



Long-term mercury dynamics in UK soils

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ABSTRACT

A model assuming first-order losses by evasion and leaching was used to evaluate Hg dynamics in UK soils since 1850. Temporal deposition patterns of Hg were constructed from literature information. Inverse modelling indicated that 30% of 898 rural sites receive Hg only from the global circulation, while in 51% of cases local deposition exceeds global. Average estimated deposition is $16 \mu\text{g Hg m}^{-2} \text{a}^{-1}$ to rural soils, $19 \mu\text{g Hg m}^{-2} \text{a}^{-1}$ to rural and non-rural soils combined. UK soils currently hold 2490 tonnes of reactive Hg, of which 2140 tonnes are due to anthropogenic deposition, mostly local in origin. Topsoil currently releases 5.1 tonnes of Hg^0 per annum to the atmosphere, about 50% more than the anthropogenic flux. Sorptive retention of Hg in the lower soil exerts a strong control on surface water Hg concentrations. Following decreases in inputs, soil Hg concentrations are predicted to decline over hundreds of years.

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1. Introduction

Soil mercury accounts for 75% of the biogeochemically-active element (Mason and Sheu, 2002) and at sufficiently high levels can exert toxic effects towards microbes, invertebrates and plants (Rundgren et al., 1992; Tipping et al., 2010a). The soil is both a sink and a source for atmospheric mercury, accumulating the metal by wet and dry deposition, and releasing Hg^0 by evasion (Grigal, 2002). The latter is an important process in controlling atmospheric levels, and the evolution of soil Hg levels as inputs change over time (Pirrone et al., 2008).

A significant issue at national or regional scales is the source of deposited Hg (Pai et al., 1999). Globally, with the pollution of remote locations in mind, long-range transport of Hg^0 in the atmosphere before oxidation and deposition is the main focus, but at smaller scales local emissions and their speciation are important. Anthropogenic emissions are roughly equally divided between Hg^0 , which can directly join the global circulation, and other forms of Hg (reactive gaseous mercury, RGM, and particulate mercury Hg_p) which deposit closer to the source (Mason et al., 1994). The importance of short-distance transport and deposition is evident from the spatial distribution of soil Hg pools. Thus, Nater and Grigal (1992) reported systematic declines in soil Hg with distance from industry in the Great Lakes states. Observations for the UK from

national scale monitoring programmes show substantial spatially auto-correlated variability in soil Hg contents (Tipping et al., 2011), apparently dependent upon proximity to urban and industrial areas.

Here we address the dynamics of soil Hg, in an attempt to account for the observed distribution of UK soil pools. Our approach was to attribute these to past deposition by accounting for the key loss processes of leaching (Hg bound to DOM) and evasion. Two sources of Hg deposition were distinguished, the global atmospheric pool of the metal, and local anthropogenic emissions of RGM and Hg_p . At each soil site, the amount of Hg required to be deposited over time to achieve observed Hg pools in topsoils was calculated, using plausible historical variations in Hg inputs. Model outputs were compared with measured Hg deposition, accumulation in deeper soil, and surface water concentrations, and with model predictions of deposition to the UK. The model was used to estimate future soil Hg dynamics under different scenarios.

The findings were expected to provide information about UK Hg in soils, its likely turnover rates and its contribution to Hg^0 in the global circulation by re-emission. This would help in assessing likely toxic effects and the suitability of emission controls, especially with respect to the Critical Loads approach (UBA, 2004).

2. The atmospheric deposition of mercury

Knowing how much Hg is being deposited and the history of deposition are key to understanding how Hg in soil has developed.

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Here we review information relevant to the UK, and propose historical deposition patterns.

2.1. Forms of atmospheric mercury

Mercury has a complex atmospheric chemistry, unlike other metals that are transported as aerosols (Schroeder and Munthe, 1998). Three principal forms of atmospheric Hg are distinguished; elemental mercury Hg^0 , reactive gaseous mercury (RGM) which comprises HgCl_2 and HgBr_2 and possibly organic forms, and particulate Hg (Hg_p). The term total gaseous mercury (TGM) refers to the sum of gaseous elemental Hg^0 (GEM) and RGM. Inputs to the global circulation of Hg^0 come in about equal parts from natural sources (volcanoes, geothermally active areas, the mineral matter of uncontaminated soils; Gustin et al., 2008), direct anthropogenic inputs, and re-emission of previously-deposited Hg (Mason et al., 1994; Schroeder and Munthe, 1998). Because it is relatively unreactive, Hg^0 has an atmospheric residence time of the order of one year and therefore it can spread globally. It is lost from the atmosphere after conversion to RGM or Hg_p both of which are deposited rapidly as wet or dry deposition. Details of atmospheric oxidation processes are still actively researched (Hynes et al., 2009). The RGM and Hg_p forms can also enter the atmosphere directly, as anthropogenic emissions. Pacyna and Pacyna (2002) estimated that 53% of total global emissions are as Hg^0 , and data quoted by Travnikov and Ilyin (2005) indicate a similar value for European countries, with a value of 52% for the UK. In 2005, UK emissions comprised 53% Hg^0 (Dore et al., 2007).

