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Review article

Mercury speciation, transformation, and transportation in soils, atmospheric flux, and implications for risk management: A critical review

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ABSTRACT

Mercury (Hg) is a potentially harmful trace element in the environment and one of the World Health Organization's foremost chemicals of concern. The threat posed by Hg contaminated soils to humans is pervasive, with an estimated 86 Gg of anthropogenic Hg pollution accumulated in surface soils worldwide. This review critically examines both recent advances and remaining knowledge gaps with respect to cycling of mercury in the soil environment, to aid the assessment and management of risks caused by Hg contamination. Included in this review are factors affecting Hg release from soil to the atmosphere, including how rainfall events drive gaseous elemental mercury (GEM) flux from soils of low Hg content, and how ambient conditions such as atmospheric O_3 concentration play a significant role. Mercury contaminated soils constitute complex systems where many interdependent factors, including the amount and composition of soil organic matter and clays, oxidized minerals (e.g. Fe oxides), reduced elements (e.g. S^{2-}), as well as soil pH and redox conditions affect Hg forms and transformation. Speciation influences the extent and rate of Hg subsurface transportation, which has often been assumed insignificant. Nano-sized Hg particles as well as soluble Hg complexes play important roles in soil Hg mobility, availability, and methylation. Finally, implications for human health and suggested research directions are put forward, where there is significant potential to improve remedial actions by accounting for Hg speciation and transportation factors.

1. Introduction

Mercury (Hg) contaminated soils pose a risk to global public health, with Hg being listed as one of the 'ten leading chemicals of concern' (WHO, 2017). In 2013, the United Nations (UN) introduced the 'Minamata Convention on Mercury', which aims for a more global effort in managing the risk of Hg to human health and the environment. Signed by 128 countries (UNEP, 2016), it entered into effect in 2017 (Selin et al., 2018). This concerted action, if successful, will have huge implications for public health for decades to come; however, there are many hurdles in the way to achieving this goal.

The global amount of Hg mass accumulated in soils is very large; assumed to be in the range of 250–1000 Gg (Obrist et al., 2018). Although Hg occurs naturally in soils from geologic sources (Obrist et al., 2018), or through natural events such as forest fires and volcanic eruptions (Ermolin et al., 2018), a significant proportion of that Hg is attributed to anthropogenic influences, with an estimated 86 Gg of anthropogenic Hg emissions now accumulated in surface soils (UNEP, 2009). The Hg contamination issue is exemplified by the Elbe floodplain, Germany, where it has been estimated that as much as 2 Gg of Hg may have accumulated in this region alone (Wallschläger, 1996). Anthropogenic mercury pollution has been brought about by the

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Table 1
Properties of specific Hg compounds (Armstrong, 1990; Beckers and Rinklebe, 2017; Chem-Supply, 2018; Gaffney and Marley, 2014; NCBI, 2018; NRC, 2000; Pohanish, 2017; Toxnet, 2019; US EPA, 1997; Wang et al., 2012; Zheng et al., 2012).

Species	Name	Solubility in water (g/L)	Henry's law constant (atm-m^3/mol)	Vapor pressure (Pa)	Density (g/cm^3)
Hg^0	Elemental mercury	5.6×10^{-5} to 6.1×10^{-5} (25 °C) 49.6×10^{-6} (20 °C)	7.1×10^{-3} to 8.7×10^{-3} (25 °C) 7.3×10^{-3} (20 °C)	0.27 (25 °C) 0.16 (20 °C)	13.53 (25 °C)
$HgCl_2$	Mercuric chloride	28.6 to 73.3 (25 °C) 66 to 69 (20 °C)	7.1×10^{-10} (25 °C) 3.6×10^{-10} (20 °C)	9.00×10^{-3} (20 °C)	5.43 (25 °C)
$HgSO_4$	Mercuric sulfate	Decomposes to mercury oxide sulfate and sulfuric acid	–	–	6.47 (25 °C)
Hg_3O_6S (isomer)	Mercury oxide sulfate	0.03 (16 °C)	–	–	6.44
HgS	Mercury sulfide	2×10^{-24} (25 °C)	–	–	8.1 (25 °C)
HgO	Mercuric oxide	0.043 to 0.053 (25 °C)	7.1×10^{-7} (25 °C)	9.20×10^{-12} (25 °C)	11.14 (25 °C)
$HgBr_2$	Mercuric bromide	6.12 to 6.77(25 °C)	–	–	6.03 (25 °C)
HgI_2	Mercuric iodide	0.06 (25 °C)	–	–	6.36 (25 °C)
$Hg(NO_3)2$	Mercuric nitrate	20	–	–	4.30 (25 °C)
Hg(CN)2	Mercuric cyanide	93 (14 °C)	–	–	4.00 (25 °C)
CH_3HgCl	Methyl mercuric chloride	< 0.10 (21 °C)	4.7×10^{-7} (25 °C) 3.8×10^{-7} (15 °C)	1.10 to 1.76 (25 °C)	4.06 (20 °C)
$(CH_3)_2Hg$	Dimethyl mercury	Slightly soluble. Most sources state “negligible”, NRC (2000) reports 1 g/L (21 °C)	6.4×10^{-3} to 7.6×10^{-3} (25 °C)	8.31×10^3 (25 °C) 7.80×10^3 (23.7 °C)	3.19 (20 °C)
CH_3HgOH	Methyl mercuric hydroxide	1 to 10 (21 °C)	–	0.90	–

chloralkali process, cement production, mining and smelting, artisanal small-scale gold mining, coal burning, and oil refining, which together emit huge quantities of Hg to the environment (Science Communication Unit, 2013), on the order of 2 Gg per year (UNEP, 2009).

Mercury can exist in soils as a number of different states and species, each possessing different properties (Table 1), adding further complexity to the assessment of contaminated sites. Mercury has three oxidation states: (1) Hg(0) (elemental mercury), (2) Hg(I) (mercurous), or (3) Hg(II) (mercuric). Of these, the mercurous form is not stable under typical environmental conditions and, therefore, is rarely observed. Because of high volatility and susceptibility to oxidation, metallic Hg^0 is typically only present in the atmosphere. It can, however, be found as a free phase in soils heavily polluted by anthropogenic activities (i.e. spillages) involving this form of Hg. Deposition of atmospheric Hg^0 to soils occurs over large spatial and temporal scales (Lin et al., 2010), whereas atmospheric Hg^{2+} is nearly always quickly disposed to the pedosphere by wet or dry deposition. Once introduced in soils, Hg in the oxidized mercuric Hg^{2+} state readily forms inorganic mercuric salts and minerals, such as $HgCl_2$, HgO, or HgS, or, under appropriate conditions, organo-Hg compounds. The mercuric compounds are unlike most other metallic compounds in that they tend to involve in covalent bonding, rather than ionic bonds. Organo-Hg compounds are defined by their C—Hg covalent bonding. It is the existence of this bond that differentiates organo-Hg compounds from inorganic Hg compounds that are bound to organic matter (US EPA, 1997). The various Hg species found in soils have distinct properties, which affect their fate and transport in soils (Table 1).

Risk assessment and long-term management of different Hg species is imperative for successfully managing risk and, therefore, successful implementation of the Minamata Convention. Recent notable reviews on Hg in the environment have addressed the atmosphere and atmospheric interfaces (Ariya et al., 2015), and Hg cycling in the various environmental spheres (Beckers and Rinklebe, 2017). The objective of the present review was to garner a better understanding of how Hg behaves in soils, so that we may better identify, assess, and manage/mitigate Hg risks at contaminated sites. Much research has been published on the topic of Hg in recent years, and particularly on its behavior in soils (Fig. 1). This review aimed to further our understanding of Hg behavior by considering pertinent studies that have focused on the following areas: (1) advances in Hg analysis and its various species in soils, (2) the exchange of Hg between soil and the atmosphere, (3) how Hg in its various forms interacts with the constituents of soil under different environmental conditions (e.g. redox, pH, and in the presence of other metals), (4) the transformation of Hg species in soil, and (5) the transportation potential of Hg in soil. Based on the review's findings, current needs for reliable environmental Hg risk assessment including implications for authorities and future challenges are discussed. This is of great interest for a wide international audience, including environmental scientists and managers, applied ecologists, environmental and technical engineers, and regulatory authorities.

2. Analysis of Hg in soils

2.1. Quantification of soil Hg

Mercury concentrations in soil are usually quantified in the laboratory from soil extracts involving concentrated acids (Sparks et al., 1996). Detection of soil Hg concentration is usually performed in the laboratory because handheld portable devices are not yet sufficiently sensitive to detect the environmentally relevant Hg concentrations of heterogeneous soils in the field. Portable X-ray Fluorescence deceives can only detect Hg levels above 20 mg/kg (Miller et al., 2013). Recently, a handheld XRF method was described that achieved a detection limit of 7.4 mg/kg (Brent et al., 2017). Such handheld XRF methods may be sufficient for screening soils at generic assessment criteria levels, but remain incapable of providing the low-level detection necessary for detailed quantitative risk assessment.

Typically, laboratory soil sample preparation methods before metal extraction will include homogenization, drying at < 60 °C, crushing, sieving, grinding, and mixing as necessary to homogenize the sample. While such methods reduce analytical variability, precautions should be taken in the case of Hg analysis so as to prevent the loss of volatile species such as Hg^0, or to cause irreversible chemical change such as precipitation of insoluble species or change in valence state (US EPA, 2007a). Adequate sample preparation, for example by following established protocols such as US EPA Method 3051 (US EPA, 2007a), will generally reduce losses during sample preparation to acceptable levels.

