Global observations and modeling of atmosphere-surface exchange of elemental mercury: a critical review

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Abstract

Reliable quantification of air-surface fluxes of elemental Hg vapor (Hg^0) is crucial for understanding mercury (Hg) global biogeochemical cycles. There have been extensive measurements and modeling efforts devoted to estimating the exchange fluxes between the atmosphere and various surfaces (e.g., soil, canopies, water, snow, etc.) in the past three decades. However, large uncertainties remain due to the complexity of Hg^0 bidirectional exchange, limitations of flux quantification techniques and challenges in model parameterization. In this study, we provide a critical review on the state of science in the atmosphere-surface exchange of Hg^0. Specifically, the advancement of flux quantification techniques, mechanisms in driving the air-surface Hg exchange, and modeling efforts are presented. Due to the semi-volatile nature of Hg^0 and redox transformation of Hg in environmental media, Hg deposition and evasion are influenced by multiple environmental variables including seasonality, vegetative coverage and its life cycle, temperature, light, moisture, atmospheric turbulence, the presence of reactants (e.g., O_3, radicals, etc.) that drive the physicochemical process of Hg in the media where Hg^0 exchange occurs. However, the effects of these processes on flux have not been fundamentally and quantitatively determined, which limits the accuracy of flux modeling.

In this study, we compile an up-to-date global observational flux database and discuss the implication of flux data on the global Hg budget. Mean Hg^0 fluxes obtained by micrometeorological measurements did not appear to be significantly greater than the fluxes measured by dynamic flux chamber methods over unpolluted surfaces (p=0.16, one-tailed, Mann-Whitney *U* test). The spatiotemporal coverage of existing Hg^0 flux measurements is highly heterogeneous with large data gaps existing in multiple continents (Africa, South Asia, Middle East, South America and Australia). The magnitude of the evasion flux is strongly enhanced by human activities, particularly at contaminated sites. Hg^0 flux observations in East Asia are comparatively larger in magnitude than the rest of the world, suggesting substantial reemission of previously deposited mercury from anthropogenic sources. The Hg^0 exchange over pristine surfaces (e.g., background soil and water) and vegetation needs better constrains for global analyses of the atmospheric Hg budget. The existing knowledge gap and the associated research needs for future measurements and modeling efforts for the air-surface exchange of Hg^0 are discussed.

1. Introduction

Mercury (Hg) is a global pollutant of broad concern due to its toxicity, bioaccumulation characteristics and adverse health effects (Driscoll et al., 2013), especially in its methylated forms such as monomethyl-mercury (CH_3Hg) species and dimethyl-mercury ($(CH_3)_2Hg$) (Clarkson and Magos, 2006). Fish consumption has been identified as the primary pathway for human exposure to CH_3Hg (Mergler et al., 2007; Mason et al., 2012), while the exposure through rice cultivated in areas with Hg pollution (e.g., mining and smelting areas) also poses a risk (Feng et al., 2008a; Zhang et al., 2010). To protect human health and the environment from the adverse effects of mercury, a global treaty "Minamata Convention for Mercury" that regulates Hg emission reduction from anthropogenic sources has been signed by 128 countries since October 2013 (UNEP Minamata Convention, 2014). Emission of Hg into the atmosphere occurs from both natural processes and human activities. The release of Hg from natural surfaces has been estimated to account for two thirds of global emissions (Fig. 1). However, this estimate is subject to large uncertainty because of the challenges in quantifying the flux and in understanding the mechanisms involved in the exchange process of elemental mercury vapor (Hg^0) (Selin, 2009; Zhang et al., 2009; Gustin, 2011; Zhang et al., 2012a).

Hg emitted from anthropogenic sources include all atmospheric species: gaseous elemental Hg (Hg^0, GEM), gaseous oxidized Hg (GOM), and particulate bound Hg (PBM) (Pacyna et al., 2006; AMAP/UNEP, 2013), while evasion derived from the Earth's surfaces is dominated by GEM (Gustin, 2011). Owing to the high deposition velocities of GOM and PBM (1-2 orders higher than GEM) (Zhang et al., 2009), GOM and PBM are readily deposited locally and regionally while GEM is subject to long-range transport (e.g., hemispheric scale) and can deposit remotely from the emission sources (Lindberg et al., 2007; Gustin and Jaffe, 2010). Atmospheric Hg continuously goes through deposition and re-emission cycles while undergoing physical and chemical transformations (Lin and Pehkonen, 1999).

Extensive efforts have been devoted to understanding the spatial and temporal pattern of Hg^0 exchange flux. Geogenically Hg-enriched surfaces and anthropogenically polluted sites are strong Hg emission sources (Kocman et al., 2013). Emissions from natural sources and from previously deposited Hg^0 on substrate surfaces are not analytically distinguishable using current measurement techniques (cf. Section 2). Direct measurement of Hg^0 flux from the background surfaces is difficult due to small vertical Hg^0 concentration gradients (therefore low flux) (Zhu et al., 2015a). Since the first application of a stainless steel dynamic flux chamber for Hg^0 flux measurements over background lakes and soils in the 1980s (Schroeder et al., 1989; Xiao et al., 1991), significant advancements in the experimental approaches (e.g., dynamic flux chambers, micrometeorological methods, the $Hg^0/^{222}Rn$ flux ratio, enriched isotope tracer methods, the open-path laser optical spectroscopic method, and the Hg^0/CO ratio) have been made (Sommar et al., 2013a). However, a standard protocol for Hg^0 flux quantification does not exist (Gustin, 2011; Zhu et al., 2015b), which complicates the comparison and interpretation of flux data reported in the literature (cf. Section 4).

In this study, we present a comprehensive review on the global observation of Hg^0 flux in peer-reviewed literature, and provide a state-of-the-science assessment on the air-surface exchange of Hg^0. Specifically, the advancement of flux quantification techniques, physicochemical factors driving the exchange process, existing field data of Hg^0 flux, and modeling efforts for scaling up the measured flux for global assessment are synthesized. Furthermore, the spatial and temporal characteristics of Hg^0 flux, as well as the underlying influencing factors are investigated. Key knowledge gaps, future directions for field measurements, and development of new-generation air-surface exchange model for Hg^0 flux are discussed. Compared to the assessment by Agnan et al. (2016) that summarized air-surface exchange of Hg over terrestrial systems, our work differs in: (1) the approaches in data compilation and synthesis, (2) the coverage of flux data over different land uses (11 categories covering whole earth surfaces versus 6 categories of terrestrial surfaces in Agnan et al., 2016), (3) the inclusion of mechanistic discussion on flux quantification approaches and air-surfaces exchange processes, (4) the inclusion of flux modeling approaches and scale-up of flux data for global cycle implications, and (5) the inclusion of more up-to-date field data and exclusion of laboratory data in the synthesis.

2. Advances in Hg^0 flux quantification methods

The theory and application of Hg^0 flux measurement techniques have been documented extensively (Zhang et al., 2009; Gustin, 2011; Sommar et al., 2013a). Here we focus on the developments, advantages and disadvantages, and comparability and uncertainties of different flux quantification techniques. DFCs, micrometeorological techniques (MM), and bulk methods (e.g., the $Hg^0/^{222}Rn$ flux ratio, enriched isotope tracers) are the mostly widely applied approaches for surface-atmosphere Hg^0 flux quantification (Schroeder et al., 1989;

Xiao et al., 1991; Kim and Lindberg, 1995; Kim et al., 1995; Cobos et al., 2002; Amyot et al., 2004; Olofsson et al., 2005; Obrist et al., 2006; Bash and Miller, 2008; Lin et al., 2012; Slemr et al., 2013; Zhu et al., 2013c), of which DFCs and MM techniques account for >95% of all observations documented to date (cf. Section 4). Open-path laser optical spectroscopic (LIDAR) method and Hg^0/CO ratio were applied to estimate Hg emission from area/regional sources (e.g., LIDAR: mining areas, industrial plants, geothermal sites; Hg^0/CO ratio: continental level atmospheric Hg transport) (Aldén et al., 1982; Edner et al., 1991; Sjöholm et al., 2004; Jaffe et al., 2005; Fu et al., 2015a). There has not been a standardized protocol for any of the techniques (e.g., instrumentation set-up, operation parameters) (Gustin, 2011; Zhu et al., 2015b). Recent collocated measurements and uncertainties analysis emphasized the importance of method standardization and processing of field data acquired by the measurement systems (Fritsche et al., 2008b; Converse et al., 2010; Zhu et al., 2015a). Application of the appropriate flux measurement technique depends on the scalar detection accuracy, sensor response frequency and level of automation (Sutton et al., 2007). The traditional standard procedure of sampling ambient air Hg^0 is by enhancement collection onto traps containing gold (Fitzgerald and Gill, 1979; Slemr et al., 1979). A wide-spread continuous Hg^0 monitor is the automated dual channel, single amalgamation, cold vapor atomic fluorescence analyzer (Model 2537, Tekran Instruments Corp.), which relies on this principle. The certified detection limit is < 0.1 ng m^{-3}. However, the pre-concentration procedure takes ≥2.5 min and therefore real-time, high-frequency data acquisition is not possible (Gustin, 2011; Fu et al., 2012b; Gustin et al., 2013; Gustin et al., 2015). Later on, monitoring ambient air Hg^0 with relative higher frequency (up to 1 Hz) was achieved by using Lumex RA-915+ Zeeman atomic absorption spectrometry (AAS) analyzer operating without pre-concentration. However, the instrument has a detection limit of ~1 ng m^{-3} and therefore is preferred for industrial level studies but applicable under ambient Hg^0 concentrations (Holland, 2005). More recently, a high frequency (25 Hz) cavity ring-down spectroscopy (CRDS) sensor has been deployed for Hg^0 concentration measurement, but it has a higher detection limit (> 0.35 ng m^{-3}) and suffers from the sensor's baseline drifting and interferences with O_3 (Fain et al., 2010; Pierce et al., 2013). Another laser technique, the laser-induced fluorescence sensor, has been designed and successfully applied for up to one day continuous measurement with improved detection limit (~15 pg m^{-3}) (Bauer et al., 2002; Bauer et al., 2014). However, both methods have not yet been proven for application in long-term field measurements. The coupling with a commercial instrument (e.g., Tekran® 2537) renders continuous and unattended flux measurements by DFC or MM techniques to be accomplished and are most widely deployed over various surfaces (cf. Section 4). However, this implementation is associated with a significant cost because the expense of the Hg^0 analyzer normally exceeds the rest of flux measurement system.

2.1 Dynamic flux chambers

The DFC method (footprint generally <0.1 m^2) is a frequently used Hg^0 flux measurement technique over soils, water surfaces, and low-stand grass due to its relatively low cost, portability, and versatility (Sommar et al., 2013a). DFCs operating under steady-state (Xiao et al., 1991; Carpi and Lindberg, 1998) and non-steady-state conditions (Rinklebe et al., 2009) are used in Hg research with the former configuration being by far the most common. Dynamic flux bags (DFB) have been applied for flux measurements over tall grass and tree branches (Zhang et al., 2005; Graydon et al., 2006; Poissant et al., 2008). Laboratory mesocosms probing whole ecosystem Hg exchange have also been attempted; a 180×10^3 L chamber ($7.3 \times 5.5 \times 4.5$ m^3) was deployed for quantifying soil-plant-atmosphere flux (Gustin et al., 2004; Obrist et al., 2005; Stamenkovic and Gustin, 2007; Stamenkovic et al., 2008). Construction materials such as fluorinated ethylene propylene (FEP) films and quartz have been recommended for DFCs due to its high actinic light transmittance and low blank (Kim and Lindberg, 1995; Carpi et al., 2007; Lin et al., 2012). Reported DFCs volumes and flushing flow rates range from 1 to 32 L and 1.5 to 20 L min^{-1}, resulting in a turnover time (TOT) ranging from 0.1 to 14 min (Eckley et al., 2010; Zhu et al., 2011). Using DFCs, the Hg^0 flux is calculated as:

