Atmospheric Environment 43 (2009) 5853–5864

Contents lists available at ScienceDirect

Atmospheric Environment

journal homepage: www.elsevier.com/locate/atmosenv

Review

A review of current knowledge concerning dry deposition of atmospheric mercury

Leiming Zhang [a,*], L. Paige Wright [b], Pierrette Blanchard [a]

[a] Air Quality Research Division, Science and Technology Branch, Environment Canada, 4905 Dufferin Street, Toronto, Ontario M3H 5T4, Canada
[b] 1122 Bavlie Ave., Ottawa, Ontario K1H 8H1, Canada

ARTICLE INFO

Article history:
Received 11 May 2009
Received in revised form
20 August 2009
Accepted 20 August 2009

Keywords:
Air–surface exchange
Dry deposition velocity
Measurement and modelling
Speciated mercury

ABSTRACT

The status of the current knowledge concerning the dry deposition of atmospheric mercury, including elemental gaseous mercury (Hg^0), reactive gaseous mercury (RGM), and particulate mercury (Hg_p), is reviewed. The air–surface exchange of Hg^0 is commonly bi-directional, with daytime emission and nighttime deposition over non-vegetated surfaces and vegetated surfaces with small leaf area indices under low ambient Hg^0 conditions. However, daytime deposition has also been observed, especially when the ambient Hg^0 is high. Typical dry deposition velocities (V_d) for Hg^0 are in the range of 0.1–0.4 cm s^{-1} over vegetated surfaces and wetlands, but substantially smaller over non-vegetated surfaces and soils below canopies. Meteorological, biological, and soil conditions, as well as the ambient Hg^0 concentrations all play important roles in the diurnal and seasonal variations of Hg^0 air–surface exchange processes. Measurements of RGM deposition are limited and are known to have large uncertainties. Nevertheless, all of the measurements suggest that RGM can deposit very quickly onto any type of surface, with its V_d ranging from 0.5 to 6 cm s^{-1}. The very limited data for Hg_p suggest that its V_d values are in the range of 0.02–2 cm s^{-1}.

A resistance approach is commonly used in mercury transport models to estimate V_d for RGM and Hg_p; however, there is a wide range of complexities in the dry deposition scheme of Hg^0. Although resistance-approach based dry deposition schemes seem to be able to produce the typical V_d values for RGM and Hg^0 over different surface types, more sophisticated air–surface exchange models have been developed to handle the bi-directional exchange processes. Both existing and newly developed dry deposition schemes need further evaluation using field measurements and intercomparisons within different modelling frameworks.

1. Introduction

Mercury (Hg) bioaccumulation in fish (primarily as methylmercury) has caused health problems in humans as well as wildlife (Clarkson, 1990; Wolfe et al., 1998). The issue has spurred an increased interest in understanding the Hg cycling among the atmospheric, terrestrial, and aquatic systems over the past few decades. Substantial knowledge on atmospheric Hg has been gained through field and laboratory studies and numerical modelling exercises, as summarized in several review papers published over the past two decades. These reviews include general overviews of atmospheric Hg (Lindqvist and Rodhe, 1985; Schroeder and Munthe, 1998); the chemistry of atmospheric Hg (Lin and Pehkonen, 1999); and the sources and sinks of atmospheric Hg (Schroeder et al., 1989; Gustin and Lindberg, 2005; Gustin et al., 2008). The scientific uncertainties in atmospheric Hg models were discussed in Lin et al. (2006) and comparisons of several different Hg models can be found in Ryaboshapko et al. (2007a,b) and Bullock et al. (2008).

Quantifying Hg total (dry + wet) deposition is needed in order to reduce the large gaps that exist in the global Hg mass balance estimates (Mason and Sheu, 2002). Quantifying Hg deposition and attributing the sources of the deposited Hg are also necessary for the development of policies relating to the control of Hg emissions (Lindberg et al., 2007). Monitoring networks with properly chosen monitoring stations can provide more accurate estimates of wet deposition at regional scales than Hg transport models since wet deposition is collected directly from the precipitation chemistry data; these networks will also improve dry deposition estimates substantially if the ambient concentrations of speciated Hg are monitored (Keeler and Dvonch, 2005; Mason et al., 2005).

* Corresponding author. Tel.: +1 416 739 5734; fax: +1 416 739 4281.
E-mail address: leiming.zhang@ec.gc.ca (L. Zhang).

1352-2310/$ – see front matter
doi:10.1016/j.atmosenv.2009.08.019

Atmospheric Hg is commonly grouped into three forms: gaseous elemental mercury (Hg^0), reactive gaseous mercury (RGM), and particulate mercury (Hg_p). The dry deposition of Hg^0 has been frequently omitted since it is considered to be a highly emitted species (Ryaboshapko et al., 2007a). However, some studies suggest that Hg^0 can contribute substantially to total Hg dry deposition over vegetated surfaces since Hg^0 makes up >95% of the total atmospheric Hg (see discussions in Lindberg et al., 2007; Gustin et al., 2008). Current estimates of the global Hg balance suggest that there appear to be too many sources of Hg and not enough sinks (Gustin and Lindberg, 2005). The uncertainties in the dry deposition estimates are also much larger than in the wet deposition estimates (Mason and Sheu, 2002; Lindberg et al., 2007). In order to improve the total Hg dry deposition estimates, a dry deposition model that includes all three forms of Hg is needed, especially considering that, under certain circumstances, dry deposition can contribute to the total deposition as much as, or even more than wet deposition (e.g., Munthe et al., 2004; Sakata et al., 2006; Graydon et al., 2008).

The concept of the dry deposition velocity (V_d) has been used for decades in numerical transport models as well as in monitoring networks to associate a chemical species' mass flux density to the surface with its ambient concentrations, i.e., a species' flux is a product of its V_d and its concentration; and V_d values are commonly estimated using a dry deposition scheme/parameterization (Wesely and Hicks, 2000). Developing a dry deposition scheme for Hg that can be used for regional/global Hg models requires a quantitative understanding of the depositional processes of the three forms of Hg (Hg^0, RGM, and Hg_p) over different surface types and under different meteorological, biological and soil conditions. Previous reviews have presented thorough summaries on the general knowledge of the deposition and emission processes (Schroeder and Munthe, 1998; Gustin and Lindberg, 2005; Lindberg et al., 2007; Gustin et al., 2008); what is lacking is a database of V_d values from available measurements. The purpose of this paper is to build such a database through a detailed review of the published measurements. Note that many of the published measurements did not provide dry deposition velocities and, wherever possible, their values have been calculated in this paper based on the flux and concentration data presented in the original publications. To gain a complete picture of the dry deposition processes, a brief review of the measurement methods and of the existing dry deposition schemes that have been applied to the Hg species is also given here.

2. Measurements

2.1. Measurement methods

There are a number of experimental methods used in the measurement of atmospheric Hg deposition. These include enclosure methods, such as the dynamic flux chambers (DFC); the use of surrogate surfaces (SS); and micrometeorological methods, including the modified Bowen ratio (MBR) method, the aerodynamic (AER) method, and the relaxed eddy accumulation (REA) method. Litterfall and throughfall methods have also been used as a rough estimation of dry deposition (e.g., Munthe et al., 2004; Graydon et al., 2008).

Enclosure methods employ flux chambers to measure the fluxes. DFCs have been used in studies over wetlands in Quebec (Poissant et al., 2004a; Zhang et al., 2005), Ontario (Graydon et al., 2006), and the Florida Everglades (Marsik et al., 2005); over a deciduous forest in Tennessee (Kuiken et al., 2008) and a boreal forest in Ontario (Hintelmann et al., 2002); over background soils across the United States (Ericksen et al., 2006); and over lakes and forest soils in southwestern Sweden (Xiao et al., 1991). A laboratory gas exchange chamber was used to examine a variety of natural soils collected from several locations across the United States (Xin and Gustin, 2007).

A surrogate surface is a surface composed of a material such as Teflon or polysulfone that is placed at a site for a designated amount of time, allowing Hg to dry deposit onto it (Sakata and Marumoto, 2004). The surface is then washed and analyzed, similar to leaf wash methods (Rea et al., 2000). Surrogate surfaces and water surface samplers have been used to measure Hg at a hardwood forest site in Michigan (Rea et al., 2000); over pastureland in rural and suburban Nevada (Lyman et al., 2007); over wetlands in Florida (Marsik et al., 2007); and over arid lands in south central New Mexico (Caldwell et al., 2006). They have also been used to observe dew over wetland vegetation in Florida (Malcolm and Keeler, 2002); and to measure Hg_p on a rooftop in Tokyo (Sakata and Marumoto, 2004). Leaf washes were also performed in the hardwood forest in Michigan for comparison to the results from the surrogate surfaces (Rea et al., 2000).

Micrometeorological methods have been used to measure fluxes for several different surface types. The MBR method has been used over wetlands in Quebec (Poissant et al., 2004b) and Florida (Lindberg et al., 2002b); over deciduous forests and their background soils within the Walker Branch Watershed (Kim et al., 1995; Lindberg et al., 1998); over a young pine plantation in Tennessee and a boreal forest floor in Sweden (Lindberg et al., 1998); over grassland sites in the United States, Austria, and Switzerland (Lindberg and Stratton, 1998; Obrist et al., 2006; Fritsche et al., 2008a,b; Mao et al., 2008); as well as over pavement surfaces in Indiana (Lindberg and Stratton, 1998). The REA method (also known as the conditional sampling method) is a relatively new method that has been used for flux measurements of RGM over snow in Alaska (Lindberg et al., 2002a; Skov et al., 2006); over modified soil surfaces in Sweden (Olofsson et al., 2005); over a leafless hardwood forest in Connecticut (Bash and Miller, 2007); and over a cornfield in Minnesota (Cobos et al., 2002). The aerodynamic method has been used to measure total gaseous Hg fluxes over a rice paddy field in Korea (Kim et al., 2003); Hg^0 fluxes over a snow surface in Nunavut (Cobbett et al., 2007); an agricultural field in Ontario (Cobbett and Van Heyst, 2007); and subalpine grassland sites in Austria and Switzerland (Fritsche et al., 2008a,b).

An additional measurement method, the so called $^{222}Rn/Hg^0$ method, has recently been used for the measurement of Hg^0 fluxes at a grassland site in Seebodenalp, Switzerland and at an urban site in Basel, Switzerland (Obrist et al., 2006). It was used during periods with a stable nocturnal boundary layer and was found to be an effective method for the measurement of Hg^0 fluxes in situations where the atmospheric conditions are non-turbulent, the fluxes are small, or the surface is highly heterogeneous (Obrist et al., 2006).

