



Concentrations and geographic distribution of selected organic pollutants in Scottish surface soils



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ABSTRACT

Concentrations of selected persistent organic pollutants (POPs) representing three chemical classes (polycyclic aromatic hydrocarbons (PAH), polybrominated diphenyl ethers (PBDE) and polychlorinated biphenyls (PCB) and the organic pollutant diethylhexyl phthalate (DEHP), were determined in surface soil samples (0–5 cm) collected at 20 km grid intersects throughout Scotland over a three-year period. Detectable amounts of all chemical classes and most individual congeners were present in all samples. There were no consistent effects of soil or vegetation type, soil carbon content, pH, altitude or distance from centres of population on concentrations which exhibited extreme variation, even in adjacent samples. It is concluded that soil POPs and DEHP concentrations and associated rates of animal and human exposure were highly variable, influenced by multiple, interacting factors, and not clearly related to local sources but possibly related to wet atmospheric deposition and the organic carbon content of the soil.

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1. Introduction

The nature of anthropogenic pollution in Northern Europe has changed in recent decades. The deleterious effects of atmospheric pollution and the release of raw sewage or poorly treated effluent into water courses and coastal waters has, in general, been drastically reduced. This is a direct result of the introduction and enforcement of stricter European Union and International Legislation such as the Water Framework Directive, the Urban Wastewater Directive, the Air Quality Directive and the Stockholm Convention on Persistent Organic Pollutants amongst others. Although point source inputs in many areas have been minimised, there is now an increasing interest in the ecological effects of diffuse pollution, where pollutants, often at relatively low concentrations, impact the environment as a result of low impact point sources or at remote locations distant to known point sources. A wide range of synthetic chemicals, many of which are highly persistent, continue to be released into the environment. Some of these compounds have the

potential to cause a range of biological effects, including endocrine disruption and carcinogenesis. Many of them are non-polar and have a high affinity to organic matter and enter the terrestrial environment as a result of anthropogenic activities such as the burning of fossil fuels, managed rotational burning of vegetation to promote productive regeneration of heather moorland habitats utilised by grouse and sheep, manufacturing processes, waste disposal and the recycling of sewage sludge to land as a fertiliser. Some compounds, such as the Polycyclic Aromatic Hydrocarbons (PAHs), are present to some extent additionally as a result of natural processes such as wildfires and biogenesis (Wilcke et al., 1999; Thiele and Brümmer, 2002; Atanassova and Brümmer, 2004).

This study reports the concentrations of Polycyclic Aromatic Hydrocarbons (PAHs), Polybrominated Diphenyl Ethers (PBDEs), Polychlorinated Biphenyls (PCBs) and the plasticiser diethylhexyl phthalate (DEHP) in Scottish soils. For the purposes of this study PAHs, PCBs and PBDEs will be referred to collectively as Persistent Organic Pollutants (POPs). These organic pollutants are generally persistent in the environment and classified as semi-volatile organic compounds (SVOCs). Such pollutants have previously been shown to have an impact on environmental quality close to their place of manufacture, use or disposal, or additionally, distant from point sources via redistribution processes such as long-range atmospheric transport (LRAT).

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¹ Dr. Stewart Rhind was tragically killed in a car accident in March 2013. His co-authors would like to dedicate this paper to him as a highly regarded colleague and friend.

LRAT of POPs and DEHP can occur either in the gaseous phase or via binding of the pollutant to atmospheric particulates, depending upon the physicochemical properties of the individual pollutants and the prevailing atmospheric conditions (Xie et al., 2005; Teil et al., 2006; Wang et al., 2008; Zeng et al., 2009; Fu and Kawamura, 2010; Salapasidou et al., 2011). Direct release into the atmosphere and subsequent wet or dry deposition is thought to be one of the most important means of entry of such compounds into the terrestrial environment (Cai et al., 2008). Once deposited, the strong sorption of these compounds to soil organic matter and their poor water solubility results in poor mobility and bioavailability. In previous studies DEHP, PBDEs, PCBs and PAH concentrations in urban air and soil samples have generally been found at higher concentrations than in samples from rural or remote sites (Wang et al., 2008; Rudel and Perovich, 2009), however, as a result of atmospheric transport and deposition, many of these compounds are now ubiquitous in the environment and can be found, often at relatively low concentrations, in remote locations.

A number of studies have indicated that atmospheric deposition is positively correlated with rainfall and negatively correlated to temperature. In colder conditions, such as at higher altitudes or latitudes, partitioning to aerosols/particulate phase is enhanced, resulting in greater particle scavenging and higher washout to terrestrial surfaces during precipitation (Thuren and Larsson, 1990; Teil et al., 2006; Callén et al., 2008; Wang et al., 2011). In the soil, compounds can exist either, to a small extent, in soil pore water or adsorbed or partitioned onto soil organic matter. In geographic areas where levels of precipitation (rain and snow) are relatively high and the soils have high levels of organic carbon environmentally stable pollutants will persist and accumulate. The concentrations of POPs and DEHP retained on soils may therefore be related to both input concentrations, the overlying vegetation and to the organic carbon content of the soil which may be regarded as an index of the ability of a soil to retain a non-polar pollutant once it has been deposited. The west coast and higher areas of Scotland have the highest levels of precipitation in the United Kingdom with a twenty year (1981–2010) average rainfall of >3000 mm and a prevalence of organic rich soils compared to the east coast of Scotland which has an average rainfall of 800–1000 mm. Detailed climate maps showing air and ground temperature, rainfall and snow data are available from the UK Meteorological (Met) Office (Met Office, 2013).