$$F = \frac{Q\left(C_{out} - C_{in}\right)}{A} \quad (1)$$

where F is the Hg^0 flux (ng m^{-2} h^{-1}), Q is the DFC internal flushing flow rate (m^{-3} h^{-1}), A is the DFC footprint, C_{out} and C_{in} are the Hg^0 concentrations at the DFC outlet and inlet, respectively. Eq. (1) relies on mass balance calculations of two C_{out} and two C_{in} measurements alternately and assumes that the surface shear velocity over the DFC footprint is uniform and therefore results in a constant flux spatially over the wetted surface. Distinct Hg^0 fluxes have been observed using DFCs of different design shapes under similar environmental conditions (Eckley et al., 2010). Lin et al. (2012) investigated the internal flow field and Hg^0 concentration distribution in two commonly designed DFCs (i.e., rectangular and dome-shaped chambers) and showed that the airstream inside the DFCs is not uniform and the surface shear flow is divergent over the footprint, resulting in a non-uniform Hg^0 concentration gradient over the substrate surface. Eckley et al. (2010) systematically investigated effects of fabrication material, footprint, chamber dimensions including port positions, and flushing flow rates on the measured Hg^0 flux by DFCs. Consistent with previous studies, the flushing flow rate is among the most influential factor that if varied may induce up to one order of magnitude differences in the observed fluxes (Gustin et al., 1999; Wallschläger et al., 1999; Gillis and Miller, 2000a; Lindberg et al., 2002c; Zhang et al., 2002). Computational fluid dynamic modeling of DFC mass transfer has indicated that a smaller diffusion resistance at a higher flushing flow rate yielded a higher measured flux. However, due to the non-uniform internal Hg^0 concentration gradient, the measured Hg^0 flux from substrates may change unpredictably when the flushing flow rate varies (Eckley et al., 2010; Lin et al., 2012), which should be taken into consideration when the fluxes obtained by DFCs of different designs and flushing flow rates cannot be directly compared. Another limitation of DFCs is the isolation of chamber internal substrate surface from ambient conditions. This excludes the effect of atmospheric turbulence and therefore may cause a large uncertainty when using DFC data as in-put for scale-up estimations. Lin et al. (2012) proposed an aerodynamically-designed chamber (NDFC) which enables producing a uniform surface friction velocity to link with ambient shear condition to rescale to the ambient flux using Eq. (2), which allows the utilization of the ambient surface shear condition rather than artificial steady flushing flow rate to calculate the flux:

$$F = \frac{Q\left(C_{out} - C_{in}\right)}{A} \cdot \frac{k_{mass(a)}}{k_{mass(DFC)}} \qquad (2)$$

where $k_{mass(a)}$ is the overall mass transfer coefficient under ambient conditions, and $k_{mass(DFC)}$ is the overall mass transfer coefficient in the DFC measurement area.

In addition to the uncertainties caused by varying flushing flow rates, altered short and long wave radiation balances within DFCs resulting in a modified micro-environment were found to bias the observed fluxes (Zhu et al., 2015a). DFC flux is measured through intermittent sampling of ambient and chamber air for Hg^0 analysis using a single detector (Lindberg et al., 2002c), which assumes that ambient Hg^0 variability is negligible during air sampling. At locations where significant variation in Hg^0 concentrations exist (e.g., sites with anthropogenic emission sources), Eckley et al. (2011a) proposed a data assimilation protocol: $|\Delta C_{oi}| > |\Delta C_{ii}|$ should be valid for each of the calculated fluxes, otherwise the flux should be rejected (ΔC_{oi} is the difference between C_{out} and the average of two C_{in} which before and after taking C_{out}, while ΔC_{ii} is the difference between above two C_{in}). The concern about influencing plant physiology restricts the deployment of small DFCs to short term field measurements over the same vegetated plot. Given the small footprint and that Hg^0 fluxes over terrestrial surfaces are profoundly variable in space and time, replication of DFC measurements are thus preferred but often not carried out.

2.2 Micrometeorological methods

MM methods differ in measurement principles and the spatial scale of flux footprints compared to DFCs and have the capability of measuring ecosystem-scale (typically hectare scale) fluxes under undisturbed conditions and represent preferred flux quantification techniques over vegetated landscapes. MM techniques for background Hg^0 flux measurements are currently comprised of the relaxed eddy accumulation method (REA), the aerodynamic gradient method (AGM), and the modified Bowen-ratio method (MBR). The preferred MM technique, eddy covariance (EC), is a direct flux measurement method without any applications of empirical constants, requires a fast response (~10 Hz) gas analyzer, and has not been successfully implemented for regular Hg^0 flux measurements (Aubinet et al., 2012). Recently, Pierce et al. (2015) reported the first field trial of CRDS-EC flux measurements over Hg-enriched soils with a flux detection limit of 32 ng m^{-2} h^{-1}, offered the opportunity to high frequently monitoring Hg^0 flux from Hg-enriched surfaces. However, the present CRDS-EC technique must be further advanced for Hg^0 flux measurement at most, if not all, background sites. Sommar et al. (2013a) and Zhu et al. (2015b) detailed the theory, deployment, and computation of existing MM approaches for measuring Hg^0 fluxes. Gradient methods rely on quantifying the vertical concentration gradient (two or more sampling heights), turbulent parameters (AGM) or scalar concentration gradient (MBR), and scalar EC-fluxes. A major advantage of REA method is that the REA up- and down-draft sampling conducted at one height overcomes the uncertainties associated with: (1) footprint differences due to two height samplings in using gradient methods, and (2) possible oxidation/reduction introduced through the forming or loss of Hg^0 between the two heights. On the other hand, the analytical requirement for REA is more stringent than for the gradient methods, especially under low turbulent conditions, increasing the demand on the precision of the sampling and chemical analysis (Zhu et al., 2015a).

The REA method has been deployed for flux measurements for agricultural lands, forest canopies, wetlands, and urban settings (Cobos et al., 2002; Olofsson et al., 2005; Bash and Miller, 2009; Osterwalder et al., 2016; Sommar et al., 2013b). The AGM method has been used over grasslands, agricultural lands, saltmarshes, landfills, and snow (Lee et al., 2000; Kim et al., 2001; Kim et al., 2003; Cobbett et al., 2007; Cobbett and Van Heyst, 2007; Fritsche et al., 2008c; Fritsche et al., 2008b; Baya and Van Heyst, 2010). The MBR method has been set up in grasslands, forest floors, agricultural lands, lakes, wetlands, and snow (Lindberg et al., 1992; Lindberg et al., 1995b; Lindberg et al., 1998; Lindberg and Meyers, 2001; Lindberg et al., 2002b; Brooks et al., 2006; Fritsche et al., 2008b; Converse et al., 2010). The theoretical and application requirements of micrometeorology are less restricted for large areas of uniform vegetation (or soil) in flat landscapes, where an atmospheric surface layer develops and the horizontal flux variability is low in the absence of pollution plumes and the flux above the surface remains constant with height (Wesely and Hicks, 2000). Under these turbulent exchange conditions, the flux acquired at the measurement height resembles the actual flux at the surface under measurement. There are several potential causes that can invalidate the above assumptions. For instance, the advection of Hg^0 from nearby sources to the measurement site may occur. It is known that local point sources of Hg^0 can affect MM measurements downwind (Bash and Miller, 2007). Loubet et al. (2009) estimated such advection errors in NH_3 gradient fluxes resulting in 2.1% to 52% of vertical fluxes at a monitoring site at 810m downwind of an NH_3 source (a farm building) implying a significant error contribution from advection. Large variation of Hg^0 fluxes measured by MBR methods were also reported at nighttime as a result of advection in the Nevada STORM project, however, the errors have not been quantified (Gustin et al., 1999). For Hg^0 fluxes over forest canopies, the influence of within-canopy source and sink terms on the net ecosystem flux has not been evaluated. Multiple heights gradients/REA measurements is needed to resolve the true flux. Since there is not a reliable sensitive Hg^0 sensor at a high measurement frequency, an empirical multiplication factor or proxy scalar is required for computing all MM-Hg^0 fluxes (e.g., relaxation coefficient β derived from a selected proxy scalar for REA, eddy diffusivity K_H derived from sensible heat for AGM, and proxy scalar such as sensible heat, CO_2, and H_2O flux for MBR) (Lindberg et al., 1995a; Edwards et al., 2005; Baya and Van Heyst, 2010; Zhu et al., 2015b). These empirical factors may introduce uncertainties when the proxy scalar value is small, which frequently occurs during dawn and dusk and under the condition of low atmospheric turbulence. Proxy scalar inferred relaxation coefficients (β_{CO_2} , β_{T_s} , and β_{H_2O}) were typically not significantly different (~0.56) during a campaign above wheat agricultural land, while all β values were highly variable when the corresponding scalar flux was close to zero (Gronholm et al., 2008; Sommar et al., 2013b). Converse et al. (2010) reported Hg^0 fluxes over a wetland meadow using collocated AGM and MBR methods for four campaigns during an entire year. They found comparable fluxes in the summer, while source/sink characteristics reversed between the two methods in fall and winter. Zhu et al. (2015b) found that the AGM and MBR methods observed similar Hg^0 fluxes when absolute sensible heat flux was >20 W m^{-2}; and the agreement was not satisfactory when the absolute sensible heat flux was <20 W m^{-2}. Rejecting flux data collected under low turbulence conditions can bias the integrated flux over time (Mauder and Foken, 2004); and adequate data rejection and correction approaches need to be developed (Aubinet et al., 2012).

2.3 Comparability of flux measured by micrometeorological and chamber methods

Limited efforts have been devoted to understanding the flux disparity caused by different flux measurement techniques. The Nevada STORMS project was the first attempt using eleven collocated measurements (7 DFC methods and 4 gradient-based MM methods) to simultaneously quantify Hg^0 fluxes from Hg-enriched bare soils in September 1997 (Gustin et al., 1999; Lindberg et al., 1999; Poissant et al., 1999; Wallschläger et al., 1999). In the campaign, the mean fluxes obtained using MM methods were three times greater than those obtained by DFCs (Fig. 2a). One possible reason for the low observed fluxes by DFC was the small flushing flow rates (corresponding TOT: 1.1-24 min) that were not sufficient in eliminating the accumulated Hg^0 in the DFC and subsequently suppressed Hg^0 evasion. Later, Gustin and coworkers extended the study at the same site using a 1-L polycarbonate DFC (TOT: 0.2 min) (Engle et al., 2001) and a MBR method (Gustin et al., 1999) in October

1998 (Gustin, 2011). Although the MBR method showed substantial flux variability, DFC and MM fluxes were not significantly different (p>0.05) for dry and wet diel flux cycles (Fig. 2b). Two challenges in comparing MM and DFCs fluxes in these studies were the site heterogeneity (1.2-14.6 μg Hg g^{-1} in soil) and the footprint differences. The footprint of MM methods was estimated to be 40-70 m upwind from the sampling sites (50-200 m^2) while DFC covered only 0.12-0.3 m^2 (Gustin et al., 1999). Recently, an integrated field Hg^0 flux methods intercomparison project measured Hg^0 flux from a background homogenized agricultural field (~45 ng Hg g^{-1}) using REA, AGM, MBR, a polycarbonate NDFC (TOT: 0.47 min), and a traditional quartz DFC (TDFC, TOT: 0.32 min) (Fu et al., 2008a; Zhu et al., 2015a, b). Overall, the MM fluxes showed highly dynamic temporal variability while the DFCs followed a gradual diel cycle similar to those for temperature and solar irradiance. REA observed a broader flux distribution similar to NH_3 and CH_4 fluxes observed by MM techniques (Beverland et al., 1996; Moncrieff et al., 1998; Nemitz et al., 2001). The median fluxes obtained by REA, AGM, and MBR were not significantly different (Friedman two-way analysis, $\chi^2 = 1.29 < \chi^2_{p=0.05} = 5.99$). Over a three-week period, NDFC obtained a comparable mean flux with AGM and MBR which are approximately three times that of the TDFC flux, implying that NDFC potentially reduced the uncertainty using real atmospheric boundary shear condition to rescale (Lin et al., 2012). However, the correlation between NDFC/TDFC and MBR fluxes were weak because of the high variability of MM fluxes (Fig. 2c). Pierce et al. (2015) observed comparable mean fluxes using simultaneous measurements of CRDS-EC, MBR and DFC (849 ng m^{-2} h^{-1}, 1309 ng m^{-2} h^{-1}, and 1105 ng m^{-2} h^{-1}, respectively) over Hg-enriched soils, similar flux patterns were recorded from CRDS-EC and MBR.