2.2. Contemporary Hg deposition to the UK

Rowland et al. (2010a) reported a volume-weighted mean concentration of 3.3 ng Hg l^{-1} in wet deposition to 10 rural UK sites over the period 2005–2009. An overall mean deposition of $2.8 \mu\text{g m}^{-2} \text{ a}^{-1}$ was derived, with ranges of $2.5\text{--}3.5 \mu\text{g m}^{-2} \text{ a}^{-1}$ among years and $2.1\text{--}4.3 \mu\text{g m}^{-2} \text{ a}^{-1}$ among sites. The authors acknowledged that these concentrations and deposition rates are surprisingly low, offering as explanations the choice of remote sites without local Hg sources, a possible decline in global emissions of Hg, and analytical or technical factors. Due to the lack of monitoring of RGM, this UK rural network did not permit estimates of Hg dry deposition. Miller et al. (2005) estimated wet deposition fluxes of $3\text{--}7 \mu\text{g m}^{-2} \text{ a}^{-1}$ to rural sites in the north-east of North America, and suggested that an equal amount might come from dry deposition. A higher average concentration of 23 ng Hg l^{-1} in bulk deposition was reported by Yang et al. (2002) for a single remote rural site in Scotland, for one year (1997–1998), with a deposition flux of $35.9 \mu\text{g m}^{-2} \text{ a}^{-1}$. Yang et al. (2009) reported annual average concentrations for a site in central London to fall from 76.0 ng l^{-1} in 1999 to 43.8 ng l^{-1} in 2005 (fluxes $45.3\text{--}15.0 \mu\text{g m}^{-2} \text{ a}^{-1}$).

Lee et al. (2001) conducted a modelling study using reported anthropogenic emissions from UK and other European countries, combined with knowledge and assumptions about long-distance transport, and the atmospheric generation of RGM. They estimated wet deposition of Hg to be 9.9 tonnes for 1998, which corresponds to an average deposition rate of $41 \mu\text{g m}^{-2} \text{ a}^{-1}$ to the UK land surface, but this would include all sites, not just rural remote ones. Ilyin et al. (2008) modelled deposition to Europe for 2006, based on inputs from both anthropogenic and “natural” emissions, and reported an average rate for the UK of $14 \mu\text{g Hg m}^{-2} \text{ a}^{-1}$.

2.3. Past deposition

The amount of circulating Hg in the atmosphere has increased by about a factor of three over the last 100–200 years (Mason et al.,

1994), consistent with sediment records of lakes and peat bogs in remote locations (Swain et al., 1992; Biester et al., 2007). However, larger increases are shown by the sediments of lakes closer to emission sources (Landers et al., 1998; Lockhart et al., 2000). For four remote undisturbed UK lakes, Yang and Rose (2003) found that ratios of present to past deposition rates were between two and four, but there were substantial variations in the estimated pre-industrial fluxes of Hg. In two cases the inputs were 4.5 and $6 \mu\text{g Hg m}^{-2} \text{ a}^{-1}$, while in the other two they were much greater, with values of 22 and $40 \mu\text{g Hg m}^{-2} \text{ a}^{-1}$. Farmer et al. (2009) studied peat cores from different parts of Scotland and reported deposition rates of $\sim 4 \mu\text{g Hg m}^{-2} \text{ a}^{-1}$ in the pristine past, rising to maximum rates of $50\text{--}200 \mu\text{g Hg m}^{-2} \text{ a}^{-1}$ in the 20th century. The estimates of historical deposition for the UK are clearly not fully consistent, but several of the sites give pre-industrial rates reasonably in accord with the general historical rate of $3\text{--}3.5 \mu\text{g Hg m}^{-2} \text{ a}^{-1}$ given by Biester et al. (2007), while rates estimated for the 20th century are higher.

2.4. Historical Hg deposition scenarios for modelling UK soils

Based on the above information, we propose a simple pattern of annual Hg deposition to the UK land surface over time (Fig. 1). Mercury deposition is considered to come from “global” and “local” sources. From modelling with STOCHEM (Yang et al., 2010) we assume global deposition to be dominated by wet deposition, characterised by a single average concentration of Hg, so the global flux at any point in the UK is proportional to annual rainfall. Present-day values are assumed to be three times those in the pristine past. We assumed that the increase in rainfall Hg concentration began in 1850 and was linear until 1950, after which it remained constant at 5 ng l^{-1} .

The main local UK sources are given by UK National Atmospheric Emissions Inventory (NAEI: <http://www.naei.org.uk>) as combustion (energy and transformation), iron and steel production and waste, although considerable amounts of mercury may have been added to agricultural land with fertilisers, lime, and manure (Fergusson, 1990). Our historical trend was constructed by assuming no locally-sourced deposition before 1850, then a linear rise to 1960–1970, then a fall to the present-day value. The ratio of the 1960–1970 value to that in 2000 is set to 60:7, based on the tonnage of UK anthropogenic emissions for 1970 and 2000 published by NAEI, and the approximately linear decline between those years. The same pattern is assumed to apply to non-UK local sources.

The same global and local patterns are assumed for all sites. As already stated, spatial variation in the global inputs is assumed to be due only to variations in rainfall. Local inputs are considered to

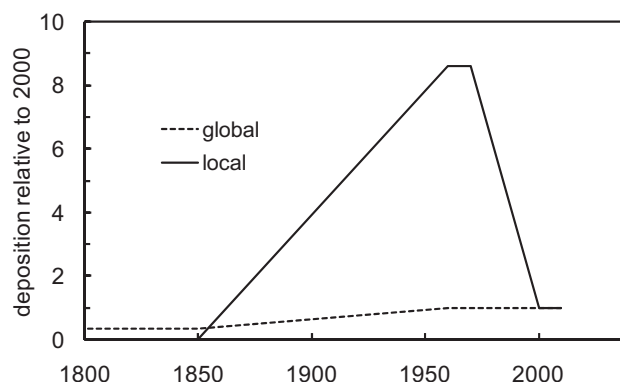


Fig. 1. Assumed patterns of deposition, relative to a contemporary value of unity.

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