Each measurement method mentioned above has its share of both positive and negative aspects. DFCs can provide immediate results and are relatively simple to use, but they can alter local conditions such as the temperature, humidity and turbulence (Cobos et al., 2002; Fritsche et al., 2008a). Some enclosure methods may encounter condensation building up inside the bag during the nighttime (Zhang et al., 2005). It has also been determined that the fluxes measured using enclosure methods are lower than those measured using micrometeorological methods (Gårdfeldt et al., 2001). The AER approach requires measurements at various heights, thereby increasing the amount of data analysis required (Cobos et al., 2002). The MBR method provides relatively accurate results but is complicated by the fact that it requires measurements of a second gas in addition to Hg (Cobos et al., 2002). Both AER and MBR method require a horizontal fetch (distance from the leading edge) which is typically 20–100 times the measurement height.

The REA method is attractive since it requires measurements at only one height and for only one gas (Olofsson et al., 2005). Unfortunately, its accuracy can be decreased by small gradients. Drawbacks to the use of surrogate surfaces include the need to cover the surfaces during rain events, thus creating gaps in the data collection (Marsik et al., 2007); the difficulty in determining the speciation of Hg (gaseous vs. particulate) (Rea et al., 2000); and the uncertainties associated with the fact that these surfaces are not real life and therefore cannot accurately duplicate every aspect of deposition.

It is worth noting that the degree of uncertainty in the measurement varies considerably with the measurement method used. For example, Fritsche et al. (2008a) measured Hg^0 air–surface exchange over grassland using two micrometeorological methods simultaneously. The average V_d for the vegetation period was 0.09 cm s^{-1} with the AER method, but only 0.03 cm s^{-1} with the MBR method. Results from a dry deposition campaign, which measured Hg dry deposition using a number of different methods, will probably shed some light on this topic (Dry Deposition Measurement Intercomparison and Workshop, University of Michigan, August 2008).

2.2. Hg^0 measurements

There have been a fair number of field studies that have measured Hg^0 fluxes over various surface types using several of the different measurement methods discussed in Section 2.1. These studies have identified the phenomenon of the bi-directional air–surface exchange of Hg^0 (see reviews by Schroeder and Munthe, 1998; Gustin et al., 2008). Downward fluxes have been observed to be the major pathway in some field studies (e.g., Lee et al., 2000; Obrist et al., 2006; Fritsche et al., 2008a,b), while upward fluxes have been found to be the dominant exchange in many others (see reviews in Schroeder et al., 2005; Gustin et al., 2006).

Fig. 1 is a schematic description of a resistance model designed for the discussions in Section 3. This scheme can help us to better understand the observed bi-directional exchange features. Atmospheric Hg^0, after being transported down to the canopy level and passing through a thin quasi-laminar sublayer, can either deposit onto the leaf surfaces (cuticular uptake), enter into the leaf stomata (stomatal uptake), or be transported further down to the soil surface and deposit there. At the same time, the Hg stored in the soil, the leaf stomata, and the leaf cuticle can be released under certain conditions, creating a bi-directional exchange over the soil surface and through the leaf surface. The fluxes measured above the canopy are the net effects of the deposition and emission processes described above. For surfaces with no canopy (bare soil, snow, water), only one exchange process exists with no additional complication of canopy leaf uptake/emission.

Theoretically, a 'surface exchange velocity' should be defined in order to quantify the bi-directional surface exchange processes, e.g., a positive value represents a downward flux while a negative value represents an upward flux. However, the majority of mercury transport models treat the upward (emission) and downward (deposition) fluxes separately; and both of these two processes need to be parameterized in these models. Thus, the definition of dry deposition is still used here and it only represents the downward flux process. This section focuses on quantifying the dry deposition of Hg^0, in particular, its V_d values over different surface types, the diurnal and seasonal patterns of V_d, as well as the effects of wetness and meteorological conditions on the observed V_d. Emissions are only briefly discussed, when needed, to explain the observed V_d patterns.

A summary of published V_d values for Hg^0 is presented in Table 1. Some of these V_d values have been published while others have been calculated in this study using the published values of the measured dry deposition fluxes and concentrations. The standard deviation of V_d has also been estimated from the standard deviation of the fluxes and the concentrations according to Boas (1996):

$$\sigma V_d = V_d \times \left[\left(\frac{\sigma F}{F}\right)^2 + \left(\frac{\sigma C}{C}\right)^2\right]^{\frac{1}{2}}$$

The V_d values from some of the studies could not be generated due to insufficient information (Rea et al., 2002 and several of their earlier studies; Kim and Kim, 1999; Cobos et al., 2002; Poissant et al., 2004a; Ericksen et al., 2005; Caldwell et al., 2006; Gabriel et al., 2006; Gustin et al., 2006; Narukawa et al., 2006; Sakata et al., 2006 and several of their earlier studies; Marsik et al., 2007; Nguyen et al., 2008). These studies have therefore not been included in Table 1, although some of them have been included in the discussions below where applicable.

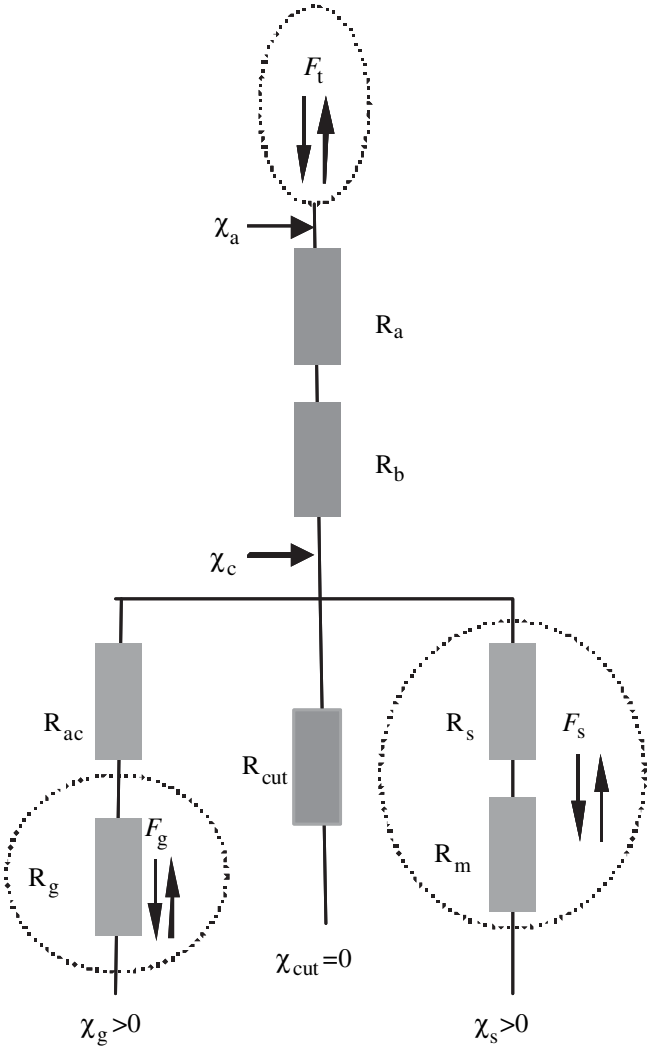

Fig. 1. Scheme of a resistance model including bi-directional exchange through stomata and over soil surface. R_i is the resistance terms including R_a, aerodynamic; R_b, quasi-laminar; R_{ac}, in-canopy aerodynamic; R_g, soil; R_{cut}, cuticle; R_s, stomatal resistance; and R_m, mesophyll resistance associated with stomatal resistance. χ_a is concentration at a reference height above the canopy; χ_c, χ_s and χ_g are canopy, stomatal and soil compensation points, respectively. F_t is overall flux at the reference height, and F_s and F_g are fluxes through stomata and above soil surface, respectively.

Table 1
A summary of V_d values for Hg^0 (mean ± one standard deviation, or range values in bracket). Note that some measurements also included reactive gaseous mercury, see TGM (=Hg^0 + RGM) in the 'Species' column.