Fig. 3 showed the comparisons of Hg^0 fluxes measured by MM methods and DFCs from relative homogeneous landscapes reported in the literature (each data represent campaign/site-based average flux which constitutes hourly, daily, and seasonal flux, cf. Section 4, substrate total Hg < 0.3 μg Hg g^{-1}). MM methods yield a broader Hg^0 flux range compared to DFCs methods, consistent with the field campaigns using collocated measurements (Zhu et al., 2015b). MM mean flux is higher than DFCs flux by a factor of approximately two, which may be a result by the fact that a large fraction of DFC measurements utilized a relative low TOT underestimating the surface flux. However, a Mann-Whitney U test indicated that the differences between the two methods are not significant (p=0.16, one-tailed). Probabilities of the two data sets showed positive skewness

(4.2 and 3.9 for MM and DFCs, respectively) and kurtosis (19.6 and 27.2) caused by those high flux observations, likely resulting from asymmetrical data distribution as well as differences in measurement sites and periods. The flux data of MM methods in Fig. 3 were mostly obtained from agricultural fields (33%) and grasslands (36%) while the data of DFC methods were mainly from background sites (68%); and MM measurement generally covered a longer period (weeks to year) compared to DFC measurements which lasted a much shorter period (hours, days to a few weeks). Typically, significant Hg^0 evasion is observed during daytime, while deposition, bidirectional exchange, or mild emission occurs at nighttime (cf. Section 4). Agnan et al. (2016) summarized MM and DFC fluxes observed in laboratory and during field campaigns over terrestrial substrates, and found that the observed median MM flux (-0.01 ng m^{-2} h^{-1}, n=51) was statistically smaller than the median DFC flux (0.5 ng m^{-2} h^{-1} and 1.75 ng m^{-2} h^{-1} for flushing flow rate ≤ 2 L min^{-1} and > 2 L min^{-1}, $p < 0.05$). They suggested that an elevated flushing flow rate generated a partial vacuum inside DFC and created an artificial Hg^0 flux from the soil even at <2 L min^{-1}. However, this hypothesis is not supported by the large Hg^0 concentration gradient (inside and outside the DFC) formed at low flushing flow rate (Zhang et al., 2002; Eckley et al., 2010). An alternative explanation is that MM measurements were predominantly deployed for background vegetated surfaces while DFCs were mainly applied for soil surfaces, the difference in the source/sink characteristics over vegetation and bare soils might have caused the differences in the median fluxes.

3. Factors influencing air-surface Hg^0 exchange

3.1 Air-soil Hg exchange

Meteorological parameters (e.g., solar radiation, soil/air temperature, atmospheric turbulence), soil substrate characteristics (e.g., Hg content, soil moisture, organic matter, porosity, and microbial activity), and ambient air characteristics (e.g., Hg^0 and O_3 concentration) can influence the air-surface exchange of Hg^0. Changes of these factors force two controlling processes: (1) the formation of evaporable Hg^0, and (2) the mass transfer of Hg^0. Solar radiation has been found to be highly positively correlated with soil Hg^0 flux (Carpi and Lindberg, 1997; Boudala et al., 2000; Zhang et al., 2001; Gustin et al., 2002; Poissant et al., 2004a; Bahlmann et al., 2006), which is generally regarded as enhancing Hg^{II} reduction and therefore facilitating Hg^0 evasion (Gustin et al., 2002). Actinic light spectral analysis suggested UV-B can reduce Hg^{II} to Hg^0 over soil, while UV-

A and visible light have a much lower enhancement (Moore and Carpi, 2005; Choi and Holsen, 2009b). Temperature is an important factor promoting Hg^0 evasion, typically following the Arrhenius equation (Carpi and Lindberg, 1997; Poissant and Casimir, 1998; Gustin et al., 2002). However, the Arrhenius relationship cannot explain the Hg^0 flux spikes at sub-zero temperatures, implying that other mechanisms such as the expansion and contraction of liquid fraction in soil substrates occurred (Corbett-Hains et al., 2012). Atmospheric turbulence (i.e., wind and surface friction velocity) is another factor that drives the Hg^0 release from soil (Lindberg et al., 1999; Wallschläger et al., 1999). Increased turbulence enhances Hg^0 mass transfer and promotes Hg^0 desorption from soil (Gustin et al., 1997; Lindberg et al., 2002c; Zhang et al., 2002; Eckley et al., 2010; Lin et al., 2012).

Soil types, soil moisture and Hg content in soil are also important factors influencing observed Hg^0 flux (Xu et al., 1999; Kocman and Horvat, 2010; Lin et al., 2010a). Lindberg et al. (1999) observed that rainfall and irrigation enhances soil Hg^0 emission by an order of magnitude. Subsequent studies supported that adding water to dry soil promotes Hg reduction and that water molecules likely replace soil Hg^0 binding sites and facilitates Hg^0 emission. In saturated soil, Hg emission is suppressed because the soil pore space is filled with water, which hampers Hg mass transfer (Gillis and Miller, 2000b; Gustin and Stamenkovic, 2005). Pannu et al. (2014) investigated Hg^0 flux over boreal soil by manipulating soil moisture, a maximum flux was observed at 60% soil moisture (water filled pore space), whereas flux became inhibited at 80%. Repeated rewetting experiments showed a smaller increase in emission, implying “volatizable” Hg^0 needs to be resupplied by means of reduction and dry deposition after a wetting event (Gustin and Stamenkovic, 2005; Song and Van Heyst, 2005; Eckley et al., 2011b). Soil organic matter (SOM) have a strong affinity with Hg^0 and form stable complexes with Hg^{II} (Grigal, 2003; Skyllberg et al., 2006), and therefore diminish soil Hg^0 efflux (Yang et al., 2007). Mauclair et al. (2008) measured Hg^0 flux from sand (0.5 μg Hg g^{-1}) spiked with humic substances; and found that Hg^0 flux decreased sharply by incremental addition of up to 0.1% of humic matter. Higher soil porosity and enhanced soil disturbance have been suggested to facilitate Hg^{II} reduction and Hg^0 transfer from soil (Fu et al., 2012a; Bash and Miller, 2007). Small soil grain size, clay and silt occurrence with higher surface area and Hg content, showed higher Hg^0 flux (Edwards, et al., 2001; Gustin et al., 2002). Microbial induced reduction can enhance Hg^0 evasion but to a lesser extent (Fritsche et al., 2008a; Choi and Holsen, 2009b). Higher flux has also been observed by increasing soil pH value (Yang et al., 2007).

Elevated ambient Hg^0 concentration has been found to suppress Hg^0 flux by reducing the Hg^0 concentration gradient at the interfacial surfaces (Xin and Gustin, 2007). At locations where ambient Hg concentration is high (e.g., mining sites, landfills), deposition is predominately observed despite of influence of meteorological factors (Bash and Miller, 2007; Wang et al., 2007b; Zhu et al., 2013c). Atmospheric O_3 has been found to induce not-yet-understood chemical processes that enhance Hg^0 emission from soil in the dark (Zhang et al., 2008). Laboratory experiments have shown that Hg^0 flux from soils with Hg^{II} as the dominant species can be enhanced by 1.7 to 51 times in the presence of O_3 (50-70 ppb), and be decreased by >75% over Hg^0-amended soils (Engle et al., 2005). Environmental factors interact naturally (e.g., irradiation and temperature), which can impose synergistic and antagonistic effects on forcing Hg^0 flux changes (Gustin and Stamenkovic, 2005). Fig. 4 shows the individual effects and synergism between solar radiation, air temperature, and water content on Hg^0 flux from a typical low organic content soil (~1.5 wt %) (Lin et al., 2010a). The three individual factors enhance flux by 90%-140%, while two-factor synergetic effect accounts for additional 20%-30% enhancement.

3.2 Air-vegetation Hg^0 exchange

Vegetation alters air-ecosystem Hg^0 flux through (1) changing environmental variables at ground surfaces (e.g., reducing solar radiation, temperature, and friction velocity) (Gustin et al., 2004), and (2) providing active surfaces for Hg uptake. Deforestation followed by biomass harvesting can increase forest floor irradiation and temperature and therefore enhanced Hg^0 emission (Carpi et al., 2014; Mazur et al., 2014). Carpi et al. (2014) reported forest floor soil fluxes of -0.73±1.84 and 0.33±0.09 ng m^{-2} h^{-1} from intact New England and Amazon forest floors, respectively. Substantial emission fluxes at 9.13±2.08, 21.2±0.35 ng m^{-2} h^{-1} were observed after deforestation, suggesting forest coverage effectively reduced ground floor Hg^0 emission. More importantly, air-plant interaction increases the complexity of air-terrestrial Hg exchanges; and the role of vegetation as a source or a sink of atmospheric Hg has been in debate in the literature. Lindberg et al. (1998) observed a significant Hg^0 emission from forest canopies in Tennessee and Sweden (10-300 and 1-4 ng m^{-2} h^{-1}). They estimated annual Hg^0 emission from global forest to be 800-2000 tons; and emphasized the need for a re-assessment on this potentially important source. Based on the observed Hg presence in xylem sap (Bishop et al., 1998), plant has been hypothesized as a conduit for releasing geospheric Hg to the atmosphere (Leonard et al., 1998a, b). Subsequent models simply treated plant emission as a function of evapotranspiration rate (Xu et al., 1999; Bash et al., 2004; Gbor et al., 2006; Shetty et al., 2008). However, more recent measurements suggested that air-surface exchange of Hg^0 is largely bidirectional between air and plant and that growing plants act as a net sink (Ericksen et al., 2003; Stamenkovic et al., 2008; Hartman et al., 2009). Stable Hg isotope tracer studies have shown that Hg in soils cannot be translocated from roots to leaf due to the transport barrier at the root zone (Rutter et al., 2011b; Cui et al., 2014), suggesting that the source of Hg in the leaf is of atmospheric origin.

Hg concentration in foliage is generally influenced by the level of ambient air Hg^0 (Ericksen et al., 2003; Frescholtz et al., 2003; Ericksen and Gustin, 2004; Millhollen et al., 2006a; Fay and Gustin, 2007a; Niu et al., 2011). Climate factors (e.g., solar irradiation, temperature), biological factors (e.g., leaf age, plant species), and ambient air components (e.g., CO_2) also significantly influence foliar Hg^0 flux (Rea et al., 2002; Millhollen et al., 2006a; Millhollen et al., 2006b; Fay and Gustin, 2007a; Bushey et al., 2008; Stamenkovic and Gustin, 2009; Rutter et al., 2011a). For instance, the higher Hg concentration found at the bottom aged leaf suggests the influence of longer exposure time (Bushey et al., 2008) over an immediate source from soil (Frescholtz et al., 2003). Stomatal and non-stomatal (e.g., cuticle) processes are both viable pathways for bidirectional Hg exchange (Stamenkovic and Gustin, 2009). Stomatal process may play a predominant role as Hg accumulated on cuticle surface was generally <10% of total Hg content in the leaf (Rutter et al., 2011a; Laacouri et al., 2013). Solar radiation, temperature, and CO_2 concentrations regulating plant stomatal activity may therefore affect Hg uptake and gas exchange. For instance, high air-vegetation Hg^0 fluxes observed during daytime show deposition, opposite to daytime evasion observed over other terrestrial surfaces (cf. Section 4.3) (Stamenkovic et al., 2008). In addition, Hg in leaf has been shown to be assimilated into leaf biomass during the growing stage (Bash and Miller, 2009), suggesting that Hg uptake occurs with plant assimilation metabolism.

It has been proposed that Hg in leaf can be classified as two forms: (1) exchangeable Hg which can be re-emitted back to the atmosphere, and (2) biologically assimilated Hg retained in the leaf (Rutter et al., 2011a). However, whether or not Hg^0 can be oxidized after uptake into tissue, and the possibility of assimilated Hg being reemitted from the leaf (e.g., reduction of leaf retained Hg^{II} or un-oxidized Hg^0 originally from ambient air) remain unclear. Many studies observed a so-called "compensation point" denoting the interfacial concentration of Hg that drives the concentration gradient for bidirectional air-vegetation exchange of Hg^0 (Hanson et al., 1995;

Poissant et al., 2008; Bash and Miller, 2009). However, the hypothesis of a compensation point does not explain the accumulation of Hg in the vegetation pool. Recent Hg isotopic fractionation studies show promise for exploring air-leaf Hg exchange mechanisms. Demers et al. (2013) reported a kinetic mass dependent fractionation (MDF, δ^{202}Hg) of -2.89‰ during air-leaf Hg exchange from air to leaf. The result indicated that uptake of atmospheric Hg by the leaf occurs, and the deposited Hg is likely to be chemically bonded in the leaf with sulfur and nitrogen functional groups in enzymes within stomatal cavities (Rutter et al., 2011a), rather than with carboxylic ligands on the leaf surface. Another important finding is that negative mass independent fractionation (MIF, Δ^{199}Hg) of Hg of -0.19‰ to -0.29‰, correlated well with Hg^{II} photochemical reduction by low molecular mass organic matter with sulfur-containing ligands (Zheng and Hintelmann, 2010). This implies that the Hg reemission may result from re-volatilization of chemically-bounded Hg in the leaf. However, they did not rule out the potential influence of PBM and GOM that deposit on the leaf, which may undergo partial uptake by the plant with the remaining being reemitted back to the atmosphere.

