



Constraining speciated mercury dry deposition models with urban observations in eastern China

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Abstract. Atmospheric dry deposition constitutes a major pathway of terrestrial mercury (Hg) loading globally, yet estimates of its fluxes remain highly uncertain, particularly in regions with elevated particulate matter (PM) concentrations. This study employed long-term surrogate surface measurements to characterize speciated atmospheric Hg deposition at an urban site in eastern China. Over the period of July 2020 to June 2021, measured total atmospheric mercury (TAM) dry and wet deposition fluxes amounted to 29.19 and 7.29 $\mu\text{g m}^{-2} \text{yr}^{-1}$, respectively, corresponding to a dry-to-wet ratio of 4.0. The observed gaseous oxidized mercury (GOM) dry deposition flux over the same period accounted for 17% of the total dry deposition. These refined flux measurements provided observational constraints for speciated Hg dry deposition modeling. Applying a site-specific correction factor of 2.21 to denuder-based GOM concentrations avoided a 46% underestimation of the mean GOM dry deposition flux. The explicit inclusion of coarse particulate-bound mercury (PBM) increased the deposition velocity by a factor of 4.5, and revealed that coarse particles accounted for 80% of total PBM dry deposition. Replacing the reference leaf area index (LAI) with site-specific values reflecting local vegetation phenology better captured bidirectional gaseous elemental mercury (GEM) exchange, including net emission during certain summer periods. The refined simulations further showed that PBM and GEM co-dominated Hg dry deposition at this site, unlike many less PM-polluted regions where GEM plays a more dominant role. This observationally constrained framework improves speciated atmospheric Hg dry deposition modelling and reduces uncertainty in global Hg budget estimates.



1 Introduction

Since the Minamata Convention entered into force in 2017, the focus of global mercury (Hg) research has shifted from characterizing emissions and atmospheric levels to evaluating the measurable environmental benefits of emission control measures (Evers et al., 2016; UN Environment, 2019). Atmospheric deposition flux observation and simulation provide a direct linkage between atmospheric Hg loading onto terrestrial and aquatic ecosystems, where deposited inorganic Hg may be retained, transformed into methylmercury, and subsequently bioaccumulated through food webs (Driscoll et al., 2013; Lyman et al., 2020). Therefore, evaluation and improvement of Hg deposition flux measurement and modeling methods are essential for closing regional Hg budgets and assessing the ecological responses to Hg emission reductions (Obrist et al., 2018; Selin et al., 2018). Compared with atmospheric Hg concentration and wet deposition measurements, however, Hg dry deposition flux remains less constrained (Zhang et al., 2019).

Atmospheric Hg is commonly operationally classified into gaseous elemental mercury (GEM), gaseous oxidized mercury (GOM), and particulate-bound mercury (PBM) (Schroeder and Munthe, 1998; Driscoll et al., 2013; Gustin et al., 2016; Wright et al., 2016). GEM generally accounts for more than 90% of total atmospheric Hg with a relatively long atmospheric lifetime, from several months to more than one year, owing to its low solubility and low chemical reactivity (Schroeder and Munthe, 1998; Lindberg et al., 2007; Wright et al., 2016). By contrast, GOM and PBM are more readily removed from the atmosphere through wet and dry deposition because of their higher solubility and reactivity, with atmospheric lifetime of hours to days for GOM and days to weeks for PBM (Schroeder and Munthe, 1998; Lindberg et al., 2007; Driscoll et al., 2013; Lyman et al., 2020). These differences in speciation strongly regulate the magnitude and spatiotemporal pattern of Hg deposition, as well as the relative importance of wet and dry deposition pathways (Zhang et al., 2019).

Hg wet deposition can be quantified directly by measuring Hg concentrations in rainfall and precipitation depth, and long-term monitoring networks have provided valuable constraints on regional wet Hg loading (Prestbo and Gay, 2009; Sprovieri et al., 2017). Dry deposition flux, on the other hand, is more difficult to measure because of its dependence on near-surface turbulence, land-cover characteristics, Hg speciation, and surface exchange processes (Zhang et al., 2009; Wright and Zhang, 2015). Micro-meteorological methods, dynamic flux chambers, and surrogate surfaces have been adopted for measuring Hg dry deposition fluxes, but their wider application is limited by high instrumental costs, dependence on chamber designs, and surface material representativeness, respectively (Wright et al., 2016; Zhang et al., 2019; Lyman et al., 2020). The surrogate surface approaches are more poised for wide application in evaluating the effectiveness of the Minamata Convention, given their low costs and ease of standardization (Gustin et al., 2024). The Aerohead samplers with cation exchange membranes (CEMs) are the most promising method for determining GOM dry deposition fluxes (Lyman et al., 2009; Huang et al., 2012; Huang and Gustin, 2015a; Gustin et al., 2024). The artificial turf surrogate surface (ATSS) is designed to measure bulk dry deposition flux of the total atmospheric mercury (TAM = GEM + GOM + PBM) (Hall et al., 2017). However, a key limitation of the ATSS method is that it does not include the process of stomatal GEM uptake by plant leaves, since the artificial turf has no stomata (Sommar et al., 2013; Huang et al., 2014; Gustin et al., 2024).



60 While observations are essential, they only provide snapshots at discrete locations. Understanding long-term, region-wide patterns requires improved process-based modeling of speciated atmospheric Hg dry deposition (Zhang et al., 2023). The basic principle of dry deposition flux models is to multiply atmospheric Hg concentrations by their corresponding deposition velocities (Seigneur et al., 2004; Zhang et al., 2009; Zhang et al., 2012; Wright and Zhang, 2015; Wright et al., 2016; Zhang et al., 2016). For GOM dry deposition estimation, the primary uncertainty arises from ambient concentration measurements, 65 as the widely used denuder-based monitoring systems may underestimate GOM under the interferences from atmospheric constituents such as ozone and water vapor (Lyman et al., 2010; Huang and Gustin, 2015b; Gustin et al., 2019). For PBM, because coarse particles have higher gravitational settling velocities and contribute substantially to dry deposition, routine measurements that represent only fine PBM may introduce site- and season-dependent biases when a fixed coarse-particle fraction is assumed (Fang et al., 2012; Feddersen et al., 2012; Zhang et al., 2016). For GEM, net deposition is inherently 70 difficult to quantify because deposited GEM can be re-emitted from surfaces, necessitating the application of bidirectional exchange models which are parameterized by surface Hg compensation points, land cover types, and leaf area index (LAI) (Wright and Zhang, 2015; Zhang et al., 2016; Zhu et al., 2016; G. Zhang et al., 2021).

Global efforts have been made to evaluate the effectiveness of the Minamata Convention, such as the Multi-Compartment Hg Modeling and Analysis Project (MCHgMAP) (Dastoor et al., 2025). However, a lack of atmospheric Hg dry deposition 75 observations limits the mechanistic optimization and observational constraint of the dry deposition schemes in Hg chemical transport models. Observation and refined simulation remain critically scarce in East Asia where elevated anthropogenic emissions, complex aerosol pollution, and strong seasonal meteorological variations may substantially affect Hg speciation and deposition processes (Fu et al., 2016; Zhang et al., 2019; Tang et al., 2020; Lyman et al., 2020). As part of the Speciation and Transformation of Atmospheric Mercury in a Polluted region (STAMP) campaign (Zhang et al., 2024; Wang et al., 2026), 80 long-term observations of GOM and TAM dry deposition fluxes, as well as Hg wet deposition flux and speciated ambient Hg concentrations, were measured in this study at an urban site in eastern China. The objectives of this study were to: (1) characterize the wet and dry deposition fluxes of atmospheric Hg; (2) evaluate the performance of existing Hg dry deposition models against field observations; and (3) improve model estimates with better parameterization schemes. This study fills a critical gap in atmospheric Hg deposition research in East Asia and offers an updated deposition flux modeling framework to 85 support the effectiveness evaluation of the Minamata Convention.

2 Materials and methods

2.1 Site and campaign description

Field sampling for this study was conducted on the rooftop of the School of Environment building at Nanjing University (NJU, 32.117 °N, 118.954 °E, 28 m above sea level), located in northeastern Nanjing and operationally defined as an urban site. As 90 one of the most economically developed cities in eastern China, Nanjing is characterized by a dense population, developed industries and high emissions of air pollutants. The city exhibits a subtropical monsoon climate with pronounced seasonal



variations, as well as relatively high rainfall levels throughout the year. As part of the STAMP campaign conducted from summer 2018 to summer 2021 with a gap during the COVID-19 pandemic, GOM and TAM dry deposition fluxes were measured by the surrogate surface methods. Daily precipitation samples were collected and analyzed for Hg concentration. Moreover, we employed offline measurements of reactive mercury (RM = GOM + PBM) and size-fractionated PBM, and online monitoring of PM_{2.5}, PM₁₀ and meteorological parameters.

2.2 Sampling and analytical methods

2.2.1 Sampling methods for Hg deposition fluxes

In this study, GOM and TAM dry deposition fluxes were measured by surrogate surfaces with CEMs (Pall Corporation, P/N MSTGS3R) and artificial turfs (Astro Turf®, Grass Worx, LLC), respectively. Three aerodynamic polyoxymethylene disks (triplicate Aerohead samplers) were deployed with CEMs. The configuration of the samplers was designed to avoid air turbulence. The membranes were exposed to the ambient atmosphere through the circular opening (115 mm in diameter) at the base of the sampler, thus preventing contamination by rainwater or particle deposition (Lyman et al., 2009). The majority of the sampling process was completed within a time frame of approximately one week, with a range of 6–14 days. A total of 98 cycles of samples were collected in the study. Triplicate turf sampling devices were deployed (Hall et al., 2017), with each consisting of two layers. The upper layer was the artificial turf and the lower layer was the runoff sampling bottles, which were connected by a connector in the middle. During precipitation events, the runoff from the artificial turf is collected in the lower bottle, and subsequent analysis of the turf and the penetrating water is conducted for Hg content to calculate the total deposition flux of TAM. A total of 73 samples were collected during the observation period of the study. Daily precipitation samples for Hg wet-deposition measurements were collected using an APS-3B automatic precipitation sampler (Changsha Xianglan Scientific Instrument Co., China). A sampling day was defined from 09:00 to 09:00 the following day. Three replicate samples were prepared for each collection. A total of 297 precipitation samples were collected from November 2018 to July 2021, except from January to June 2020, when precipitation sampling was suspended because of COVID-19 control measures.

2.2.2 Analytical methods for membrane and liquid samples

Membrane, turf, and liquid samples were analyzed in accordance with the U.S. EPA method 1631 (Revision E). Hg in solid samples was dissolved using bromine monochloride (BrCl) (6% (v/v) for membrane samples, and 4% (v/v) for turf), and liquid samples were prepared by adding 0.5% (v/v) BrCl. The solutions were transferred to glass vials with a volume of 40 mL. Hydroxylamine hydrochloride (NH₂OH·HCl) and stannous chloride (SnCl₂) were then added in sequence, followed by a brief period of shaking. Hg in the solutions was measured using the Tekran 2600 system. The r^2 values of the calibration curve exceeded 0.999 for each analysis.

For GOM, the dry-deposition flux was calculated from the Hg mass collected on the CEM:



$$F_{GOM} = \frac{M}{AT} \quad (1)$$

where F_{GOM} is the GOM dry deposition flux $\text{ng m}^{-2} \text{h}^{-1}$; M is the mass of Hg collected on the CEM (ng); A is the membrane area directly exposed to the atmosphere (approximately 0.0104 m^2); and T is the sampling duration (h).

125 For TAM, the dry deposition flux was calculated as:

$$F_{TAM(dry)} = \frac{M_{ATSS} + M_{through} - M_{rain}}{AT} \quad (2)$$

where $F_{TAM(dry)}$ is the TAM dry-deposition flux ($\text{ng m}^{-2} \text{h}^{-1}$); M_{ATSS} is the mass of Hg retained on the artificial turf (ng); $M_{through}$ is the mass of Hg in the runoff collected beneath the turf (ng); M_{rain} is the mass of Hg contained in an equivalent volume of ambient precipitation during the corresponding sampling period (ng); A is the projected surface area of the artificial turf
130 (m^2); and T is the sampling duration (h).

The annual or seasonal volume-weighted mean (VWM) Hg concentration was calculated by:

$$VWM = \frac{\sum_{i=1}^n C_i \times PD_i}{\sum_{i=1}^n PD_i} \quad (3)$$

where C_i and PD_i are the Hg concentration (ng L^{-1}) and precipitation depth (mm) of precipitation event i , respectively.

The annual or quarterly wet deposition flux was calculated by:

$$F_{wet} = \sum_{i=1}^n C_i \times PD_i \quad (4)$$

where F_{wet} is the total Hg wet deposition flux over a period of time.