Surface	Location	Season	Species	Method	V_d (cm s^{-1})	Condition	Remarks	Reference
Background soils								
Mixed forest soil	Walker Branch, TN, USA	Summer 1993 Fall 1993	Hg^0	MBR	0.03 ± 0.024 0.009		Measurements during adverse conditions omitted, emission occurred 70% of the time	Kim et al., 1995
Coniferous forest soil	Lake Gardsjon, Sweden	Spring 1994	Hg^0	MBR	0.015–0.041[a] 0.031–0.064[a]	Dry soil Wet soil	Daytime data, emission occurred 60% of the time	Lindberg et al., 1998
Pine forest soil	Yellowstone National Park		TGM	DFC	0.003 ± 0.0007[a]	Nighttime	Mean fluxes showed soil emission at many other sites across USA	Ericksen et al., 2006
Deciduous forest floor litters	Standing Stone State forest, TN	Spring 2004	TGM	DFC	0.002 ± 0.005[a]	Daytime	Emission occurred 80% of the time	Kuiken et al., 2008
Coniferous forest soil	Lake Gardsjon, Sweden	Winter 1988 Spring 1989	TGM	DFC	0.011 ± 0.01[a] 0.01 ± 0.008[a]	Daytime Day and night	Deposition was observed when $T < 3$ °C and emission was observed at $T = 10$ °C	Xiao et al., 1991
Substrates and natural soil	Various locations, USA	Laboratory	Hg^0	GEC	0.01 ± 0.012[a] 0.01 ± 0.008[a] 0.02 ± 0.007	Low C, night High C, day High C, night	Higher deposition under high ambient concentration (C) and dark condition	Xin and Gustin, 2007
Forest Canopies								
Hardwood foliage seeding	Greenhouse	Laboratory	Hg^0	GEC	0.012–0.015[a]		Deposition at high Hg^0 air concentration, compensation points defined	Hanson et al., 1995
Mature deciduous	Walker Branch, TN, USA	Summer-Fall 1993	Hg^0	MBR	0.4 ± 0.3 1.3 ± 1.8	Dry canopy Wet canopy	Daytime data, site close to local sources, emission more frequent than dry deposition	Lindberg et al., 1998
Pine plantation	Wartburg, TN, USA	Fall 1994	Hg^0	MBR	0.35–0.55[a] 0.26[a]	Dry canopy Dew canopy	Daytime data, data for dry deposition were non-significant, emission more frequent	
Quaking aspen foliage	Nevada, USA	2000 2001 Summer 2001	Hg^0	SS SS GEC	$4.7 \pm 2.2 \times 10^{-4}$ $5.6 \pm 3.3 \times 10^{-4}$ 0.0003–0.2		Hg in foliar tissue originated from atmospheric deposition	Ericksen et al., 2003
Quaking aspen foliage	Nevada, USA		Hg^0	GEC	0.002–0.048[a]	Various ambient and soil Hg content	Emission at background ambient concentration (C), deposition at higher C	Ericksen and Gustin, 2004
Red maple forest	Connecticut, USA	Spring 2006	TGM	REA	1.88[a]	High concentration	Deposition observed when adjacent field tilled and manure spread	Bash and Miller, 2007
Agricultural field								
Rice paddy field, seedlings	Kang Hwa Island, Korea	Spring 2001 Spring 2002	Hg^0	AER	0.22 0.28		Deposition mostly occurred during early morning or late evening	Kim et al., 2003
Biosolids amended field	Ontario, Canada	Fall 2004	Hg^0	AER	0.05 ± 0.005[a]		Peak deposition during biosolids application event	Cobbett and Van Heyst (2007)
Bare paddy field	Kang Hwa Island, Korea	Spring 2001	TGM	AER	0.26 ± 0.01[a]		Emission during daytime and deposition during nighttime	Kim et al., 2002
Grasslands								
Subalpine grassland	Seebodenalp, Switzerland	Summer 2004	Hg^0	$^{222}Rn/Hg^0$ MBR MBR	0.003 ± 0.005[a] 0.03 ± 0.007[a] 0.03 ± 0.004[a]	Stable night Turbulent night Day & night	Net deposition (emission < deposition)	Obrist et al., 2006
Subalpine grassland	Fruebel, Switzerland	2005–2006	Hg^0	AER MBR	0.09 0.03	Vegetation period for both methods	Uncontaminated area; deposition dominated	Fritsche et al., 2008a
Subalpine grassland	Fruebel, Switzerland	Summer 2006	Hg^0	AER MBR	0.1 ± 0.16 0.04		Uncontaminated area; deposition dominated; diurnal pattern found	Fritsche et al., 2008b
Flat grassland	Neustift, Austria	Summer 2006	Hg^0	AER MBR	0.05 ± 0.16 0.01		Uncontaminated area; deposition dominated; diurnal pattern found	
Wetlands								
Mixed vegetation, soil, water	Quebec, Canada	Late summer 2002	Hg^0	MBR	0.19 (0.01–1.8)		Large net daytime deposition observed when site impacted by regional Hg sources	Poissant et al., 2004b
Cattail (uprooted, floating) – flooded	Florida Everglades, USA	Winter 1998	Hg^0	MBR	0.14 ± 0.13	Daytime, abnormal conditions	Mostly emission during daytime, fluxes close to zero during nighttime	Lindberg et al., 2002b

Table 1 (*continued*)

Surface	Location	Season	Species	Method	V_d (cm s^{-1})	Condition	Remarks	Reference
River bulrush – flooded	Quebec, Canada	Late summer 2003	Hg^0	DFB	0.02 ± 0.018[a]		Calculated from mean values of all data (from both emission and deposition)	Zhang et al., 2005
Sawgrass, cattail marsh	Florida Everglades, USA	Summer 2000	Hg^0	DFC	0.02 ± 0.07[a]	Nighttime, wet season	Mostly emission during daytime and deposition during nighttime	Marsik et al., 2005
Salt marsh	Connecticut, USA	Spring 1998 Summer 1998	TGM	AER	0.006 ± 0.135[a] 0.05 ± 0.181[a]	Early growth full foliage	Deposition dominated during full foliage period, even during daytime	Lee et al., 2000
Contaminated sites								
Landfill	Nan-Ji-Do, Korea	Spring 2000	TGM	MBR	5.01 ± 5.45 1.26 ± 1.52	Impacts from vents Not impacted	Strong emission and deposition observed	Kim et al., 2001
Landfill	Bang Chun, Korea	January 2004	TGM	AER	0.51 ± 1.39[a]		Emission occurred 58% of the time	Nguyen et al., 2008
Mining area	Guizhou, China	Winter 2003 winter 2004	TGM	DFC	0.05–0.14[a]	Range of mean value at 5 sites	Net deposition at 3 of the 5 sites, higher deposition over crops and/or close to sources	Wang et al., 2007a
Mining area	Guizhou, China	Winter 2002, summer 2004	TGM	DFC	0.0–0.17[a]	Range of mean value at 8 sites	Net emission at majority of the 8 sites	Wang et al., 2007b
Snow and water								
Snowpack	Alert, Nunavut, Canada	Winter–spring 2005	Hg^0	AER	0.00025 ± 0.0025[a] 0.00008 ± 0.0008[a]	Julian day 31–62 Julian day 31–174	Calculated from mean values of all data (from both emission and deposition)	Cobbett et al., 2007
River	Knobesholm, Sweden	Summer 1999	TGM	DFC	0.012 ± 0.014[a]	Nighttime	Weak deposition only occurred during night	Gårdfeldt et al., 2001
Seawater	Fjord Gullmaren, Sweden	Summer 1997, summer 1998	TGM	DFC	0.003–0.006[a]	Nighttime	Weak deposition only occurred during night	

[a] V_d values were not reported in the original paper and were calculated using only deposition fluxes whenever possible; AER: Aerodynamic Method; DFB: Dynamic Flux Bag; DFC: Dynamic Flux Chamber; GEC: Gas Exchange Chamber; LW: Leaf Wash; MBR: Modified Bowen Ratio; REA: Relaxed Eddy Accumulation; RMC: Refluxing Mist Chamber; SS: Surrogate Surface; WSS: Water Surface Sampler.

In the following sections, deposition over background soils, vegetated canopies (including some wetlands covered by vegetation), contaminated sites, and snow and water surfaces are discussed separately. Earlier studies that focused on emission fluxes over naturally Hg-enriched soils are not discussed here (see Gustin and Lindberg, 2005 and references therein). There are also a few studies conducted at urban sites over different surfaces (e.g., bare soil, grass, pavement) (Kim and Kim, 1999; Gabriel et al., 2006; Obrist et al., 2006). Data from Kim and Kim (1999) showed higher concentrations and fluxes than some contaminated sites, but data from Gabriel et al. (2006) and Obrist et al. (2006) showed little difference from background sites. Thus, only the study of Kim and Kim (1999) is included in the 'Contaminated sites' section.

2.2.1. Background soils

Measurements of Hg^0 fluxes over background forest soils have frequently shown the dominance of emission over deposition. A few studies listed in Table 1 (Kim et al., 1995; Lindberg et al., 1998; Kuiken et al., 2008) have suggested that emission occurred 60–80% of the time. Ericksen et al. (2006) measured Hg^0 fluxes over background soils of agricultural, desert, grassland, mixed and pine forest ecosystems; the mean fluxes were upward (emission) under dark and light conditions at all locations except for the soil at the pine forest under dark conditions for which a low mean downward (deposition) flux was observed. Gabriel et al. (2006) observed net emission fluxes over bare soil in an urban area during every season and net emission or deposition fluxes over pavement depending on the season.

V_d values over background soils are generally smaller than 0.05 cm s^{-1}. If only the downward flux data are considered, V_d values are typically on the order of 0.01 cm s^{-1}. There is evidence that soil moisture enhances the soil Hg emission fluxes (e.g., Gustin and Stamenkovic, 2005; Ericksen et al., 2006); thus, the net deposition fluxes should be decreased under wet soil conditions. However, under conditions when Hg^0 emission was minimal, the soil wetness could also increase the deposition of Hg^0. For example, Lindberg et al. (1998) observed that the V_d was ~50% higher over wet soils than over dry soils.

Diurnal variations have been found for V_d over soil surfaces in several studies. For example, data from Kim et al. (1995) showed small bi-directional fluxes in the morning, peak emissions near the mid-afternoon, and insignificant exchange during the nighttime. Similar diurnal patterns have also been found in several other studies (Cobos et al., 2002). These phenomena also agree qualitatively with results from a laboratory study which showed emission during light conditions and deposition in the dark under low Hg^0 air concentrations (Xin and Gustin, 2007). Few of the studies covered multiple seasons. A study by Gabriel et al. (2006) showed quite large seasonal variations in the emission/deposition fluxes over soil and pavement surfaces in an urban area.

The net flux from emission and deposition depends on many factors, e.g., meteorological conditions (solar radiation, temperature, wind speed), soil conditions (soil Hg content, moisture, pH, and organic matter), and the ambient Hg^0 concentration; therefore it is not possible to generalize the diurnal and seasonal patterns to all locations. For example, while daytime emission and nighttime deposition were observed under low Hg^0 air concentrations, deposition was the major pathway in both light and dark conditions if the ambient concentration of Hg^0 was sufficiently high (Xin and Gustin, 2007). The day/night emission/deposition pattern also varied from month to month and from location to location (e.g., Gustin et al., 2006). Nevertheless, these different diurnal and seasonal patterns were identified to be caused by different dominant factors (e.g., soil moisture, temperature, light intensity). Our

handling of Hg emission and deposition within the Hg budget will be improved if the effects of these factors on the net flux can be quantified.

2.2.2. Vegetated canopies (including wetlands)

V_d values over vegetated canopies (forest canopies, grasslands, agricultural fields, and wetlands) shown in Table 1 cover a large range, e.g., 0.0–2.0 cm s^{-1}. This might be explained by the fact that both deposition and emission intensities vary substantially with meteorological, biological, soil, and ambient conditions. The largest V_d values would most probably be for the conditions with the strongest deposition and the weakest emission.

