Mercury in the Free Troposphere and Bidirectional Atmosphere-Vegetation Exchanges – Insights from Maïdo Mountain Observatory in the Southern Hemisphere Tropics

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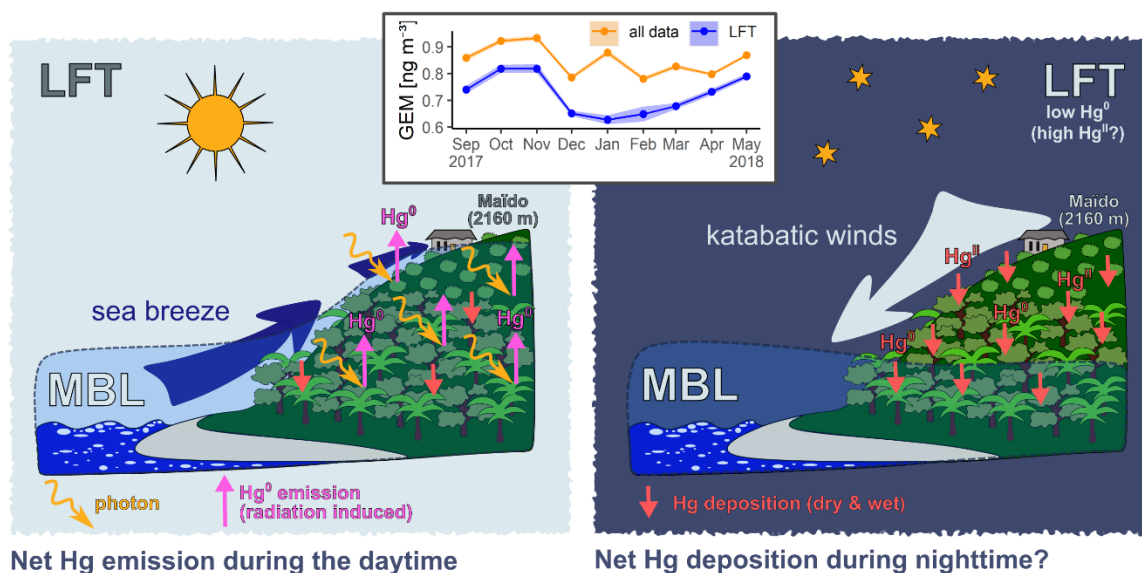
Abstract

Atmospheric mercury (Hg) observations in the lower free troposphere (LFT) can give important insights into Hg redox chemistry and can help constrain Hg background concentrations on a regional level. Relatively continuous sampling of LFT air, inaccessible to most ground-based stations, can be achieved at high-altitude observatories. However, such high-altitude observatories are rare, especially in the Southern Hemisphere (SH), and atmospheric Hg in the SH LFT is unconstrained. To fill this gap, we continuously measured gaseous elemental mercury (GEM; hourly) and reactive mercury (RM; integrated over ~6-14 days) for 9 months at Maïdo mountain observatory (2160 masl) on remote Réunion Island (21.1°S, 55.5°E) in the tropical Indian Ocean. GEM exhibits a marked diurnal variation characterized by a midday peak (mean: 0.95 ng m^{-3}; SD: 0.08 ng m^{-3}) and a nighttime low (mean: 0.78 ng m^{-3}; SD: 0.11 ng m^{-3}). We find that this diurnal variation is likely driven by the interplay of important GEM photo-reemission from the islands' vegetated surfaces (i.e. vegetation + soil) during daylight hours (8 – 22 ng m^{-2} h^{-1}), boundary layer influences during the day, and predominant LFT influences at night. We estimate GEM in the LFT based on nighttime observations in particularly dry airmasses and find a notable seasonal variation, with LFT GEM being lowest from December to March (mean 0.66 ng m^{-3}; SD: 0.07 ng m^{-3}) and highest from September to November (mean: 0.79 ng m^{-3}; SD: 0.09 ng m^{-3}). Such a clear GEM seasonality contrasts the weak seasonal variation reported for the SH marine boundary layer, but goes in line with modeling results, highlighting the added

value of continuous Hg observations in the LFT. Maïdo RM is 10.6 pg m^{-3} (SD: 5.9 pg m^{-3}) on average, but RM in the cloud-free LFT might be about twice as high, as weekly-biweekly sampled RM observations are likely diluted by low-RM contributions from the boundary layer and clouds.