Exposure to bioavailable POPs and DEHP is known to perturb a wide range of physiological systems, including the reproductive (Colborn et al., 1993; IEH, 1999), immune (Vine et al., 2000) and thyroid systems (Hansen, 1998; Langer et al., 1998; Meeker et al., 2007). Amongst other effects, some pose a risk of carcinogenicity, particularly the higher molecular weight PAHs such as benzo[a]pyrene (Xue and Warshawsky, 2005). PCBs, PBDEs, PAHs and DEHP are known to act as endocrine disrupting compounds (EDCs). EDCs are exogenous substances that alter the function(s) of the endocrine system causing adverse health effects in an organism, or its progeny (IPCS/WHO, 2002; Kortenkamp et al., 2011).

Such links between exposure and effect may be apparent only through epidemiological studies in which the incidences of certain conditions or diseases are linked statistically. Studies of this type suggest that low level exposures to EDCs may underlie long-term trends such as declining sperm count in humans and increased incidences of breast and testicular cancer (Toppari et al., 1996; Swan et al., 2000; Gray et al., 2009). However, the relationship between exposure and effect is highly complex and poorly understood because organisms are generally exposed to multiple compounds, some of which may be metabolised to form toxic or non-toxic products and which may interact at individual very low, apparently harmless, concentrations to exert a range of effects (Rajapakse

et al., 2002; Crofton et al., 2005; Kortenkamp, 2007; Kortenkamp et al., 2011). Assessment of effects is further complicated by the fact that many of them, such as perturbed gene or protein expression (Ghanem et al., 2006; Fowler et al., 2008), are subtle and not easily observed. Such effects of individual and multiple compounds can be investigated in laboratory-based studies and linked to epidemiological observations but the value of all of these studies remains limited if they are not related to real world levels and patterns of exposure to relevant compounds. This current study aims to provide information on background concentrations of POPs and DEHP to which relevant organisms may be exposed.

There are a few studies reporting gross geographic variations in soil levels of some POPs on a continental or global scale (Meijer et al., 2003; Hassanin et al., 2004). Other surveys have focussed on single soil types (Dreyer et al., 2005; Maliszewska-Kordybach et al., 2008), single classes of POPs (Creaser et al., 1989; Bracewell et al., 1993; Dreyer et al., 2005; Maliszewska-Kordybach et al., 2008) or on heavily polluted soils (Cai et al., 2008) but little is known of the pattern of variability in concentrations on a national scale and how they are influenced by multiple factors, including soil and vegetation types, proximity to sources and chemical class. The aim of this study was to characterise soil concentrations of selected POPs and DEHP as an index of environmental pollutant concentrations and to relate these measurements to factors that might affect patterns of distribution, including soil and vegetation type, altitude and proximity to cities and associated production of pollutants, or to the sea, and chemical class. Knowledge of these profiles will contribute to the understanding of patterns of POPs and DEHP exposure in humans and animals and associated potential risk.

A further objective was to determine the validity of surface soil pollutant concentrations as an index of exposure in view of the fact that bio-accumulation of some chemicals from soils by grazing animals may depend, in part, on exposure through inhaled air (Withey and Karpinski, 1985) as well as through ingestion of soil during feeding (Norstrom, 2002).

2. Material and methods

2.1. Sample collection

Representative soil samples (0–5 cm depth) were collected at 20 km grid intersects throughout Scotland in stainless steel rings (7.5 cm diameter × 5 cm depth). This component of the soil was considered likely to be exposed to the greatest degree of contamination resulting from atmospheric deposition of POPs and DEHP. It also represented the soil component to which most animals might be exposed, either because they live either above ground, where they may ingest soil or inhale re-volatilised POPs and DEHP (e.g. grazing animals), or because they live within the soil where they may be exposed through ingestion, inhalation or direct contact (e.g. soil invertebrates). The sites varied considerably in distance from the nearest metalled road. In the more agricultural and populated areas of Scotland, some sites were within 50 m of a public road whereas in the more remote parts of North Scotland, they were up to 10 km from roads that also have very low traffic usage.

Samples were collected from different regions of Scotland, during spring and early summer, over a three year period (year 1: Central and East; year 2: South and West; year 3: North and West) All samples were wrapped in dichloromethane (DCM) washed aluminium foil and stored at –20 °C. The samples were freeze dried and sieved (2 mm) to remove stones and large particles, and stored in glass jars before analysis.

2.2. Sample classification and characterisation

The sample sites were categorised according to soil type (brown earth; gley; peat; podzol; alluvial soil; ranker; Robertson (1984)) and dominant vegetation type (arable; unimproved grassland; improved grassland; dry and moist heath; mire and wet heath; conifer plantation) in the area around each sampling point, pH, carbon content (%), altitude above sea level (m), and distance from the centre of the nearest city of >250,000 people (<40 km; 40–80 km; >80 km).

Soil total organic carbon content was determined for each sample using an automated Duma combustion procedure (Pella and Colombo, 1973) using a flash EA 1112 Elemental Analyser (Thermo Finnigan, Italy) and soil pH was determined using the method of McLean (1982).

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