Earlier studies suggest that many gaseous species (e.g., inorganic species commonly considered in air-quality models) tend to deposit faster to canopies with larger leaf area indices, smaller minimum stomatal resistances, and larger roughness lengths than to other canopies (Wesely and Hicks, 2000; Zhang et al., 2003). This theory also seems to apply to Hg^0, as shown in Table 1. For example, measurements over three different forests (Lindberg et al., 1998; Bash and Miller, 2007) showed the V_d to be 0.4 cm s^{-1}, on average, over dry canopies, and 3–5 times higher over wet forests or forests with high ambient concentration conditions. Note that the small V_d values for the forest canopies shown in Table 1 were from measurements conducted using individual tree leaves rather than entire forest canopies. Typical V_d values were around 0.2–0.3 cm s^{-1} over agricultural fields, smaller than the 0.2 cm s^{-1} measured over wetlands, and smaller than the 0.1 cm s^{-1} over grasslands. Apparently, V_d values over forests can be as much as 2–5 times higher than over other vegetated canopies. The very high V_ds over forests can be explained by their large leaf area indices and large roughness lengths, and the fairly high V_ds over agricultural fields might be explained by their small stomatal resistances and large roughness lengths.

Canopy wetness seems to enhance Hg^0 deposition. The V_d was observed to be three times higher when the canopy was wet than dry over a mature deciduous forest canopy (Lindberg et al., 1998). Marsik et al. (2005) also observed deposition fluxes immediately after precipitation followed by emission fluxes while the canopy dried by solar radiation. Fritsche et al. (2008b) have suggested that Hg^0 co-deposits with dew at night. The increased deposition over wet canopies is probably caused by aqueous-phase chemistry which converts Hg^0 to other mercury species.

Strong diurnal variations in the Hg^0 air–surface exchange over vegetation have been observed. Two known factors that have caused the diurnal patterns include: (1) when the levels of turbulence were the weakest during the night and the strongest in the early afternoon with two transition periods (changing between stable and unstable surface layers) in the morning and in the evening (Marsik et al., 2005); and (2) when emissions from the soils were minimal during the night and were the strongest in the daytime as discussed in Section 2.2.1. The third possible factor is the bi-directional exchange of Hg^0 through the leaf stomata. At this stage, it is not clear under what environmental conditions the leaf stomata is a source of Hg rather than a sink. The Hg^0 content stored in the leaf stomata should play a major role. Zhang et al. (2006) showed that ozone deposited to the forest canopies the fastest during the first several hours of the morning (when the stomata were just opening) and then the deposition processes slowed down. They attributed the slower deposition at later times to the ozone accumulation within the leaf stomata. It is speculated whether a similar theory might also apply to Hg^0 stomatal uptake. The three possible factors suggested above seem to explain the diurnal patterns of the Hg^0 exchange commonly observed over vegetated surfaces: very weak deposition during the night, somewhat higher deposition in the early morning, more frequent emission than deposition during the rest of the day, and deposition again in the late evening (Lindberg et al., 1998, 2002b; Kim et al., 2002, Kim et al., 2003; Marsik et al., 2005).

Since the amount of Hg^0 deposition also depends strongly on its ambient concentration (Ericksen and Gustin, 2004), leaf area index and leaf age (Lee et al., 2000), and other biological and meteorological conditions (as discussed above), some measurements might not follow exactly the typical diurnal pattern discussed above. For example, Lee et al. (2000) observed a dominance in deposition at a salt marsh wetland, during the full foliage period, during both the daytime and the nighttime. Obrist et al. (2006) and Fritsche et al. (2008a,b) also observed net deposition during the day and night over several grasslands. Deposition has also been observed to be dominant when the ambient Hg^0 concentrations are high, similar to the findings over background soils discussed in Section 2.2.1. It has been frequently reported that local sources enhanced Hg^0 deposition (Lindberg et al., 1998; Kim and Kim, 1999; Poissant et al., 2004b; Marsik et al., 2005; Bash and Miller, 2007; Cobbett and Van Heyst, 2007). This is probably because local sources increased the ambient Hg^0 concentration and thereby increased its deposition.

The majority of the studies listed in Table 1 did not cover multiple seasons and thus, it is difficult to generalize a seasonal pattern for the observed V_d values. Nevertheless, the limited data seem to suggest ahigher V_d in the summer or the full growing season than during the other seasons. At a salt marsh site, the V_d was almost one order of magnitude higher during the full foliage season than during the early growth season (Lee et al., 2000). Deposition was the net pathway for Hg^0 during the active vegetation periods over a grassland site, whereas during the snow-covered period, the mean exchange was emission (Fritsche et al., 2008b).

2.2.3. Contaminated sites

Flux measurement studies have been done over landfill (Kim et al., 2001; Nguyen et al., 2008), mining (Kotnik et al., 2005; Wang et al., 2007a,b), waste repository (Olofsson et al., 2005), and other polluted locations (see references in Nguyen et al., 2008). Net emission fluxes have been observed at the majority of the contaminated areas (Kim and Kim, 1999; Olofsson et al., 2005; Wang et al., 2007b, also see summaries in Nguyen et al., 2008). Non-negligible deposition fluxes have also been observed, and have sometimes exceeded the emission fluxes under certain conditions. For example, Wang et al. (2007a) observed net deposition at three out of five sites in a mining area, mostly at sites covered by vegetation.

V_d values over contaminated sites have been calculated from a few studies (Table 1). Typical V_d values calculated from the depositional flux data collected at multiple locations within a mining district are in the range between 0.05 and 0.2 cm s^{-1}. These values are consistent with values found at vegetated locations with background Hg. They are, however, higher than the V_ds found over bare soils. The high ambient Hg^0 concentration and the possible inclusion of RGM in the collected data could have contributed to the relatively large V_d values. The average V_d calculated from the flux and concentration data collected at a landfill site is 0.5 cm s^{-1} (Nguyen et al., 2008). This value seems very high, but is still within the range of observed values over other areas. In contrast, Kim et al. (2001), at another landfill site, observed a V_d value of ~5 cm s^{-1}. This extremely high V_d value might have been caused by its proximity to a nearby vent. The typical V_d values (0.05–0.5 cm s^{-1}) at the contaminated sites discussed above suggest that a large percentage of Hg^0 might deposit back to the surface within the vicinity of the contaminated area.

2.2.4. Snow and water surfaces

Flux measurements over snow surfaces are very limited. The quick decrease in the ambient Hg^0 concentration after polar sunrise, the atmospheric mercury depletion event first noted by Schroeder et al. (1998) at Alert, Canada, was found to be caused by the photo-chemistry induced conversion of Hg^0 to fast depositing Hg species (RGM and Hg_p) rather than by the direct deposition of Hg^0. Using the aerodynamic gradient method, Cobbett et al. (2007) found that Hg^0 deposition was dominant during polar night while both deposition and emission were equally important during polar day. V_ds calculated using all of the data (both emission and deposition) were very low (0.00025 cm s^{-1} during polar night and even smaller during other times); however, V_ds for individual samples were higher than 0.01 cm s^{-1}. Schroeder et al. (2005) also observed nighttime depositional fluxes over fresh snow above background soil; however, the net flux was emission over snow above Hg-enriched soils. The limited data discussed above suggest that snow surfaces can be a small net sink for Hg^0 during dark conditions.

Measurements of Hg^0 fluxes over water surfaces, for both sea and inland water, frequently show the dominance of emission over deposition (Schroeder et al., 2005 and references therein). Mean fluxes observed at a number of inland water surfaces in Ontario, Quebec, and Nova Scotia all show emission fluxes (Schroeder et al., 2005). Xiao et al. (1991) also observed emission fluxes during both the daytime and the nighttime over four forest lakes. Gårdfeldt et al. (2001) observed daytime emission and nighttime deposition over both seawater and inland water; and the nighttime deposition was very weak with V_d values being on the order of 0.001–0.01 cm s^{-1}. These data suggest that deposition is only a minor concern and only during the nighttime; it is the emission processes that have to be addressed carefully.

2.3. RGM and Hg_p

Limited measurement data are available for RGM deposition (Table 2). These data are known to have large uncertainties due to: (1) the very low air concentration of RGM (i.e., two orders of magnitude smaller than Hg^0) and the limitations in the instruments' detection limits; (2) the frequent use of surrogate surfaces; (3) the small vertical gradients in its concentration; and (4) the effects of fast chemical reactions and advections from local sources. Factors (3) and (4) above are particularly important when quantifying RGM fluxes using micrometeorological methods (AER, MBR, REA).

Nevertheless, all of the measurements point to high rates of deposition for RGM, with typical V_d values of 0.5–6 cm s^{-1}. These high V_d values are consistent with its high solubility and reactivity. During a field campaign over a wetland in Quebec, Canada, Poissant et al. (2004b) observed a median value of 7.6 cm s^{-1}. It is suspected that a nearby local Hg source could have contributed partly to this high value through advection. Skov et al. (2006) observed both upward and downward RGM fluxes using the REA method over a snow surface in Barrow, Alaska after polar sunrise. The average V_d values using all of the data are in the range of the typical values observed at other places. However, by excluding the upward flux data, the average V_d values were ~10 cm s^{-1} during two spring seasons. They attributed the upward flux to the chemical formation of RGM at or near the snow surface. In addition to the effects from

Table 2
A summary of V_d values (cm s^{-1}) for RGM and Hg_p.

Surface	Location	Season	Species	Method	V_d (cm s^{-1})	Condition	Remarks	Reference
Forest Canopies								
Deciduous forest	Walker Branch, TN, USA	Fall 1992 Summer 1995	RGM	MBR	5.0–6.0	Daytime	Only 3 gradient samples	Lindberg and Stratton, 1998
Deciduous forest	Pellston, MI, USA	Early Fall 1996	RGM	LW	0.1–0.5	Long-term fluxes to leaf surfaces	Assumes all Hg washed from surfaces is RGM	Rea et al., 2000
Deciduous forest	Pellston, MI, USA	Early Fall 1996	Hg_p	LW & SS	1.2 ± 1.7	Long-term fluxes to leaf surfaces	Assumes all Hg washed from surfaces is Hg_p	
Grasslands								
Grassland	Indiana, USA	Summer 1993	RGM	MBR	0.4	Daytime	Only 1 gradient sample	Lindberg and Stratton, 1998
Rural-pastureland and sagebrush	Nevada, USA	2005–2006	RGM	SS	1.72 ± 0.19		Data during rain period were not included	Lyman et al., 2007
Suburban – sage - and rabbitbrush	Nevada, USA	2005–2006	RGM	SS	1.52 ± 0.58			
Suburban - sage- and rabbitbrush	Nevada, USA	Summer 2005	RGM + Hg_p	LW	0.03 ± 0.02			
Irrigated grass with sagebrush	Nevada, USA	Summer 2005	RGM + Hg_p	LW	0.16 ± 0.10			
Wetlands								
Sawgrass and cattail	Florida Everglades, USA	Late Winter 1999	RGM	WSS	0–1.6	Dew sample	Dry deposition enhanced by dew	Malcolm and Keeler, 2002
Mixed vegetation, soil, water	Quebec, Canada	Late summer 2002	RGM	MBR	7.6 (0.012–73)		Site impacted by local Hg sources	Poissant et al., 2004b
Mixed vegetation, soil, water	Quebec, Canada	Late summer 2002	Hg_p	MBR	2.1 (0.001–43.1)			
River bulrush–flooded	Quebec, Canada	Late summer 2003	RGM	DFB	0.19 ± 0.42[a]		Condensation inside bag at night	Zhang et al., 2005
River bulrush–flooded	Quebec, Canada	Late summer 2003	Hg_p	DFB	0.02 ± 0.12[a]			
Snow								
Snowpack	Barrow, Alaska	Spring 2001	RGM	REA	~1	Polar sunrise (~3–5 h darkness)	Mercury depletion event	Lindberg et al., 2002a
Snowpack	Barrow, Alaska	Spring 2001 Spring 2002 Spring 2003 2001–2004	RGM	REA	1.54 (4.16) 5.17 (12.31) <0 (9.45) 1.17 (9.63)		Values in () are from downward fluxes only Surface resistance close to zero	Skov et al., 2006

advection, this chemical reaction at the upper levels is also suspected to be the cause of the extremely high V_d values.