For the extraction of total-Hg, soil samples are typically digested with aqua regia (HCl/HNO_3) using microwave assisted digestion, because of its ability to dissolve cinnabar (HgS). Soil samples containing large amounts of organic material may be digested in HNO_3/H_2SO_4 and then diluted with BrCl solution to destroy any remaining organic material (US EPA, 2002). The total-Hg in the extract is then commonly detected using cold vapor atomic florescence spectrometry (CV-AFS)

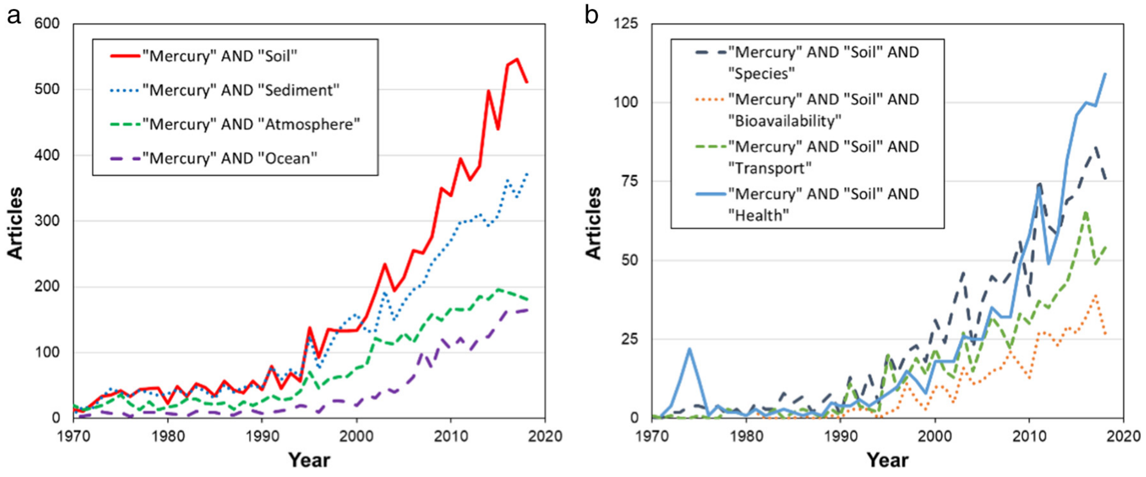

Fig. 1. Articles by year returned from a search of titles, abstracts or keywords in Scopus (a) mercury in different environmental media, and (b) different topics for mercury in soil (searched on 13 February 2019).

(US EPA, 2002) or inductively coupled plasma - mass spectrometry (ICP-MS) (Frentiu et al., 2013; US EPA, 1994). However, traditional laboratory techniques are time consuming and expensive. Table 2 lists some alternative laboratory techniques involving reduced cost or labor that have recently been reported in the literature. For instance, an extraction method for CV-ASS analysis proposed by Almeida et al. (2016) is reportedly much quicker than other described procedures, with the additional advantage of not requiring expensive reagents for Hg extraction (Almeida et al., 2016). The CV-μCCP-OES procedure has been put forward by Frentiu et al. (2013) for its improved analytical performance, simplicity, and reliance on low cost instrumentation. CV-ICP-OES has also been noted for offering several advantages, such as reduced analysis interference, rapid turnover of samples, low detection limits, and a wide calibration range. These features have made this technique an appealing alternative for the determination of total-Hg in soil samples (Hellings et al., 2013).

For methyl-Hg analysis, extraction of soil samples is more complicated. For example, extraction may involve digestion with acidic KBr and $CuSO_4$ solution to extract organo-mercury from inorganic complexes, followed by dichloromethane (DCM) to extract methyl-Hg, and then back-extraction into aqueous solution by Ar purging. The aqueous solutions are ethylated in a purge vessel using sodium tetraethylborate ($NaB(Et)_4$) to convert methyl-Hg to the volatile methylethyl-Hg species. In this method, CH_3Hg and total recoverable CH_3Hg are synonymous (US EPA, 1998). The organo-Hg content can then be detected by using various techniques. An inter-laboratory study was conducted by Kodamatani et al. (2017) to compare detection by three separate analytical methods, which were: (1) high-performance liquid chromatography-chemiluminescence (HPLC-CL), (2) gas chromatography-electron capture (GC-ECD); and (3) gas chromatography-atomic fluorescence spectrometry (GC-AFS); of these, the third is the most commonly utilized approach in analytical laboratories. It was revealed that, in general, all three methods returned comparable Methyl-Hg results for all soil and sediment samples tested. However, it was noted that the HPLC-CL method performed poorly in analyzing samples with high sulfur contents (Kodamatani et al., 2017). Solid phase micro extraction (SPME) has also been successfully coupled with GC atomic emission detection (GC-AED) for the determination of organo-Hg once in the aqueous phase (Mothes and Wennrich, 2000).

2.2. *Determination of Hg species*

Methods exploited to identify different Hg species and fractions in soils include (1) sequential extraction, (2) thermo-desorption, and (3) X-ray absorption techniques.

2.2.1. *Sequential extraction*

Sequential extraction is an operationally defined and indirect approach in which the soil is successively extracted using different reagents, thereby releasing Hg fractions of different reactivity. Many multiple step sequential extraction procedures have been developed, some specifically for the characterization of Hg species. Table 3 lists some of these approaches, and the rationale presented for each method. Because of lack of specificity, researchers will often classify sequential extraction fractions based on relative environmental mobility, or availability (Frohne and Rinklebe, 2013). Reis et al. (2016) classified the mobility of Hg in different sequential extraction fractions as either high, medium or low. In general, highly mobile Hg involves water-soluble Hg species, such as dissolved Hg or soluble inorganic or organic complexes (Section 6.1), or exchangeable species that are only weakly sorbed to soil surfaces (Section 4.1). Medium mobility Hg entails acid soluble or reducible species, for example, associated with oxides. Low mobility Hg is often associated with oxidizable species, such as non-labile organic carbon, and other insoluble species/minerals such as HgS, which are not extracted in the other fractions.

It should be considered that some other researchers have found sequential leaching methods to be lacking in extracting the precise Hg species of interest. Extractant liquids used by sequential leaching techniques may actually affect Hg speciation, thus giving misrepresentative results (Kim et al., 2000). For instance, Hall and Pelchat (2005a, 2005b) found that $HgCl_2$ and HgO could predictably be extracted using $Ca(NO_3)_2$ and hydroxylamine hydrochloride reagents, respectively. However, when these species were mixed in a silicate matrix these species were observed in other extraction steps too. Their finding suggests a degree of undesirable Hg resorption that may affect the accuracy of any derived speciation data. Therefore, Hall and Pelchat (2005a, 2005b) recommended that sequential leaching results for typical soil samples (i.e. < 1 mg/kg) should normally be viewed with caution, particularly for the initial liable fractions. On the other hand, they reported that HgS was always extracted in the final extraction step (i.e. aqua regia), whether extracted alone or mixed with a granitic matrix (Hall and Pelchat, 2005b). Because of this limitation, researchers will often classify sequential leaching fractions based on relative environmental mobility, or availability. In other words, rather than determining specific Hg species, sequential leaching provides classification of Hg species in terms of behavior or mobility.

2.2.2. *Thermodesorption techniques*

Mercury thermodesorption (TD) techniques rely on the difference in the volatility of different Hg species in order to separate species. Quantitative detection of Hg after conversion to Hg^0 is usually

Table 2
Analytical approaches for the detection of Total-Hg and methyl-Hg in soil.

Hg form	Detection technique	Sample preparation	Typical extraction time (min)	Reported detection limit (μg/kg)	Detection limited calculated as:	Reference(s)
Total-Hg	CV AAS	Pyrolysis or aqua regia digestion	180	0.5	n/a	(Almeida et al., 2016; Biester and Nehrke, 1997)
Total-Hg and Methyl-Hg	HPLC CV AFS	Ultrasonic extraction with KOH/CH_3OH 25%, and UV oxidation	180	51 (ng/dm^3 solution)	Net signal equal to 3× standard deviation of the blank*	(Almeida et al., 2016; Ramalhosa et al., 2001)
Methyl-Hg	HPLC PCO CV AFS	Extraction in 10% NaCl, 50% H_2SO_4 with pre-concentration using a C18 column	45	40	Signal to noise ratio of 3:1	(Almeida et al., 2016; Eiden et al., 1997)
Total-Hg	Gold film mercury analyzer	Acid digestion in 9:1 HNO3:HCl	115	n/a	n/a	(Almeida et al., 2016; Mudroch and Kokotich, 1987)
Total-Hg	CV AAS	Extraction using Universol® 40%, 60 °C and CH_3Hg oxidation with 0.1% KMnO4	30	70	3σ criterion**	(Almeida et al., 2016)
Total-Hg	CV-μCCP-OES	Microwave acid digestion with aqua regia	45	4.8	3σ criterion**	(Frentiu et al., 2013)
Total-Hg	CV-ICP-OES	Microwave acid digestion with 3:1 HNO_3:HCl	10	3	3σ criterion**	(Hellings et al., 2013)

Note: * Calibration curves in the range of 100–800 ng/dm^3 and based on the amount (or concentration) necessary to yield a net signal equal to three times the standard deviation of the blank ** 3σ criterion is three times the standard deviation of 10 independently measured blanks divided by the calibration sensitivity. Acronyms: CV AAS = cold vapor atomic absorption spectrometry; HPLC CV AFS = High Performance Liquid Chromatography Cold Vapor Atomic Fluorescence Spectrometry; HPLC PCO CV AFS = high performance liquid chromatography post column oxidation—cold vapor atomic fluorescence spectrometry; CV-μCCP-OES = cold vapor capacitively coupled plasma microtorch optical emission spectrometry; CV-ICP-OES = cold vapor inductively coupled plasma optical emission spectrometry.