2.2.3 Online monitoring methods

Speciated atmospheric Hg was measured by the Tekran 2537X/1130/1135 system at a 2-hr operating cycle (L. Zhang et al., 2021). During the first half of each cycle, GEM was sampled and analyzed by cold vapor atomic fluorescence spectroscopy
140 (CVAFS) at a temporal resolution of 5 min, while GOM and PBM were collected with KCl-coated denuder and quartz filter, respectively. In the second part of the cycle, PBM and GOM were thermally decomposed into GEM sequentially for analysis by the 2537X analyzer. Quality control procedures were performed according to the guidelines set forth by the Atmospheric Mercury Network (AMNet, <http://nadp.slh.wisc.edu/AMNet/docs.aspx>). Quartz filters, KCl-coated denuders, and soda lime cartridges were replaced bi-weekly. Routine calibrations were carried out every 25 hours for Tekran 2537X with its internal
145 permeation source and semi-annually with a manual calibration using the Tekran 2505 external permeation source.

$PM_{2.5}$ and PM_{10} concentrations were measured using the 1405-D TEOMTM ambient particle continuous monitor (Thermo Scientific Co.) at a temporal resolution of 5 min. Meteorological parameters were measured by an automatic weather station (Campbell Scientific Co., Ltd), including wind speed (WS, m s^{-1}), wind direction (WD, °), solar radiation (SR, W m^{-2}), air



temperature (T, °C), relative humidity (RH, %), air pressure (P, kPa) and precipitation depth (Precip, mm), with a temporal
150 resolution of 10 min.

2.2.4 Membrane-based offline methods for RM

To obtain substantially more accurate and refined RM concentrations, PBM concentrations in different particle-size ranges
were determined using a micro-orifice uniform deposit impactor (MOUDI, Model 110NR, TSI ®) with PTFE membranes,
capturing 11 distinct particle size ranges (>18, 10–18, 5.6–10, 3.2–5.6, 1.8–3.2, 1.0–1.8, 0.56–1.0, 0.32–0.56, 0.18–0.32, 0.1–
155 0.18, and 0.056–0.1 µm). All the membrane samples were analyzed using Tekran 2600 as described in Section 2.2.2.

2.2.5 Quality assurance and quality control (QA/QC)

The Tekran data were screened using zero-air blanks, gold-cartridge agreement, calibration response, cycle completeness, and
post-maintenance stability. The two gold cartridges were required to agree within 10%. Glassware was cleaned according to
the procedure described by Lyman et al. (2009). All liquid samples were analyzed in duplicate, and duplicate analyses were
160 accepted when the relative percent difference (RPD) was ≤15%. Duplicate pairs exceeding this criterion were reanalyzed.
Instrument cleaning and matrix-spike recovery checks were performed after every 12 samples. An analytical batch was
accepted when the blank concentration was ≤1 ng L⁻¹ and matrix-spike recovery was within 85–115%. Blank correction was
performed separately for each deposition sample. The validated dataset included 309 precipitation samples, 165 artificial-turf
samples, and 256 CEM samples. The corresponding blank concentrations were subtracted from digestion concentrations before
165 calculating Hg mass and deposition flux.

Method detection limits (MDLs) were determined for each sample matrix and expressed as deposition-flux equivalents. The
concentration-based MDLs were converted using the corresponding sample volume, effective collection area, and sampling
duration. The resulting MDLs were 0.01 ng m⁻² h⁻¹ for Hg wet deposition samples, 0.25 ng m⁻² h⁻¹ for TAM dry deposition
measured using artificial turfs, and 0.10 ng m⁻² h⁻¹ for GOM dry deposition measured using CEMs.

170 The precision of field-replicate measurements was evaluated only for sampling periods with three valid blank-corrected
replicate fluxes. The median CV was 16.2% for the artificial-turf measurements, with a range of 4.1–54.4%, and 13.9% for
the CEM measurements, with a range of 0.4–51.3%. CV values above 30% were flagged for further review of possible blank
contamination, precipitation-correction errors, sample loss, or other analytical anomalies.

The analytical reliability of the CEM measurements was further supported by an interlaboratory comparison, which yielded
175 an average reproducibility RSD of 9 ± 4% (Wang et al., 2026). Interlaboratory comparison supported the analytical reliability
of the CEM measurements, with an average reproducibility RSD of 9 ± 4% (Wang et al., 2026). The uncertainty of the
deposition fluxes includes analytical precision, blank correction, sample area, sampling duration, precipitation volume, and
surrogate-surface representativeness. Although the QA/QC results support the analytical reliability of the measurements,
absolute fluxes remain subject to sampling uncertainty, particularly because artificial turf does not reproduce the stomatal
180 uptake and re-emission processes of natural vegetation.



2.3 Methods for modeling speciated Hg dry deposition

Our previous review work (Zhang et al., 2023) has systematically evaluated the existing framework of dry deposition schemes for speciated atmospheric Hg, and performed sensitivity analysis to identify the key parameters for estimating dry deposition velocities. Therefore, in this section, only crucial principles of the methods were introduced in detail. More information is available in Zhang et al. (2023).

2.3.1 Multiple resistance model for GOM dry deposition

The multiple resistance model is the most commonly used approach for parameterizing the deposition velocity of gaseous substances. The GOM dry deposition flux is estimated as the product of GOM concentration and dry deposition velocity (Zhang et al., 2003):

$$F_{GOM} = C_Z \times V_d \quad (5)$$

where C_Z represents the GOM concentration at the reference height and V_d represents the dry deposition velocity. Based on multiple resistance theory, V_d is defined as:

$$V_d = (R_a + R_b + R_c)^{-1} \quad (6)$$

where R_a , R_b , and R_c represent the aerodynamic resistance, the quasi-laminar sublayer resistance above the canopy and the canopy resistance, respectively. R_a , R_b , and R_c depend on meteorology and land use category (LUC) and their expressions were discussed in detail in previous dry deposition studies (Wesely et al., 2002; Zhang et al., 2003; Zhang et al., 2012). The uncertainties of R_a and R_b are generally small, while that of R_c is usually substantial (Huang et al., 2012; Zhang et al., 2019).

The canopy resistance (R_c) is calculated as:

$$R_c = \left(\frac{1 - W_{st}}{R_{st} + R_m} + \frac{1}{R_{ac} + R_g} + \frac{1}{R_{cut}} \right)^{-1} \quad (7)$$

where W_{st} is a surface-wetness factor describing the suppression of stomatal uptake under wet-canopy conditions; R_{st} is the stomatal resistance; R_m is the mesophyll resistance; R_{ac} is the in-canopy aerodynamic resistance; R_g is the ground resistance; and R_{cut} is the cuticular resistance.

Among these terms, R_g and R_{cut} are parameterized according to the solubility and chemical reactivity of the depositing gas. Their values for a target species (i) are scaled from the corresponding resistances of SO_2 and O_3 as:

$$\frac{1}{R_{g,cut}(i)} = \frac{\alpha(i)}{R_{g,cut}(SO_2)} + \frac{\beta(i)}{R_{g,cut}(O_3)} \quad (8)$$

where α and β are empirical numbers determined by the solubility and half-redox reactivity of chemical species (Zhang et al., 2002; Zhang et al., 2003). In this study, $\alpha = \beta = 2$ for GOM (Huang et al., 2017), and $\alpha = 0$ and $\beta = 0.1$ for GEM (Zhang et al., 2012) were applied.



As mentioned previously, GOM concentrations measured by the Tekran system could be biased low because of the KCl-coated denuders (Lyman et al., 2010; Huang and Gustin, 2015b; Gustin et al., 2019). A correction factor was used based on the comparison between Tekran and the reactive mercury active system (RMAS) in Zhang et al. (2024) during the STAMP campaign. The GOM concentration measured by RMAS was significantly correlated with that measured by Tekran, with a regression slope of 2.21 when the intercept was fixed to zero ($r^2 = 0.60$, $p < 0.001$, $n = 22$) (Zhang et al., 2024). Therefore, the Tekran GOM concentration was corrected as follows before being used to calculate GOM dry deposition fluxes:

$$C_{GOM, corrected} = 2.21 \times C_{GOM, Tekran} \quad (9)$$

where C_{GOM} stands for the GOM concentration.

2.3.2 Dynamic particle-size-regarded model for PBM dry deposition

The PBM dry deposition flux can be calculated according to (Zhang et al., 2016):

$$F_{PBM} = C_1 V_{d1} + C_2 V_{d2} = C_1 \left(V_{d1} + \frac{f}{1-f} V_{d2} \right) \quad (10)$$

where C_1 and C_2 are the mass concentrations of PBM in fine and coarse particles, respectively, and V_{d1} and V_{d2} are the corresponding dry deposition velocities. f is the mass fraction of PBM in coarse particles. The Tekran system measures PBM in fine particles ($PBM_{2.5}$), and f was assumed to be 0.3 based on available measurements of PBM size distribution in a previous study (Zhang et al., 2016). In this study, $PM_{2.5}$ and PM_{10} monitoring data were used to estimate dynamic f , which can be calculated by the distribution coefficient, named k , of the PBM between fine and coarse particles:

$$k = \frac{PBM_{2.5}}{PM_{2.5}} \bigg/ \frac{PBM_{2.5-10}}{PM_{2.5-10}} \quad (11)$$

Then f can be obtained by the following formula:

$$f = \frac{PBM_{2.5-10}}{PBM_{2.5} + PBM_{2.5-10}} = \frac{PM_{2.5-10}}{k \cdot PM_{2.5} + PM_{2.5-10}} \quad (12)$$

The distribution coefficient k was found to be positively linearly correlated to the ratio of $PBM_{2.5}$ over $PM_{2.5}$ measured by MOUDI ($r^2=0.66$, $p<0.05$; Figure 1a). $PBM_{2.5}$ and $PM_{2.5}$ measured by MOUDI were linearly correlated to $PBM_{2.5}$ measured by the Tekran system ($r^2=0.84$, $p<0.05$, with the intercept fixed to zero; Figure 1b) and $PM_{2.5}$ measured by Thermo 1405-D ($r^2=0.97$, $p<0.05$, with the intercept fixed to zero; Figure 1c), respectively. Therefore, the coefficient k could be obtained as follows:

$$k = 0.943 \times \frac{0.8989 \times PBM_{2.5}(Tekran)}{1.2348 \times PM_{2.5}(Thermo)} + 1.7289 \quad (13)$$

Consequently, dynamic f could be obtained according to the high temporal resolution data of PBM measured by Tekran and $PM_{2.5}$ and PM_{10} measured by Thermo, which are usually easy to obtain in field studies in most areas.

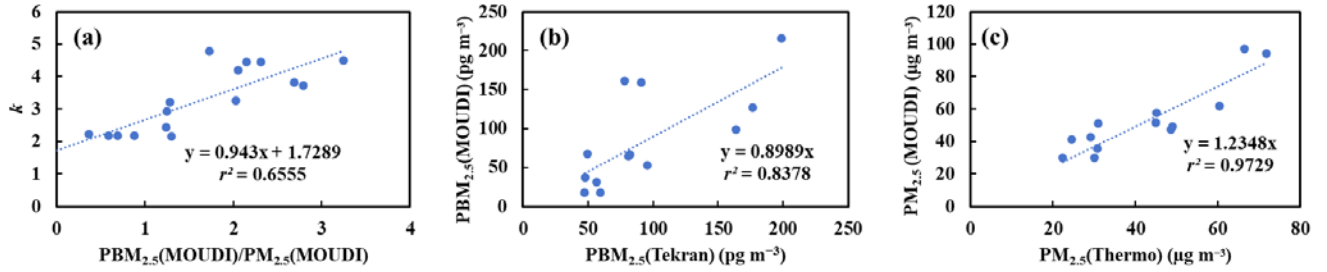


Figure 1. Relationships (a) between k and the ratio of $\text{PBM}_{2.5}$ over $\text{PM}_{2.5}$ measured by MOUDI, (b) between $\text{PBM}_{2.5}$ measured by MOUDI and by Tekran, and (c) between $\text{PM}_{2.5}$ measured by MOUDI and by Thermo.

The PBM dry deposition velocity is calculated by:

$$V_d = V_g + \frac{1}{R_a + R_s} \quad (14)$$

where V_g is the gravitational settling velocity, and is mainly determined by particle size. Assuming the particle size fits the lognormal distribution, values of 3.7×10^{-5} and 1.8×10^{-3} were obtained for fine and coarse particles, respectively. R_a is the aerodynamic resistance above the canopy and is the same as in GOM deposition modeling. R_s is the surface resistance. Detailed calculation formulas of R_s can be found in Zhang et al. (2014).