RGM were found to deposit slightly more quickly to forest canopies and snow surfaces than to grassland sites (Lindberg and Stratton, 1998; Skov et al., 2006). Another type of variation in the canopy is the difference between a surrogate surface and an actual leaf. Lyman et al. (2007) discussed the potential of RGM depositing more easily to a surrogate surface than a natural surface due to a lack of surface resistance, thereby producing higher deposition velocities.

Seasonal and diurnal variations in deposition have been found for RGM. Lindberg and Stratton (1998) found higher V_d values in the summer than in the fall and in the afternoon than in the morning over a forest canopy in Tennessee. This can be explained by the change in seasons and the variations in leaf phenology. Peaks in deposition during polar sunrise and snowmelt have been found in the Arctic by Lindberg et al. (2002a). In another study, diurnal cycles were observed and could be mostly explained by changes in the intensity of the turbulence at the surface layer (Poissant et al., 2004b).

There have been very few studies to date that have examined the deposition of RGM during both wet and dry conditions. Malcolm and Keeler (2002) found that the presence of dew significantly reduced the ambient RGM concentration, suggesting an enhancement of RGM deposition by dew events. Rain events measured by Lindberg and Stratton (1998) produced a 50% decrease in the atmospheric concentration of RGM.

Future studies could focus on reducing measurement uncertainties in the existing methods and on better understanding the diurnal and seasonal patterns through continuous measurements. The comparison of the different measurement methods is important in identifying the magnitude of the measurement uncertainties.

Very few measurements have been made for the deposition of Hg_p (Table 2). Measured V_d values are in the range of 0.02–2.0 cm s^{-1}, consistent with theoretical and field studies that have been conducted for other particle species with sizes dominating the fine fractions. Observations show that Hg_p deposits at a faster rate than Hg^0, but not as quickly as RGM (Poissant et al., 2004b). The degree of variation in the measurement methods (e.g., MBR, DFB, LW) could have contributed to this large range in observed V_ds. Another important factor is the particle size since different measurements might have collected different sizes of Hg_p. In the study by Rea et al. (2000), the measured deposition onto the leaves was higher than onto the surrogate surfaces, again suggesting the large uncertainty that arises from the use of the surrogate surface approach.

Large diurnal variations were observed with higher deposition during the daytime than nighttime in two other studies at wetland sites (Poissant et al., 2004b; Zhang et al., 2005). We are not able to discuss the differences in the deposition of Hg_p over different surfaces due to the very limited data. Considering the strong dependence of particle V_d on particle size, it is recommended that future Hg_p flux measurements include Hg_p size distribution measurements so that the data collected can be used to verify/improve existing particle deposition models with applications to Hg_p.

3. Dry deposition schemes

Numerical models have been developed to study the atmospheric cycling of Hg on regional (Pai et al., 1999; Xu et al., 2000; Petersen et al., 2001; Bullock and Brehme, 2002; Lin and Tao, 2003; Seigneur et al., 2003; Christensen et al., 2004; Dastoor and Laroque, 2004; Miller et al., 2005; Gbor et al., 2007; Pan et al., 2008) and global scales (Bergan et al., 1999; Shia et al., 1999; Seigneur et al., 2004; Christensen et al., 2004; Dastoor and Laroque, 2004; Selin et al., 2007; Strode et al., 2007). In these Hg transport models, dry deposition of Hg^0, RGM and Hg_p, as well as the natural emission of Hg^0 need to be considered. In the following, the dry deposition schemes used in the Hg transport models are briefly discussed, and recommendations under different scenarios are also given.

The resistance approach is used to simulate the dry deposition velocity of RGM in more recently developed Hg transport models, although a few earlier models simply used a constant V_d value (see discussions in Lin et al., 2006 and model intercomparison in Ryaboshapko et al. 2007a,b and Bullock et al., 2008). In many circumstances, the dry deposition of RGM is treated in the same way as nitric acid (HNO_3), a gaseous species that has been measured extensively and is commonly considered in existing dry deposition schemes developed for air-quality models (e.g., Wesely, 1989; Zhang et al., 2003). This is because both RGM and HNO_3 have high solubility and reactivity; and the very limited measurements of RGM suggest that its V_d values are similar in magnitude to those of HNO_3. Fig. 2 shows an example of modelled V_d values for RGM over different surfaces under typical daytime conditions using the model of Zhang et al. (2003), with the two scaling parameters (α and β) for RGM chosen to be the same as for HNO_3. Modelled V_ds during a typical day are 2.5–5 cm s^{-1} over forests, 1–2 cm s^{-1} over other vegetated and non-vegetated land surfaces, 1.5 cm s^{-1} over ice (or snow), and around 1 cm s^{-1} over water surfaces. These values are comparable to the values listed in Table 2. A few other studies have also suggested that the models produce similar V_ds compared with measurements (e.g., Marsik et al., 2007; Lyman et al., in press).

Lin et al. (2006) investigated RGM V_ds by assuming RGM to be $HgCl_2$ or HgO based on the dry deposition scheme of Wesely (1989). They found a factor of 2 difference in the seasonally-averaged V_d between these two species due to the differences in their respective Henry's Law constants. This implies that the speciation of RGM is important in estimating its dry deposition if using the scheme of Wesely (1989); although it will not be the case for the scheme of Zhang et al. (2003). Future studies need to compare available schemes under different scenarios (e.g., different RGM speciation, surfaces, and meteorological conditions) using quality-controlled field measurements.

The resistance approach is also commonly used for Hg_p. The deposition velocity of any particulate species mainly depends on its size distributions. Thus, the best approach for estimating the V_d of Hg_p is to use a well-developed size-resolved particle dry deposition model coupled to reasonable size distribution assumptions (Dastoor and Laroque, 2004). Alternatively, V_ds of Hg_p can be estimated from existing bulk parameterizations developed for fine particles, assuming that the fine particle fraction dominates the Hg_p concentrations (Bullock and Brehme, 2002). Future research should focus on improving the size-resolved particle dry deposition models as well as quantifying the Hg_p size distributions more accurately through field measurements. The particle dry deposition model of Zhang et al. (2001) has recently been modified by incorporating the theory simplified from Petroff et al. (2008, 2009). The aim of the modified model was to have better agreement with available field data of size-resolved V_ds over different surfaces, including forests, grassland, bare soil, and water surfaces (manuscript in preparation).

There is a wide range of complexity in the dry deposition scheme of Hg^0 in atmospheric Hg models. Some models simply neglect Hg^0 deposition as well as its natural emission since the net effect of these two competing processes is not known well (e.g., Bullock et al., 2008). Several of the models choose a small constant V_d value, especially over vegetated surfaces, assuming that the deposition over the canopy surpasses the emission from the

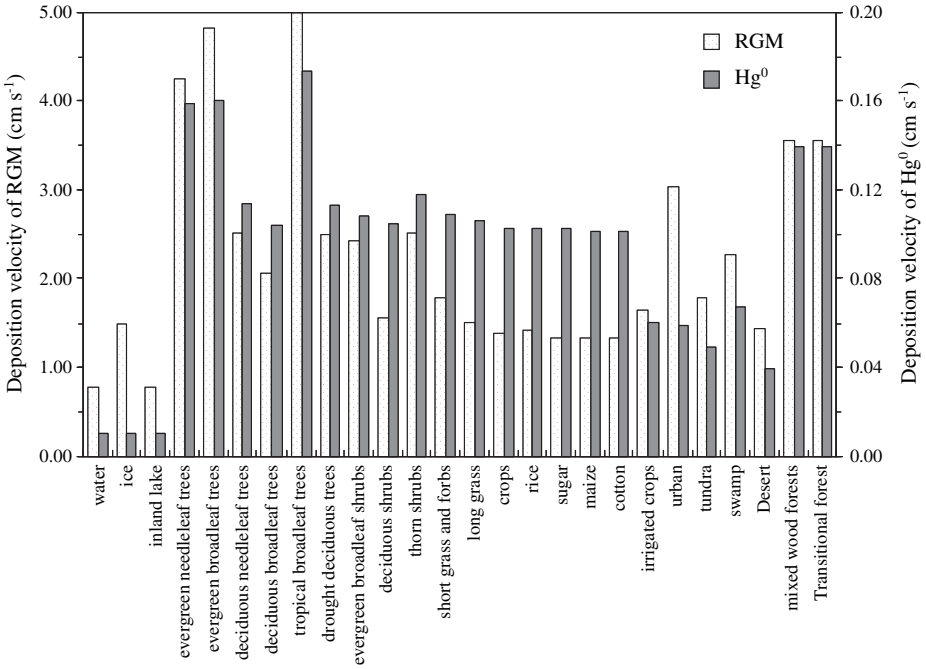

Fig. 2. Deposition velocity over different canopies calculated using the model of Zhang et al. (2003) under a typical daytime condition assuming a 6 m s^{-1} wind speed at 10 m height. The three species-dependent parameters chosen for RGM are: $\alpha = 10$, $\beta = 10$, and $R_m = 0$ s m^{-1}; and for Hg^0: $\alpha = 0$, $\beta = 0.2$, and $R_m = 500$ s m^{-1}. Note that the resistance scheme of Zhang et al. (2003) is the same as in Fig. 1 except without bi-directional exchange.

underlying soils. A few of the models use the bi-directional approach based on or modified from Xu et al. (1999), which treats emission and dry deposition separately without the constraint of concentration gradients between different surface media (Xu et al., 2000; Lin and Tao, 2003; Gbor et al., 2006).

