splendens*	2.03	4
27	06/05/2021	Moors & heathland	*Hypnum cupressiforme*	9.47	4
28	09/05/2021	Mixed forest	*Hypnum cupressiforme*	1.10	1
29	14/05/2021	Grassland	*Pleurozium schreberi*	3.76	4
30	16/05/2021	Grassland	*Rhytidiadelphus squarrosus*	9.42	7
31	-	Urban	*Pseudoscleropdium purum*	5.55	6
32	26/05/2021	Grassland	*Hylocomium splendens*	7.49	6
33	19/05/2021	Grassland	*Pleurozium schreberi*	0.00	0
34	30/05/2021	Moors & heathland	*Pleurozium schreberi*	1.29	2
35	29/05/2021	Grassland	*Pleurozium schreberi*	2.81	5
36	03/06/2021	Moors & heathland	*Hypnum cupressiforme*	24.89	5
37	07/06/2021	Urban	*Brachythecium spp*	3.25	5
38	14/06/2021	Moors & heathland	*Hylocomium splendens*	3.49	4
39	17/06/2021	Mixed forest	unidentified *pleurocarpous*	1.29	2

Location	Date sampled	Land cover	Species	Total (MP/g)	Polymer diversity
40	14/06/2021	Mixed forest	*Pleurozium schreberi*	1.61	4
41	19/06/2021	Grassland	*Pleurozium schreberi*	14.79	7
42	25/06/2021	Grassland	unidentified *pleurocarpous*	0.71	1
43	08/07/2021	Moors & heathland	*Pleurozium schreberi*	9.53	6
44	-	Moors & heathland	*Pleurozium schreberi*	5.30	4
45	21/07/2021	Moors & heathland	*Hylocomium splendens*	7.40	5
46	11/08/2021	Grassland	*Pleurozium schreberi*	4.75	5
47	30/08/2021	Grassland	unidentified *pleurocarpous*	2.83	6
48	17/08/2021	Moors & heathland	*Hylocomium splendens*	5.87	8
49	21/07/2021	Moors & heathland	*Hylocomium splendens*	5.26	8
50	31/08/2021	Grassland	*Hylocomium splendens*	7.24	7
51	04/11/2021	Grassland	*Pseudoscleropdium purum*	3.39	7
52	06/01/2022	Moors & heathland	unidentified pleurocarpous	4.47	6

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