Table 3
Sequential extraction methods employed for fractionation of Hg in soil.

Extractant(s)	Behavior classification	Attributed Hg species	Rationale for method	Reference
Deionized water	Water soluble	$HgCl_2$, $HgSO_4$	This method was developed for determining Hg species in geological solids. It was validated by robust quality assurance with reference materials, and pure compounds. Extraction time, solids-to-liquid ratio, and solvents were evaluated. Hg species are attributed based on extractions of known Hg species and XAFS analysis (Kim et al., 2003). Moreover, concentrations in 'water soluble' and 'human stomach acid'-soluble fractions have correlated well with results garnered from in vitro bioaccessibility tests.	(Bloom et al., 2003)
pH 2, 0.1 M CH_3COOH + 0.01 M HCl	"Human stomach acid"	HgO, $HgSO_4$, Hg sorbed to oxides (Fe, Mn)		
1 M KOH	Organo complexed/organo chelated	Hg bound to humics, Hg_2Cl_2, CH_3Hg		
12 M HNO_3	Strong complexed/elemental hg	Soil mineral lattice, Hg (0), Hg_2Cl_2 m-HgS w/impurities		
Aqua regia	Mercuric sulfide	HgS, HgSe m-HgS		
Deionized water	Water soluble species	$HgCl_2$	This method was developed for a typical environmental laboratory. The extraction method was evaluated with known mercury salts and minerals (HgS, $HgSO_4$, HgO, $HgCl_2$) in a prepared soil matrix.	(Gavilan-Garcia et al., 2008)
Heating to 180 °C	Elemental Hg	Amalgamated Hg		
$MgCl_2$	Exchangeable species	$HgSO_4$, HgO		
HCl	Strongly bound species	Hg bounded to Fe & Mn		
NaOH, CH_3COOH	Organic Hg	Organic Hg		
NaS	sulfide fraction	HgS		
Aqua regia	Residual Hg	HgSe and residual		
0.01 mol/L $Ca(NO_3)_2$	Soluble and exchangeable	$Hgcl_2$	The method is used by the Geological Survey of Canada (GSC). It is useful for identifying HgS; by first identify other Hg forms prior to aqua regia extraction.	(Hall and Pelchat, 2005a, 2005b)
0.1 mol/L $Na_4P_2O_7$	Labile organic component`			
0.25 mol/L NH_2OH_3HCl in 0.25 mol/L HCl	Amorphous oxide-bound	HgO, amorphous Fe and Al oxides		
1 mol/L NH_2OH_3HCl in 25% CH_3COOH	Crystalline oxide-bound	Crystalline Fe and Al oxides		
6.4 mol/L HNO_3	Non-labile organic and elemental	Hg(0)		
Aqua regia	Sulfide-bound and residual	Sulfides (HgS), oxides, some silicates, refractory organics		

performed via atomic absorption spectroscopy (AAS), and therefore, this technique is also referred to as TDAAS. The results are presented as thermo-desorption curves. Species are identified by comparison with synthetic standard reference materials (Reis et al., 2015a). Overlapping peaks can often be problematic. For example, there is often difficulty in distinguishing between $HgCl_2$ and humic acid–Hg complexes due to peak overlap (Reis et al., 2012). Alternatively, fractional classification based on behavior information can be garnered. Biester and Scholz (1997) compared thermodesorption and sequential extraction techniques for attaining Hg speciation information for soil samples collected from a former chloralkali plant, a wood preservation site, and two mining areas. The results revealed that Hg^0, non-specific matrix-bound Hg^{2+}, and HgS could be identified by thermodesorption. However, sequential extraction did not identify any specific Hg compounds, as most of the Hg was extracted in the last non-specific 'residual' fraction. Thus revealing inadequate selectivity of the extractants used. Moreover, the authors reported that sequential extraction effectiveness was influenced by organic matter and buffering carbonates (Biester and Scholz, 1997).

2.2.3. *X-ray techniques*

X-ray techniques provide the most conclusive information on Hg speciation. However, the use of X-ray techniques for obtaining Hg speciation information is typically expensive and may require samples that contain Hg concentrations that are higher than normally found in typical soils (> 100 mg/kg) (Kim et al., 2000). Synchrotron radiation-based X-ray absorption spectroscopy (XAS) can be used to garner molecular-level Hg speciation information. They are non-destructive, and require minimal sample preparation. Kim et al. (2000) investigated the speciation of Hg in heterogeneous mine tailing samples collected from a site in California, USA. To derive speciation data, XAS spectra were compared to a spectral database of crystalline Hg minerals and sorption samples. Spectra phase and amplitude differences can be observed, which are used as 'fingerprints' to identify species in heterogeneous samples (Kim et al., 2003).

2.3. *Gaseous elemental mercury (GEM) measurement*

Estimation of gaseous elemental mercury (GEM) flux from soils is often complex (Böhme et al., 2005). Researchers have made efforts to develop mobile and cost-effective field methods to allow rapid measurements, for example, portable Hg-AAS analyzers e.g. (Böhme et al., 2005; Rinklebe et al., 2009). Some of the devices enable direct and continuous quantification via Hg electrodeless discharge lamps positioned at 253.7 nm. Zeeman correction ensures accuracy, even in the presence of dusts, aerosols, absorbing gases and vapors (Sysalova et al., 2017). Measurements of soil Hg flux can also be performed using a chamber, which is placed on uncovered ground (Bohme et al., 2005; During et al., 2009; Rinklebe et al., 2009). Gas exchange chambers (GECs) can be used for precise control of studied variables, with experiments performed in the laboratory in an enclosed chamber holding the sample. The more simplistic dynamic flux chambers (DFCs) are used in the field to obtain ambient flux data from dry and wet soils, and can be used to measure non-point source Hg emissions on a wide variety of different ground surfaces (Eckley et al., 2010). Micrometeorological methods (MM) have also been developed that allow for larger spatial-scale measurements and for marginal modification of environmental conditions with the Relaxed Eddy Accumulation (REA) method, the Aerodynamic Gradient Method (AGM), or the Modified Bowen Ration (MBR) method.

3. Release of Hg from soil to the atmosphere

Volatile Hg species, such as Hg^0 and dimethyl-Hg (Table 1), have the potential to be released to the atmosphere if present in soil. Globally, average emissions of Hg are estimated to be in the order of 1 ng/m^2 per hour (Lindqvist et al., 1991; Mason et al., 1994; Schroeder and Munthe, 1998), but may be as high as 100,000 ng/m^2 per hour at highly contaminated sites (Zhang and Lindberg, 1999). Therefore, the flux at the pedosphere-atmosphere interface is an important component of global and regional Hg biogeochemical cycling (Beckers and Rinklebe, 2017; Gustin and Jaffe, 2010; Pierce et al., 2015). In the atmosphere, Hg is widely dispersed and transported over thousands of km before being redeposited to the pedosphere or other environmental spheres, thus, having wide-ranging implications on global Hg distribution (Zheng et al., 2012). Therefore, it is important to recognize the factors that may affect Hg flux from soil to the atmosphere, with the intention of managing contaminated sites in ways that consider this process.

Factors that influence Hg release from soil include Hg species and their concentration in soil; atmospheric Hg concentration (Bahlmann et al., 2006; Edwards et al., 2001; Gustin and Lindberg, 2005; Yu et al., 2018); meteorological factors (e.g. atmospheric pressure, air temperature, wind speed and turbulence, solar radiation, snow cover) (Carpi and Lindberg, 1997; Gustin et al., 2002); soil moisture content (Bahlmann et al., 2006; Gustin and Stamenkovic, 2005); soil temperature and surface characteristics (Beckers and Rinklebe, 2017); and, air mixing (Bahlmann and Ebinghaus, 2003). The interaction of different factors on Hg flux can be synergistic. For example, Lin et al. (2010) described the synergistic effects of two-factor interactions. Air temperature-soil moisture and air temperature-solar intensity were the most significant of the two factor interactions described. The effects of these combined factors were observed to be greater than the sum of each of the factors separately.

3.1. *Influence of soil conditions on Hg flux*

Lin et al. (2010) used DFC measurements (Section 2.3) to calculate Hg flux for soils containing various levels of Hg. The Hg flux was associated with soil Hg content, with a linear positive correlation ($r^2 > 0.99$), thus revealing that Hg flux from the pedosphere to the atmosphere is significantly dependent on its concentration in soil. However, it should be noted that the soil tested contained a relatively low fraction of organic carbon ($< 2\%$), and the impact of organic carbon on Hg flux levels warrants further investigation. Surface covering is also an important factor on Hg flux (Luo et al., 2016). For example, Gabriel and Williamson (2008) found that Hg flux measurements for turf grass were two times lower than bare soil; and paved ground was 22 times lower than bare soil.