Graphical abstract

1   Introduction

Atmospheric mercury (Hg) observations are unequally distributed over the globe in several ways. On the one hand, many more sampling sites exist in the Northern Hemisphere (NH) than in the Southern Hemisphere (SH) (Sprovieri et al., 2016). On the other hand, most Hg observations in either hemisphere are made at ground level and within the boundary layer. In this lowermost layer of the atmosphere, Hg concentrations are importantly affected by atmosphere-surface interactions such as local emissions and dry deposition. However, much of the long-range transport of Hg, which leads to its global distribution, does not occur within the relatively shallow boundary layer, but in the free troposphere where winds tend to be strongest and transport tends to be fastest (Travnikov, 2011). The fate of Hg in the free troposphere, detached from direct surface influences, depends strongly on chemical transformations (Travnikov, 2011) because divalent oxidized mercury (Hg^{II}), water-soluble and readily incorporated into water droplets and adsorbed onto particles, is removed from the atmosphere much more quickly than poorly soluble elemental mercury (Hg^{0}) (Schroeder and Munthe, 1998; Ariya et al., 2015; Lindberg et al., 2007).

Hg redox chemistry in the atmosphere is still subject to considerable uncertainties, and the debate on the dominant Hg oxidants in the atmosphere has been ongoing for more than two decades (Lin, 2011; Ariya et al., 2008; Lindberg et al., 2007; Dibble et al., 2020; Calvert and Lindberg, 2005; Lindqvist and Rodhe, 1985). In many studies, it was assumed that atmospheric ozone (O$_3$) and hydroxyl radicals (OH) act as dominant Hg oxidants (Lin, 2011). However, thermodynamic considerations and quantum chemistry calculations showed that the homogeneous and direct oxidation of Hg^{0} to Hg^{II} via either O$_3$ or OH is likely insignificant in the real atmosphere (Calvert and Lindberg, 2005; Dibble et al., 2020). Besides, the detection of Hg^{0} depletion events in polar regions (Schroeder et

al., 1998) led to the increasing consideration of halogens, mainly bromine (Br) radicals, as important Hg oxidants. Indeed, the widely-used mercury simulation of the GEOS-Chem chemical transport model (Selin et al., 2007) employed in recent versions a two-step Br-initiated pathway as the main Hg^0 oxidation pathway (Horowitz et al., 2017; Feinberg et al., 2022). This reaction scheme was recently updated by Shah et al. (2021), who somewhat reconciled earlier studies by introducing, among others, a two-step OH-initiated oxidation pathway alongside the abovementioned Br-induced pathway, and by introducing O_3 as a second-stage oxidant for both the Br- and OH-initiated oxidation pathways.

Despite these important new developments, Hg redox chemistry remains insufficiently constrained by observations. As Hg concentrations in the free troposphere are less sensitive to direct surface-atmosphere interactions than in the boundary layer, it could be argued that Hg observations in the free troposphere are especially valuable for constraining Hg redox chemistry. However, such observations are rare, especially in the SH. Apart from aircraft campaigns, mountain observatories are currently the only practical way of measuring Hg in the free troposphere (Travnikov, 2011). While these observatories are still subject to surface influences and often exhibit a complex variability in the origin of sampled air masses due to orographic and thermal flows (Forrer et al., 2000), they are usually able to sample air from the lower free troposphere (LFT) with regularity, especially during stable atmospheric conditions and at night (Reidmiller et al., 2010; Kleissl et al., 2007; Hahn et al., 1992; Baray et al., 2013; Collaud Coen et al., 2011). Atmospheric Hg observations from NH mountain sites such as Pic du Midi in France (Fu et al., 2016a, c; Marusczak et al., 2017), Mauna Loa in Hawaii (Carbone et al., 2016; Luippold et al., 2020), Jungfraujoch in Switzerland (Denzler et al., 2017), Mount Bachelor (Weisspenzias et al., 2007; Swartzendruber et al., 2006) in the US, Storm Peak (Obrist et al., 2008; Faïn et al., 2009) in the US, and Lulin in Taiwan (Nguyen et al., 2019, 2022; Sheu et al., 2010), among others, have provided important insights into transport and chemistry of atmospheric Hg. Meanwhile, to our best knowledge, mountain-top observations of atmospheric Hg in the SH have until now only been reported from the Chacaltaya observatory in the tropical Bolivian Andes (Koenig et al., 2021).