2.3.3 Bidirectional air-surface exchange model for GEM dry deposition

In contrast to GOM, GEM has the capacity to be reemitted into the atmosphere following deposition onto vegetated surfaces and soils. The bidirectional air-surface exchange scheme (Wright and Zhang, 2015) is employed in this study to estimate the GEM exchange flux:

$$F_{GEM} = \frac{C_{GEM} - \chi_c}{R_a + R_b} \quad (15)$$

$$\chi_c = \left(\frac{\chi_c}{R_a + R_b} + \frac{\chi_{st}}{R_{st} + R_m} + \frac{\chi_g}{R_{ac} + R_g} \right) \left(\frac{1}{R_a + R_b} + \frac{1}{R_{st} + R_m} + \frac{1}{R_{ac} + R_g} + \frac{1}{R_{cut}} \right)^{-1} \quad (16)$$

where χ_c represents the GEM concentration at the canopy top (ng m^{-3}). R_a and R_b have the same meanings as in GOM modeling. R_{st} is the stomatal resistance (s m^{-1}) and was calculated using a sunlit/shade stomatal resistance sub-model (Zhang et al., 2002). R_m is the mesophyll resistance and the values are 500 s m^{-1} for GEM and 0 for GOM in this study (Zhang et al., 2012). R_{ac} , R_g and R_{cut} are the in-canopy aerodynamic, soil and cuticle resistance, respectively, which are the same as those used for GOM modeling and are discussed in detail in Zhang et al. (2003). χ_a is the ambient GEM concentration ($\mu\text{g m}^{-3}$). χ_{st} and χ_g are the stomatal and ground surface compensation points ($\mu\text{g m}^{-3}$), respectively, and their calculation formulas can be found in Zhang et al. (2016). When C_{GEM} is higher than χ_c , the flux is positive, representing a downward flux. Otherwise, the



flux is upward, representing a net emission. The value of χ_c at which the flux changes direction is defined as the canopy compensation point, χ_{cp} .

260 3 Results and discussion

3.1 Characterization of Hg deposition fluxes based on observations

3.1.1 GOM dry deposition flux measurements

As shown in Figure 2, the GOM dry deposition flux observed at the NJU site from July 2020 to June 2021 was $5.01 \mu\text{g m}^{-2} \text{yr}^{-1}$, with an average flux of $0.57 \text{ ng m}^{-2} \text{h}^{-1}$. This value accounted for approximately 17 % of the observed TAM dry deposition flux in this study ($29.19 \mu\text{g m}^{-2} \text{yr}^{-1}$). Although GOM represented only a minor fraction of atmospheric Hg, with an annual mean concentration of 15.63 pg m^{-3} , corresponding to about 0.5 % of the TAM concentration, its contribution to dry deposition was substantial because of its high reactivity and efficient deposition. Long-term observations from 2018 to 2021 showed that weekly mean GOM dry deposition fluxes in Nanjing ranged from 0.11 to $2.84 \text{ ng m}^{-2} \text{h}^{-1}$. The mean fluxes before and after the COVID-related data gap were very similar, at 0.67 ± 0.46 and $0.66 \pm 0.45 \text{ ng m}^{-2} \text{h}^{-1}$, respectively, indicating no clear long-term shift in GOM dry deposition during the observation period. During the representative annual period, seasonal mean fluxes were 0.76, 0.41, 0.57, and $0.54 \text{ ng m}^{-2} \text{h}^{-1}$ in spring, summer, autumn, and winter, respectively. The spring maximum was consistent with the highest seasonal GOM concentration in spring (Table 1), whereas the summer minimum was likely associated with frequent precipitation and humid conditions, which enhanced wet scavenging and reduced the amount of GOM available for dry deposition. Similar springtime enhancements and summertime reductions have been reported in Florida, Korea, and the western United States (US), where seasonal variations were linked to changes in GOM concentration, wind speed, humidity, and precipitation scavenging (Han et al., 2016; Sather et al., 2013; Huang et al., 2017; Peterson et al., 2012). It should be noted that the GOM concentration was measured by Tekran, the widely used denuder-based monitoring systems which could suffer from underestimation with interferences from ozone and moisture (Lyman et al., 2010; Gustin et al., 2019), and partially caused the inconsistency of seasonal variation between the concentration and the dry deposition flux.

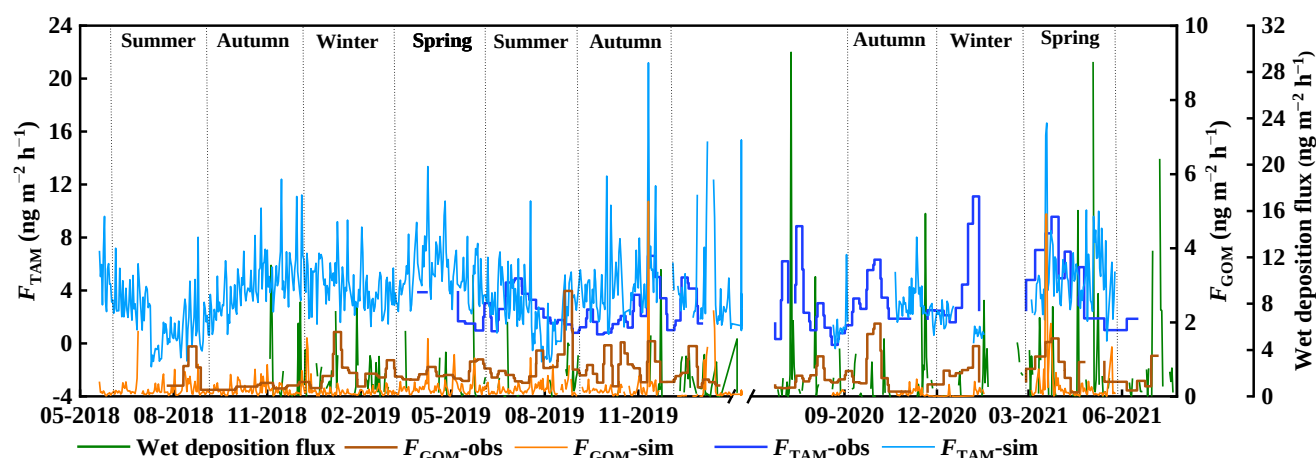


Figure 2. Observed and simulated dry deposition fluxes of TAM and GOM, together with wet deposition fluxes. Model-estimated fluxes are shown as daily averages.

Table 1. Observed results of speciated Hg concentrations by Tekran at NJU

Season	GEM (ng m ⁻³)	GOM (pg m ⁻³)	PBM (pg m ⁻³)
Spring	3.17±1.06	19.97±28.49	92.80±173.37
Summer	2.97±1.00	14.26±21.79	41.34±109.97
Autumn	3.15±1.32	15.43±20.37	96.95±204.36
Winter	3.23±1.36	11.83±17.23	126.01±302.98
Annual	3.12±1.19	15.63±22.87	86.78±207.38

Compared with measurements at other sites globally with the CEM surrogate surfaces (Table 2), the GOM dry deposition flux at NJU was higher than that of a suburban site in Florida ($0.14 \text{ ng m}^{-2} \text{ h}^{-1}$), but lower than four urban sites in the western US ($0.8\text{--}1.3 \text{ ng m}^{-2} \text{ h}^{-1}$). The GOM dry deposition fluxes in most rural, remote, and coastal sites in the western and southern US ranged from 0.05 to $0.5 \text{ ng m}^{-2} \text{ h}^{-1}$, with exceptions for sites in California and Nevada ($0.5\text{--}2.9 \text{ ng m}^{-2} \text{ h}^{-1}$). Year-long observations of GOM dry deposition fluxes remain extremely rare (Zhang et al., 2019). One notable early attempt by Peterson et al. (2012) found the annual mean GOM dry deposition fluxes to be $0.4\text{--}1.5 \mu\text{g m}^{-2} \text{ yr}^{-1}$ at three sites in Florida, much lower than that in this study, which was likely attributable to the relatively low annual mean GOM concentrations ($4.8\text{--}6.6 \text{ pg m}^{-3}$, measured by RMAS). The GOM dry deposition flux in this study was close to the median value of global observation results, but considerably higher than the simulated results, as summarized by Zhang et al. (2019). One example is the study of Zhang et al. (2016) for 24 North American sites during 2009–2014, in which the multi-year mean flux was estimated to be $0.2\text{--}4.0$



295 $\mu\text{g m}^{-2} \text{yr}^{-1}$ based on GOM concentrations measured by the Tekran system. The underestimation of the GOM concentration by the denuder-based method (McClure et al., 2014) has significant impact on the dry deposition simulation.

Table 2. GOM dry deposition fluxes measured by CEM surrogate surfaces

Site	Site type	Period	Flux ($\text{ng m}^{-2} \text{h}^{-1}$)	Reference
Nanjing, China	Urban	2020.7–2021.6	0.57	This study
Florida, USA	Suburban	2007.9–2008.9	0.14	Weiss et al., 2011
CO99, Colorado, USA	Urban	2017.8–2019.8	1.3±1.0	Sather et al., 2021
NM95, New Mexico, USA	Urban	2017.8–2019.8	1.0±0.8	Sather et al., 2021
NM97, New Mexico, USA	Urban	2017.8–2019.8	0.8±0.6	Sather et al., 2021
NM98, New Mexico, USA	Urban	2017.8–2019.8	0.8±0.6	Sather et al., 2021
Four sites in Texas, USA	Rural	2011.9–2012.9	0.1±0.0 – 0.3±0.1	Sather et al., 2014
Georgia, USA	Rural	2007.9–2008.9	0.22	Weiss et al., 2011
OLF, Florida, USA	Coastal	2006.10–2008.10	0.1±0.1	Lyman et al., 2009
Three sites in Florida, USA	Rural	2009.7–2010.7	0.05±0.06 – 0.20±0.18	Peterson et al., 2012
YRK, Georgia, USA	Rural	2006.10–2008.10	0.2±0.2	Lyman et al., 2009
OLF, Florida, USA	Coastal	2012.6–2014.3	0–0.5	Huang et al., 2017
Western Maryland, USA	Remote	2009.9–2010.10	0.35	Castro et al., 2012
CCRS, California, USA	Remote (high-elevation)	2013	1.2±0.9	Huang and Gustin, 2015a
CO96, Colorado, USA	Remote (high-elevation)	2017.8–2019.8	0.3±0.1	Sather et al., 2021
Five sites in Nevada, USA	Remote (high-elevation)	2013	2.2±1.0 – 2.9±1.5	Huang and Gustin, 2015a
NV02, Nevada, USA	Rural	2006.10–2008.10	2.2±0.5	Lyman et al., 2009
NV98, Nevada, USA	Suburban	2006.10–2008.10	1.0±0.8	Lyman et al., 2009
Four sites in California and Nevada, USA	Remote	2013	0.5±0.4 – 1.0±0.8	Huang and Gustin, 2015a
Zeppelin (Arctic)	Remote	2019.3–2019.7	0.35	Osterwalder et al., 2021



3.1.2 TAM dry deposition flux measurements

300 The period from July 2020 to June 2021 was used to characterize the annual TAM dry deposition flux at NJU in this study (29.19 $\mu\text{g m}^{-2} \text{yr}^{-1}$). The mean TAM dry deposition fluxes during this period were 4.23, 2.53, 2.82, and 3.49 $\text{ng m}^{-2} \text{h}^{-1}$ in spring, summer, autumn, and winter, respectively. Long-term artificial-turf observations from 2019 to 2021 further showed that the TAM dry deposition flux varied widely from -0.96 to $14.99 \text{ ng m}^{-2} \text{h}^{-1}$, with an overall mean of $3.25 \text{ ng m}^{-2} \text{h}^{-1}$ (Figure 2). The episodic enhancements in spring and winter were likely related to the high GOM concentration in spring and
305 the high PBM concentration in winter (Table 1), whereas the summer minimum could be due to the stronger GEM re-emission under high temperature or high solar radiation conditions. Compared with results from other artificial or natural turf surrogate-surface measurements (Table 3), the Nanjing flux was higher than most sites except for two industrially influenced Michigan sites, where the fluxes reached 6.42 and $5.38 \text{ ng m}^{-2} \text{h}^{-1}$, respectively (Hall et al., 2017).

Table 3. TAM dry deposition fluxes measured by artificial or natural turf surrogate surfaces

Site	Site type	Period	Flux ($\text{ng m}^{-2} \text{h}^{-1}$)	Reference
Nanjing, China	Urban	2020.7–2021.6	3.33	This study
Botanical Gardens, Ann Arbor, MI, USA	Suburban	2008/8/4–2008/9/13	1.56	Hall et al., 2017
Dearborn, MI, USA	Urban	2007/7/18–2007/8/8	6.42	Hall et al., 2017
Detroit-Fort Street, MI, USA	Urban	2007/7/18–2007/8/8	5.38	Hall et al., 2017
FCP, IL, USA	Urban	2011/6/9–2011/7/3	1.39	Lynam et al., 2014
JCS, IL, USA	Rural	2011/6/9–2011/7/3	1.25	Lynam et al., 2014
ERM, IL, USA	Rural	2011/6/9–2011/7/3	2.64	Lynam et al., 2014
JPC, IL, USA	Rural	2011/6/9–2011/7/3	3.19	Lynam et al., 2014
SCL, IL, USA	Rural	2011/6/9–2011/7/3	2.64	Lynam et al., 2014
Nilwood, IL, USA	Rural	2011/6/9–2011/7/3	2.50	Lynam et al., 2014
BVD, IL, USA	Rural	2011/6/9–2011/7/3	2.08	Lynam et al., 2014
RSL, IL, USA	Rural	2011/6/9–2011/7/3	2.08	Lynam et al., 2014
Central Illinois, USA	Mixed rural/urban	2011/6/9–2011/7/3	2.08 ± 0.69	Lynam et al., 2013
Florida TMDL, USA	Mixed coastal/urban/rural	2009–2010	0.83–2.92	Hall et al., 2013

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The annual TAM dry deposition flux observed in this study are comparable to those simulated by Pirrone and Keating (2010) using the GEOS-Chem model for East Asia ($30 \mu\text{g m}^{-2} \text{yr}^{-1}$), which was closely linked to the high atmospheric Hg emission



intensity in this region. In contrast, the multi-year mean TAM dry deposition fluxes estimated by Zhang et al. (2016) from speciated atmospheric Hg concentration observations at North American sites ranged from 5.1 to 14.3 $\mu\text{g m}^{-2} \text{yr}^{-1}$, far lower than the annual average at the NJU site. This discrepancy likely stems from the combination of higher dry deposition fluxes driven by elevated GEM and PBM concentrations, and the underestimation of GOM concentrations at the AMNet monitoring sites that suppresses the estimated dry deposition fluxes, which will be further discussed in Section 3.2.