3.2. *Influence of ambient conditions on Hg flux*

Mercury flux from low concentration soils (i.e. < 100 ng/kg) is associated with weather conditions (Fig. 2), as was explained by Briggs and Gustin (2013), building on (Gustin and Stamenkovic, 2005). It should be noted that the Hg flux associated with such soils is relatively low (i.e. < 20 ng/m^2 per hour) but is significant on the global scale. The proposed mechanism suggests six scenarios (A–F) during rainfall events: (A) Initial infiltration water will desorb Hg^0 from soils and displace gaseous elemental mercury (GEM) to the atmosphere as a pulse; (B) as rainwater further infiltrates the subsurface, freely available Hg^{2+} species in the soil can be dissolved to the aqueous phase, and Hg^0 sorbed to soil may desorb to a GEM phase, but Hg flux will be prohibited as soils become water saturated; (C) after rainfall cessation, Hg flux is increased by the increase of available energy (i.e. solar radiation and higher soil temperature), and any abiotic or biotic Hg reduction processes; (D) however, as the soil dries, Hg flux will be reduced as Hg movement is conducted by upward water vapor movement, which brings dissolved Hg^{2+} and GEM to the surface; (E) in vegetated areas, Hg movement will also be conducted by transpiration; (F) finally, if the soil becomes effectively dry, Hg flux is significantly reduced.

Hg flux is known to vary in daily patterns (Rinklebe et al., 2010). It is long established that Hg flux from soils is influenced by daily

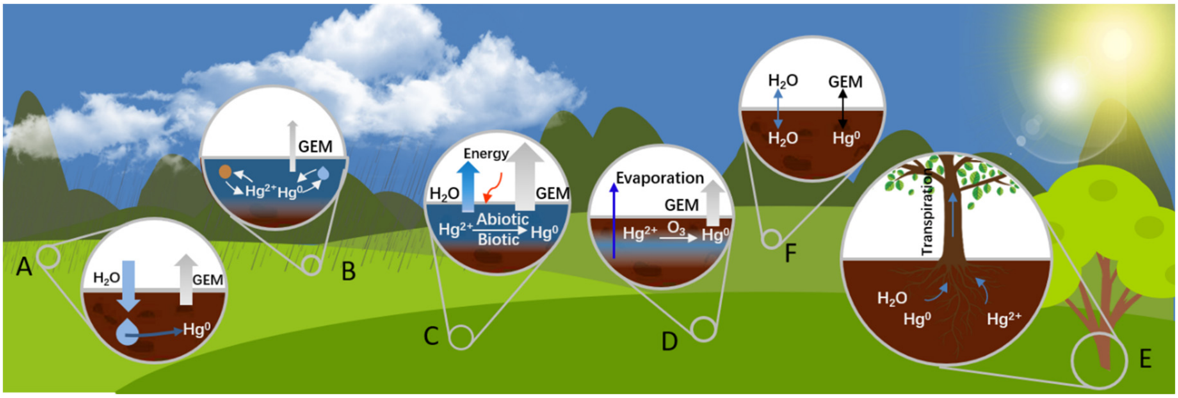

Fig. 2. Conceptual model of six stages of Hg flux from low Hg concentration soils associated with rainfall events (based on Briggs and Gustin (2013)).

fluctuations in solar radiation intensity (Carpi and Lindberg, 1997; Poissant and Casimir, 1998). More recently, it has been suggested that ambient atmospheric chemistry, especially oxidants such as ozone (O_3), also has a considerable role in daily Hg flux levels. For instance, recurring daily fluctuations in Hg flux from dry soils was observed under dark conditions and constant temperature (Zhang and Lindberg, 1999). Engle et al. (2005) reported a > 75% decrease in Hg flux from Hg^0 contaminated media in the presence of up to 70 ppb O_3 in air, as compared to the presence of oxidant free air. This is expected because volatile Hg^0 is converted to the non-volatile Hg^{2+} state. On the other hand, a 1.7-51 × Hg flux increase was observed from Hg^{2+} contaminated media. Although an exact mechanism for this could not be put forward, the authors suggested that O_3 oxidation of the sulfide component of HgS might have freed Hg to be reduced to Hg^0 by S species. It is becoming clearer that atmospheric O_3 is a significant influencing factor in Hg flux from dry Hg^{2+} contaminated soils; potentially more so than solar radiation intensity. It is interesting to note that because average O_3 concentrations in the atmosphere now are twice as high as they were 100 years ago, this has significant implications for Hg flux from soils (Engle et al., 2005; Rinklebe et al., 2010). Chemical reduction and oxidation rates for Hg in the presence of atmospheric oxidants also require investigating, including determination of which oxidants are important (AMAP/UNEP, 2013).

4. Interactions between Hg and soil

The pedosphere is deemed a net sink of Hg, primarily due to Hg taken in by plants being deposited on soils (Obrist et al., 2018). Mercury-soil systems involve complex systems, with Hg tending to be immobilized in surficial soil due to its affinity for mineral surfaces and bonding to organic matter. For typical concentrations in soils, Hg^{2+} tends to form stable complexes with, OH−, Cl−, and S containing functional groups of organic ligands (Powell et al., 2004). Relationships between various soil properties and Hg are discussed in the section below so that determinants of Hg behavior can be identified at contaminated sites and, therefore, remediation strategies can be better targeted to manage risk.

4.1. Hg-soil sorption

Mercury is known to be relatively immobile, as compared to many other metals in soil, as it can bind strongly with soil constituents. A number of studies have been carried out examining the competitive sorption and selectivity sequences of various heavy metals by various soils. As an exemplar, Seo et al. (2008) explored the sorption potential of Hg and six other metals to a wetland soil. In batch mono-metal experiments (at pH = 6), the seven metals were ordered by adsorptive capacity (mg/g) as follows: Pb (25.4) ≫ Hg (6.4) > Cr (4.9) > Cd (2.9) ⩾ Cu (2.6) ⩾ Zn (2.4) ≫ As (0.8). Based on multi metal adsorption they were ordered as Hg (3.0) > Cr (1.1) > Cu (0.6) ⩾ Cd (0.4) ≈ Pb (0.4) ≫ As (0.02) ≈ Zn (0.02). Thus, it was apparent that the adsorption capacity for Hg remained strong in the presence of the other metals, whereas that for Pb was significantly lowered (Seo et al., 2008). Antoniadis et al. (2017b) showed that Hg sorption was not related to the presence of any other potentially toxic elements at a highly contaminated former mining area in Germany. Liao et al. (2009) considered the adsorption of Hg in different types of soils, with Sharkey clay having greater Hg sorption capacity than Olivier loam, which was itself greater than Windsor sand. It was also revealed that Hg sorption in each case was rapid and strongly irreversible, with freely available Hg typically being < 1%. The binding of Hg in soils is due to its tendency to bind with soil organic matter or soil matrix surfaces. Mercury can be regarded as a highly immobile metal in most soils due to highly stable complex formation (Liao et al., 2009). Mercury speciation also has a significant effect on the amount binding to soil. For instance, Hg^0 has a typical soil water partition coefficient of ~1000 mL/g, whereas Hg^{2+} can range between 24,000 and 270,000 mL/g with a typical value of ~60,000 mL/g (US EPA, 1997).

4.1.1. Sorption to soil organic matter

Soil organic matter is a system of progressively decomposing organic compounds (Lehmann and Kleber, 2015), being byproducts of the biogeochemical degradation of plants and animals. Organic matter consists of different types of substances such as: high molecular-weight hydrophobic compounds, hydrophobic neutral organic matter, and low-molecular weight compounds that are more hydrophilic (Stevenson, 1994). Soil organic matter has a particularly high affinity for Hg^{2+}. Therefore, elevated total-Hg is often associated with organic rich soils, such as forest soils, peaty soils, or rice paddy fields (Beckers and Rinklebe, 2017). Mercury can bind to organic matter in a number of ways. Organic matter functional groups, such as hydroxyl, carboxylic, aromatic and S-containing ligands may facilitate cationic Hg^{2+} binding (Sysalova et al., 2017). Mercury in soil is particularly inclined to form covalent bonds with any available reduced S active sites (Reis et al., 2015b). The reason for this was ascribed by Mousavi (2015) to the phenomenon of polarizability. The hard and soft acids and bases rule predicts that soft acids and soft bases will have strong interactions. Because S containing thiol functional groups act as a soft base and Hg^{2+} is a polarizable soft acid, the binding is strong. Oxygen containing alcohol and carboxylic acid functional groups and N containing amine groups act as hard bases and, therefore, do not bind with Hg as strongly. However, S bearing functional groups of humic acids can quickly become saturated with Hg, therefore, most Hg^{2+} bound organic matter is found to be associated with O or N containing functional groups (Gismera et al., 2007). Because of such binding, the amount and type organic matter content in soil can significantly affect soil Hg solubility, mobility and, toxicity (Sysalova et al., 2017). Chai et al. (2012) reported stable interactions between soil humic acids and Hg, attributed to the abundance of O containing ligands. Humic acids have high complex stability potential, thereby causing a decrease in Hg mobility (Aijun et al., 2006), whereas Hg bound to fulvic acids is considered a more labile form Wallschlager et al. (1998).

The effect of soil organic matter on Hg sorption can also be influenced by human practices. Dai et al. (2013) considered Hg distribution of arable and natural unfarmed soils in the historic Hg mining area of Wanshan in Guizhou, China. It was stated that Hg was introduced to the study area via contaminated irrigation water, with Hg bound to particulate matter. For natural unfarmed soils, positive correlations between Hg levels and the amount of organic matter were observed; however, no such correlations were observed for worked arable soils. Farming cultivation practices generally decrease soil organic matter and increases air exposure. In the case of rice paddy soils, farming practices minimize exposure to air, and may reduce the fraction of large soil aggregates and increase soil organic matter content. Therefore, where there is a source of Hg introduction, concentrations of Hg in rice paddy soils can significantly increase during long-term rice cultivation (D. Yin et al., 2016).