3.3 Air-water Hg exchange

The bulk methods, DFC, and MM methods have been utilized in air-water Hg^0 flux measurements. Bulk methods is the most widely utilized approach for the oceanic surface (>80% of the field data, Table 1). Sommar et al. (2013a) summarized the methodologies of the bulk method, which is generally controlled by both kinetic (overall mass transfer coefficient, *k*) and thermodynamic (partial pressure related concentration gradients) forcing (Wanninkhof, 1992; Wanninkhof et al., 2009; Kuss et al., 2009; Kuss, 2014):

$$F = k \times \left(DGM - GEM / H_T^{'} \right) = 0.31 \times U_{10}^{2} \times \left(\frac{v}{600 \times D_{Hg^0}} \right)^{-0.5} \times \left(DGM - GEM / H_T^{'} \right) \qquad (3)$$

where DGM is the dissolved gaseous Hg concentration in the surface water film, GEM is the near surface gas Hg^0 concentration, $H_T^{'}$ is the dimensions Henry's law constant, U_{10} is the wind speed at 10m, v is the water kinematic viscosity, and D_{Hg^0} is the Hg^0 diffusion coefficient in water. Fig. 5 shows air-surface exchange processes and transformation of DGM in the water phase. From a kinetic point of view, the overall mass transfer coefficient of Hg^0 is described by a molecular diffusivity in the water and gas film. Since the mass transfer boundary layer of water has a much greater resistance than the gaseous layer for sparingly soluble Hg^0, the overall mass transfer coefficient is limited by the water transfer velocity (Eq. 3) (Kim and Fitzgerald, 1986). Surface wind speed is an important driving force enhancing the mass transfer coefficient in water (Qureshi et al., 2011b), D_{Hg^0} has been experimentally determined as a function of temperature (T, Kelvin) for freshwater ($D_{Hg^0}^{fresh} = 0.0335e^{-18.63/RT}$, R represents gas constant) and seawater ($D_{Hg^0}^{sea} = 0.0011e^{-11.06/RT}$) (Kuss, 2014).

Processes controlling the concentration of DGM in surface water directly regulates air-water Hg^0 flux. Photochemically induced Hg^{II} reduction is the predominant pathway of DGM formation in surface water (Amyot et al., 1994; Amyot et al., 1997a; Amyot et al., 1997b; Costa and Liss, 1999; Lalonde et al., 2001; Zhang and Lindberg, 2001; Feng et al., 2004). Zhang (2006) summarized the Hg photochemical redox chemical process. Eq. (4) resembles a simplified scheme of gross photo-reactions governing the DGM pool in surface waters (O'Driscoll et al., 2006; O'Driscoll et al., 2008; Qureshi et al., 2010):

$$Hg_{reducible} + photoreductants \rightleftharpoons DGM + photooxidants \qquad (4)$$

Fe^{III} has been reported to enhance sunlit photo-reduction in natural water (Lin and Pehkonen, 1997; Zhang and Lindberg, 2001). Complexes of Fe^{III}-natural organic ligands were hypothesized to undergo photolysis to form reactive intermediates (e.g., organic free radicals) capable of reducing Hg^{II}. Dissolved organic matter (DOM) serving as electron donor and complexation agent in the natural water is the most important precursor for formation of photo-reductants (Ravichandran, 2004; Vost et al., 2011; Zhang et al., 2011). Similarly, irradiation derived photo-oxidants may oxidize DGM simultaneously and reduce Hg evasion from water. Reactive radicals (e.g., $\cdot O_2$, $\cdot OH$) produced through DOM, NO_3^- photolysis have been identified as possible oxidants (Lin and Pehkonen, 1997; Zhang and Lindberg, 2001; Zhang et al., 2012b). In addition, Cl^- has been reported to enhance photo-oxidation by stabilizing the oxidative products ($HgCl_n^{2-n}$) and facilitating oxidation via formation of highly oxidizing ligand (Cl_2^-) (Yamamoto, 1996; Lalonde et al., 2001; Sun et al., 2014). Secondary radicals (e.g., $CO_3^{\bullet -}$) can sometimes act as a photo-oxidant (He et al., 2014).

Field studies also observed DGM and Hg^0 flux peaks in the nighttime, suggesting the importance of dark reduction (O'Driscoll et al., 2003; Zhang et al., 2006b; Fu et al., 2013b). Both dark abiotic and biotic redox transformations have been suggested to be involved (Fig. 5). Although dark abiotic reduction takes place mainly in the anoxic environment (Gu et al., 2011; Zheng et al., 2012), it also occurs in the oxic condition at a lower reaction rate (Allard and Arsenie, 1991). Natural organic matter shows reducing, oxidizing, and complexing properties with Hg in the anoxic environment due to its diversity functional groups (e.g., thiols group, quinones and semiquinone groups, carboxyl group) (Gu et al., 2011; Zheng et al., 2012; Zheng et al., 2013). Although aqueous liquid Hg droplets can be rapidly oxidized in oxygenated chloric water, DGM is unable to be oxidized under such conditions (Amyot et al., 2005).

Biological redox transformation is another important DGM cycling pathway. Ariya et al. (2015) reviewed the biological processes in Hg redox transformation, which contains phototrophic and chemotrophic Hg redox processes. Aquatic algae, cyanobacteria, and diatoms involved phototrophic Hg reduction was positively correlated with photosynthetic activities, which is likely a bio-detoxification process (Ben-Bassat and Mayer, 1975; Kuss et al., 2015). In addition, photo-reactivation of DOM and Fe^{III} facilitates Hg^{II} reduction through algae (Deng et al., 2009). Kuss et al. (2015) reported that cyanobacteria-light synergetic and photochemical transformation equally contributed to ~30% DGM production in the Baltic Sea, while low-light production contributed ~40%, highlighting the importance of biotic reduction. Two pathways have been identified for Hg^{II} reduction by bacteria. The first is reduction by Hg-resistant microorganisms where Hg^{II} is reduced in the cell's cytoplasm by mercuric reductase and transported out as Hg^0 (Barkay et al., 2003); the other is Hg^{II} reduced by Hg-sensitive dissimilatory metal-reducing bacteria utilizing iron and/or manganese as a terminal electron acceptor during respiration (Wiatrowski et al., 2006). Intracellular oxidation is supposed to be mediated by oxidase (Siciliano et al., 2002), while extracellular thiol functional groups on cell membrane also show capabilities in oxidizing Hg^0 under anoxic environment (Colombo et al., 2013; Hu et al., 2013). A review of genetic-based microbial Hg redox transformation can be found in Lin et al. (2011).

3.4 Air-snow Hg exchange

Schroeder et al. (1998) reported episodes of unexpected low Hg^0 concentrations in the Arctic air during spring time, so-called atmospheric mercury depletion events (AMDEs), through an array of photochemically initiated oxidation by halogens (Lindberg et al., 2002a; Sommar et al., 2007; Moore et al., 2014). The phenomena was finally confirmed widespread in the coastal Polar Regions. During AMDEs, a large amount of surface layer Hg^0 is oxidized and deposited in snowpack via GOM and PBM dry deposition (Steffen et al., 2008). The deposited Hg onto snow can be rapidly re-volatilized back to the atmosphere via photochemical Hg^{II} reduction on snow or in melted snow (Dommergue et al., 2003; Faïn et al., 2007; Kirk et al., 2006). Photo-reduction is the predominant pathway for Hg reemission from snow as inferred by Hg isotope fractionation signatures (Sherman et al., 2010). The reduction rate was found to be linearly correlated with UV intensity (Lalonde et al., 2002; Mann et al., 2015b), while Cl^- showed an inhibiting effect on the photo-reduction (Section 3.3) (Steffen et al., 2013). Oxidation and reemission of Hg^0 occurred simultaneously with the presence of oxidants (e.g., $\cdot OH$, Cl, and Br) formed through photolysis (Poulain et al., 2004). Nighttime elevated GEM in snow air was observed at Station Nord, Greenland, likely a result from dark formation of reducing radicals (e.g., $HO_2\cdot$) (Ferrari et al., 2004). Temperature is another factor enhancing Hg emission from snow by changing the solid and liquid water ratio (Mann et al., 2015a). Hg^0 flux from snow surface in the temperate regions has rarely been investigated (Faïn et al., 2007). Field data collected in Ontario and Northern New York confirmed that photo-reduction is the predominant pathway in enhancing Hg^0 emission (Lalonde et al., 2003; Maxwell et al., 2013). A positive correlation between Hg^0 fluxes and temperature has also been found (Maxwell et al., 2013). Hg^0 flux over snow cover under forest canopy was found to be smaller compared to those found in open field, possibly caused by lower light under the canopy (Poulain et al., 2007).

4. Global observation of atmosphere-biosphere Hg exchange

4.1 Data sources, extraction and processing

A comprehensive database of global observation of Hg^0 flux over terrestrial and oceanic surfaces is compiled from the field observed data reported in peer-reviewed literature; and the fluxes over water surfaces calculated using two-film gas exchange model based on *in-situ* measured DGM are also included. For those studies that measured TGM (Hg^0 + GOM), the measured flux is regarded as Hg^0 flux because of the small fraction of GOM in TGM measurement (GOM/GEM < 2% in general, Gustin and Jaffe, 2010, Sprovieri et al., 2010; Fu et al., 2015b), therefore Hg^0 and TGM are not discriminable for Hg vapor analyzer during a typical concentration measurement period (5 mins sampling, 1.0-1.5 L min^{-1}) in flux sampling. As complete time-series flux datasets are not available in the literature, each data point included in the database corresponds to the arithmetic mean of the flux observed during each campaign, with the campaign period lasting up to one year. For those studies that periodically (e.g., weekly) measured seasonal fluxes at the same site, the average flux of all of the campaigns was used. A summary of Hg^0 flux data documented in a total of 172 peer reviewed articles are presented in Table 1, which were obtained using the DFCs (85.6%), MM (7.9%), $Hg^0/^{222}Rn$ flux ratio (0.3%), and enriched isotope tracers (0.1%) methods, or estimated using the two-film gas exchange model (6.1%). Based on the landscape characteristics and surface Hg contents, the flux datasets are assigned to 11 categories. Classification of background soils (e.g., open field bare soil and forest ground soils with little perturbation by human activities) follows the corresponding literature definition. Soil Hg content of ≤0.3 μg g^{-1}, which representing terrestrial background level (Agnan et al., 2016), was applied as the threshold for background soil in case no classification was assigned in the original article. Hg contaminated sites were divided into natural enriched and anthropogenic contaminated sites based on the Hg sources. The remaining flux data were categorized into 9 classes according to the land uses and ecosystem types (Table 1). It is important to recognize that the Hg^0 fluxes represent the experimental and modeling results using diverse methodologies with campaign periods of different durations. Given the reasonably large flux sample sizes, the flux statistics (e.g., mean, median) from multiple studies for different landscapes are compared. It should be noted that the flux reported in laboratory studies and field experiments utilizing treated/untreated substrates are *not* included in the database. Instead, the implications of those studies are discussed in terms of the environmental effects of Hg^0 exchange mechanisms (cf. Section 3).

4.2 Global database of earth surfaces-atmosphere Hg^0 flux

Table 1 summarizes the statistics of Hg^0 fluxes measured to date. The site characteristics where Hg^0 flux measurements were performed are highly diverse. Most studies were devoted to flux investigations over natural Hg-enriched sites (~38.2%) and background surfaces (~18.4%). Direct field measurements over terrestrial surfaces accounts for 94.1% (n=811) of the data, only 5.9% (n=51) of the data represents oceanic fluxes. In terms of substrate Hg contents, measurements at contaminated sites (natural Hg-enriched and anthropogenic polluted) consisted of 44.9% of the datasets, motivated by extensive emission at these sites which may cause local and regional atmospheric pollution. For unpolluted terrestrial surfaces, most measurements were carried out over background soils (37%, n=159), while only a few studies directed to the forest foliage and above canopy flux (n=8). DFC methods are suitable for bare soil and low vegetated surface, covering 97% of the data over background soils. The remaining datasets are observations of ecosystem flux using MM methods, which require relatively more complex instrumentation and experimental efforts in the field (Gustin, 2011; Aubinet et al., 2012; Sommar et al., 2013a).