3.1.3 Hg wet deposition flux measurements

During the representative annual period from July 2020 to June 2021, the average Hg wet deposition flux was 0.83 $\text{ng m}^{-2} \text{h}^{-1}$ and a volume-weighted mean precipitation Hg concentration of 6.29 ng L^{-1} . The annual precipitation depth during this period was 1158 mm. As shown in Figure 2, the Hg wet deposition flux was highly episodic and seasonally uneven. Spring contributed 37% of the annual total (7.29 $\mu\text{g m}^{-2}$), followed by summer (29%), autumn (19%), and winter (14%). Spring and summer together accounted for about two thirds of annual wet deposition, consistent with the concentration of precipitation during the warm, monsoon-influenced seasons (75% of annual rainfall). Winter showed the highest precipitation Hg concentration (11.93 ng L^{-1}) but the lowest deposition flux due to limited precipitation (87.6 mm). The high winter concentration likely reflected weak dilution and elevated PBM (125.85 pg m^{-3}), while high spring RM concentrations (GOM: 19.97 pg m^{-3} ; PBM: 92.80 pg m^{-3}) enhanced scavenging and deposition relative to summer. These results indicate that the seasonal variation in the Hg wet deposition flux at NJU was primarily controlled by precipitation depth, while atmospheric RM abundance played a secondary role by influencing precipitation Hg concentrations.

Hg wet deposition at Nanjing was intermediate relative to observations worldwide, being lower than those at most urban or suburban Asian sites including Chongqing (Wang et al., 2012), Xiamen (Xu et al., 2015), Tokyo (Sakata et al., 2005), Seoul (Eom et al., 2021), and Incheon (Eom et al., 2021), but generally higher than those at most rural, coastal or remote Asian sites including Mt. Changbai (Fu et al., 2016), Mt. Damei (Eom et al., 2021), Mt. Waliguan (Eom et al., 2021), Chongming Island (Tang et al., 2020), and Jeju Island (Eom et al., 2021). Global observations show that the precipitation depth generally exerts a stronger first-order control on the annual Hg wet deposition than the precipitation Hg concentration (Sprovieri et al., 2017; Pearson et al., 2019). Humid coastal and monsoon-influenced regions with abundant precipitation and anthropogenic activities, such as southeastern China and the eastern US, tend to exhibit relatively high Hg wet deposition fluxes (Prestbo and Gay, 2009; Lombard et al., 2011; Xu et al., 2015). The precipitation amount primarily determines the large-scale climatic potential for Hg wet deposition, whereas the precipitation Hg concentration modulates deposition according to regional and local atmospheric Hg loading. The high wet deposition fluxes in polluted urban area, such as Chongqing and Seoul, were driven by the markedly high precipitation Hg level (Wang et al., 2012; Eom et al., 2021), while the low Hg concentration at the remote Amsterdam Island contributed to limited wet deposition fluxes (Tassone et al., 2023).

Despite the relatively abundant precipitation in Nanjing, dry deposition remained the dominant pathway of atmospheric Hg input at NJU. During July 2020–June 2021, the dry-to-wet deposition ratio was 4.0. Co-located direct measurements at other global sites show that this ratio can vary substantially by climatic regimes and site types. At two relatively polluted (urban or



industrial) sites in Michigan, the ratios of measured dry over wet deposition fluxes were 2.5 and 1.9, respectively (Hall et al., 2017). In contrast, summer measurements across coastal, urban, and rural sites in Florida yielded the ratios of 0.17–0.91, indicating a greater contribution from wet deposition in the relatively humid conditions (Hall et al., 2013). Direct measurements at a suburban site and a rural site in Washington State found the dry-to-wet ratios to be 1.2 and 0.33, respectively (Beutel et al., 2021). Although differences in sampling duration and surrogate-surface type could compromise the comparison, the large discrepancies could still embody that the dry-to-wet deposition ratio is strongly site dependent, with the wet deposition generally more dominant at rural or remote sites under humid and precipitation-rich conditions, whereas dry deposition could dominate the polluted urban and industrial environments, especially with high PM concentrations.

3.2 Optimization of speciated Hg dry deposition simulations

3.2.1 Impacts of the ambient concentration on GOM dry deposition simulation

Accurate simulation of GOM dry deposition is highly related to the measured GOM concentrations in the ambient air. In our previous work (Zhang et al., 2024), also as part of the STAMP campaign, simultaneous Tekran and RMAS measurements were performed for ambient GOM concentrations, and a significant correlation between results from the two methods was observed. A correction factor of 2.21 for RMAS over Tekran was extracted and applied in this study to get the corrected GOM concentrations from the continuous Tekran monitoring. With the ambient concentration corrected, the GOM dry deposition fluxes were simulated. As shown in Figure 3, the estimated fluxes agree well with the observations, yielding a regression slope of 0.96 (uncentered $r^2 = 0.84$ with the intercept fixed to zero). Without correction, the average simulated deposition flux would be underestimated by 46%. In comparison, the application of a correction factor of 3 recommended by previous studies (Huang et al., 2017; Gustin et al., 2021a) resulted in an evident positive bias for NJU (with a regression slope of 1.69). Therefore, site-specific correction for the GOM concentration could reduce biases in simulating its dry deposition fluxes. The correction factor was found to vary with the ozone concentration, the moisture in the air, the PBM/GOM ratio, and the fraction of halogenated RM compounds (Zhang et al., 2024).

The uncertainty of GOM dry deposition simulation could also be associated with the chemical composition. GOM consists of multiple oxidized Hg compounds with species-dependent collection efficiencies on the KCl-coated denuders of Tekran (Huang et al., 2013). GOM speciation also affects the modelled deposition velocity through the solubility- and reactivity-related scaling parameters α and β . Values of $\alpha = \beta = 2$ have been recommended for some GOM compounds based on HgCl_2 and $\text{Hg}(\text{OH})_2$ properties (Lyman et al., 2007), whereas other studies have adopted $\alpha = \beta = 10$ to represent more soluble and reactive behaviour (Marsik et al., 2007; Zhang et al., 2009; Castro et al., 2012; Zhang et al., 2012; Yu et al., 2013). Because ambient GOM is a mixture whose composition varies over time, a single set of α and β values could not fully represent its deposition behaviour (Huang and Gustin, 2015b; Huang et al., 2017). Overall, the comparison demonstrates that correcting measurement bias is essential for reproducing the magnitude of GOM dry deposition, whereas further improvements in event-scale model performance require better constraints on GOM chemical speciation and species-dependent deposition parameters.

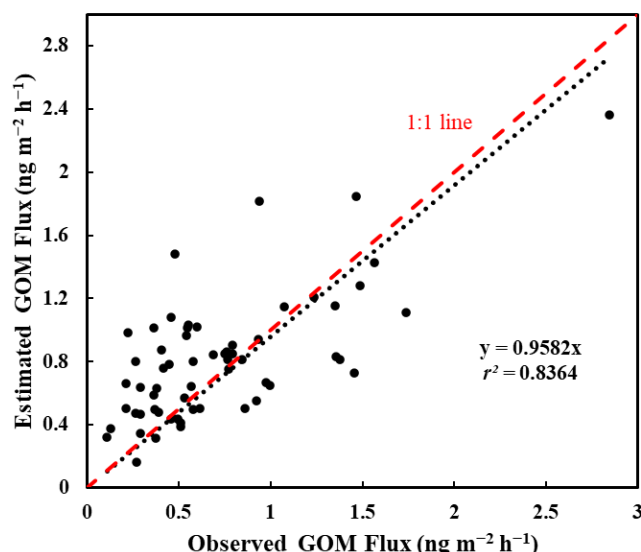


Figure 3. Zero-intercept linear regression between observed and simulated GOM dry deposition fluxes.

3.2.2 Impacts of particle size on PBM dry deposition simulation

Particle size is key to the simulation of PBM dry deposition. In this study, the multi-year mean $\text{PBM}_{2.5}$ deposition velocity was estimated to be 0.19 cm s^{-1} , whereas the inclusion of PBM on coarse particles increased it by 4.5 times. This is consistent with size-fractionated studies showing that coarse PBM could dominate the dry deposition despite their smaller mass fraction (Fang et al., 2012; Zhu et al., 2014; Zhang et al., 2016). The fraction of PBM on coarse particles was set to 0.3 based on these findings for dry deposition simulation (Zhang et al., 2016). However, the fraction (f) is not consistent and varies significantly among different types of sites. Therefore, in this study, we compared the fixed- f scheme to a dynamic- f scheme as described in Section 2.3.2. As demonstrated in Figure 4, the dynamic- f scheme yielded a multi-year mean PBM dry deposition flux of $2.04 \pm 2.26 \text{ ng m}^{-2} \text{ h}^{-1}$ at NJU, significantly lower than the estimate ($2.63 \pm 3.53 \text{ ng m}^{-2} \text{ h}^{-1}$) by fixing f to 0.3. Under the dynamic- f scheme, coarse particles had a deposition velocity of approximately 2.29 cm s^{-1} , more than an order of magnitude higher than that of fine particles, and consequently contributed to 80% of the total PBM dry deposition. More pronounced seasonal variation was captured under the dynamic- f scheme. The f value was relatively low in summer (0.23 ± 0.14) and winter (0.19 ± 0.13), while approaching 0.3 in spring and autumn. The wintertime low PBM deposition velocity alleviated its impacts on the terrestrial surface despite of its high ambient concentration level.

Previous global observations showed substantial spatiotemporal variability in PBM size distributions across urban, coastal, and source-influenced environments (Landis et al., 2002; Feddersen et al., 2012; Fang et al., 2012; Tang et al., 2019), implying that a coarse-particle fraction derived from one site may not be transferable to other atmospheric conditions. Therefore, PBM dry deposition simulation requires both explicit representation of coarse particles and a temporally resolved coarse-particle fraction. The dynamic- f approach developed here reduces the bias associated with a fixed f while retaining the dominant role



of coarse particles in PBM deposition. Since PBM dry deposition is a major component of the total deposition at NJU, the new
400 scheme provides an important advancement towards the subsequent evaluation of TAM dry deposition.

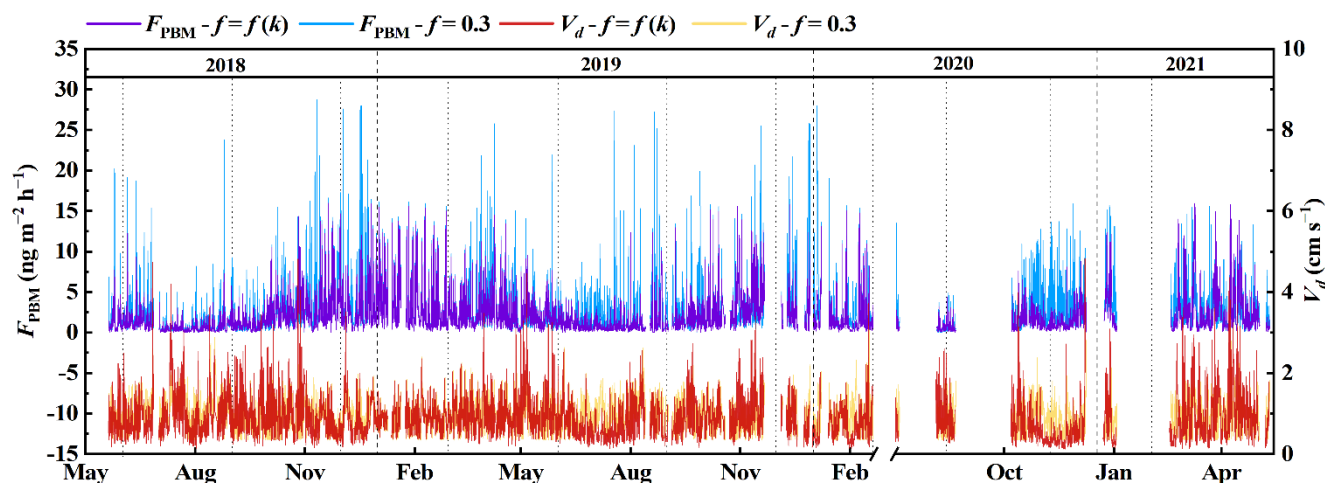


Figure 4. Estimated PBM dry deposition fluxes and deposition velocities under different parameterization schemes.