Bash et al. (2007) proposed a compartmentalized modelling framework in which Hg dry deposition and emission were coupled through storage and concentration gradients across the surface media. It is encouraging to see that such a framework has been put in place in the modelling framework of CMAQ (Community Multiscale Air-Quality model) (Bash and Bullock: A dynamic bi-directional air–surface exchange model for mercury in CMAQ. Presented at the 9th International Conference on Mercury as a Global Pollutant, Guiyang, China, June 7–12, 2009). This new scheme is theoretically advanced over all of the previous approaches and is expected to provide more reasonable emission and deposition budgets if all of the necessary input parameters are available. Due to its complexity, the implementation of this scheme into other Hg transport models will not be so straightforward when compared to other simpler schemes. It is recommended that this scheme be evaluated under different modelling frameworks to assess its performance. It should be kept in mind that a more sophisticated scheme usually needs to make more assumptions, especially considering our limited knowledge on both deposition and natural emission processes.

Another approach to the treatment of Hg^0 dry deposition is to modify the existing big-leaf models by including the compensation points over soil and leaf stomata, similar to the approach used in dealing with ammonia deposition (e.g., Sutton et al., 1998). Such a simple approach includes the consideration of concentration gradients between different surface media and is an advance over the traditional approach; however, it is not as sophisticated as the approach recently developed by Bash and Bullock mentioned above. Fig. 1 is an example of a scheme modified from the big-leaf model of Zhang et al. (2003) by changing the downward flux to a bi-directional exchange over the soil surface and through the leaf stomata. The flux at a reference height above the canopy can be calculated as:

$$F_t = -\frac{(\chi_a - \chi_c)}{R_a + R_b}$$

The overall canopy compensation point (χ_c) can be calculated as a function of all of the resistance terms and the compensation points over the soil (χ_g) and over the stomata (χ_s):

$$\chi_c = \left[\frac{x_a}{R_a + R_b} + \frac{x_s}{R_s + R_m} + \frac{x_g}{R_{ac} + R_g}\right]\left[\frac{1}{R_a + R_b} + \frac{1}{R_s + R_m} + \frac{1}{R_{ac} + R_g} + \frac{1}{R_{cut}}\right]^{-1}$$

What is needed in this modified model is the definition of two new parameters (χ_g and χ_s) since all of the resistance terms are available from the original model. Factors such as soil Hg content, soil moisture, temperature, and light intensity should be considered when formulating χ_g. Knowledge of χ_s at the present stage is still very limited (Graydon et al., 2006, and references therein). Note that many published air–surface exchange data of Hg^0, if reanalyzed, could provide some information on compensation points. Numerical sensitivity tests can also shed some light on this topic.

Recent studies by Gustin and colleagues suggest that for low Hg-containing ecosystems (soil Hg < 0.1 μg g^{-1}), vegetation is a significant sink for atmospheric Hg (Gustin et al.: evolution of thought on terrestrial ecosystems as a net source or sink for atmospheric Hg. Presented at the 9th International Conference on Mercury as a Global Pollutant, Guiyang, China, June 7–12, 2009). Note that the above Hg budget also includes deposition of RGM and Hg_p. Based on this new knowledge and the knowledge of Hg^0 air–surface exchange discussed above, some recommendations are made below on the treatment of Hg^0 dry deposition in regional/global Hg transport models. It is assumed that a Hg transport model already includes Hg emissions from anthropogenic sources and

from naturally Hg-enriched surfaces, but does not necessarily include natural emissions from background soils.

The simplest approach for handing Hg^0 dry deposition in an Hg transport model would be to use a big-leaf resistance dry deposition scheme, the same approach that is used for RGM. If the Hg transport model does not include the natural Hg emissions from background soils, the modelled V_d should be designed to be smaller than the values presented in Table 1; however, if natural emissions are included, the modelled V_d should be on the same order as, or slightly higher than, those values shown in Table 1. Fig. 2 shows an example of modelled V_d values for Hg^0, under typical daytime conditions, using the model of Zhang et al. (2003). Modelled V_ds during a typical day are 0.1–0.18 cm s^{-1} over forests, ~0.1 cm s^{-1} over other vegetated surfaces, ~0.05 cm s^{-1} over non-vegetated land surfaces, and ~0.01 cm s^{-1} over water and ice. Compared to the values in Table 1, these values are slightly lower over vegetated surfaces but are similar over non-vegetated surfaces. These values can be treated as the upper-end values if natural emissions are not included in the model and can be treated as conservative values if natural emissions are included.

A second approach would be to use a bi-directional approach, either the one similar to that of Xu et al. (1999) or the one proposed in Fig. 1. The approach described in Fig. 1 can be used in Hg transport models with or without natural emissions. Upward fluxes represent emissions and downward fluxes represent depositions. If natural emissions are already included in a Hg transport model, one can simply discard all of the upward fluxes to avoid the double counting of natural emissions. In this case, the scheme just serves as a dry deposition scheme, but only allows the dry deposition process to happen when the ambient concentration is higher than the compensation point. The most theoretically complete (and complex) approach among all of the available schemes would be the one recently developed by Bash and Bullock (Presented in Guiyang, China, June 7–12, 2009). In this approach, a soil–vegetation–air transfer model is used to handle natural emissions and depositions for all three Hg forms. All the above approaches need further evaluation using field measurements and also need to be evaluated through model intercomparisons within different modelling frameworks.

4. Conclusions

Our understanding on the air–surface exchange of the atmospheric mercury has been gradually progressing from qualitative to quantitative knowledge over the past two decades. However, large uncertainties still exist in both measurement and modelling studies. The measurement uncertainties from using different measurement methods are still quite large, e.g., larger than a factor of 2 in the calculated dry deposition velocity (e.g., Fritsche et al., 2008a). Existing dry deposition schemes also have uncertainties of similar orders of magnitude (e.g., Lin et al., 2006).

Meteorological, biological, and soil conditions, ambient concentrations, and atmospheric chemistry all play important roles in the air–surface exchange of atmospheric mercury. Bi-directional exchange of Hg^0 has been frequently observed over non-vegetated surfaces or vegetated surfaces with small leaf area indices, although deposition process may dominate over full-grown canopies (i.e., with a large leaf area index). The amount of deposition of Hg^0 to vegetated surfaces can be substantial considering its high ambient conditions and its significantly large dry deposition velocities (0.1–0.4 cm s^{-1}). The majority of measurements suggest that RGM can deposit very quickly onto any type of surface, with its deposition velocities ranging from 0.5 to 6 cm s^{-1}. Although Hg_p dry deposits much more slowly than RGM, it might deposits faster than Hg^0 (Poissant et al., 2004b). Existing resistance-approach based dry deposition schemes seem to be able to produce the typical V_d values for RGM and Hg^0 over different surface types. More sophisticated air–surface exchange models are available and are expected to produce more reasonable mercury emission/deposition budgets once knowledge on all of the needed inputs is available.

There is a need for direct flux and concentration measurements of speciated mercury over different types of surfaces and under different scenarios in order to improve our understanding on the topic of mercury air–surface exchange and to provide data for evaluating the Hg dry deposition schemes. Measurement uncertainties need to be reduced by improving the chemical instruments and/or by developing new measurement methods. Dry deposition schemes need further evaluation using field measurements and intercomparisons within different model frameworks. It is also recommended that different approaches for parameterizing the bi-directional air–surface exchange of Hg^0 should be developed. This type of model can be best evaluated if simultaneous measurements of fluxes above a canopy and over the soil surfaces below the canopy are available together with information related to meteorological, biological, and soil conditions.

Acknowledgements

Leiming Zhang greatly appreciates very helpful discussions at various occasions with the following people: Jesse Bash, O. Russell Bullock Jr., Xinbin Feng, Che-Jen (Jerry) Lin, Frank Marsik, Laurier Poissant and Xiaohong Xu.

References

Bash, J.O., Bresnahan, P.A., Miller, D.R., 2007. Dynamic surface interface exchanges of mercury: a review and compartmentalized modeling framework. Journal of Applied Meteorology and Climatology 46, 1606–1618.
Bash, J.O., Miller, D.R., 2007. A note on elevated total gaseous mercury concentrations downwind from an agriculture field during tilling. Science of the Total Environment 388, 379–388.
Bergan, T., Gallardo, L., Rodhe, H., 1999. Mercury in the global troposphere: a three-dimensional model study. Atmospheric Environment 33, 1575–1585.
Boas, M.L., 1996. Mathematical Methods in the Physical Sciences, second ed. John Wiley and Sons Inc, Canada.
Bullock Jr., O.R., Brehme, K.A., 2002. Atmospheric mercury simulation using the CMAQ model: formulation description and analysis of wet deposition results. Atmospheric Environment 36, 2135–2146.
Bullock, O.R., Atkinson, D., Braverman, T., 2008. The North American mercury model intercomparison study (NAMMIS): study description and model-to-model comparisons. Journal of Geophysical Research 113. doi:10.1029/2008JD009803.
Caldwell, C.A., Swartzendruber, P., Prestbo, E., 2006. Concentration and dry deposition of mercury species in arid south central New Mexico (2001–2002). Environmental Science and Technology 40, 7535–7540.
Christensen, J.H., Brandt, J., Frohn, L.M., Skov, H., 2004. Modelling of mercury in the arctic with the Danish Eulerian Hemispheric model. Atmospheric Chemistry and Physics 4, 2251–2257.
Clarkson, T.W., 1990. Mercury – an element of mystery. New England Journal of Medicine 323, 1137–1139.
Cobbett, F.D., Steffen, A., Lawson, G., Van Heyst, B.J., 2007. GEM fluxes and atmospheric mercury concentrations (GEM, RGM and Hgp) in the Canadian Arctic at Alert, Nunavut, Canada (February–June 2005). Atmospheric Environment 41, 6527–6543.
Cobbett, F.D., Van Heyst, B.J., 2007. Measurements of GEM fluxes and atmospheric mercury concentrations (GEM, RGM and Hgp) from an agricultural field amended with biosolids in Southern Ont., Canada (October 2004–November 2004). Atmospheric Environment 41, 2270–2282.
Cobos, D.R., Baker, J.M., Nater, E.A., 2002. Conditional sampling for measuring mercury vapour fluxes. Atmospheric Environment 36, 4309–4321.
Dastoor, A.P., Laroque, Y., 2004. Global circulation of atmospheric mercury: a modelling study. Atmospheric Environment 38, 147–161.
Ericksen, J.A., Gustin, M.S., Schorran, D.E., Johnson, D.W., Lindberg, S.E., Coleman, J.S., 2003. Accumulation of atmospheric mercury in forest foliage. Atmospheric Environment 37, 1613–1622.
Ericksen, J.A., Gustin, M.A., 2004. Foliar exchange of mercury as a function of soil and air mercury concentrations. Science of the Total Environment 324, 271–279.
Ericksen, J.A., Gustin, M.S., Lindberg, S.E., Olund, S.D., Krabbenhoft, D.P., 2005. Assessing the potential for re-emission of mercury deposited in precipitation from arid soils using a stable isotope. Environmental Science and Technology 39, 8001–8007.