Fig. 6 shows the box and whisker plots of Hg^0 fluxes. As seen, the categorized data exhibit substantial data variability and positive skewness. Many campaigns focus only on daytime flux (cf. Section 4.3.2) and therefore the median flux in each category is a more appropriate statistic for comparison. The medians of Hg^0 fluxes for the 11 site categories follow the order: grasslands < forest foliage & canopy level < background soils < wetlands < seawater < snow < freshwater < urban settings < agricultural fields < anthropogenically contaminated surfaces < natural Hg-enriched surfaces (Table 1). A clear increase in flux from background to contaminated sites suggests the strong influence of substrate Hg contents on Hg^0 flux. Median fluxes from contaminated sites are two orders of magnitude greater than those over other surfaces; such source strength significantly enhances local and regional atmospheric Hg concentration. Fluxes over vegetative surfaces (grasslands, forest foliage and canopy level), mixed vegetated waters (wetlands) are lower than those over background soils and open water (freshwater and seawater), supporting that vegetation reduces Hg emission by masking ground floor evasion and/or plant uptake. The fluxes at human perturbed urban settings and over agricultural fields were higher than the fluxes over undisturbed earth surfaces, likely a result of the reemission of legacy Hg deposition. Most surfaces showed Hg^0 emission; approximately 25% of measurements over vegetated surfaces showed net Hg deposition (Fig. 6).

Results of frequency analysis of the mean Hg^0 fluxes for each land cover are presented in Fig. 7. While the mean Hg^0 flux from background soils have a large range (-51.7 to 33.3 ng m^{-2} h^{-1}), ~90% of the flux data range from -5 to 10 ng m^{-2} h^{-1}. Similar patterns are also evident for freshwater, oceans, grasslands, and wetlands. The occasional high emission and deposition fluxes are mainly due to short sampling duration (e.g., mid-day flux) or extreme atmospheric Hg^0 concentration events caused by local/regional sources. Comparatively, fluxes over agricultural fields and in urban settings show a much larger range and a lower kurtosis. Strong Hg^0 evasion was observed at contaminated sites (>97% of total observations showed evasion), although extremely high deposition also occurred in the presence of high ambient Hg^0 and atmospheric subsidence (Bash and Miller, 2007; Zhu et al., 2013c). Most measurements over snow (87%) show evasion; these studies were carried out in the Polar Regions and focused on Hg reemission from snow after AMDEs. The distribution of Hg^0 fluxes of air-foliage and canopy level exchange showed that half of the measurements (n=4) gave a net emission, while the mean flux is not significantly different from zero (p=0.24, ANOVA).

4.3 Spatial distribution and temporal variation of global Hg^0 flux data

4.3.1 Spatial distribution

Fig. 8 shows the box and whisker plots of Hg^0 flux from four relatively homogeneous surfaces (background soils, agricultural fields, grasslands, and freshwater) observed in different regions. Worldwide fluxes measurements were unevenly distributed, most studies were conducted in North American and East Asia, which limits global representativeness. Hg^0 fluxes observed in East Asia are consistently higher compared to those measured in Europe, North and South America, Australia, and South Africa ($p < 0.05$, ANOVA, except freshwater). This can be explained by the greater anthropogenic emission and re-emission of Hg deposition (Selin et al., 2007; Selin et al., 2008; Lin et al., 2010b; Smith-Downey et al., 2010). The fluxes over freshwater bodies in Europe are somewhat higher than those measured in East Asia (6.5 vs. 4.6 ng m^{-2} h^{-1}, p=0.40, ANOVA). These Europe data were obtained mostly prior to 2002 (n=9) or during summer time and daytime (n=8) (Schroeder et al., 1989; Xiao et al., 1991; Lindberg et al., 1995b; Gårdfeldt et al., 2001; Feng et al., 2002). The earlier measurements were also subject to higher blank and larger extent of photo-reduction and evaporation. These complicating factors could have yielded higher fluxes.

4.3.2 Diurnal and seasonal patterns

Fig. 9 displays the general diel variation of Hg^0 fluxes measured by DFC and DFB methods. Fluxes are typically higher during daytime and lower at nighttime from soil, mines, water and snow surfaces, where Hg^0 can be formed through photo-reduction. As discussed in Section 3, the observed diel variations are in agreement with results from laboratory controlled studies: higher irradiance and temperature promoted Hg^0 reduction and evasion in the daytime, which formed a “dome-shaped” diel flux pattern for most of the earth surfaces (e.g., soils, mine, water, and snow). On the contrary, greater deposition during daytime and evasion/near-zero-flux at nighttime have been frequently observed from foliage, possibly facilitated by the uptake through the stomata that exhibit higher stomatal conductivity during the daytime.

Seasonally, higher evasion flux occurs in the warm season and smaller exchange is observed in the cold season. For example, the seasonal data from Choi and Holsen (2009a) showed a higher evasion from forest floor soil in the Adirondack Mountain (New York, USA) in the summer (1.46 ng m^{-2} h^{-1}) shifted to insignificant exchange in the winter (0.19 ng m^{-2} h^{-1}). Similar trends were also found in agricultural soils, freshwater, and mine surfaces (Fu et al., 2010a; Eckley et al., 2011b; Zhu et al., 2011). However, an opposite low flux in spring and summer was observed in a pristine background deciduous forest floor (Hartman et al., 2009; Kuiken et al., 2008a), which likely due to drier conditions and lower irradiation resulted from intense leaf cover. Observed diurnal and seasonal patterns may also be influenced by vegetative surface changes and meteorological characteristics (Bash and Miller, 2009; Fritsche et al., 2008b; Lee et al., 2000; Sommar et al., 2015). For example, Sommar et al. (2015) reported seasonal flux observations over a wheat-corn rotation cropland using REA measurements, an unexpected low flux was observed in the summer during the corn growing stage (median: -6.1 ng m^{-2} h^{-1}) due to the uptake by the corn leaf (leaf area index 2.7-3.6), which is similar to the flux (-6.7 ng m^{-2} h^{-1}) observed in the winter and much lower than the wheat canopy flux (13.4 ng m^{-2} h^{-1}) in early spring. The limited availability of seasonal data in peer-reviewed literature does not allow a thorough assessment of seasonal characteristics of different terrestrial surfaces. It is important to recognize that the modified landscapes and vegetative biomass growing cycle caused by seasonal changes (e.g., change of LAI in deciduous forest, growing season of forest ecosystem, etc.) may significantly modify the flux characteristics. More data, especially measurements using consistent techniques over a longer campaign period (e.g., 1 year or longer), are needed for addressing the seasonal variability of Hg^0 exchange and better estimating the annual exchange from vegetative surfaces. To accomplish such measurements, automation of flux quantification apparatus is also required.

4.4 Source and sink characteristics of natural surfaces in the context of global Hg budget

Based on the findings of the above data synthesis, the implications regarding the role of natural surfaces in modifying global Hg budget are discussed as followed.

Background soils and water are important diffuse sources of Hg^0. Although Hg^0 flux observed over background soil (1.3 ng m^{-2} h^{-1}) and unpolluted water bodies (2.8 and 2.5 ng m^{-2} h^{-1} for fresh and seawater) may appear mild (Table 1), the annual emission from these two types surfaces accounted for 64% of total atmospheric Hg emission because of their large areal coverage globally (Pirrone et al., 2010). For example, it has been estimated that bare soil releases ~550 Mg yr^{-1} (Selin et al., 2008; Pirrone et al., 2010) and surface ocean releases 2000-2900 Mg yr^{-1} of Hg^0 globally (Fig. 1) (Mason et al., 2012; AMAP/UNEP, 2013). Agnan et al. (2016) projected terrestrial background surfaces flux ~129 Mg yr^{-1} using compiled laboratory and field measured Hg^0 flux, however, the uncertainty various from -829 (37.5th percentile) to 1037 (62.5th percentile) Mg yr^{-1}. Constraining the uncertainties on Hg^0 emission from these diffuse sources will greatly improve the accuracy of the global Hg budget. Global Hg^0 evasion from soil is mainly based on the empirical relationship between flux, temperature and irradiation (cf., Section 5), which needs mechanistic refinement. Air-seawater exchange estimated by global models is subject to the uncertainties in: (1) mechanisms of aqueous redox transformation and the associated kinetic parameters, and (2) Hg^0 mass transfer rates as determined by surface fiction velocity (Qureshi et al., 2011a). Kinetic parameters of these processes largely rely on limited field data without experimental verification (AMAP/UNEP, 2013) and require further investigation. Parameterization of Hg^0 flux using field data and redox transformation rate constants in soil and water are critical to reduce the uncertainty in future studies.

Contaminated surfaces are intensive local Hg^0 sources. Hg evasion from contaminated surfaces (Fig. 6 and Fig. 7) has been recognized as an important input contributing to the regional atmospheric Hg budget (Ferrara et al., 1998b; Kotnik et al., 2005). Hg^0 fluxes from contaminated point sources have been extensively investigated by using the LIDAR technique, which is by far the most effective experimental approach to spatially resolve the Hg^0 air-surface exchange at contaminated sites. Ferrara et al. (1998a) measured the spatial distribution TGM concentration and TGM flux from the world's largest Hg mine, the Almadén Hg mine, in Spain. TGM concentrations and fluxes were estimated to be 0.1-5 µg m^{-3} and 600-1200 g h^{-1} in fall, 1993, above the village of Almadén. Several attempts have been made to quantitatively estimate atmospheric Hg input in mining areas. Gustin et al. (2003) and Wang et al. (2005) applied a log-linear correlation between the flux and substrate Hg contents and solar irradiance. Eckley et al. (2011b) computed annual Hg emission from two active gold mines (up to 109 kg year^{-1}) using flux measurements and statistically derived the empirical relationship between fluxes and meteorological variables based on Geographical Information System (GIS) data. Similarly, Kocman and Horvat (2011) obtained ~51 kg year^{-1} emission from the Idrijca River catchment, a former Hg mine, using field measurement and GIS data. In total, annual Hg emission from global contaminated surfaces was estimated to be ~82 Mg via modeling of fluxes from more than 3000 Hg contaminated sites comprising Hg mining, non-ferrous metal production, precious metal processing, and various polluted industrial sites (Kocman et al., 2013), which are emitted from a very limited surface area thus can pose a strong environmental impact to the local area surrounding the contaminated sites.

Areas impacted by human activities exhibit elevated Hg^0 reemission. The median evasion flux over human urban settings and agricultural fields is 5-10 times higher than the value over background soils (Table 1). Direct anthropogenic Hg input and atmospheric Hg deposition explain the enhanced reemission. Natural surfaces near the anthropogenic point sources (e.g., power plant, Pb-Zn smelter, chlor-alkali plant) generally showed higher soil Hg content due to atmospheric Hg deposition (Lodenius and Tulisalo, 1984; Li et al., 2011; Zheng et al., 2011; Guédron et al., 2013). A fraction of these deposited Hg can be swiftly reemitted back to the atmosphere (Fu et al., 2012a; Eckley et al., 2015). Newly deposited Hg to soil, aquatic systems and snow packs in the Polar Regions can also be readily converted to Hg^0 and reemitted (Amyot et al., 2004; Poulain et al., 2004; Ericksen et al., 2005). Eckley et al. (2015) observed soil Hg^0 flux near a large base-metal smelter (Flin Flon, Manitoba, Canada) and reported a net deposition during operation (-3.8 ng m^{-2} h^{-1}) and elevated emission (108 ng m^{-2} h^{-1}) after operation ceased. To date, the source and sink characteristics of surfaces impacted by human activities have not been adequately investigated. Future investigations should be coordinated toward spatially resolving the Hg^0 exchange over human impacted surfaces for better quantifying the reemission budget of legacy Hg.

Fluxes over vegetated surfaces likely a sink but large uncertainties remain. Data of Hg^0 fluxes over foliage and forest canopy showed a small net emission (median: 0.7 ng m^{-2} h^{-1}) with substantial variability (Figs. 6 and 7). Extrapolating the median of database including laboratory flux from leaves, field measured leaves and forest floor flux, Agnan et al. (2016) predicted terrestrial forest act as a net Hg^0 sink (-59 Mg yr^{-1}). There have been conflicting reports regarding the role of forest ecosystems as Hg sources or sinks at global scale (Lindqvist et al., 1991; Lindberg et al., 1998; Frescholtz and Gustin, 2004; Fay and Gustin, 2007a; Fay and Gustin, 2007b; Hartman et al., 2009; Cui et al., 2014). Laboratory studies have suggested that plants are a net sink for atmospheric Hg through leaf assimilation (Millhollen et al., 2006a; Stamenkovic and Gustin, 2009; Rutter et al., 2011b; Cui et al., 2014). Using Hg concentrations in plant tissues and net primary productivity as a proxy for atmospheric Hg deposition, Obrist (2007) estimated plants remove ~1024.2 Mg yr^{-1} Hg globally (foliage contributed 237.6 Mg yr^{-1}). Fu et al. (2016) estimated that global litterfall contributes to 1232 Mg yr^{-1} of Hg deposition, throughfall contributes to 1338 yr^{-1} of Hg deposition, and the forest floor evades ~381 Mg yr^{-1} of Hg into the atmosphere. Hg content in forest soil is comparatively higher than the concentration found in bare soil due to the input via litterfall and wet Hg deposition (Blackwell and Driscoll, 2015a, b; Obrist et al., 2011); and ~90% boreal forest soil Hg was believed to be originated from litterfall input (Jiskra et al., 2015). These studies suggest that the forest ecosystem is likely a large atmospheric Hg sink, although these bulk proxy methods are not sufficiently sophisticated to resolve the global Hg mass balances. Synchronized, long-term observations of air canopy flux and litterfall/throughfall deposition are useful in helping to understand the source and sink characteristics of forests.