3.2.3 Influence of vegetation parameterization on GEM dry deposition simulation

Vegetation phenology is crucial to simulation of GEM dry deposition or bidirectional exchange. The underlying-surface-type-
405 based reference LAI scheme (Zhang et al., 2003) could be largely different from the local LAI scheme in reality. Therefore, in this study, we applied both the reference scheme (LAI.1) and the local scheme of NJU (LAI.2) to GEM dry deposition simulation. With improvements of both GOM and PBM dry deposition modelling incorporated (as described in Sections 3.2.1 and 3.2.2), the TAM dry deposition flux was simulated and compared with the measurements at NJU (Figure 5). It can be seen that replacing the reference LAI scheme with the NJU-specific LAI scheme increased the uncentered r^2 of the zero-intercept
410 regression from 0.80 to 0.87, and reduced the pronounced overestimation during the period of late September to early November. The largest effects occurred during March–April and September–November, corresponding to the vegetation green-up and senescence periods in Nanjing, respectively.

The simulated mean TAM dry deposition with local LAI scheme was $5.05 \pm 2.54 \text{ ng m}^{-2} \text{ h}^{-1}$, 46.8 % higher than the observed mean. However, it should be noted that the ATSS method for TAM dry deposition flux measurements has its limitation. The
415 stomatal and cuticular uptake of GEM by vegetation is not reflected by the artificial surrogate surface. In the scheme of Wright and Zhang (2015), LAI regulates leaf area, stomatal and cuticular uptake, in-canopy resistance, and the canopy compensation point, thereby affecting both the magnitude and direction of GEM exchange. Sensitivity analysis was performed to investigate the impacts (Figure 6). When stomatal and cuticular uptake pathways were effectively removed, the TAM dry deposition was underestimated in spring and summer but tended to be overestimated in autumn. The contrasting seasonal response indicates
420 that vegetation does not represent as a constant one-way sink for GEM. Stomatal and cuticular uptake enhances downward GEM exchange fluxes, whereas under warm and high-radiation conditions, increased surface compensation points can favor



re-emission (Wright et al., 2016; Zhang et al., 2019). Accordingly, net GEM emission occurred in summertime in the optimized simulation. In the sensitivity tests (Figures 6a and 6b), replacing the modelled GOM dry deposition with the corresponding observations slightly changed the zero-intercept regression slope, from 0.79 to 0.82, while the uncentered r^2 did not change significantly. The comparison indicates that the simulation of GOM dry deposition has achieved enough accuracy to represent the real situation. Overall, the applications of the dynamic- f scheme for PBM and the local LAI scheme for GEM better capture the seasonal pattern of the observed TAM dry deposition fluxes.

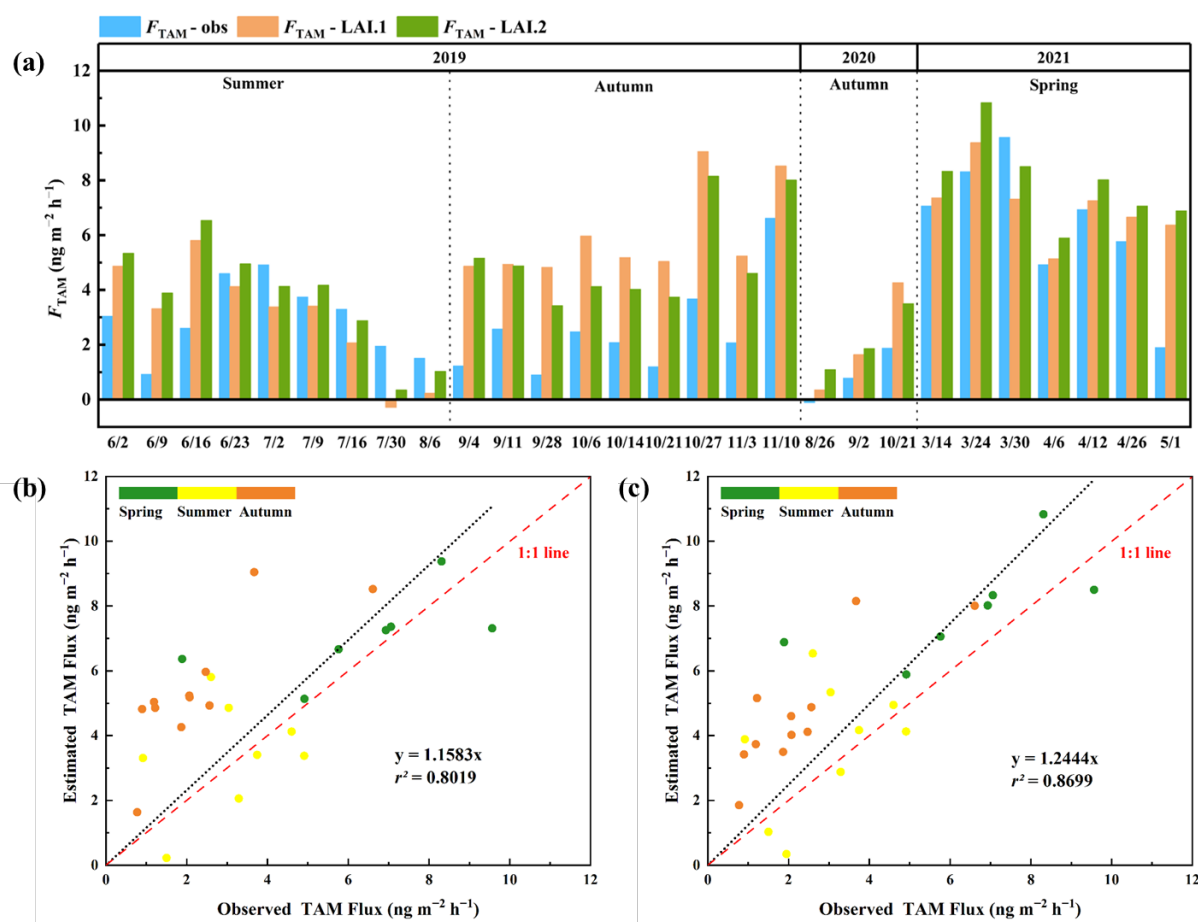


Figure 5. Comparison of (a) observed and simulated TAM dry deposition fluxes under two LAI schemes, and the results of zero-intercept linear regression based on (b) the reference LAI scheme (LAI.1) from Zhang et al. (2003), and (c) the local LAI scheme for NJU (LAI.2).

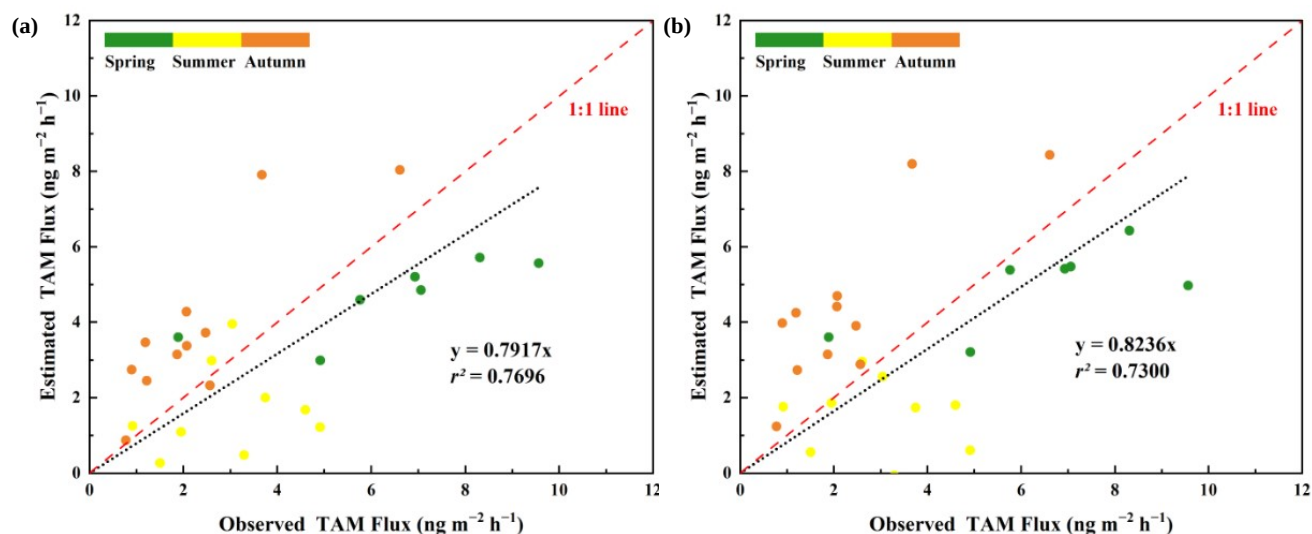


Figure 6. Zero-intercept linear regressions between observed and simulated TAM dry deposition fluxes: (a) simulation with plant stomatal and cuticular uptake pathways effectively removed by setting both R_{st} and R_{cut} to 108 s m^{-1} ; and (b) same as (a), but using observed GOM fluxes for the TAM dry deposition estimate.

3.3 Speciated Hg deposition flux characteristics in urban eastern China

According to the optimized simulation results, both Hg dry deposition velocities and fluxes exhibited pronounced variability among chemical forms and across seasons. As shown in Table 4, the mean dry deposition velocities during the study period (April 2018 to May 2021 with the COVID-19 pandemic gap) followed the order $\text{PBM} > \text{GOM} \gg \text{GEM}$. The PBM deposition velocity was approximately 1.4 times that of GOM and more than 40 times that of GEM, reflecting the strong gravitational settling of particles, particularly the coarse fraction. The corresponding mean dry deposition fluxes for GEM, GOM, and PBM were estimated to be 2.23 ± 3.60 , 0.88 ± 3.77 , and $2.37 \pm 5.39 \text{ ng m}^{-2} \text{ h}^{-1}$, respectively. The contribution of each form to dry deposition is determined jointly by atmospheric abundance and deposition efficiency. The high ambient concentration of GEM compensates for its very low deposition velocity, whereas PBM combines elevated concentrations with efficient particle deposition. The highest and lowest TAM dry deposition fluxes were found in spring ($7.77 \pm 8.84 \text{ ng m}^{-2} \text{ h}^{-1}$) and summer ($2.52 \pm 4.67 \text{ ng m}^{-2} \text{ h}^{-1}$), respectively. The springtime flux was characterized by the high GEM dry deposition, due to the relatively high deposition velocity during this season and enhanced vegetation uptake. In summer, the GEM deposition velocity approached zero on average, and episodic negative GEM fluxes occurred (Figure 7), indicating that high temperature and solar radiation triggered surface Hg re-emission, offsetting the GEM dry deposition. PBM dry deposition was also lowest in summer but increased markedly in winter. Since the winter coarse-particle fraction remained relatively low, this enhancement was primarily associated with increased PBM mass concentration. GOM showed a different seasonal behaviour, with the highest dry deposition flux in spring and the lowest in winter. The seasonal composition of Hg dry deposition shifted from a larger



GEM contribution in spring toward enhanced PBM deposition in winter, whereas weak GEM uptake or net re-emission together with low PBM loading suppressed total dry deposition in summer.