To fill data gaps in the SH in general and the SH LFT more specifically, we continuously measured from September 2017 to May 2018 (9 months) atmospheric Hg at Maïdo mountain observatory (2160 masl), a high-altitude regional GAW station on Réunion Island (21.1°S, 55.4°E) in the tropical Indian Ocean (Baray et al., 2013). Atmospheric Hg was sampled in the form of gaseous elemental mercury (GEM; atmospheric Hg^0) with a high time resolution (every 15 minutes), and in the form of reactive mercury (RM; atmospheric Hg^{II}) with a lower time resolution (integrated over ~6-14 days). Our measurement period overlapped with the OCTAVE project ("Oxygenated Compounds in the Tropical Atmosphere: Variability and Exchanges", http://octave.aeronomie.be), dedicated to the study of oxygenated organic compounds in tropical regions (Rocco et al., 2020). This is further complemented by regular and continuous observations of atmospheric trace gases such as carbon dioxide (CO_2), methane (CH_4), carbon monoxide (CO), and ozone (O_3) at the observatory (Baray et al., 2013; Zhou et al., 2018; Duflot et al., 2019).

Here, we 1) give an overview of the 9 months of continuous GEM and RM observations at Maïdo observatory in the tropical Indian Ocean. We 2) derive and discuss a time series of GEM in the LFT and estimate RM in the LFT. Finally, with the help of ancillary data and the FLEXPART-AROME lagrangian dispersion model, we 3) explore

possible drivers for remarkably pronounced GEM diurnal cycles at Maïdo and address the potential role of GEM

105   photo-reemission from the island's vegetated surface.

2   Methods

2.1   Site description

110   Maïdo observatory (21.0792°S, 55.38°E) lies at an altitude of 2160 masl on the remote Réunion Island in the

tropical Indian Ocean, about 700 km east of Madagascar (see Figure 1a). Réunion is a relatively small island

(~2512 km^2) with a complex orography due to its volcanic origin (Gillot and Nativel, 1989). The volcano "Piton

de la Fournaise" to the east of the island is still very active, with on average one eruption every 7 - 10 months

(Stieltjes and Moutou, 1989; Villeneuve and Bachèlery, 2006). The island, especially its mountain slopes, is

115   densely covered by evergreen forest, while its beach strips are highly populated (population density for 2022: 358

inhabitants per km^2). Maïdo observatory is directly surrounded by shrublands, while lush tropical vegetation is

found further to the northeast, in the Cirque de Mafate (a densely vegetated volcanic caldera). To the west of

Maïdo lies a high Tamarisk forest, which also dominates the coastal region where the closest urban areas of Saint

Paul and Le Port (105,000 and 40,000 inhabitants, 13 and 15 km from the observatory, respectively) are located

120   (Rose et al., 2019).

The local atmospheric circulation of Réunion Island is complex and a result of both orographic and thermal flows

(Lesouëf et al., 2011; Foucart et al., 2018). Orographic flows are induced by the island's rugged relief, which

represents an environmental obstacle to the trade winds (east-south-easterlies on average). Upon encountering

Réunion Island, these air masses can either rise (orographic lifting regime) or they can flow around the island, in

125   which case an overturning loop can be observed in the northwest of the island (Foucart et al., 2018). Thermal

flows, which are driven by differential heating and cooling of the islands' surface, follow an important diurnal

cyclicity (see Figure 1b). Nocturnal radiative surface cooling creates a cold downwash (or katabatic wind) along

the slopes of the island (Baray et al., 2013), which usually leads to cloudless nights in the mountain regions. After

sunrise, radiative heating typically generates a sea breeze circulation and upslope winds (or anabatic winds), which

130   are accompanied by cumulus clouds (Lesouëf et al., 2011).

Maïdo observatory samples mostly air from the LFT during nighttime due to the abovementioned katabatic winds

that develop after sunset and mostly manifest as easterlies (Figure 1). During the daytime, the observatory is

importantly influenced by the planetary boundary layer (PBL) of the island as well as the marine boundary layer

(MBL) of the surrounding ocean, brought to the observatory by a sea breeze circulation and anabatic winds, which

135   usually manifest as westerlies (Lesouëf et al., 2011). When the daytime sea breeze weakens on the west coast,

moist air masses can also originate from the nearby Cirque de Mafate to the northeast, or get advected from the

windward (eastern) side of the island by strong south-easterly trade winds (Lesouëf et al., 2011, 2013; Tulet et al.,

2017).