5. Modeling of air-surface Hg^0 exchange flux

A summary of recent modeling efforts on estimating natural emission is presented in Table 2. For air-foliage Hg^0 exchange, earlier parameterizations (S1, Table 2) calculate the flux as a function of the evapotranspiration rate based on soil-root-stem-foliage transpiration stream. It is assumed that Hg is passed through the soil-root interface and then transferred into foliage in as complexes with organic ligands (Moreno et al., 2005a; Moreno et al., 2005b; Wang et al., 2012). However, root uptake is unlikely to occur (Cui et al., 2014). Hg isotopic signatures between air and foliage (Demers et al., 2013; Yin et al., 2013), and air-foliage flux measurements (Graydon et al., 2006; Gustin et al., 2008) suggest that: (1) the exchange is bidirectional, and (2) atmospheric Hg uptake by foliage is the major pathway for Hg accumulation. Therefore, a bidirectional flux scheme building on the compensation point (S2, Table 2) is perhaps more scientifically sound and mathematically robust. For air-soil Hg^0 exchange, in addition to the bidirectional resistance scheme (S3), statistical relationships have been developed based on measured fluxes and observed environmental factors such as air/soil temperature, solar radiation, soil moisture and soil Hg content (S1-S2, Table 2), which tends to be site-specific and oversimplifies the influence of environmental factors (Wang et al., 2014). For air-water flux simulations, the two-film diffusion model is widely used by incorporating surface storage and aqueous Hg redox chemistry (Bash et al., 2007; Strode et al., 2007). Bash (2010) suggested a pseudo-first kinetic water photo-redox scheme in CMAQ simulations with bidirectional Hg exchange. Strode et al. (2007) parameterized the reduction rate as the product of local shortwave solar radiation, net primary productivity, and a scaling parameter in GEOS-Chem. Soerensen et al. (2010) updated the surface ocean redox reactions in GEOS-Chem, and added a term for dark oxidation, and suggested new linear relationships between the total solar radiation, net primary productivity, and photo-oxidation rate coefficient, photoreduction coefficient, and biotic reduction coefficient.

Using the S1 scheme (Table 2), the range of simulated air-foliage fluxes were 0 to 5 ng m^{-2} h^{-1} in North America (Bash et al., 2004) and 0 to 80 ng m^{-2} h^{-1} in East Asia (Shetty et al., 2008). Changing the modeling approach to resistance based models with a compensation point assumption (S2 scheme), the range was -2.2 to -0.7 ng m^{-2} h^{-1} (Wang et al., 2014). Zhang et al. (2012a) reported the annual Hg^0 uptake by foliage was 5-33 μg m^{-2} with the S2 scheme, similar to the litterfall Hg flux measured at Mercury Deposition Network Sites. For air-soil exchange, model-estimated fluxes ranged from 0 to 25 ng m^{-2} h^{-1} using the S1 and S2 scheme (Bash et al., 2004; Gbor et al., 2006; Shetty et al., 2008; Kikuchi et al., 2013), comparable to the 0-20 ng m^{-2} h^{-1} using the S3 scheme (Wang et al., 2014). For air-water exchange, the model-estimated fluxes were 1-12 ng m^{-2} h^{-1} (Bash et al., 2004; Shetty et al., 2008; Bash, 2010; Wang et al., 2014), consistent with measured fluxes (Table 1).

Future development of Hg^0 flux models require mechanistic understanding of air-surface exchange processes. Presently, in the bidirectional resistance scheme, the stomatal compensation point is treated as a constant value (Bash, 2010; Wang et al., 2014) or calculated as following in Wright and Zhang (2015):

$$\chi_{st} = 8.204 \frac{8.9803 \times 10^9}{T} \cdot \Gamma_{st} \cdot e^{-\frac{8353.8}{T}} \qquad (5)$$

Where T is the temperature of stomata/surface, and Γ_{st} is the emission potential of the stomata. Γ_{st} is an empirical input value and suggested as 5-25 ng m^{-3} depending on the specific land use. Battke et al., 2005; Heaton et al., 2005; and Battke et al., 2008 reported that plants have the ability to reduce the Hg^{II} to Hg^0 in foliar cells through reducing ligands (e.g., NADPH). To propose a more physically robust modeling scheme, the redox processes in foliage and the role of ligands on Hg uptake need to be better understood. The finding that Hg^0 can pass through the soil-root interface under artificial laboratory conditions (Moreno et al., 2005b) needs to be carefully verified in the field.

Another area that requires advancement is the determination of the Hg^{II} reduction rate (Scholtz et al., 2003;Bash, 2010;Wang et al., 2014) and the hypothetical parameter Γ_{st} (Wright and Zhang, 2015) in soil. It is well known that Hg^{II} can be reduced by natural organic acids via biotic/abiotic reduction (Zhang and Lindberg, 1999; Zheng et al., 2012). Experimental investigations showed that O_3 is important in controlling Hg emissions from substrates (Engle et al., 2005). However, the kinetic description of these process is fundamentally unknown. The pseudo-first reduction rate constant of Hg^{II} has been assumed to be in the range of 10^{-11} to 10^{-10} s^{-1} (Scholtz et al., 2003; Qureshi et al., 2011a). Under laboratory conditions at 100 W m^{-2} and 32±7 °C, the pseudo-first reduction rate was estimated to be $2\text{-}8\times10^{-10}$ m^2 s^{-1} w^{-1} basing on 2 mm soil depth (the maximum depth for light penetration in soil) (Quinones and Carpi, 2011). Si and Ariya (2015) reported a photo-reduction rate of Hg^{II} in the presence of alkanethiols to be $3\text{-}9\times10^{-9}$ m^2 s^{-1} w^{-1}. Other than these kinetic information, kinetic measurements for Hg^{II} reduction in the absence of light will enable additional mechanistic parameterization of Hg evasion models for soil and vegetative surfaces.

6. Conclusions and future perspectives

Understanding the air-surface exchange of Hg^0 has been steadily advancing since the mid-1980s. A substantial amount of data exists, but with large uncertainty and data gaps in Africa, South and Central Asia, Middle East, South America and Australia. Fundamentally, flux measurement approaches (e.g., MM and DFCs) are different and individual flux measurement data are not directly comparable. The Hg^0 flux data compiled in this study represent the current state of understanding that requires continuous updates. Hg^0 fluxes in East Asia are statistically higher than the values observed in other world regions, suggesting reemission of atmospheric deposition or a strong anthropogenic influence. Hg^0 exchange over weak diffuse sources (e.g., background soil and water) and vegetation need better constrains for global analysis of the atmospheric Hg budget through extensive on-site measurements and fundamental mechanical studies (e.g., redox transformation rate constant, mass transfer diffusivity). Although predominate factors controlling Hg^0 fluxes have been identified, the effects of those factors on flux have not been fundamentally and quantitatively determined for different surfaces, which limits the accuracy of flux modeling. Based on the synthesis in this study, the following knowledge gaps need to be addressed:

(1) Improving the temporal resolution and sensitivity of Hg^0 flux measurements. Insufficient temporal resolution and sensitivity in the detection of ambient Hg have limited our capability in accurately determining the air-surface exchange of Hg^0. Development of high temporal resolution and sensitive sensors for determining Hg^0 concentration gradients is of prime importance in improving flux data quality and in reducing uncertainties in the global assessment of the Hg budget. Such advancement will also open up new opportunities to explore fundamental exchange mechanism in response to the changes in environmental factors.

(2) Standardization of Hg^0 flux measurement techniques and establishment of a data comparison strategy. Hg^0 flux measurement uncertainties from using different techniques remains large, a standardized method is useful to compare fluxes obtained from various techniques. Fundamental studies are needed to compare contemporary Hg^0 flux quantification methods. Experimental approaches that synchronize the measured fluxes by different methods are also suggested in order to build empirical database for method inter-comparisons. Achieving this will largely reduce the uncertainty in the Hg budget estimation and greatly improve comparability of flux data reported by different research groups.

(3) Fundamental investigation on the environmental processes driving Hg exchange. Although flux responses to environmental parameters (e.g., irradiance, precipitation, temperature rising) are qualitatively defined in a statistical sense, the processes driving Hg^0 exchange need to be understood fundamentally. Recent advancement on isotopic tracing techniques (e.g., enriched Hg isotope tracers and stable Hg isotopic fractionation data) may offer mechanistic insights and new data should be incorporated into modeling analyses.

(4) Long-term measurement of Hg^0 fluxes at representative sites. There is a substantial data gap in the current Hg^0 flux database in terms of geographical coverage and land use type. Forests are most likely an overlooked sink for atmospheric Hg^0. However, few field campaigns have been conducted at forest sites. In addition, current flux databases are mainly obtained from short-term campaigns. It is unclear how global changes (e.g., climate change, global Hg emission reduction from anthropogenic sources) will force Hg^0 flux changes over different surfaces. There is presently no network of fluxes measurements at global monitoring sites. Continuous

observation of flux is useful for providing a more comprehensive database for scale-up estimation.

(5) Development and improvement of air-surface exchange models for Hg. The present state of development of air-surface exchange models does not allow appropriate process analysis due to a lack of fundamental understanding in the chemical and mass transfer processes of evasion and deposition. Existing air-surface Hg^0 flux schemes incorporate over-simplified chemical schemes with not-yet verified kinetic parameters. In addition, the interactions between Hg^{II} and organic matters in the natural environment, as well as the interfacial transfer of different Hg species over various surfaces, have significant knowledge gaps. Studies addressing these gaps are critically needed and will benefit not only the measurement approaches but also the model parameterization for estimating the global air-surface exchange of Hg.

Acknowledgements

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Table 1: A statistical summary of field *in situ* observed Hg^0 flux reported in the literatures.

Landscapes	Hg^0 flux (ng m^{-2} h^{-1})				*N*	References[b]
	Mean	Median	Min (*min*)[a]	Max (*max*)[a]		
Background soil	2.1	1.3	-51.7 (*-51.7*)	33.3 (*97.8*)	159	*(1)*
Urban settings	16.4	6.2	0.2 (*-318*)	129.5 (*437*)	29	*(2)*
Agricultural fields	25.1	15.3	-4.1 (*-1051*)	183 (*1071*)	59	*(3)*
Forest foliage & canopy level	6.3	0.7	-9.6 (*-4111*)	37.0 (*1000*)	8	*(4)*
Grasslands	5.5	0.4	-18.7 (*-989.6*)	41.5 (*870*)	38	*(5)*
Wetlands	12.5	1.4	-0.3 (*-375*)	85 (*677*)	23	*(6)*
Freshwater	4.0	2.8	-0.3 (*-18.2*)	74.0 (*88.9*)	93	*(7)*
Sea water	5.9	2.5	0.1 (*-2.7*)	40.5 (*46.0*)	51	*(8)*
Snow	5.7	2.7	-10.8 (*-2160*)	40 (*720*)	15	*(9)*
Natural enriched surfaces	5618	226	-5493 (*-9434*)	239200 (*420000*)	329	*(10)*
Anthropogenically contaminated surfaces	595	184	-1.4 (*-286.2*)	13700 (*13700*)	58	*(11)*

Notes: [a]. Min/Max are campaign/site-based average flux, while *(min)/(max)* represent lowest/largest instantaneous flux; [b] References: see **Appendix A**.