455 **Table 4. Modelling results of speciated and total Hg dry deposition at NJU during the study period (April 2018 to May 2021 with the COVID-19 pandemic gap)**

Parameter	Season	GEM	GOM	PBM	TAM
Deposition velocity (cm s ⁻¹)	Spring	0.03±0.03	0.61±0.33	0.96±0.63	
	Summer	0.001±0.03	0.66±0.39	0.83±0.52	
	Autumn	0.01±0.01	0.54±0.25	0.88±0.54	
	Winter	0.02±0.005	0.60±0.30	0.75±0.46	
	Annual	0.02±0.02	0.60±0.32	0.86±0.55	
Deposition flux (ng m ⁻² h ⁻¹)	Spring	3.74±4.11	1.29±5.23	2.74±3.59	7.77±8.84
	Summer	0.72±3.66	0.82±1.36	0.98±1.21	2.52±4.67
	Autumn	1.73±2.29	0.83±4.44	2.51±3.95	5.07±6.96
	Winter	2.71±3.45	0.57±2.49	3.19±9.28	6.47±10.87
	Annual	2.23±3.60	0.88±3.77	2.37±5.39	5.49±8.36

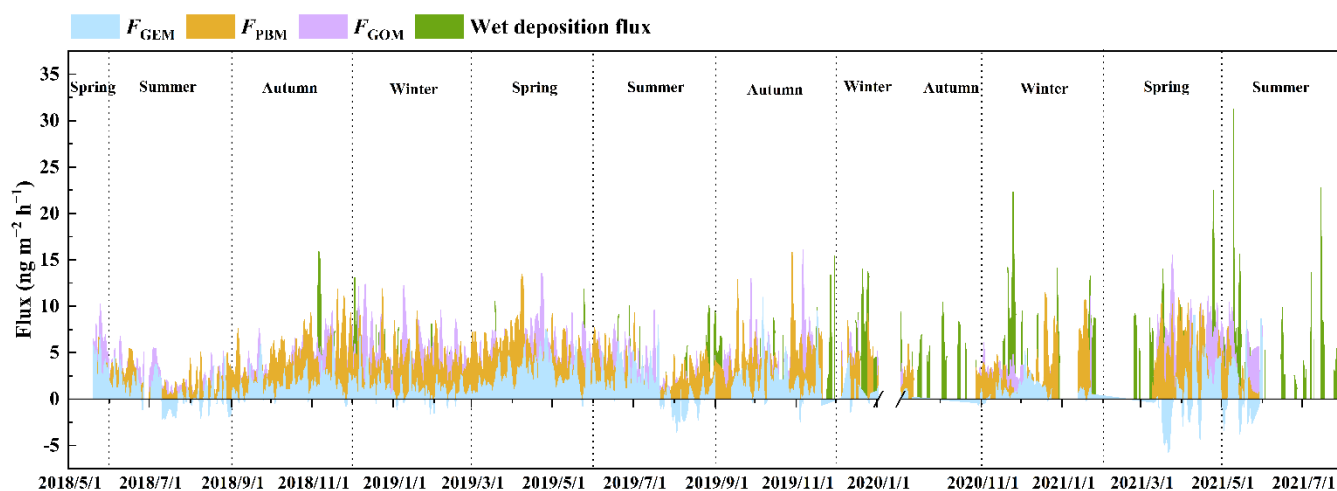


Figure 7. Composition of speciated Hg deposition fluxes during the study period.



460 The speciated dry deposition pattern at NJU, a typical urban site in eastern China, differs largely from the modelling results for North America based on the AMNet Hg concentration data (Zhang et al., 2012; Zhang et al., 2016). PBM and GEM were the main sources of Hg dry deposition fluxes at NJU, while the much lower PBM concentration level in North America led to the dominance of GEM in the total dry deposition benefited mainly from vegetation uptake (Zhang et al., 2016). The enhanced importance of PBM in urban eastern China reflected the strong dependence of dry deposition on particle size distribution.

465 Previous studies that did not explicitly represent coarse PBM may underestimate the PBM contribution to total dry deposition (Wright et al., 2016). Size-resolved observations from polluted regions emphasize that particle-size distribution can strongly regulate PBM dry deposition (Tang et al., 2019).

Consistent with the observations, dry deposition was the dominant atmospheric Hg sink at NJU, with a modelled dry-to-wet deposition ratio of 6.6. At the global terrestrial scale, the Global Mercury Assessment 2018 indicates that this ratio is in the

470 range of 1.2–2.2 across most regions, whereas wet deposition exceeds dry deposition over the oceans (UN Environment, 2019). Substantial departures from this average global pattern occur among different climatic regimes and land-cover types. Across 23 predominantly deciduous forest sites in the eastern US, litterfall Hg deposition alone had a mean dry-to-wet deposition ratio of 1.3 (Risch et al., 2012). At the Lulin background forest in East Asia, the eight-year mean total dry-to-wet ratio reached 2.8, and GEM contributed 78 % of the modelled dry deposition because of the dense forest canopy and its much higher atmospheric

475 abundance than GOM and PBM (Nguyen et al., 2019). An even stronger GEM-dominated pathway has been identified in the interior Arctic tundra, where ecosystem-scale mass-balance and isotope measurements attributed 70% of the total Hg input to atmospheric GEM deposition, with only minor contributions from wet deposition and atmospheric Hg depletion events (Obrist et al., 2017).

Global modelling further identifies tropical South America, central Africa, Southeast Asia, and East Asia as major terrestrial

480 GEM deposition regions because of high vegetation activity and elevated atmospheric GEM concentrations (Obrist et al., 2018). GOM dry deposition is more important at high-elevation grasslands and savannas, where enhanced oxidation, free-tropospheric influence, stronger solar radiation, and atmospheric turbulence increase GOM concentrations and surface transfer (Zhang et al., 2019). When GOM-rich air coincides with strong convection and precipitation, wet scavenging becomes increasingly important, as demonstrated at tropical and windward mountain sites (Obrist et al., 2018; Nguyen and Sheu, 2019).

485 PBM deposition is enhanced in urban, industrial, and agricultural regions with heavy PM pollution, particularly in East Asia (Zhang et al., 2019). CMAQ-Hg simulations over the southern China showed that PBM contributed 19% of the total Hg dry deposition and 33% of wet deposition (Liu et al., 2019). NJU in this study, therefore, represents a polluted East Asian urban regime in which atmospheric Hg input is strongly shifted toward dry deposition, with GEM and PBM jointly controlling the annual dry deposition budget, while RM abundance and precipitation regulate the magnitude of wet deposition.

490 4 Conclusions

This study combined long-term surrogate-surface observations with process-based modelling to characterize atmospheric Hg deposition and constrain key uncertainties in speciated dry deposition estimates at an urban site in eastern China. During July



2020–June 2021, the observed TAM dry and wet deposition were 29.19 and 7.29 $\mu\text{g m}^{-2} \text{yr}^{-1}$, respectively, yielding a dry-to-wet deposition ratio of 4.0. The observations demonstrate that dry deposition dominates atmospheric Hg input into the terrestrial surface at NJU despite the abundant precipitation of the subtropical monsoon climate. Hg wet deposition exhibited pronounced seasonality governed primarily by precipitation depth, with RM abundance further modulating precipitation Hg concentrations.

Model optimization was performed to improve speciated Hg dry deposition estimates. First, correcting the low bias in the denuder-based GOM concentrations using a site-specific RMAS/Tekran correction factor of 2.21 brought simulated GOM dry deposition fluxes into close agreement with observations. Second, explicitly including coarse particles with a dynamic scheme increased the PBM deposition velocity by a factor of 4.5, with coarse particles contributing 80% of total PBM dry deposition. Third, localizing vegetation phenology by replacing the reference LAI with NJU-specific values, together with the coarse PBM consideration, improved the model–observation regression of TAM dry deposition. Together, these refinements indicate that accurate simulation of Hg dry deposition in polluted eastern Chinese cities requires constraining GOM measurement bias and composition, PBM particle-size distribution, and GEM bidirectional exchange with local vegetation phenology.

Our results offer several advances over previous studies. Earlier observations in East and Southeast Asia were conducted in more heavily polluted regions and generally reported higher atmospheric Hg concentrations and deposition fluxes (Fu et al., 2016; Ma et al., 2015; Wright et al., 2016). The TAM dry-deposition flux observed at NJU was lower than several earlier values reported for urban and industrialized Asian sites, reflecting the combined effects of emission reductions, shifts in source composition, regional transport, and differences in meteorological conditions and measurement approaches. Previous studies have largely focused on GEM concentrations or wet deposition, whereas simultaneous observations linking atmospheric Hg species to surface dry-deposition processes remain scarce, particularly for urban sites in eastern China (Zhang et al., 2019). By combining atmospheric Hg measurements with turf-based deposition observations and model calculations, this study helps constrain the relationship between atmospheric composition and surface Hg input.

Several limitations should be noted. The Aerohead method may overestimate GOM dry deposition under the high particle-loading conditions at NJU, since particle-associated Hg can interfere with the collection of nominally gaseous Hg, although this bias was not quantified and is unlikely to alter the main conclusion given that GOM represents only a small fraction of TAM dry deposition. In addition, the artificial-turf method does not reproduce stomatal exchange and therefore underestimate Hg uptake by vegetated surfaces. Comparisons with simulations excluding stomatal uptake confirm that the observations remain useful for constraining non-stomatal deposition. Finally, the single urban site and approximately two-year observation period limit extrapolation to other locations and times, but they do not undermine the internal comparisons among Hg species, deposition pathways, and model parameterizations.

Code and data availability

The dataset related to this article is available at <https://doi.org/10.5281/zenodo.22958565> (Yuan et al., 2026).



525 **Author contributions**

RY: data curation, formal analysis, validation, visualization, writing (original draft, review, and editing). GCZ: investigation, data curation, methodology, formal analysis, writing (original draft). LZ: conceptualization, methodology, formal analysis, writing (original draft, review, and editing), supervision. HZ: investigation, data curation. PSZ: methodology, investigation. WKZ: investigation. HTZ: investigation. ZW: project administration, resources. QGW: supervision. YZ: supervision.

530 **Competing interests**

The contact author has declared that none of the authors has any competing interests.

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References

- Beutel, M. W., DeSilva, L., and Amegbletor, L.: Direct Measurement of Mercury Deposition at Rural and Suburban Sites in Washington State, USA, *Atmosphere*, 12, 35, <https://doi.org/10.3390/atmos12010035>, 2021.
- Castro, M. S., Moore, C., Sherwell, J., and Brooks, S. B.: Dry deposition of gaseous oxidized mercury in western Maryland,
540 *Sci. Total Environ.*, 417–418, 232–240, <https://doi.org/10.1016/j.scitotenv.2011.12.044>, 2012.
- Dastoor, A., Angot, H., Bieser, J., et al.: The Multi-Compartment Hg Modeling and Analysis Project (MCHgMAP): mercury modeling to support international environmental policy, *Geosci. Model Dev.*, 18, 2747–2860, <https://doi.org/10.5194/gmd-18-2747-2025>, 2025.
- Driscoll, C. T., Mason, R. P., Chan, H. M., Jacob, D. J., and Pirrone, N.: Mercury as a global pollutant: sources, pathways, and
545 effects, *Environ. Sci. Technol.*, 47, 4967–4983, <https://doi.org/10.1021/es305071v>, 2013.
- Eom, S., Lee, H., Kim, J., Park, K., Kim, Y., Sheu, G.-R., Gay, D. A., Schmeltz, D., and Han, S.: Potential sources, scavenging processes, and source regions of mercury in the wet deposition of South Korea, *Sci. Total Environ.*, 762, 143934, <https://doi.org/10.1016/j.scitotenv.2020.143934>, 2021.
- Evers, D. C., Keane, S. E., Basu, N., and Buck, D.: Evaluating the effectiveness of the Minamata Convention on Mercury:
550 principles and recommendations for next steps, *Sci. Total Environ.*, 569–570, 888–903, <https://doi.org/10.1016/j.scitotenv.2016.05.001>, 2016.
- Fang, G. C., Zhang, L., and Huang, C. S.: Measurements of size-fractionated concentration and bulk dry deposition of atmospheric particulate-bound mercury, *Atmos. Environ.*, 61, 371–377, <https://doi.org/10.1016/j.atmosenv.2012.07.052>, 2012.



- 555 Feddersen, D. M., Talbot, R., Mao, H., and Sive, B. C.: Size distribution of particulate mercury in marine and coastal atmospheres, *Atmos. Chem. Phys.*, 12, 10899–10909, <https://doi.org/10.5194/acp-12-10899-2012>, 2012.
- Fu, X., Yang, X., Lang, X., Zhou, J., Zhang, H., Yu, B., Yan, H., Lin, C.-J., and Feng, X.: Atmospheric wet and litterfall mercury deposition at urban and rural sites in China, *Atmos. Chem. Phys.*, 16, 11547–11562, <https://doi.org/10.5194/acp-16-11547-2016>, 2016.
- 560 Gratz, L. E., Keeler, G. J., Morishita, M., Barres, J. A., and Dvonch, J. T.: Assessing the emission sources of atmospheric mercury in wet deposition across Illinois, *Sci. Total Environ.*, 448, 120–131, <https://doi.org/10.1016/j.scitotenv.2012.11.011>, 2013.
- Gustin, M. S., Dunham-Cheatham, S. M., and Zhang, L.: Comparison of 4 methods for measurement of reactive, gaseous oxidized, and particulate-bound mercury, *Environ. Sci. Technol.*, 53, 14489–14495, <https://doi.org/10.1021/acs.est.9b04648>,
565 2019.
- Gustin, M. S., Dunham-Cheatham, S. M., Huang, J., Lindberg, S., and Lyman, S. N.: Development of an understanding of reactive mercury in ambient air: a review, *Atmosphere*, 12, 73, <https://doi.org/10.3390/atmos12010073>, 2021a.
- Gustin, M. S., Dunham-Cheatham, S. M., Osterwalder, S., Magand, O., and Dommergue, A.: What is the utility of measuring gaseous Hg^{II} dry deposition using Aerohead samplers?: A review, *Sci. Total Environ.*, 907, 167895, <https://doi.org/10.1016/j.scitotenv.2023.167895>, 2024.
570
- Gustin, M. S., Dunham-Cheatham, S. M., Zhang, L., Lyman, S., Choma, N., and Castro, M.: Use of membranes and detailed HYSPLIT analyses to understand atmospheric particulate, gaseous oxidized, and reactive mercury chemistry, *Environ. Sci. Technol.*, 55, 893–901, <https://doi.org/10.1021/acs.est.0c07876>, 2021b.
- Gustin, M. S., Pierce, A. M., Huang, J., Miller, M. B., Holmes, H. A., and Loria-Salazar, S. M.: Evidence for different reactive
575 Hg sources and chemical compounds at adjacent valley and high-elevation locations, *Environ. Sci. Technol.*, 50, 12225–12231, <https://doi.org/10.1021/acs.est.6b03339>, 2016.
- Hall, N. L., Dvonch, J. T., Barres, J. A., Marsik, F. M., and Keeler, G. J.: Applications of a turf surrogate surface sampling technique in measuring mercury dry deposition in Florida, USA, *E3S Web Conf.*, 1, 03008, <https://doi.org/10.1051/e3sconf/20130103008>, 2013.
- 580 Hall, N. L., Dvonch, J. T., Marsik, F. J., Barres, J. A., and Landis, M. S.: An artificial turf-based surrogate surface collector for the direct measurement of atmospheric mercury dry deposition, *Int. J. Environ. Res. Public Health*, 14, 173, <https://doi.org/10.3390/ijerph14020173>, 2017.
- Han, J. S., Seo, Y. S., Kim, M. K., Holsen, T. M., and Yi, S. M.: Total atmospheric mercury deposition in forested areas in South Korea, *Atmos. Chem. Phys.*, 16, 7653–7662, <https://doi.org/10.5194/acp-16-7653-2016>, 2016.
- 585 Huang, J. and Gustin, M. S.: Uncertainties of gaseous oxidized mercury measurements using KCl-coated denuders, cation-exchange membranes, and nylon membranes: humidity influences, *Environ. Sci. Technol.*, 49, 6102–6108, <https://doi.org/10.1021/acs.est.5b00098>, 2015b.