4.1.2. Sorption to soil matrices

Reis et al. (2016) described how Hg might sorb onto a soil matrix surfaces in two ways, either by ‘non-specific’ or ‘specific’ sorption. Cation exchange drives non-specific sorption, resulting in ‘outer-sphere complexes’. The cation exchange process occurs rapidly and is considered reversible. In the case of specific adsorption, stable complexes are formed in which Hg diffuses inward to form ‘inner-sphere complexes’. These are considered slow non-reversible processes (Bradl, 2004; Reis et al., 2016). Dissolution mechanism may cause Hg bound to soil matrices to become available. Dissolution may be brought about by the presence of complexing or chelating agents present in organic matter, for instance, produced by plant roots (exudates) or mycorrhiza. It is possible that dissolution will also occur due to reductive dissolution of Fe oxides. The effect of organic ligands on Hg sorption by mineral colloids in soils, in terms of the precise mechanisms and sorption kinetics, remains somewhat unclear. Recently, Yang and Ok (2017) explored Hg sorption by non-crystalline Al hydroxides under different pH conditions in the presence of selected organic ligands such as S containing cysteine, glycine, and citric acid. It was determined that Hg^{2+} sorption by the control sample, cysteine and glycine systems was mediated by specific surface complexation, whereas ligand exchange in the citric acid system was predominant. The Hg adsorption was observed to be initially rapid, and the amount of sorption decreased with increasing pH, except in the presence of higher concentrations of cysteine.

Soil clay content has an important role in soil-Hg binding (Biester et al., 2002b; Boszke et al., 2008). For instance, rice paddy soils, which are characterized by their clayey as well as organic content, are susceptible to high Hg levels (R. Yin et al., 2016). It has been reported that clay Hg sorption capacity can reach ~1000 mg for each 1 $\mathrm{cmol_c}$ per kg soil (Antoniadis et al., 2017a). Therefore, elevated Hg levels are often associated with more clayey soils. Coufalík et al. (2012) divided soil samples from northern Mongolia into different particle size fractions. The greatest Hg levels were discovered in the finest sized fraction. The authors attributed the greater Hg sorption to the clay's high specific surface area (Coufalík et al., 2012) and high cation exchange capacity (CEC). Antoniadis et al. (2017a) suggested that 2:1 clay minerals such as illites have greater capacity to sorb Hg than 1:1 clay minerals such as kaolinite. The expandable 2:1 type clay minerals including smectites have greater capacity still (Antoniadis et al., 2017a). According to the Pauling electronegativity theory, Hg is likely to form covalent bonds with O atoms of clay lattices. Because of this favorable bonding mechanism, Hg is more readily immobilized than most other potentially toxic elements in soils (Antoniadis et al., 2017b). Clay in soil can also be an effective sorbent of organic matter. It is suggested that increased Hg sorption capacity in clayey soils may also relate to binding with organic matter.

Thermodesorption of Portuguese soils revealed a major peak at 125–250 °C ascribed to either HgCl2, Hg bound to Fe oxides, or Hg bound to humic substances (Reis et al., 2015a, 2015b). This implies that the Hg in the sample may have been sorbed to the matrix mineral surfaces, or to organic matter. The precise species could not be distinguished by this technique. In fact, Hg organic matter complexes can themselves be sorbed to soils matrix mineral surfaces - forming organo-mineral Hg complexes – thus simultaneously existing as different forms. In this case, it was reported that Fe oxides represented a large fraction of the soil (~10%), whereas the organic matter content was low (~0.5%), and there were no suspected sources of chloride. Therefore, it was assumed that the Hg would likely be associated with Fe oxides. Thermodesorption analysis of weathered Amazonian soils by do Valle et al. (2006) revealed peaks attributed to Hg^0 release at ~150 °C and peaks at higher temperatures attributed to various mercuric salts. For Hg that is released between 150 and 250 °C, the non-specific term “matrix-bound Hg” is often used. This is because desorption of Hg sorbed to mineral surfaces (e.g. Fe oxides) cannot easily be distinguished from Hg sorbed to organic matter (Biester and Scholz, 1997).

4.2. Influence of soil properties on Hg speciation and behavior

The physico-chemical parameters of soil, including pH, cation exchange capacity (CEC), soil grain size distribution, organic matter type and content and the presence of different clay types and fractions, may affect its interaction with Hg, and, thus be influential on Hg fate and transport. Chen et al. (2016) reported that soil total-Hg levels at a former goldmining area near Beijing, China, were correlated to soil CEC ($R = -0.408$, $P < 0.005$), but not to pH and soil organic matter. It should be noted that clay and organic matter content would usually contribute CEC to soils. Mercury may exist as available labile species or it can be in immobilized forms. Xu et al. (2014) found only a small amount (< 0.2%) of total-Hg at the Tidermans padding area in Sweden to be a freely available water-soluble form. The vast majority of the Hg was bound to the various constituents of the soil (Xu et al., 2014). As such, traditional total-Hg based assessments of soils may overlook such complexities (Li et al., 2008).

Sequential extraction of soil samples by Chen et al. (2016) again revealed freely available Hg accounting for only a small proportion of total-Hg (0.3%), with high residual fractions. The high pH of the soil may have facilitated the binding of Hg to Fe-Mn oxides. Lucjan Rozanski et al. (2016) tested bioaccessibility of Hg in soils with Pentetic acid (DTPA) extraction. These soils contained large amounts of clay and amorphous Fe oxides, and only low amounts of bioavailable Hg (Lucjan Rozanski et al., 2016). Retention of Hg^{2+} in weathered Amazonian soils was reported to be highly affected by soil pH (Miretzky et al., 2005), as Hg^{2+} could be released from soils under acidic conditions (Coufalík et al., 2012). Various researchers have undertaken correlation analyses between sequential extraction fraction and soil properties. Lucjan Rozanski et al. (2016) found significant correlations between soil organic carbon (SOC) content and water leachable Hg, suggesting that SOC was holding Hg in a freely available form, whereas soil clay content was associated with a less mobile fraction with tighter binding. Li et al. (2008) also revealed a negative relationship between easily extractable Hg and clay content. Chen et al. (2016) assessed soils around former gold mines and a reservoir near Beijing, China, and found that carbonate, and organic-bound Hg fractions had strong correlation with soil pH and organic carbon. The authors also stated that the formation of Fe-Mn oxide-bound Hg was increased by the high pH conditions.

In anaerobic Hg contaminated soils, the microbial reduction of sulfate can lead to the formation of mercuric sulfide (HgS) (Devai et al., 2005). HgS is a dimorphous mercuric compound found in mineral forms known as cinnabar (α-HgS), or metacinnabar (β-HgS). HgS is usually encountered in the cinnabar form in nature. For instance, Kim et al. (2004) reported that HgS at former Hg mining sites were in the cinnabar form. However, where soils had historically been exposed to temperatures in excess of 600 °C due to ore processing, samples were found to contain large amounts of metacinnabar. This was ascribed to reconstructive HgS phase transformation processes occurring at temperatures above 345 °C. Recently, direct HgS (metacinnabar) formation has been demonstrated under aerobic conditions via linear Hg^{2+} thiolate complexes in cysteine solution (Enescu et al., 2016). This finding is relevant to soils where the most significant source of sulfide is organosulfides.

Mercury residing in soils as HgS (cinnabar or metacinnabar) is considered chemically stable, even over geological time periods. Revis et al. (1989) revealed that Hg in floodplain soils in Tennessee, USA, was mostly (85%) in the HgS form. The high percentage of HgS was brought about by sulfate reduction occurring in the soils. HgS is the predominant form of Hg found in many gold mining waste ores and calcines (Lowry et al., 2004) due to typically elevated S contents (Reis et al., 2010). Because of its low solubility, low bioavailability and stability in anaerobic conditions, it is considered the least toxic Hg species in soil (Sysalova et al., 2017). For this reason, researchers have considered adding reduced S to Hg contaminated soils in order to immobilize Hg within the soil. To this aim, O'Connor et al. (2018c) successfully tested an S modified biochar, a carbon rich product of biomass pyrolysis, as a soil amendment to immobilize Hg in contaminated soil. Selenium has also drawn attention as it may reduce Hg uptake in the aerial portions of plants. This is thought to be due to insoluble Hg–Se complexes forming in plant root zones, thus inhibiting Hg metabolization and/or translocation (H. Zhang et al., 2012).

5. Chemical transformations of Hg in the soil environment

Mercury in soil can be converted between different chemical forms depending on the physico-chemical soil conditions. This has implications for contaminated soils under long-term management post remediation. Potential routes of chemical transformation must be identified as one of the requirements for obtaining a robust decision framework for risk-based management of contaminated sites.

5.1. Oxidation of Hg in soil

The Hg^{2+} cation is more commonly found in soils than Hg^0. This is related to Hg^0 being volatile and susceptible to oxidation to Hg^{2+} under aerobic condition. This was exemplified at a former gold mining site in Brazil that had been contaminated by metallic Hg pollution. After fifty years, the majority of Hg at the site was in the form of oxidized Hg^{2+}, which had stabilized within the soil by sorption to soil minerals (Fe, Mn and Al oxides) or organic matter (Durao et al., 2009). A number of factors may influence Hg^0 oxidation, however, identifying these specifically is challenging. Windmoller et al. (2015) studied the Hg^0 oxidation process experimentally. They added Hg^0 and $HgCl_2$ to three surface soil samples of pH 5.6–5.8 containing 1.6–2.7% OM and monitored the Hg transformation via thermodesorption. After maintaining the samples in ambient temperature or low temperature (4 °C) dark conditions a large portion of Hg^0 was oxidized to Hg^{2+} (~36–88%). The oxidation process was reported to occur via Hg^{1+} as two first-order rate reactions. Oxidation of Hg^0 to Hg^{1+} was rapid ($t_{1/2}$ = 2–5 days), and ~43–139 times faster than the subsequent oxidation of Hg^{1+} to Hg^{2+} ($t_{1/2}$ = 107–270 days). Based on the finding it is curious that mercurous forms of Hg are not more frequently reported.