Table 2: A comparison of natural surface mercury flux models

	General models	**Description**	**References**
Foliage	S1: $F = ECs$	E: transpiration rate (g m^{-2} s^{-1})	Xu et al., 1999; Bash et al., 2004; Shetty et al., 2008; Gbor et al., 2006
		Cs: Hg^0 in soil water (ng g^{-1})	
	S2: $F_{st/cu} = \frac{\chi_{st/cu} - \chi_c}{R_{st/cu}}$	$\chi_{st/cu}$: stomatal/cuticular compensation point (ng m^{-3})	Zhang et al., 2009;Bash, 2010;Wang et al., 2014;Wright and Zhang, 2015
		$F_{st/cu}$: air-cuticular/stomatal flux (ng m^{-2} s^{-1})	
		χ_c: compensation point at the air-canopy (ng m^{-3})	
		$R_{st/cu}$: resistance between air-cuticular/stomatal (m s^{-1})	
Soil	S1: $\log F = -\frac{\alpha}{T} + \beta \log(C) + \gamma R + \varepsilon$	T: soil temperature (°)	Xu et al., 1999; Bash et al., 2004; Gbor et al., 2006; Shetty et al., 2008; Selin et al., 2008
		C: soil Hg concentration (ng g^{-1})	
		R: solar radiation (W m^{-2})	
	S2: $\frac{F}{C} = \alpha T + \beta R + \delta\Theta + \delta TR + \cdots$	T: soil temperature (°)	Lin et al., 2010a; Kikuchi et al., 2013
		C: soil Hg concentration (ng g^{-1})	
		R: solar radiation (W m^{-2})	
		Θ: soil moisture (%)	
	S3: $F = \frac{\chi_s - \chi_c}{R_g + R_{ac}}$	χ_s: soil compensation point (ng m^{-3})	Zhang et al., 2009; Bash, 2010; Wang et al., 2014; Wright and Zhang, 2015
		χ_c: compensation point at the air-soil (ng m^{-3})	
		R_g: resistance between air-soil (m s^{-1})	
		R_{ac}: In-canopy aerodynamic resistance (m s^{-1})	
Water	$F = \frac{\chi_w - \chi_c}{R_w + R_a}$	χ_w: water compensation point (ng m^{-3})	Xu et al., 1999; Bash et al., 2004; Gbor et al., 2006; Shetty et al., 2008; Bash, 2010; Wang et al., 2014
		χ_c: air Hg^0 concentration (ng m^{-3})	
		R_w: liquid side resistance (m s^{-1})	
		R_a: air side resistance (m s^{-1})	

Fig. 1. The most recent Hg reservoirs and global atmosphere Hg inventory illustrating the exchange flux between atmosphere and earth surfaces. Adapted from Selin, 2009; Gustin and Jaffe, 2010; Soerensen et al., 2010; Corbitt et al., 2011; Mason et al., 2012; AMAP/UNEP, 2013.

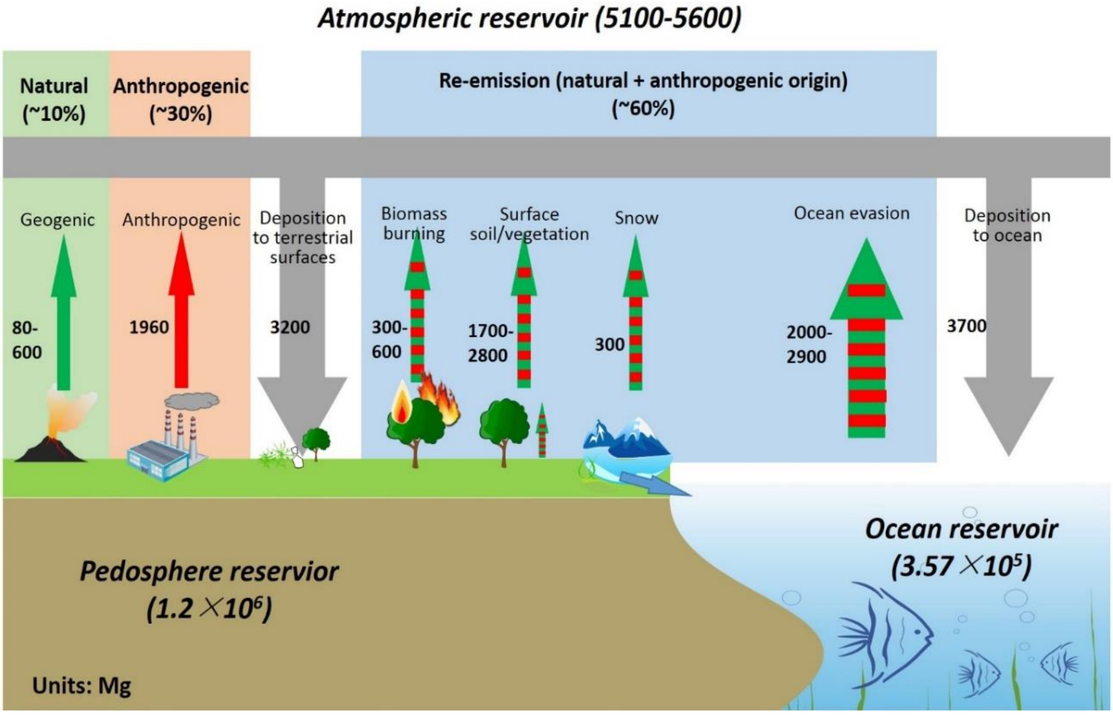


Fig. 2. Field collocated DFCs and MM techniques observed fluxes from two intercomparison studies: (a). Inlaid scatter plot of averaged 4-MM flux vs. averaged 7-DFC flux (TOT: 1.1-24 min) in Hg-enriched Nevada STORMS site in September, 1997 (Gustin et al., 1999); (b). Diel evolution of Hg^0 flux measured using a 1L polycarbonate-DFC (TOT: 0.2 min) and MBR method at same Nevada STORMS site in October, 1998 (Gustin, 2011); (b). Scatter plot of DFC with traditional/novel designs (TDFC/NDFC) vs. MBR Hg^0 flux obtained in Yucheng Intercomparision project (Zhu et al., 2015b).

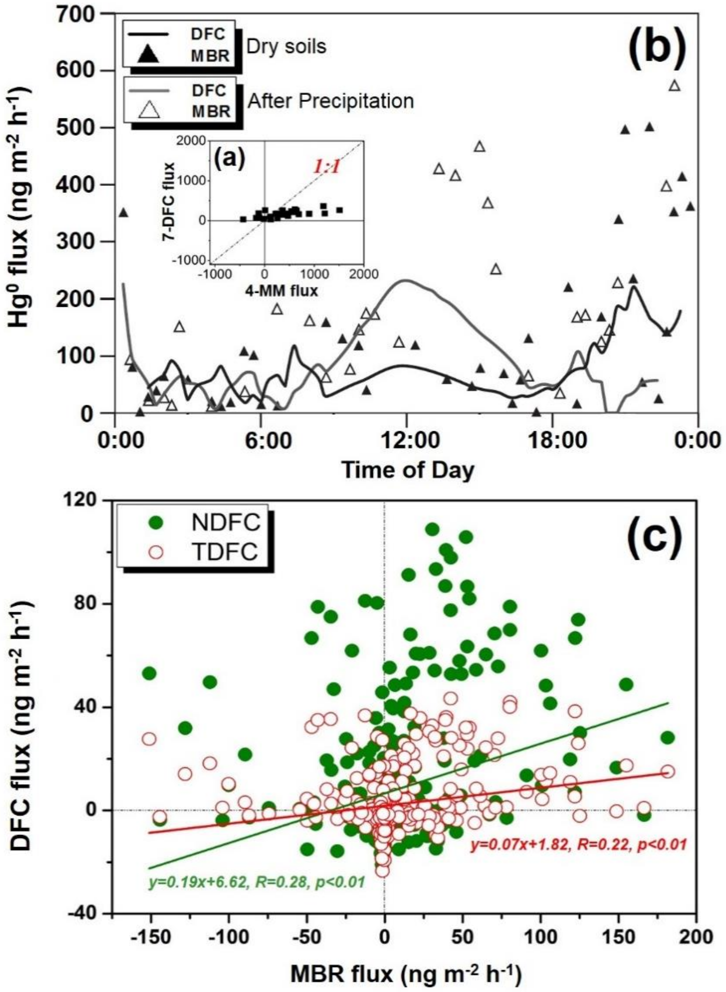


Fig 3. Box and whisker plots of Hg^0 fluxes measured by MM methods and DFC methods. (Flux data including measurements from background soils, agricultural fields, grasslands, and wetlands at substrate total Hg lower than 0.3 µg Hg g^{-1}, data source: Table 1. The two box horizontal border lines indicate 25th and 75th percentiles, whiskers represent 10th and 90th percentiles, and outliers (green circles) indicate 5th and 95th percentiles from bottom to top. Red line and black line indicate mean and median flux).

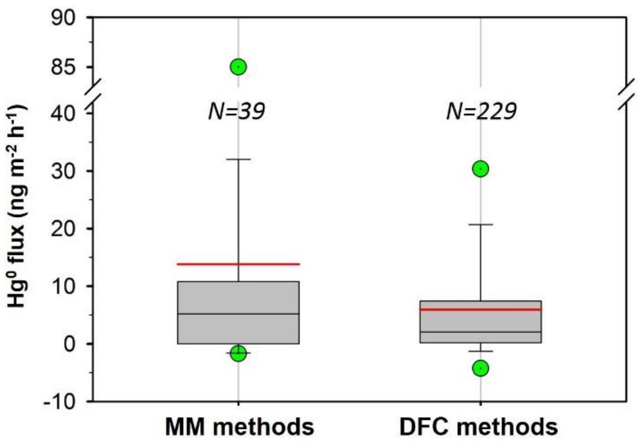


Fig. 4. 4-D graphical visualization of the synergetic effect of air temperature, soil water content, and solar radiation on the measured Hg^0 flux from soil (Lin et al., 2010a).

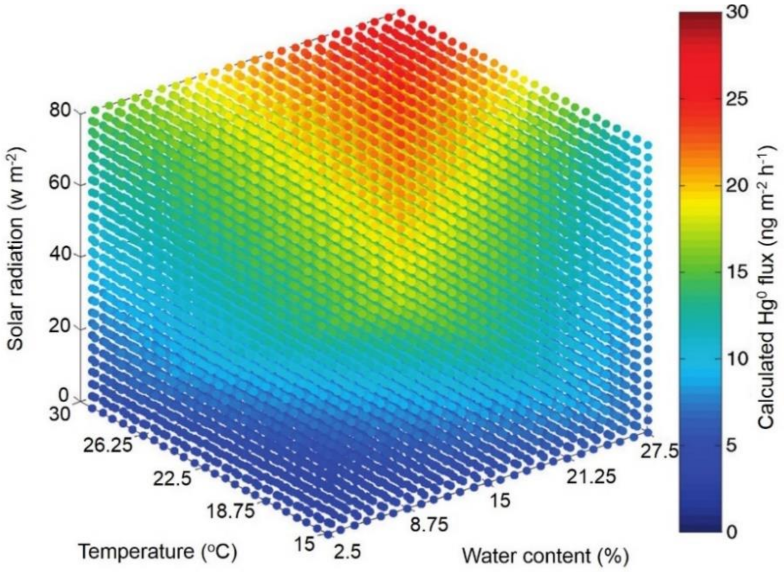


Fig. 5. Conceptual view of DGM cycling in water and mass transfer process across the atmosphere-water interface.

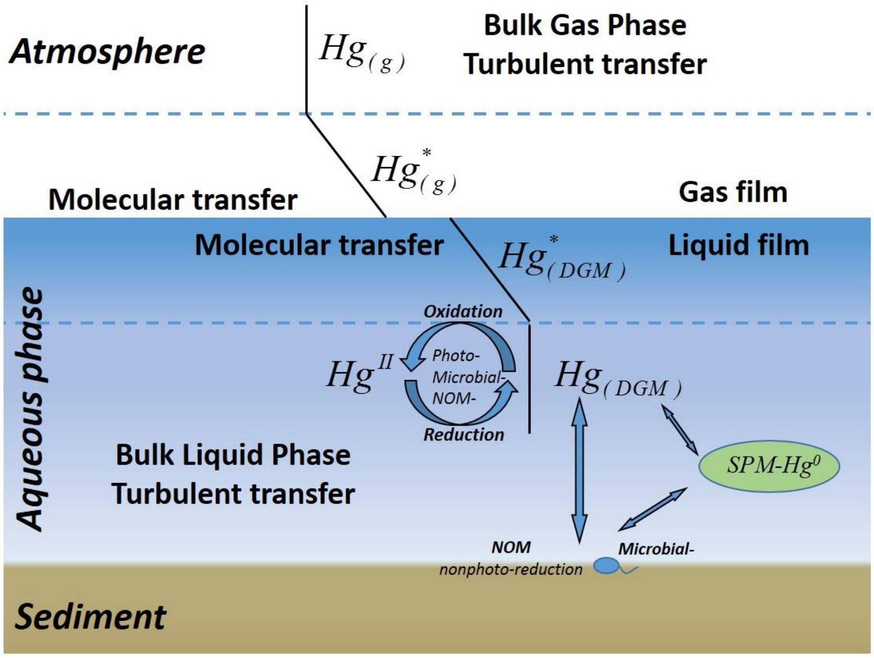


Fig. 6. Box and whisker plots of global field observed Hg^0 fluxes obtained from various landscapes. (Data Source: Table 1. Red line and black line indicate mean and median flux).