- Huang, J. and Gustin, M. S.: Use of passive sampling methods and models to understand sources of mercury deposition to high elevation sites in the western United States, *Environ. Sci. Technol.*, 49, 432–441, <https://doi.org/10.1021/es502836w>, 2015a.
- Huang, J., Choi, H.-D., Landis, M. S., and Holsen, T. M.: An application of passive samplers to understand atmospheric mercury concentration and dry deposition spatial distributions, *J. Environ. Monit.*, 14, 2976–2982, <https://doi.org/10.1039/C2EM30514C>, 2012.
- Huang, J., Kang, S., Tang, W., He, M., Guo, J., Zhang, Q., Yin, X., and Tripathee, L.: Contrasting changes in long-term wet mercury deposition and socioeconomic development in the largest city of Tibet, *Sci. Total Environ.*, 804, 150124, <https://doi.org/10.1016/j.scitotenv.2021.150124>, 2022.
- Huang, J., Lyman, S. N., Hartman, J. S., and Gustin, M. S.: A review of passive sampling systems for ambient air mercury measurements, *Environ. Sci.-Process. Impacts*, 16(3), 374–392, <https://doi.org/10.1039/C3EM00501A>, 2014.
- Huang, J., Miller, M. B., and Weiss-Penzias, P.: Comparison of gaseous oxidized Hg measured by KCl-coated denuders, and nylon and cation exchange membranes, *Environ. Sci. Technol.*, 47, 7307–7316, <https://doi.org/10.1021/es4012349>, 2013.
- Huang, J., Miller, M. B., Edgerton, E., and Sexauer Gustin, M.: Deciphering potential chemical compounds of gaseous oxidized mercury in Florida, USA, *Atmos. Chem. Phys.*, 17, 1689–1698, <https://doi.org/10.5194/acp-17-1689-2017>, 2017.
- Jones, C. P., Lyman, S. N., Jaffe, D. A., Allen, T., and O'Neil, T. L.: Detection and quantification of gas-phase oxidized mercury compounds by GC/MS, *Atmos. Meas. Tech.*, 9, 2195–2205, <https://doi.org/10.5194/amt-9-2195-2016>, 2016.
- Landis, M. S., Vette, A. F., and Keeler, G. J.: Atmospheric mercury in the Lake Michigan Basin: influence of the Chicago/Gary urban area, *Environ. Sci. Technol.*, 36, 4508–4517, <https://doi.org/10.1021/es011216j>, 2002.
- Lindberg, S., Bullock, R., Ebinghaus, R., Engstrom, D., Feng, X., Fitzgerald, W., Pirrone, N., Prestbo, E., and Seigneur, C.: A synthesis of progress and uncertainties in attributing the sources of mercury in deposition, *Ambio*, 36, 19–32, [https://doi.org/10.1579/0044-7447\(2007\)36\[19:ASOPAU\]2.0.CO;2](https://doi.org/10.1579/0044-7447(2007)36[19:ASOPAU]2.0.CO;2), 2007.
- Liu, J., Wang, L., Zhu, Y., Lin, C.-J., Jang, C., Wang, S., Xing, J., Yu, B., Xu, H., and Pan, Y.: Source attribution for mercury deposition with an updated atmospheric mercury emission inventory in the Pearl River Delta Region, China, *Front. Environ. Sci. Eng.*, 13, 2, <https://doi.org/10.1007/s11783-019-1087-6>, 2019.
- Lombard, M. A. S., Bryce, J. G., Mao, H., and Talbot, R.: Mercury deposition in southern New Hampshire, 2006–2009, *Atmos. Chem. Phys.*, 11, 7657–7668, <https://doi.org/10.5194/acp-11-7657-2011>, 2011.
- Lynam, M. M., Hall, N. L., and Dvonch, J. T.: Spatial patterns in wet and dry deposition of atmospheric mercury to Illinois watersheds, *E3S Web Conf.*, 1, 03007, <https://doi.org/10.1051/e3sconf/20130103007>, 2013.
- Lynam, M. M., Dvonch, J. T., Hall, N. L., Morishita, M., and Barres, J. A.: Spatial patterns in wet and dry deposition of atmospheric mercury and trace elements in central Illinois, USA, *Environ. Sci. Pollut. Res.*, 21, 4032–4043, <https://doi.org/10.1007/s11356-013-2011-4>, 2014.
- Lyman, S. N., Cheng, I., Gratz, L. E., Weiss-Penzias, P., and Zhang, L.: An updated review of atmospheric mercury, *Sci. Total Environ.*, 707, 135575, <https://doi.org/10.1016/j.scitotenv.2019.135575>, 2020.



- Lyman, S. N., Gustin, M. S., Prestbo, E. M., and Marsik, F. J.: Estimation of dry deposition of atmospheric mercury in Nevada by direct and indirect methods, *Environ. Sci. Technol.*, 41, 1970–1976, <https://doi.org/10.1021/es062323m>, 2007.
- Lyman, S. N., Gustin, M. S., Prestbo, E. M., Kilner, P. I., Edgerton, E., and Hartsell, B.: Testing and application of surrogate
625 surfaces for understanding potential gaseous oxidized mercury dry deposition, *Environ. Sci. Technol.*, 43, 6235–6241, <https://doi.org/10.1021/es901192e>, 2009.
- Lyman, S. N., Jaffe, D. A., and Gustin, M. S.: Release of mercury halides from KCl denuders in the presence of ozone, *Atmos. Chem. Phys.*, 10, 8197–8204, <https://doi.org/10.5194/acp-10-8197-2010>, 2010.
- Ma, M., Wang, D., Du, H., Sun, T., Zhao, Z., and Wei, S.: Atmospheric mercury deposition and its contribution of the regional
630 atmospheric transport to mercury pollution at a national forest nature reserve, southwest China, *Environ. Sci. Pollut. R.*, 22, 20007–20018, <https://doi.org/10.1007/s11356-015-5152-9>, 2015.
- Mao, H., Ye, Z., and Driscoll, C. T.: Meteorological effects on Hg wet deposition in a forested site in the Adirondack region of New York during 2000–2015, *Atmos. Environ.*, 168, 90–100, <https://doi.org/10.1016/j.atmosenv.2017.08.058>, 2017.
- Marsik, F. J., Keeler, G. J., and Landis, M. S.: The dry-deposition of speciated mercury to the Florida Everglades:
635 measurements and modeling, *Atmos. Environ.*, 41, 136–149, <https://doi.org/10.1016/j.atmosenv.2006.07.032>, 2007.
- McClure, C. D., Jaffe, D. A., and Edgerton, E. S.: Evaluation of the KCl denuder method for gaseous oxidized mercury using HgBr_2 at an in-service AMNet site, *Environ. Sci. Technol.*, 48, 11437–11444, <https://doi.org/10.1021/es502545k>, 2014.
- Nguyen, L. S. P. and Sheu, G.-R.: Four-year measurements of wet mercury deposition at a tropical mountain site in central Taiwan, *Aerosol Air Qual. Res.*, 19, 2043–2055, <https://doi.org/10.4209/aaqr.2019.05.0250>, 2019.
- 640 Nguyen, L. S. P., Zhang, L., Lin, D.-W., Lin, N.-H., and Sheu, G.-R.: Eight-year dry deposition of atmospheric mercury to a tropical high mountain background site downwind of the East Asian continent, *Environ. Pollut.*, 255, 113128, <https://doi.org/10.1016/j.envpol.2019.113128>, 2019.
- Obrist, D., Agnan, Y., Jiskra, M., Olson, C. L., Colegrove, D. P., Hueber, J., Moore, C. W., Sonke, J. E., and Helmig, D.: Tundra uptake of atmospheric elemental mercury drives Arctic mercury pollution, *Nature*, 547, 201–204, <https://doi.org/10.1038/nature22997>, 2017.
- 645 Obrist, D., Kirk, J. L., Zhang, L., Sunderland, E. M., Jiskra, M., and Selin, N. E.: A review of global environmental mercury processes in response to human and natural perturbations: Changes of emissions, climate, and land use, *Ambio*, 47(2), 116–140, <https://doi.org/10.1007/s13280-017-1004-9>, 2018.
- Osterwalder, S., Dunham-Cheatham, S. M., Ferreira Araujo, B., Magand, O., Thomas, J. L., Baladima, F., Pfaffhuber, K. A., Berg, T., Zhang, L., Huang, J., Dommergue, A., Sonke, J. E., and Gustin, M. S.: Fate of springtime atmospheric reactive
650 mercury: concentrations and deposition at Zeppelin, Svalbard, *ACS Earth Space Chem.*, 5, 3234–3246, <https://doi.org/10.1021/acsearthspacechem.1c00299>, 2021.
- Pearson, C., Howard, D., Moore, C., and Obrist, D.: Mercury and trace metal wet deposition across five stations in Alaska: controlling factors, spatial patterns, and source regions, *Atmos. Chem. Phys.*, 19, 6913–6929, <https://doi.org/10.5194/acp-19-6913-2019>, 2019.
- 655



- Peterson, C., Alishahi, M., and Gustin, M. S.: Testing the use of passive sampling systems for understanding air mercury concentrations and dry deposition across Florida, USA, *Sci. Total Environ.*, 424, 297–307, <https://doi.org/10.1016/j.scitotenv.2012.02.031>, 2012.
- Pirrone, N. and Keating, T. (Eds.): Hemispheric Transport of Air Pollution 2010, Part B: Mercury, Air Pollution Studies No. 18, United Nations, New York and Geneva, 214 pp., ISBN 978-92-1-117044-3, 2010.
- 660 Prestbo, E. M. and Gay, D. A.: Wet deposition of mercury in the U.S. and Canada, 1996–2005: results and analysis of the NADP mercury deposition network (MDN), *Atmos. Environ.*, 43, 4223–4233, <https://doi.org/10.1016/j.atmosenv.2009.05.028>, 2009.
- Qin, X., Wang, X., Shi, Y., Yu, G., Zhao, N., Lin, Y., Fu, Q., Wang, D., Xie, Z., Deng, C., and Huang, K.: Characteristics of atmospheric mercury in a suburban area of east China: sources, formation mechanisms, and regional transport, *Atmos. Chem. Phys.*, 19, 5923–5940, <https://doi.org/10.5194/acp-19-5923-2019>, 2019.
- 665 Risch, M. R., DeWild, J. F., Krabbenhoft, D. P., Kolka, R. K., and Zhang, L.: Litterfall mercury dry deposition in the eastern USA, *Environ. Pollut.*, 161, 284–290, <https://doi.org/10.1016/j.envpol.2011.06.005>, 2012.
- Sakata, M. and Marumoto, K.: Wet and dry deposition fluxes of mercury in Japan, *Atmos. Environ.*, 39, 3139–3146, <https://doi.org/10.1016/j.atmosenv.2005.01.049>, 2005.
- 670 Sather, M. E., Mukerjee, S., Allen, K. L., Smith, L., Mathew, J., Jackson, C., Callison, R., Scrapper, L., Hathcoat, A., Adam, J., Keese, D., Ketcher, P., Brunette, R., Karlstrom, J., and Van der Jagt, G.: Gaseous oxidized mercury dry deposition measurements in the southwestern USA: a comparison between Texas, eastern Oklahoma, and the Four Corners area, *Sci. World J.*, 2014, 580723, <https://doi.org/10.1155/2014/580723>, 2014.
- 675 Sather, M. E., Mukerjee, S., Smith, L., Brunette, R., Karlstrom, J., and Van der Jagt, G.: Gaseous oxidized mercury dry deposition measurements in the Four Corners area, U.S.A., after large power plant mercury emission reductions, *Atmos. Pollut. Res.*, 12, 148–158, <https://doi.org/10.1016/j.apr.2020.08.030>, 2021.
- Sather, M. E., Mukerjee, S., Smith, L., Mathew, J., Jackson, C., Callison, R., Scrapper, L., Hathcoat, A., Adam, J., Keese, D., Ketcher, P., Brunette, R., Karlstrom, J., and Van der Jagt, G.: Gaseous oxidized mercury dry deposition measurements in the Four Corners area and eastern Oklahoma, U.S.A., *Atmos. Pollut. Res.*, 4, 168–180, <https://doi.org/10.5094/APR.2013.017>, 2013.
- 680 Schroeder, W. H. and Munthe, J.: Atmospheric mercury: an overview, *Atmos. Environ.*, 32, 809–822, [https://doi.org/10.1016/S1352-2310\(97\)00293-8](https://doi.org/10.1016/S1352-2310(97)00293-8), 1998.
- Seigneur, C., Vijayaraghavan, K., Lohman, K., Karamchandani, P., and Scott, C.: Global source attribution for mercury deposition in the United States, *Environ. Sci. Technol.*, 38, 555–569, <https://doi.org/10.1021/es034109t>, 2004.
- Selin, H., Keane, S. E., Wang, S., Selin, N. E., Davis, K., and Bally, D.: Linking science and policy to support the implementation of the Minamata Convention on Mercury, *Ambio*, 47(2), 198–215, <https://doi.org/10.1007/s13280-017-1003-x>, 2018.