It has been observed that soil organic matter may perform a significant role in Hg oxidation. Windmoller et al. (2015) found that large amounts of organic matter in soil favors the oxidation of Hg^0 to Hg^{2+} and hinders the reduction of Hg^{2+} to Hg^0. On the other hand, the presence of organic matter can provide substrates as electron acceptors, being oxidized to CO_2 by microbes (O'Connor et al., 2018a), and thus bringing about reducing redox conditions. Further research is needed to understand how each of these conflicting processes may dominate under different environmental conditions and in different soil types.

5.2. Chemical reduction of Hg in soil

Microbial mediated reduction of Hg from the oxidized state (Hg^{2+}) to its elemental form (Hg^0) can occur under low soil redox conditions in permanently or periodically flooded anaerobic soils (Devai et al., 2005; Frohne et al., 2012). Under these conditions, Hg^{2+} can also be precipitated as HgS, or be converted to Methyl-Hg. Therefore, an intricate understanding of environmental conditions beyond oxidation-reduction potential (ORP) is necessary to discern how Hg will transform. This is an important area where more research is needed. For example, methylation potential is highly dependent on Hg bioavailability (discussed in Section 5.3 below). Abiotic reduction of Hg^{2+} is also possible in certain circumstances. Organic acids, such as humic acids and other humic substances may perform this role (Allard and Arsenie, 1991). Chakraborty et al. (2015) found that abiotic reduction of Hg^{2+} to Hg^0 by humic substances was faster at lower pH (i.e. pH ≤ 4) and salinity levels (i.e. ≤5.0 PSU). Some reduction reactions in surface soils may be enhanced photochemically (Lin et al., 2010). Solar radiation may increase the rate of some Hg^{2+} reduction reactions, but may also increase the rate of Hg^0 oxidation reactions (Zhang and Lindberg, 1999).

5.3. Organo-Hg formation in soil

In the reducing conditions that occur in many permanently or periodically flooded soils, Hg may be biogeochemically transformed into organo-Hg forms (Frohne et al., 2012; Parks et al., 2013), of which Methyl-Hg is the most prevalent. The bioavailability of Hg species to methylating microorganisms is crucial in determining the extent of these conversions. Fernandez-Martinez and Rucandio (2014) found that the most mobile Hg fractions, such as Hg-humic or Hg-fulvic complexes, in a Ferric Lixisol soil at a former Hg mining area in Almaden, Spain, only accounted for 3.2–7.7% of the total-Hg content. However, these fractions were the most susceptible to Hg methylation. On the other hand, soils with greater amounts of larger organic matter types (i.e. larger molecular weight) are less at risk of Hg methylation because of lower bioavailability (Sipkova et al., 2016; Skyllberg et al., 2006).

The direct conversion of insoluble HgS (cinnabar) species to Methyl-Hg in anaerobic soils is generally believed to be low/insignificant (Gray et al., 2015), due to its low bioavailability. But it is now known that in certain circumstances, the mercury in HgS can be methylated by sulfate reducing bacteria (Lefebvre et al., 2007). Benoit et al. (1999) proposed that HgS methylation might occur when environmental conditions favor HgS complexation (see Section 6.1). T. Zhang et al. (2012) suggested that Hg methylation potential might be associated with geochemical intermediate products deriving from rate-limited Hg-sulfide precipitation reactions. However, the precise role of this in Methyl-Hg formation is still not well understood. Thomas et al. (2018) put forward two potential mechanisms for Hg^{2+} uptake by methylating microorganisms: (1) the passive diffusion of neutral HgS species, particularly nano sized HgS; and, (2) the energy-dependent uptake of Hg^{2+} by active transport. Regarding the former, it has been reported that nano sized HgS particles may be favored in soil when Hg binds with dissolved organic carbon species, due to a slow-down in growth rate of HgS particles (Deonarine and Hsu-Kim, 2009). Recent reports suggest that nano HgS may be bioavailable for methylation by microorganisms (T. Zhang et al., 2012).

Methyl-Hg in rice paddy soil has been reported to contaminate rice grain produce in some Hg contaminated farming areas. Rice paddies are periodically flooded by farmers, making them susceptible to very low redox conditions. It may be expected that Hg methylation in paddy soil will be influenced by pH and organic matter content. However, Zhao et al. (2016b) reported no direct impact these factors at two sites with alkaline soils. A second article revealed that net Methyl-Hg formation was significantly influenced by the recent deposition of Hg in surficial

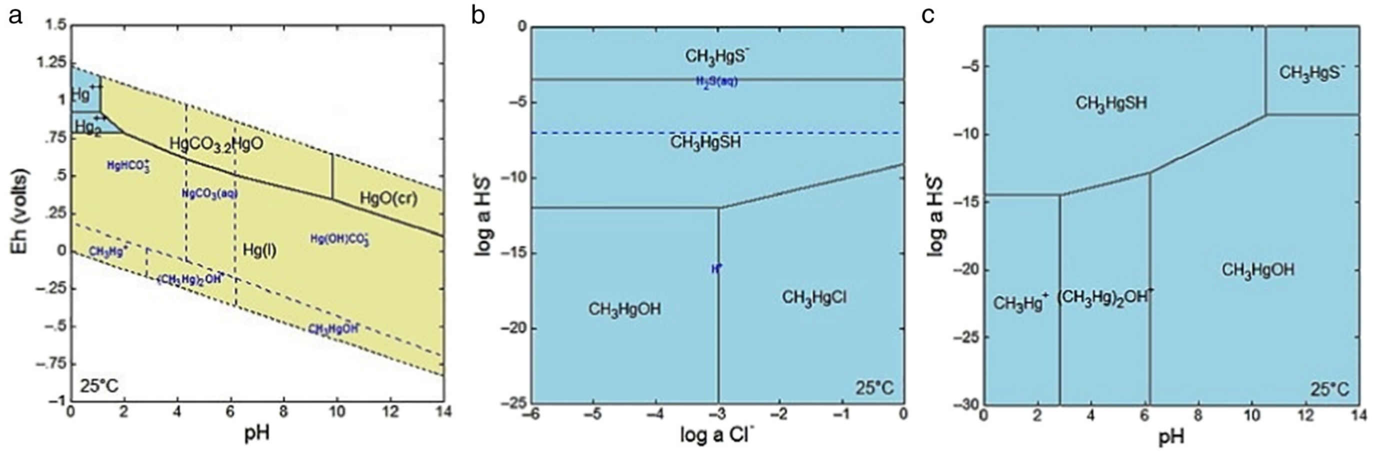

Fig. 3. Methyl-Hg aqueous complexes at 25 °C: (a) carbonate sub-system; (b) sulfide and chloride activity; and, (c) sulfide activity and pH. In yellow: solid phase stability domains; in blue: aqueous complexes stability domains. $[Hg_T] = 10^{-9}$ mol/L; $[CO_{3,T}] = 10^{-3}$ mol/L and $[Cl_T] = 10^{-3}$ mol/L (Blanc et al., 2018). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

soils under anaerobic and low pH conditions (Zhao et al., 2016a). Therefore, the Hg methylation rate in soil may also be a function of atmospheric Hg concentration and deposition processes. Han et al. (2006) also reported that organo-Hg is more likely found at sites of recent contamination with soluble Hg species, and noted that methylation is less likely at sites containing insoluble HgS forms. Tomiyasu et al. (2017) investigated the influence of organic matter decomposition stage, which was found to be correlated to organo-Hg formation. Fallen leaves were reported to deposit Hg on the soil surface and the decomposition of leaves led to Hg sorption. Despite recent advances in our knowledge of Hg methylation, it is apparent that there remain gaps in the research of Hg^{2+} reduction to Hg^0, formation of HgS, and Hg methylation.

6. Availability and mobility of Hg in soils

Transport of Hg by leaching to the subsurface and groundwater is often thought to be insignificant (Grigal, 2003; Johnson and Lindberg, 1995). However, the influence of Hg speciation on the potential transportation rates within soils has not yet been sufficiently studied. The speciation of Hg in soil may vary from site to site, or even across locations on a site (Boszke et al., 2008). For instance, Miller et al. (2013) characterized a site contaminated by the historical use of elemental Hg^0 in Tennessee, USA. The authors found Hg^0 to a depth of 3.2 m at a sampling location, and only in the top 0.3 m at another location just 12 m away, due to the differences in soil conditions. This section discusses factors that may influence Hg transport in soils and the availability of different Hg species.