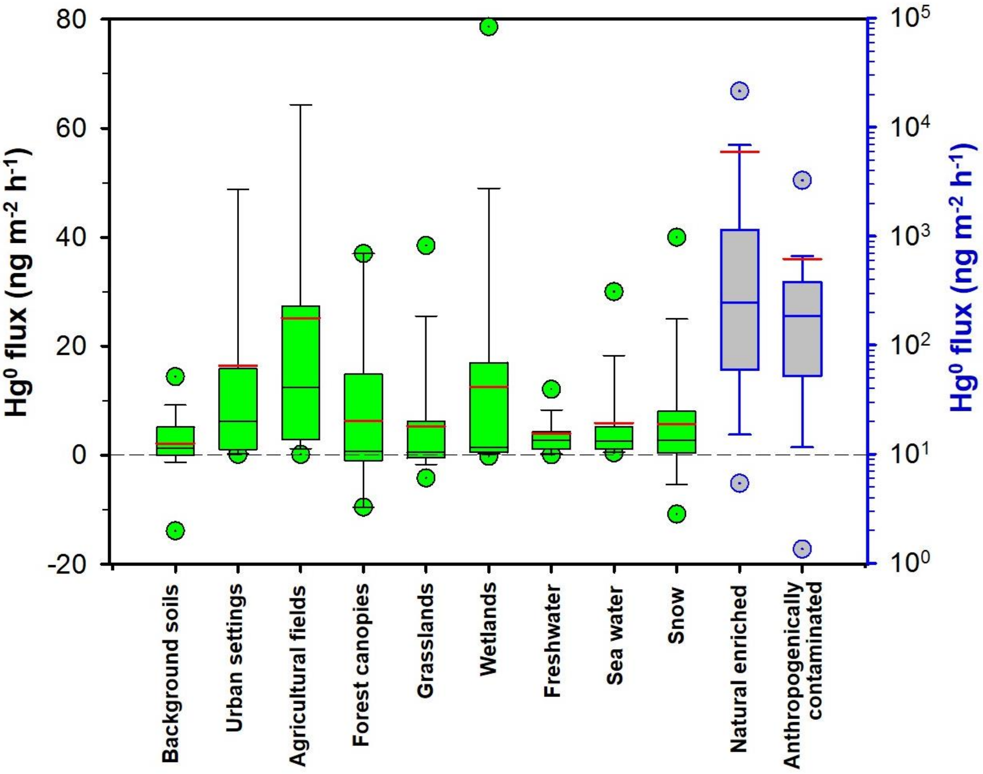


Fig. 7. Histograms of Hg^0 flux frequency distribution obtained from various earth surfaces. (Data source: Table 1).

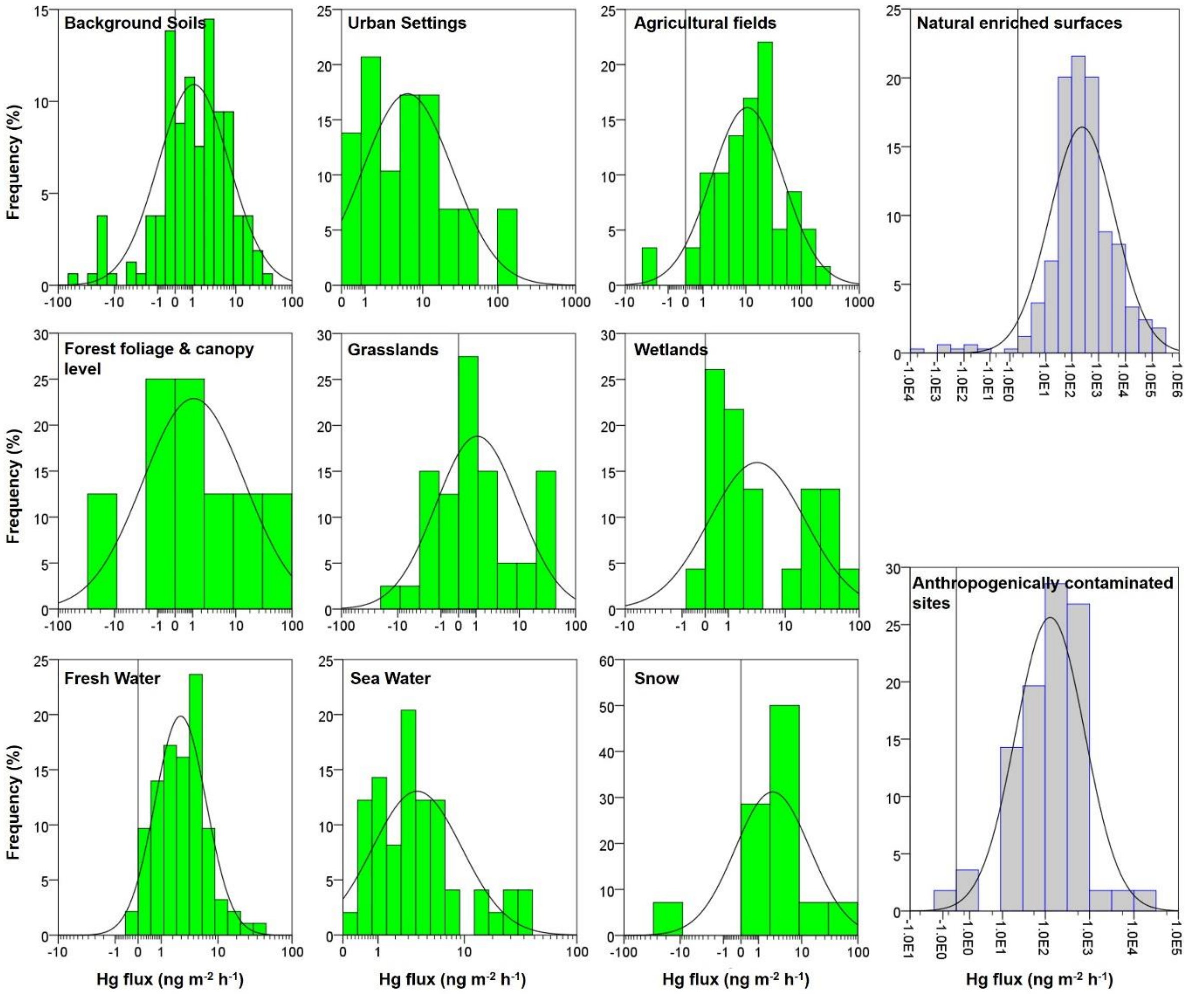


Fig. 8. Box and whisker plots of continents segregated Hg^0 flux obtained from four homogeneous surfaces (Background soils, agricultural fields, grasslands, and freshwater. Filled square block and horizontal line in box indicate mean and median flux).

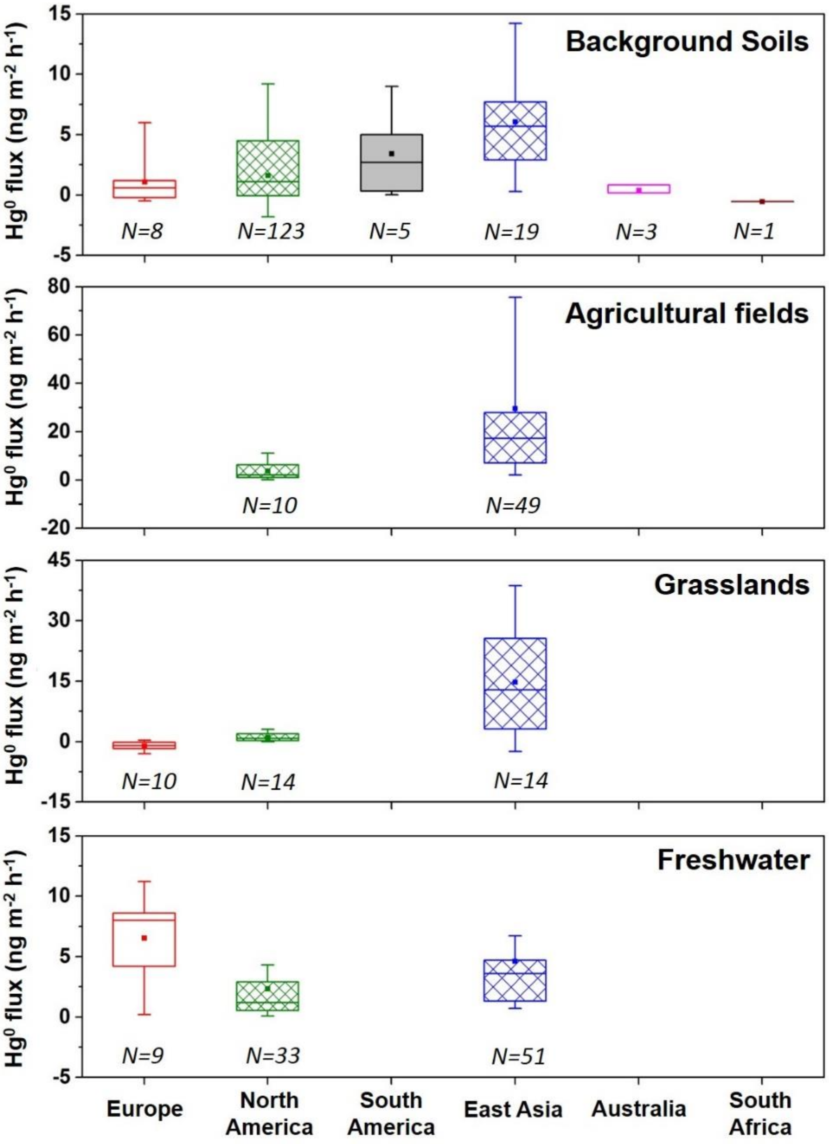
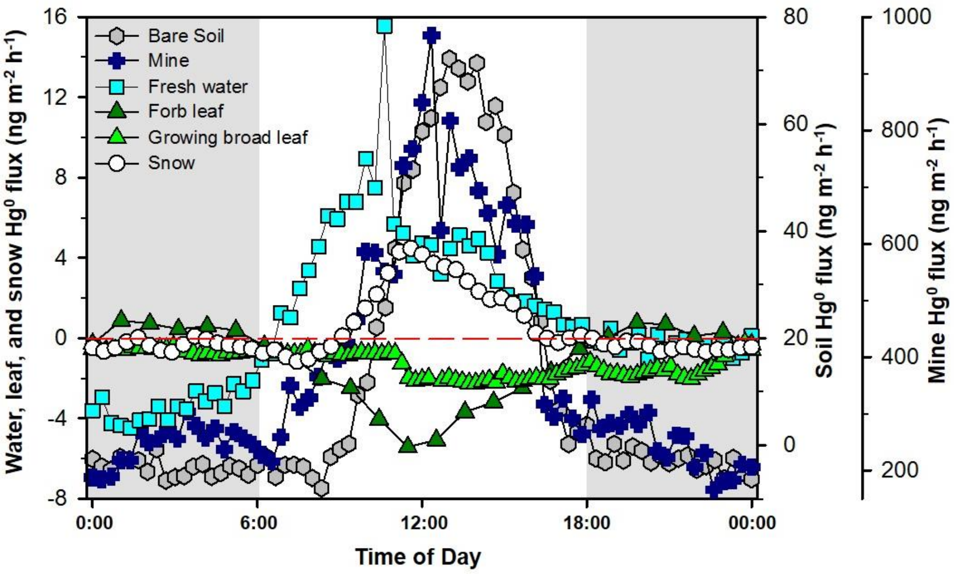


Fig. 9. Diurnal patterns of Hg^0 flux from various environmental compartments (soil, mine, freshwater, forb leaf, growing broad leaf, and snow) measured using DFC methods. (Data obtained from soil: Zhu et al., 2015b; mine: Eckley et al., 2011a; fresh water: O'Driscoll et al., 2003; forb leaf: Stamenkovic et al., 2008; growing broad leaf: Fu et al., 2016; and snow: Maxwell et al., 2013)



Appendix A: References for Table 1

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