Ericksen, J.A., Gustin, M.A., Xin, M., Weisberg, P.J., Fernandez, G.C.J., 2006. Air–soil exchange of mercury from background soils in the United States. Science of the Total Environment 366, 851–863.
Fritsche, J., Obrist, D., Zeeman, M.J., Conen, F., Eugster, W., Alewell, C., 2008a. Elemental mercury fluxes over a sub-alpine grassland determined with two micrometeorological methods. Atmospheric Environment 42, 2922–2933.
Fritsche, J., Wohlfahrt, G., Ammann, C., Zeeman, M., Hammerle, A., Obrist, D., Alewell, C., 2008b. Summertime elemental mercury exchange of temperate grasslands on an ecosystem-scale. Atmospheric Chemistry and Physics 8, 7709–7722.
Gabriel, M.C., Williamson, D.G., Zhang, H., Brooks, S., Lindberg, S., 2006. Diurnal and seasonal trends in total gaseous mercury flux from three urban ground surfaces. Atmospheric Environment 40, 4269–4284.
Gårdfeldt, K., Feng, X., Sommar, J., Lindqvist, O., 2001. Total gaseous mercury exchange between air and water at river and sea surfaces in Swedish coastal regions. Atmospheric Environment 35, 3027–3038.
Gbor, P.K., Wen, D., Meng, F., Yang, F., Zhang, B., Sloan, J.J., 2006. Improved model for mercury emission, transport and deposition. Atmospheric Environment 40, 973–983.
Gbor, P.K., Wen, D., Meng, F., Yang, F., Sloan, J.J., 2007. Modelling of mercury emission, transport, and deposition in North America. Atmospheric Environment 41, 1135–1149.
Graydon, J.A., St. Louis, V.L., Lindberg, S.E., Hintelmann, H., Krabbenhoft, D.P., 2006. Investigation of mercury exchange between forest canopy vegetation and the atmosphere using a new dynamic chamber. Environmental Science and Technology 40, 4680–4688.
Graydon, J.A., St. Louis, V.L., Hintelmann, H., Lindberg, S.E., Sandilands, K.A., Rudd, J.W.M., Kelly, C.A., Hall, B.D., Mowat, L.D., 2008. Long-term wet and dry deposition of total and methyl mercury in the remote boreal ecoregion of Canada. Environmental Science and Technology 42, 8345–8351.
Gustin, M.S., Lindberg, S.E., 2005. Terrestrial mercury fluxes: is the net exchange up, down, or neither? In: Dynamics of Mercury Pollution on Regional and Global Scales, 241–259.
Gustin, M.S., Stamenkovic, J., 2005. Effect of watering and soil moisture on mercury emissions from soils. Biogeochemistry 76, 215–232.
Gustin, M.S., Engle, M., Ericksen, J., Lyman, S., Stamenkovic, J., Xin, M., 2006. Mercury exchange between the atmosphere and low mercury containing substrates. Applied Geochemistry 21, 1913–1923.
Gustin, M.S., Lindberg, S.E., Weisberg, P.J., 2008. An update on the natural sources and sinks of atmospheric mercury. Applied Geochemistry 23, 482–493.
Hanson, P.J., Lindberg, S.E., Tabberer, T.A., Owens, J.G., Kim, K.H., 1995. Foliar exchange of mercury vapour: evidence for a compensation point. Water, Air, and Soil Pollution 80, 373–382.
Hintelmann, H., Harris, R., Heyes, A., Hurley, J.P., Kelly, C.A., Krabbenhoft, D.P., Lindberg, S., St. Louis, V.L., 2002. Reactivity and mobility of new and old mercury deposition in a boreal forest ecosystem during the first year of the METAALICUS study. Environmental Science and Technology 36, 5034–5040.
Keeler, G.J., Dvonch, T.J., 2005. Atmospheric mercury: a decade of observations in the Great Lakes. In: Dynamics of Mercury Pollution on Regional and Global Scales, 611–636.
Kim, K.-H., Lindberg, S.E., Meyers, T.P., 1995. Micrometeorological measurements of mercury vapor fluxes over background forest soils in Eastern Tennessee. Atmospheric Environment 29, 267–282.
Kim, K.-H., Kim, M.-Y., 1999. The exchange of gaseous mercury across soil–air interface in a residential area of Seoul, Korea. Atmospheric Environment 33, 3153–3165.
Kim, K.-H., Kim, M.-Y., Lee, G., 2001. The soil–air exchange characteristics of total gaseous mercury from a large-scale municipal landfill area. Atmospheric Environment 35, 3475–3493.
Kim, K.H., Kim, M.Y., Kim, J., Lee, G., 2002. The concentrations and fluxes of total gaseous mercury in a western coastal area of Korea during late March 2001. Atmospheric Environment 36, 3413–3427.
Kim, K.–H., Kim, M.-Y., Kim, J., Lee, G., 2003. Effects of changes in environmental conditions on atmospheric mercury exchange: comparative analysis from a rice paddy field during the two spring periods of 2001 and 2002. Journal of Geophysical Research 108, 4607. doi:10.1029/2003JD003375.
Kotnik, J., Horvat, M., Dizdarevič, T., 2005. Current and past mercury distribution in air over the Idrija Hg mine region, Slovenia. Atmospheric Environment 39, 7570–7579.
Kuiken, T., Zhang, H., Gustin, M., Lindberg, S., 2008. Mercury emission from terrestrial background surfaces in the eastern USA. Part I: air/surface exchange of mercury within a southeastern deciduous forest (Tennessee) over one year. Applied Geochemistry 23, 345–355.
Lee, D.S., Benoit, G., Hu, X., 2000. Total gaseous mercury concentration and flux over a coastal saltmarsh vegetation in Connecticut, USA. Atmospheric Environment 34, 4205–4213.
Lin, C.-J., Pehkonen, S.O., 1999. The chemistry of atmospheric mercury: a review. Atmospheric Environment 33, 2067–2079.
Lin, C.-J., Pongprueksa, P., Lindberg, S.E., Pehkonen, S.O., Byun, D., Jang, C., 2006. Scientific uncertainties in atmospheric mercury models I: model science evaluation. Atmospheric Environment 40, 2911–2928.
Lin, X., Tao, Y., 2003. A numerical modeling study in regional mercury budget for eastern North America. Atmospheric Chemistry and Physics 3, 535–548.
Lindberg, S.E., Hanson, P.J., Meyers, T.P., Kim, K.-H., 1998. Air/surface exchange of mercury vapour over forests – the need for a reassessment of continental biogenic emissions. Atmospheric Environment 32, 895–908.
Lindberg, S.E., Stratton, W.J., 1998. Atmospheric mercury speciation: concentration and behaviour of reactive gaseous mercury in ambient air. Environmental Science and Technology 32, 49–57.
Lindberg, S.E., Brooks, S., Lin, C.-J., Scott, K.J., Landis, M.S., Stevens, R.K., Goodsite, M., Richter, A., 2002a. Dynamic oxidation of gaseous mercury in the arctic troposphere at polar sunrise. Environmental Science and Technology 36, 1245–1256.
Lindberg, S.E., Don, W., Meyers, T., 2002b. Transpiration of gaseous elemental mercury through vegetation in a subtropical wetland in Florida. Atmospheric Environment 36, 5207–5219.
Lindberg, S., Bullock, R., Ebinghaus, R., Engstrom, D., Feng, X., Fitzgerald, W., Pirrone, N., Seigneur, C., 2007. A synthesis of progress and uncertainties in attributing the sources of mercury in deposition. Ambio 36, 19–32.
Lindqvist, O., Rodhe, H., 1985. Atmospheric mercury – a review. Tellus 27B, 136–159.
Lyman, S.N., Gustin, M.S., Prestbo, E.M., Marsik, F.J., 2007. Estimation of dry deposition of atmospheric mercury in Nevada by direct and indirect methods. Environmental Science and Technology 41, 1970–1976.
Lyman, S.N., Gustin, M.S., Prestbo, E.M., Kilner, P.I., Edgerton, E., Hartsell, B. Testing and application of surrogate surfaces for understanding potential gaseous oxidized mercury dry deposition. Environmental Science and Technology, in press.
Malcolm, E.G., Keeler, G.J., 2002. Measurements of mercury in dew: atmospheric removal of mercury species to a wetted surface. Environmental Science and Technology 36, 2815–2821.
Mao, H., Talbot, R.W., Sigler, J.M., Sive, B.C., Hegarty, J.D., 2008. Seasonal and diurnal variations of Hg^0 over New England. Atmospheric Chemistry and Physics 8, 1403–1421.
Marsik, F.J., Keeler, G.J., Lindberg, S.E., Zhang, H., 2005. Air–surface exchange of gaseous mercury over a mixed sawgrass-cattail stand within the Florida Everglades. Environmental Science and Technology 39, 4739–4746.
Marsik, F.J., Keeler, G.J., Landis, M.S., 2007. The dry-deposition of speciated mercury to the Florida Everglades: measurements and modeling. Atmospheric Environment 41, 136–149.
Mason, R.P., Sheu, G.-R., 2002. Role of the ocean in the global mercury cycle. Global Biogeochemical Cycles 16, 1093. doi:10.1029/2001GB001440.
Mason, R.P., Abbott, M.L., Bodaly, R.A., Bullock Jr., O.R., Driscoll, C.T., Evers, D., Lindberg, S.E., Murray, M., Swain, E.B., 2005. Monitoring the response to changing mercury deposition. Environmental Science and Technology, 15A–22A.
Miller, E.K., Vanarsdale, A., Keeler, G.J., Chalmers, A., Poissant, L., Kamman, N.C., Brulotte, R., 2005. Estimation and mapping of wet and dry mercury deposition across northeastern North America. Ecotoxicology 14, 53–70.
Munthe, J., Bishop, K., Driscoll, C., Graydon, J., Hultberg, H., Lindberg, S., Matzner, E., Porvari, P., Rea, P., Schwesig, D., St Louis, V., Verta, M., 2004. Input–output of Hg in forested catchments in Europe and North America. RMZ-Materials and Geoenvironment 51, 1243–1246.
Narukawa, M., Sakata, M., Marumoto, K., Asakura, K., 2006. Air–sea exchange of mercury in Tokyo Bay. Journal of Oceanography 62, 249–257.
Nguyen, H.T., Kim, K.-H., Kim, M.-Y., Shon, Z.-H., 2008. Exchange pattern of gaseous elemental mercury in an active urban landfill facility. Chemosphere 70, 821–832.
Obrist, D., Conen, F., Vogt, R., Siegwolf, R., Alewell, C., 2006. Estimation of Hg^0 exchange between ecosystems and the atmosphere using ^{222}Rn and Hg^0 concentration changes in the stable nocturnal boundary layer. Atmospheric Environment 40, 856–866.
Olofsson, M., Sommar, J., Ljungstrom, E., Andersson, M., Wangberg, I., 2005. Application of Relaxed Eddy accumulation technique to quantify Hg^0 fluxes over modified soil surfaces. Water, Air, and Soil Pollution 167, 331–352.
Pai, P., Karamchandani, P., Seigneur, C., 1999. Sensitivity of simulated atmospheric mercury concentrations and deposition to model input parameters. Journal of Geophysical Research 104, 13855–13868.
Pan, L., Carmichael, G.R., Adhikary, B., Tang, Y., Streets, D., Woo, J.-H., Frieli, H.R., Radke, L.F., 2008. A regional analysis of the fate and transport of mercury in East Asia and an assessment of major uncertainties. Atmospheric Environment 42, 1144–1159.
Petersen, G., Bloxam, R., Wong, S., Munthe, J., Kruger, O., Schmolke, S.R., Kumar, A.V., 2001. A comprehensive Eulerian modelling framework for airborne mercury species: model development and applications in Europe. Atmospheric Environment 35, 3063–3074.
Petroff, A., Mailliat, A., Amielh, M., Anselmet, F., 2008. Aerosol dry deposition on vegetative canopies. Part II: a new modelling approach and applications. Atmospheric Environment 42, 3654–3683.
Petroff, A., Zhang, L., Pryor, S.C., Belot, Y., 2009. An extended dry deposition model for aerosols onto broadleaf canopies. Journal of Aerosol Science 40, 218–240.
Poissant, L., Pilote, M., Constant, P., Beauvais, C., Zhang, H.H., Xu, X., 2004a. Mercury gas exchanges over selected bare soil and flooded sites in the bay St. Francois wetlands (Quebec, Canada). Atmospheric Environment 38, 4205–4214.
Poissant, L., Pilote, M., Xu, X., Zhang, H., Beauvais, C., 2004b. Atmospheric mercury speciation and deposition in the Bay St. François wetlands. Journal of Geophysical Research– Atmospheres 109. doi:10.1029/2003JD004364.
Rea, A.W., Lindberg, S.E., Keeler, G.J., 2000. Assessment of dry deposition and foliar leaching of mercury and selected trace elements based on washed foliar and surrogate surfaces. Environmental Science and Technology 34, 2418–2425.
Rea, A.W., Lindberg, S.E., Scherbatskoy, T., Keeler, G.J., 2002. Mercury accumulation in foliage over time in two northern mixed-hardwood forests. Water, Air, and Soil Pollution 133, 49–67.