6.1. Solubility of mercuric species

Typically, Hg in soil is mostly bound to solids, and only a small proportion partitioned to aqueous solution. For example, Liao et al. (2009) revealed that Hg binding to three types of soil was rapid and strongly irreversible, with freely available portion typically being < 1%. Typical soil-water partition coefficients for Hg in superficial soils are ~1000 mL/g for Hg^0, ~60,000 mL/g for Hg^{2+} (range of 24,000–270,000 mL/g) and ~6700 mL/g for methyl-Hg (range of 2700–31,000 mL/g) (US EPA, 1997). The extent of partitioning to soil solution will depend on Hg speciation and conditions at a site. The Hg species found in soils are usually mercuric salts or minerals. The solubility of these salts and minerals can vary widely (Table 1), and, therefore, so can their transportation potential. In typical non-saturated soil conditions, Hg^{2+} in soil solution is typically in the form of $HgCl_2$, HgClOH and $Hg(OH)_2$ (Powell et al., 2004). Gai et al. (2016) calculated that dissolved Hg^{2+} in a simulated rainwater was present as HgClOH (47%), $HgCl_2$ (39%), and $Hg(OH)_2$ (11%). Many mercury compounds dissolved in water do not disassociate (US EPA, 1997). For example, mercuric chloride ($HgCl_2$) will mostly exist as discrete molecules in aqueous solution (Earnshaw, 1997). Dimethyl-Hg is much more lipophilic than mono methyl-Hg, consisting of stable covalent bonds that do not dissociate at pH > 5.6 (Alexander et al., 2008; Fagerström and Jernelöv, 1972).

The solubility of Hg compounds will also depend on the properties of the soil solution. For instance, Jacobson et al. (2005) reported that HgS may actually be soluble in waters that contain certain dissolved organic matter (DOM) substances (e.g. fulvic acids) or in the presence of thiol ligands, or, if there is an excess of sulfide ions present. In such circumstances, insoluble HgS may form soluble complexes, such as the soluble mercuric disulfide complex. The reactions that form HgS and the disulfide complex are highly pH dependent. Moreover, dissolved Hg may interact with DOM to form solid phase HgS (Skyllberg and Drott, 2010). The presence of humic fractions may inhibit this process, where hydrophobic organic acids such as fulvic and humic acids will have a greater effect on inhibiting aggregation than more hydrophilic organic substances (Ravichandran et al., 1999).

Most organo-Hg compounds have low solubility in water (Table 1), and have no tendency to react with weak acids or bases. This is because of their low affinity for O bound to carbon. However, methylmercury hydroxide (CH_3HgOH), is highly soluble because the hydroxide group is strongly involved in hydrogen bonding (US EPA, 1997). The solubility of organo-Hg species is also highly dependent on Hg complexation with organic matter and ion exchange processes (Sipkova et al., 2016). Methyl-Hg aqueous complexes may vary under different environmental conditions. For example, Blanc et al. (2018) reported that solid phase stability domains dominate in carbonate sub-systems under various Eh and pH conditions, whereas aqueous complexes stability domains dominate under various sulfide and chloride activities or under various sulfide activity and pH (Fig. 3).

6.2. Effects of soil organic matter and clay content on Hg transport

Sorption of Hg to colloidal organic matter, such as fulvic acids and hydrophilic compounds, can increase Hg mobility in the soil profile. These species can easily become suspended and transported in porewater (Naidu and Harter, 1998; Sipkova et al., 2016). Decreased mobility has also been reported at Hg contaminated sites with high organic matter contents. For instance, the Hg content of soils amended with organic matter (vermicompost) was stable after 21 days, with mobile Hg concentration reducing from ~700 to ~200 μg/kg (Sipkova et al., 2016). This contrast can be attributed to the high sorption capacity of soil organic matter for Hg (Heeraman et al., 2001), some of which is mobile in solution, while most is immobile and part of the solid matrix. An investigation of Hg mobility in an organic matter rich peat bog in Chile revealed that organic acids in the soil's surface horizon facilitate Hg transport to deeper soils (Biester et al., 2002a). It is apparent that the mobility of Hg sorbed to organic matter is dependent upon the type of matter to which it is sorbed. Lighter molecular weight organic matter substances are more susceptible to being suspended as colloids in porewater and transported by advection (see Section 6.3). Mobility will also depend on the organic matter surface charge density. Lehmann and Kleber (2015) suggested that alkaline extracts of humic substances only provide limited information on sorbed contaminant behavior. This was ascribed to measurements of organic matter solubility in alkaline solution over- or underestimating its reactivity (i.e. electron shuttling, metal adsorption). A better understanding of contaminant mobility may be obtained by examination of a soil's entire soil organic matter, or at least the portion in soil solution, as the most relevant fraction.

The transportation of soil-bound Hg by physical processes such as soil erosion has been explored only to a limited extent. Zheng et al. (2016) addressed how different types of organic matter may affect Hg transported by surface runoff by measuring Hg in heavy and light organic matter fractions, based on density fractionation. It was found that Hg transport was dominated by the heavier fraction of organic matter, which was associated with clay minerals. The clay mineral content was itself an indirect influential factor. For normal rainfall events, an understanding of soil constituents is just as critical for understanding Hg mobility as the soil Hg concentration. Wang et al. (2016) observed that Hg in the tillage layer (0–20 cm) of a site in the North China Plain, a major grain producing area, was less than in the deeper soils. This could have been due to Hg being taken up by plants, or to downward migration of Hg into the soil subsurface. The authors stated that downward migration was unlikely due to large amounts of clay minerals. The clay content of soil may have reduced Hg transport due to Hg sorption to clay mineral surfaces, or hydrogeological conditions (i.e. reduced permeability). On the other hand, Hg may be mobilized by sorption to suspended colloidal clay minerals, which is discussed in Section 6.3.

6.3. Colloidal-Hg transport

Any Hg^{2+} in soil at a site will mostly reside immobilized to solid phases in surface soil. However, because of their small size, solid phase colloidal particles have the potential to migrate within soil pore spaces, presenting a potential pathway for the transportation of bound Hg. Colloids in the pedosphere have been defined as 0.005–5 μm diameter particles in soil (Richards et al., 2007), which may be formed of organic matter, clay minerals, metal oxides, sulfides, etc. It should be noted that dissolved organic carbon is often defined as being < 0.45 μm, which is within this definition of a colloidal fraction. The Hg binds to colloids due to high specific surface area and the presence of surface functional groups. Zhu et al. (2014) suggested that organic matter colloids, or organic matter coated on clay colloids, may be considered primary colloidal carriers of Hg, due to their abundance and strong Hg binding potential. Mineral colloidal kaolinite particles in pore water can also mobilize Hg that would otherwise be immobilized by sand particles, by a process of desorbing Hg from the sand and resorbing it onto itself (competitive sorption) and acting as a carrier in soil pores (Zhu et al., 2012).

It is long-known that DOM can influence the mobility of suspended colloids (Aiken et al., 2011). However, it has been reported only recently that nano-sized HgS particles may form in the pedosphere when Hg binds with DOM species such as humics or thiolates. The small size of HgS nanoparticles means that they may pass through a standard 0.45 μm laboratory filter. Researchers have observed solid phase HgS particles < 5 nm in diameter present in transmission electron microscopy (TEM) images, with HgS aggregates stabilized in the order of 20–200 nm (Deonarine and Hsu-Kim, 2009). EDX spectra confirmed the presence of Hg and S, and possibly Cl.

The potential for subsurface transportation of HgS nanoparticles will be greatly affected by their particle size, surface charge, and aggregation. Gai et al. (2016) synthesized HgS nanoparticles with average hydrodynamic diameters of ∼230 nm, which were polydisperse in 5 mM NaCl solution at pH 7.5. Aggregation of HgS nanoparticles was not observed in a 200 mM NaCl solution, suggesting that the 147 mg/L of organic carbon in solution may have sterically stabilized the nanoparticles. Thus, Hg interaction with DOM may constitute a mechanism of stabilization of HgS in colloidal suspension, with the metal to DOM ratio altering the precipitation kinetics, reactivity and bioavailability, as well as the transportation potential. It has been reported by Anh Le-Tuan et al. (2015) that diffusive passive sampling techniques sample only the freely available dissolved Hg fraction, and not particulate bound species. The authors used X-ray absorption spectroscopy (XAS) to determine Hg species accumulated on a diffusive gradient in thin-film (DGT) passive sampler binding layer. They reported that HgS nanoparticles were excluded; thus suggesting that the use of diffusive samplers may overlook this fraction in soil environments where HgS nanoparticles are of importance for assessing risks.

6.4. Relative mobility of different Hg species

The relative mobility of different Hg species has received little attention so far. Recently, Gai et al. (2016) considered the vertical mobility of four different Hg species by laboratory column experiments. The four Hg species considered were: (1) dissolved Hg^{2+}, (2) a prepared Hg^{2+} and DOM complex, (3) Hg^0, and (4) HgS nanoparticles. The species were applied to the surface of various porous media (sand, sand with 2% kaolin clay, a low-organic carbon natural soil, and a high-organic carbon natural soil) in columns, and subjected to simulated rainfall under semi-saturated conditions i.e. at an infiltration rate that did not fully saturate the soils. Based on effluent Hg concentrations against time, the percentage of total Hg mass immobilized in the columns was calculated for the different Hg species. The Hg^{2+} DOM species were the most mobile, and HgS nanoparticles the least mobile, yet the mobility in each case was relatively low, showing the susceptibility of Hg to bind with surface soil constituents even in sandy soil.

7. Implications for risk management

The fact that Hg is hazardous to human health is long known. The use of inorganic Hg to produce felt for the 19th century hat industry led to the characteristic neurological disorders experienced by hatters (Mahajan and Sharma, 2011). Organo-Hg compounds have proven to be even more harmful. Methyl-Hg has a high affinity for fatty tissues, and can readily accumulate to highly toxic levels (Liu et al., 2018). Organo-Hg poisoning epidemics have occurred in countries such as Iraq and Japan - where the infamous Minamata disaster occurred. At present, Hg pollution is affecting many communities around the world (Wang et al., 2012), with ~8 million people being potentially impacted (Pure Earth and Green Cross, 2016). Moreover, soil pollution in many parts of the world is pervasive (Jin et al., 2019; Peng et al., 2019; Zhang et al., 2019), with a numerous complications surrounding land management (Hou and Li, 2017). As the pharmacokinetics and toxicological effects of mercury compounds have been comprehensively reviewed elsewhere (Rice et al., 2014), this review has focused on factors that may impact risk assessment for the long-term management of different Hg species in soil, which is an imperative for successfully managing risk and, therefore, successful implementation of the Minamata Convention (World Bank, 2016).