- Sommar, J., Zhu, W., Lin, C.-J., and Feng, X. B.: Field approaches to measure Hg exchange between natural surfaces and the atmosphere—A review, *Crit. Rev. Environ. Sci. Technol.*, 43(15), 1657–1739, <https://doi.org/10.1080/10643389.2012.671733>, 2013.
- Sprovieri, F., Pirrone, N., Bencardino, M., D'Amore, F., Angot, H., Barbante, C., Brunke, E. G., Arcega-Cabrera, F., Cairns, W., Comero, S., Diéguez, M. D. C., Dommergue, A., Ebinghaus, R., Feng, X. B., Fu, X., Garcia, P. E., Gawlik, B. M., Hageström, U., Hansson, K., Horvat, M., Kotnik, J., Labuschagne, C., Magand, O., Martin, L., Mashyanov, N., Mkololo, T., Munthe, J., Obolkin, V., Ramirez Islas, M., Sena, F., Somerset, V., Spandow, P., Vardè, M., Walters, C., Wängberg, I., Weigelt, A., Yang, X., and Zhang, H.: Five-year records of mercury wet deposition flux at GMOS sites in the Northern and Southern hemispheres, *Atmos. Chem. Phys.*, 17, 2689–2708, <https://doi.org/10.5194/acp-17-2689-2017>, 2017.
- Tang, Y., Wang, S., Wu, Q., Liu, K., Li, Z., Zou, J., Hou, D., Wu, Y., and Duan, L.: Measurement of size-fractionated particulate-bound mercury in Beijing and implications on sources and dry deposition of mercury, *Sci. Total Environ.*, 675, 176–183, <https://doi.org/10.1016/j.scitotenv.2019.04.245>, 2019.
- Tang, Y., Wu, Q., Gao, W., Wang, S., Li, Z., Liu, K., and Han, D.: Impacts of anthropogenic emissions and meteorological variation on Hg wet deposition in Chongming, China, *Atmosphere*, 11, 1301, <https://doi.org/10.3390/atmos11121301>, 2020.
- Tassone, A., Magand, O., Naccarato, A., Martino, M., Amico, D., Sprovieri, F., Leuridan, H., Bertrand, Y., Ramonet, M., Pirrone, N., and Dommergue, A.: Seven-year monitoring of mercury in wet precipitation and atmosphere at the Amsterdam Island GMOS station, *Heliyon*, 9, e14608, <https://doi.org/10.1016/j.heliyon.2023.e14608>, 2023.
- UN Environment: Global Mercury Assessment 2018, UN Environment Programme, Chemicals and Health Branch, Geneva, Switzerland, ISBN 978-92-807-3744-8, 2019.
- Wang, W., Zhang, W., Zhang, L., Zhou, H., Zhang, C., Gustin, M. S., Dunham-Cheatham, S. M., Zhang, G., Zhou, P., Wang, Z., Wang, Q., and Zhao, Y.: Evaluation of an improved reactive mercury active system: uncertainties and applicability in polluted environments. *Environ. Sci. Technol.*, 60(30): 21135–21144, <https://doi.org/10.1021/acs.est.6c03802>, 2026.
- Wang, Y. M., Wang, D. Y., Meng, B., Peng, Y. L., Zhao, L., and Zhu, J. S.: Spatial and temporal distributions of total and methyl mercury in precipitation in core urban areas, Chongqing, China, *Atmos. Chem. Phys.*, 12, 9417–9426, <https://doi.org/10.5194/acp-12-9417-2012>, 2012.
- Weiss-Penzias, P. S., Gustin, M. S., and Lyman, S. N.: Sources of gaseous oxidized mercury and mercury dry deposition at two southeastern U.S. sites, *Atmos. Environ.*, 45, 4569–4579, <https://doi.org/10.1016/j.atmosenv.2011.05.069>, 2011.
- Wesely, M. L. and Hicks, B. B.: A review of the current status of knowledge on dry deposition, *Atmos. Environ.*, 34, 2261–2282, [https://doi.org/10.1016/S1352-2310\(99\)00467-7](https://doi.org/10.1016/S1352-2310(99)00467-7), 2000.
- Wesely, M. L., Doskey, P. V., and Shannon, J. D.: Deposition parameterizations for the Industrial Source Complex (ISC3) model, Argonne National Laboratory, Argonne, IL, USA, ANL/ER/TR-01/003, 128 pp., <https://doi.org/10.2172/1260839>, 2002.
- Wright, L. P. and Zhang, L.: An approach estimating bidirectional air-surface exchange for gaseous elemental mercury at AMNet sites, *J. Adv. Model. Earth Syst.*, 7, 35–49, <https://doi.org/10.1002/2014MS000367>, 2015.



- Wright, L. P., Zhang, L., and Marsik, F. J.: Overview of mercury dry deposition, litterfall, and throughfall studies, *Atmos. Chem. Phys.*, 16, 13399–13416, <https://doi.org/10.5194/acp-16-13399-2016>, 2016.
- 725 Xu, L., Chen, J., Yang, L., Niu, Z., Tong, L., Yin, L., and Chen, Y.: Characteristics and sources of atmospheric mercury speciation in a coastal city, Xiamen, China, *Chemosphere*, 119, 530–539, <https://doi.org/10.1016/j.chemosphere.2014.07.024>, 2015.
- Yu, C., Xu, Y., Yan, Y., Xiao, W., Liu, M., Cheng, M., He, W., Xu, F., and Wang, X.: Mercury and methylmercury in China's lake sediments and first estimation of mercury burial fluxes, *Sci. Total Environ.*, 770, 145338, <https://doi.org/10.1016/j.scitotenv.2021.145338>, 2021.
- 730 Yu, X., Driscoll, C. T., Huang, J. Y., Holsen, T. M., and Blackwell, B. D.: Modeling and mapping of atmospheric mercury deposition in Adirondack Park, New York, *PLOS ONE*, 8, e59322, <https://doi.org/10.1371/journal.pone.0059322>, 2013.
- Zhang, G., Zhou, X., Li, X., Wang, L., Li, X., Luo, Z., Zhang, Y., Yang, Z., Hu, R., Tang, Z., Wang, D., and Wang, Z.: Gaseous elemental mercury exchange fluxes over air-soil interfaces in the degraded grasslands of northeastern China, *Biology*, 10, 917, <https://doi.org/10.3390/biology10090917>, 2021.
- 735 Zhang, L., Blanchard, P., Gay, D. A., Prestbo, E. M., Risch, M. R., Johnson, D., Narayan, J., Zsolway, R., Holsen, T. M., Miller, E. K., Castro, M. S., Graydon, J. A., Louis, V. L. S., and Dalziel, J.: Estimation of speciated and total mercury dry deposition at monitoring locations in eastern and central North America, *Atmos. Chem. Phys.*, 12, 4327–4340, <https://doi.org/10.5194/acp-12-4327-2012>, 2012.
- 740 Zhang, L., Brook, J. R., and Vet, R.: A revised parameterization for gaseous dry deposition in air-quality models, *Atmos. Chem. Phys.*, 3, 2067–2082, <https://doi.org/10.5194/acp-3-2067-2003>, 2003.
- Zhang, L. and He, Z.: Technical Note: An empirical algorithm estimating dry deposition velocity of fine, coarse, and giant particles, *Atmos. Chem. Phys.*, 14, 3729–3737, <https://doi.org/10.5194/acp-14-3729-2014>, 2014.
- Zhang, L., Moran, M. D., Makar, P. A., Brook, J. R., and Gong, S.: Modelling gaseous dry deposition in AURAMS: a unified regional air-quality modelling system, *Atmos. Environ.*, 36, 537–560, [https://doi.org/10.1016/S1352-2310\(01\)00447-2](https://doi.org/10.1016/S1352-2310(01)00447-2), 2002.
- 745 Zhang, L., Wright, L. P., and Blanchard, P.: A review of current knowledge concerning dry deposition of atmospheric mercury, *Atmos. Environ.*, 43, 5853–5864, <https://doi.org/10.1016/j.atmosenv.2009.08.019>, 2009.
- Zhang, L., Wu, Z., Cheng, I., Wright, L. P., Olson, M. L., Gay, D. A., Risch, M. R., Brooks, S., Castro, M. S., Conley, G. D., Edgerton, E. S., Holsen, T. M., Luke, W., Tordon, R., and Weiss-Penzias, P.: The estimated six-year mercury dry deposition across North America, *Environ. Sci. Technol.*, 50, 12864–12873, <https://doi.org/10.1021/acs.est.6b04276>, 2016.
- 750 Zhang, L., Zhang, G., Zhou, P., and Zhao, Y.: A review of dry deposition schemes for speciated atmospheric mercury, *Bull. Environ. Contam. Toxicol.*, 110, 16, <https://doi.org/10.1007/s00128-022-03641-0>, 2023.
- Zhang, L., Zhou, H., Zhang, W., Zhou, H., Zhang, G., Zhou, P., Wang, Z., Wang, Q., and Zhao, Y.: Intercomparison of methods for atmospheric reactive mercury observations: evidences to interpret what we are actually measuring, *Atmos. Environ.*, 333, 120637, <https://doi.org/10.1016/j.atmosenv.2024.120637>, 2024.



- Zhang, L., Zhou, P., Cao, S., and Zhao, Y.: Atmospheric mercury deposition over the land surfaces and the associated uncertainties in observations and simulations: a critical review, *Atmos. Chem. Phys.*, 19, 15587–15608, <https://doi.org/10.5194/acp-19-15587-2019>, 2019.
- 760 Zhang, L., Zhou, P., Zhong, H., Zhao, Y., Dai, L., Wang, Q., Xi, M., Lu, Y., and Wang, Y.: Quantifying the impacts of anthropogenic and natural perturbations on gaseous elemental mercury (GEM) at a suburban site in eastern China using generalized additive models, *Atmos. Environ.*, 247, 118181, <https://doi.org/10.1016/j.atmosenv.2020.118181>, 2021.
- Zhang, Y., Song, Z., Huang, S., Zhang, P., Peng, Y., Wu, P., Gu, J., Dutkiewicz, S., Zhang, H., Wu, S., Wang, F., Chen, L., Wang, S., and Li, P.: Global health effects of future atmospheric mercury emissions, *Nat. Commun.*, 12, 3035, <https://doi.org/10.1038/s41467-021-23391-7>, 2021.
- 765 Zhou, H., Zhou, C., Hopke, P. K., and Holsen, T. M.: Mercury wet deposition and speciated mercury air concentrations at rural and urban sites across New York state: temporal patterns, sources and scavenging coefficients, *Sci. Total Environ.*, 637–638, 943–953, <https://doi.org/10.1016/j.scitotenv.2018.05.047>, 2018.
- Zhu, J., Wang, T., Talbot, R., Mao, H., Yang, X., Fu, C., Sun, J., Zhuang, B., Li, S., Han, Y., and Xie, M.: Characteristics of atmospheric mercury deposition and size-fractionated particulate mercury in urban Nanjing, China, *Atmos. Chem. Phys.*, 14, 2233–2244, <https://doi.org/10.5194/acp-14-2233-2014>, 2014.
- 770 Zhu, W., Lin, C. J., Wang, X., Sommar, J., Fu, X., and Feng, X.: Global observations and modeling of atmosphere-surface exchange of elemental mercury: a critical review, *Atmos. Chem. Phys.*, 16, 4451–4480, <https://doi.org/10.5194/acp-16-4451-2016>, 2016.