Ryaboshapko, A., Bullock Jr., O.R., Christensen, J., Cohen, M., Dastoor, A., Ilyin, I., Petersen, G., Syrakov, D., Artz, R.S., Davignon, D., Draxler, R.R., Munthe, J., 2007a. Intercomparison study of atmospheric mercury models: 1. Comparison of models with short-term measurements. Science of the Total Environment 376, 228–240.

Ryaboshapko, A., Bullock Jr., O.R., Christensen, J., Cohen, M., Dastoor, A., Ilyin, I., Petersen, G., Pacyna, J., 2007b. Intercomparison study of atmospheric mercury models: 2. Modelling results vs. long-term observations and comparison of country deposition budgets. Science of the Total Environment 377, 319–333.

Sakata, M., Marumoto, K., 2004. Dry deposition fluxes and deposition velocities of trace metals in the Tokyo metropolitan area measured with a water surface sampler. Environmental Science and Technology 38, 2190–2197.

Sakata, M., Marumoto, K., Narukawa, M., Asakura, K., 2006. Regional variations in wet and dry deposition fluxes of trace elements in Japan. Atmospheric Environment 40, 521–531.

Schroeder, W.H., Munthe, J., Lindqvist, O., 1989. Cycling of mercury between water, air, and soil compartments of the environment. Water, Air, and Soil Pollution 48, 337–347.

Schroeder, W.H., Munthe, J., 1998. Atmospheric mercury – an overview. Atmospheric Environment 32, 809–822.

Schroeder, W.H., Anlauf, K.G., Barrie, L.A., Lu, J.Y., Steffen, A., Schneeberger, D.R., Berg, T., 1998. Arctic springtime depletion of mercury. Nature 394, 331–332.

Schroeder, W.H., Beauchamp, S., Edwards, G., Poissant, L., Rasmussen, P., Tordon, R., Dias, G., Kemp, J., Van Heyst, B., Banic, C.M., 2005. Gaseous mercury emissions from natural sources in Canadian landscapes. Journal of Geophysical Research 110. doi:10.1029/2004JD005699.

Seigneur, C., Lohman, K., Vijayaraghavan, K., Shia, R.L., 2003. Contributions of global and regional sources to mercury deposition in New York State. Environmental Pollution 123, 365–373.

Seigneur, C., Vijayaraghavan, K., Lohman, K., Karamchandani, P., Scott, C., 2004. Global source attribution for mercury deposition in the United States. Environmental Science and Technology 38, 555–569.

Selin, N.E., Jacob, D.J., Park, R.J., Yantosca, R.M., Strode, S., Jaegle, L., Jaffe, D., 2007. Chemical cycling and deposition of atmospheric mercury: global constraints from observations. Journal of Geophysical Research 112, D02308. doi:10.1029/2006JD007450.

Shia, R.L., Seigneur, C., Pai, P., Ko, M., Sze, N.D., 1999. Global simulation of atmospheric mercury concentrations and deposition fluxes. Journal of Geophysical Research 104, 23747–23760.

Skov, H., Brooks, S.B., Goodsite, M.E., Lindberg, S.E., Meyers, T.P., Landis, M.S., Larsen, M.R.B., Christensen, J., 2006. Fluxes of reactive gaseous mercury measured with a newly developed method using relaxed eddy accumulation. Atmospheric Environment 40, 5452–5463.

Strode, S.A., Jaegle, L., Selin, N.E., Jacob, D.J., Park, R.J., Yantosca, R.M., Mason, R.P., Slemr, F., 2007. Air–sea exchange in the global mercury cycle. Global Biogeochemical Cycles 21 (GB1017). doi:10.1029/2006GB002766.

Sutton, M.A., Burkhardt, J.K., Guerin, D., Nemitz, E., Fowler, D., 1998. Development of resistance models to describe measurements of bi-directional ammonia surface atmosphere exchange. Atmospheric Environment 32, 473–480.

Wang, S., Feng, X., Qui, G., Fu, X., Wei, Z., 2007a. Characteristics of mercury exchange flux between soil and air in the heavily air-polluted area, eastern Guizhou, China. Atmospheric Environment 41, 5584–5594.

Wang, S., Feng, X., Qui, G., Shang, L., Li, P., Wei, Z., 2007b. Mercury concentrations and air/soil fluxes in Wuchuan mercury mining district, Guizhou, China. Atmospheric Environment 41, 5984–5993.

Wesely, M.L., 1989. Parameterization of surface resistances to gaseous dry deposition in regional-scale numerical models. Atmospheric Environment 23, 1293–1304.

Wesely, M.L., Hicks, B.B., 2000. A review of the current status of knowledge on dry deposition. Atmospheric Environment 34, 2261–2282.

Wolfe, M.F., Schwarzbach, S., Sulaiman, R.A., 1998. Effects of mercury on wildlife: a comprehensive review. Environmental Toxicology and Chemistry 17, 146–160.

Xiao, Z.F., Munthe, J., Shroeder, W.H., Lindqvist, O., 1991. Vertical fluxes of volatile mercury over forest soil and lake surfaces in Sweden. Tellus 43B, 267–279.

Xin, M., Gustin, M.S., 2007. Gaseous elemental Hg exchange with low Hg containing soils: investigation of controlling factors. Applied Geochemistry 22, 1451–1466.

Xu, X., Yang, X., Miller, D.R., Helble, J.J., Carley, R.J., 1999. Formulation of bidirectional atmosphere–surface exchanges of elemental mercury. Atmospheric Environment 33, 4345–4355.

Xu, X., Yang, X., Miller, D.R., Helble, J.J., Carley, R.J., 2000. A regional scale modeling study of atmospheric transport and transformation of mercury. I., model development and evaluation. Atmospheric Environment 34, 4933–4944.

Zhang, H.H., Poissant, L., Xu, X., Pilote, M., 2005. Explorative and innovative dynamic flux bag method development and testing for mercury air–vegetation gas exchange fluxes. Atmospheric Environment 39, 7481–7493.

Zhang, L., Gong, S., Padro, J., Barrie, L., 2001. A size-segregated particle dry deposition scheme for an atmospheric module. Atmospheric Environment 35, 549–560.

Zhang, L., Brook, J.R., Vet, R., 2003. A revised parameterization for gaseous dry deposition in air-quality models. Atmospheric Chemistry and Physics 3, 2067–2082.

Zhang, L., Vet, R., Brook, J.R., Legge, A.H., 2006. Factors affecting stomatal uptake of ozone by different canopies and a comparison between dose and exposure. Science of the Total Environment 370, 117–132.