It is established that Hg is highly attracted to certain soil constituents, particularly, organic matter and soil mineral surfaces. Mercury can form covalent bonds with the O atoms of organic matter functional groups, and can react with sulfides to form the highly stable

HgS mineral cinnabar. This makes Hg relatively immobile; under normal environmental conditions, Hg is usually only found to reside only in superficial soils, with low transportation potential to deeper soils. Therefore, it may be prudent to focus health risk assessment on surface soil exposure scenarios. However, the complexity of Hg-soil systems is increasingly being unraveled. For example, although generally small fractions of total soil Hg will partition to pore water, the extent of this partitioning will depend on Hg speciation at a site. Moreover, stable insoluble compounds such as HgS may become soluble when soil solutions contain certain DOM substances (e.g. fulvic acids) or thiol ligands, or, if there is an excess of sulfide ions present. This is due to the formation of soluble complexes, such as the mercuric disulfide complex. The reactions that form these complexes are highly pH dependent. Such complexities should be considered in detailed site specific risk assessments.

Under the chemically reducing conditions found in many permanently or periodically flooded soils, Hg may be biogeochemically transformed into the more toxic organo-Hg forms (Frohne et al., 2012). Moreover, Methyl-Hg formed in rice paddy soils, which are periodically flooded by farmers, is susceptible to being taken up into the rice grain produce, thus posing a risk to health through the food chain (Zhang et al., 2010). The bioavailability of different Hg species is a crucial factor in this process, as some Hg forms are more readily available to methylating microorganisms. For example, mobile Hg-humic or Hg-fulvic complexes tend to be susceptible to Hg methylation, whereas, Hg bound with larger organic matter types is at less at risk of methylation. The direct conversion of insoluble HgS (cinnabar) species to Methyl-Hg in anaerobic soils is generally believed to be low, but this can change when environmental conditions favor HgS complexation. Methylation potential may be associated with intermediate geochemical products deriving from rate-limited Hg-sulfide precipitation reactions, but this is still not well understood. Recent reports suggest that nano-sized HgS particles may be bioavailable for methylation by microorganisms (Thomas et al., 2018). It is also reported that organo-Hg is more likely found at sites of recent contamination (Han et al., 2006).

It is imperative that Hg contaminated land is managed in a way that Hg species and behavior are incorporated as part of risk assessment. Some nations have already adopted soil environmental quality standards that take into account Hg speciation and soil conditions (Table 4). For example, the US EPA has implemented separate screening levels for total Hg, methyl Hg, and mercuric chloride (and other Hg salts), and China introduced their 2018 soil screening values for total Hg and methyl Hg with soil pH specific levels, due to the differences in bioavailability of Hg under different pH conditions. However, there remains a need for greater consideration of bioavailability in environmental risk assessment in general (O'Connor et al., 2018b). There is also a need for remediation engineers to have a better understanding of how soil remediation methods influence contaminant speciation and transportation. For example, some researchers are now using the addition of reduced S modified sorbents in soils. As we have noted, Hg can bind strongly to reduce S species, thus immobilizing Hg in situ, with low apparent risk to receptors. However, we suggest that further attention should be paid to the soil environmental conditions, and whether or not a long-term residual risk remains after remedial actions have been completed. Residual risks may arise due to complexation, methylation or colloidal transport processes. Researchers have also recently proposed the use of increased Hg flux to clean up soils via microorganism augmentation, which facilitates Hg transformation to more volatile species. It is suggested that further attention is paid to the long-term potential changes of environmental factors such as the frequency of rainfall events and ambient conditions, so that better predictions of remediation efficacy can be made.

8. Summary and outlook

The global risk posed to humans and the wider environment by Hg contaminated soils is severe, with anthropogenic emissions of Hg to the environment being on the order of 2 Gg per year. This review has examined research published in recent years in order to garner a better knowledge of Hg behavior in soils, which may allow a better understanding and management of contaminated sites.

This review has examined recent studies regarding Hg transport processes, such as how colloidal particles have the potential to act as carriers of bound Hg within soil pores, presenting a potential pathway for Hg transportation and increasing availability. Mercury can readily bind to colloids due to their high specific surface area and the presence

Table 4
Soil guideline values for Hg in various countries.

Country	Values (mg/kg)				Guideline
Total-Hg					
Canada	6.6^{e} (Residential/parkland)	50^{e} (Industrial)	6.6^{e} (Agricultural)	24^{e} (Commercial)	(CCME, 1999)
China	8^{a} (Residential land, school, hospital, parks, etc.)	38^{a} (Industrial land, commercial land, roads, public service land, etc.)	33^{b} (Residential land, school, hospital, parks, etc.)	82^{b} (Industrial land, commercial land, roads, public service land, etc.)	(MEE, 2018b)
	0.5^{a} (Paddy field, pH ≤ 5.5)	0.5^{a} (Paddy field, 5.5 < pH ≤ 6.5)	0.6^{a} (Paddy field, 6.5 < pH ≤ 7.5)	1.0^{a} (Paddy field, pH > 7.5)	(MEE, 2018a)
	1.3^{a} (Other agricultural land, pH ≤ 5.5)	1.8^{a} (Other agricultural land, 5.5 < pH ≤ 6.5)	2.4^{a} (Other agricultural land, 6.5 < pH ≤ 7.5)	3.4^{a} (Other agricultural land, pH > 7.5)	
	2.0^{b} (pH ≤ 5.5)	2.5^{b} (5.5 < pH ≤ 6.5)	4.0^{b} (6.5 < pH ≤ 7.5)	6.0^{b} (6.5 < pH ≤ 7.5)	
USA	11^{c} (Resident soil)	46^{c} (Industrial soil)	0.033^{d} (Protection of ground water)		(US EPA, 2018)
Methyl-Hg					
China	5^{a} (Residential land, school, hospital, parks, etc.)	45^{a} (Industrial land, commercial land, roads, public service land, etc.)	10^{b} (Residential land, school, hospital, parks, etc.)	120^{b} (Industrial land, commercial land, roads, public service land, etc.)	(MEE, 2018b)
USA	7.8^{c} (Resident soil)	120^{c} (Industrial soil)	14^{d} (Protection of ground water)		(US EPA, 2018)
Mercuric chloride (and other Hg salts)					
USA	23^{c} (Resident soil)	350^{c} (Industrial soil)			(US EPA, 2018)

Note: ^{a}Risk screening value; ^{b}Risk intervention value; ^{c}Screening Levels; ^{d}Risk-based Soil Screening Level (SSL); ^{e}Soil quality guidelines for mercury.

of surface functional groups. On examining Hg flux from soils to the atmosphere we found that mechanisms have been put forward that describe gaseous Hg release associated with the progressive stages of rainfall events. The influence of ambient conditions was also found to be critical, with atmospheric chemistry, such as O_3 levels, playing a crucial role in Hg flux. Mercury contaminated soils constitute complex systems where many interdependent factors, including amount and composition of soil organic matter and clays, oxidized minerals (e.g. Fe oxides), reduced elements (e.g. S^{2-}), as well as soil pH and redox conditions affect Hg forms and transformation. Speciation influences the extent and rate of Hg subsurface transportation, which often has been assumed insignificant. Nano-sized Hg particles as well as soluble Hg complexes play important roles in Hg mobility, availability, and methylation in soils.

Considering the global scale and hazardous nature of Hg contaminated sites, the need to develop new, effective, and preferably 'green' remediation technologies to cleanup Hg contaminated soils is now urgent (O'Connor et al., 2018c). This review focused on improving the understanding of Hg behavior in soil, which may allow optimization of risk management and improved remediation approaches to protect human health. Going forward, new approaches may involve engineering soil environments so that Hg behaves in a more favorable way for its removal or immobilization. For example, researchers have recently begun to consider the use of citric acid facilitated thermal treatments for the remediation of Hg contaminated soils (Ma et al., 2015), to apply microorganisms for increased Hg flux (with capture) (Mahbub et al., 2017; Mahbub et al., 2016), and Hg biosorption methods (Dash and Das, 2015; K. Yin et al., 2016). Such approaches fit well with the current movement to promote green & sustainable remediation (GSR) practice (O'Connor et al., 2018d; Song et al., 2019; Zhang et al., 2018) in preference of resource/energy intensive traditional Hg remediation approaches such as conventional thermal treatment or soil washing (US EPA, 2007b).

Environmental pollution by Hg is a global problem. Often, authorities are sanctioned by law to conduct appropriate risk assessment and to implement practical actions that mitigate environmental hazards to acceptable risk levels (Table 4), but often without consideration of possible wider impacts (O'Connor and Hou, 2018). Wider ranging risk assessments with a view to land ecosystems (vegetation as green fodder and hay production, grazing and wild animals), in order to evaluate the potential transfer of Hg into the human food chain, should also be encouraged. This will require accurate determination of Hg species, soil constituents, fluxes, and their dynamics, since gaseous Hg exchange from soil to atmosphere is an important pathway to the environment. However, reliable estimates of Hg fluxes in certain areas around the world, and its influencing factors, remain scarce. It is recommended that further research should be implemented to assess this pathway in terms of impacts to human health.

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