While atmospheric transport to Réunion Island on the mesoscale is dominated by south-easterly trade winds

140   (Foucart et al., 2018, see also supplementary Sect. S1), transport pathways can change under the influence of

tropical cyclones developing over the South-West Indian Ocean (Tulet et al., 2021; Pohl et al., 2016), mostly from

November to April (the cyclonic season). Several cyclonic storms affected Réunion Island during our measurement period (September 2017 – May 2018), with the most noteworthy storms being Ava (~13 days, 27.12.2017 – 09.01.2018), Berguitta (~11 days, 09.01.2018 – 20.01.2018), Dumazile (~6 days, 01.03.2018 - 06.03.2018),

145 Eliakim (~7 days, 13.03.2018 – 20.03.2018) and Fakir (~5 days, 20.04.2018 – 25.04.2018) (http://www.meteo.fr/temps/domtom/La_Reunion/webcmrs9.0/anglais/index.html, last access: 10/08/2022).

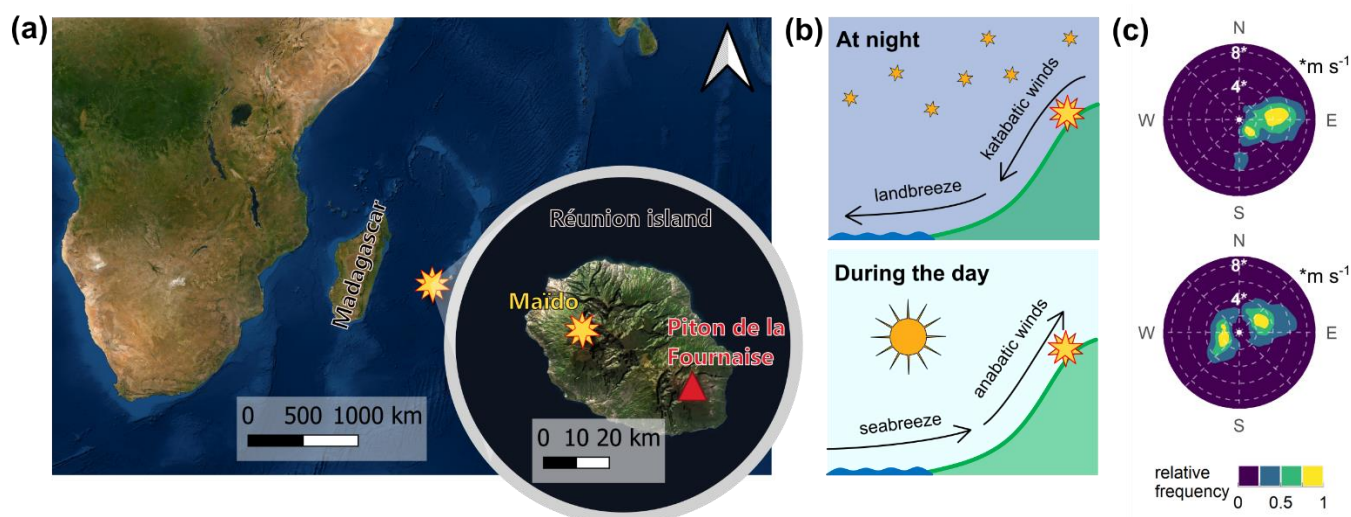



Figure 1. Site overview

150 **(a) The location of Réunion Island in the tropical Indian Ocean, and the location of Maïdo observatory (star) on Réunion Island (zoomed in image). The rugged orography of the island was represented by overlaying a digital elevation model (Jarvis et al., 2008) over an aerial image (obtained from ESRI).**

(b) Schema of simplified mesoscale circulation patterns affecting Maïdo (star), characterized by katabatic winds and mostly lower free tropospheric influences during the night (top), and by anabatic winds and boundary layer

155 **influences during the day (bottom).**

(c) Wind speed and wind direction at Maïdo (observations from July 2017 to July 2018) show important day-night differences (compare to b), with mostly fast easterly katabatic winds during nighttime (top), and a competition between easterlies and westerlies during daytime (bottom). The radial axis gives the wind speed (see dashed circles). The numbers give wind speed in m s⁻¹. The color scale is normalized, so that "1" corresponds to the most frequent

160 **combination of wind direction and wind speed.**

2.2 GEM and RM observations

Between September 2017 and June 2018, gaseous elemental mercury (GEM) was continuously measured from the instrumented platform at Maïdo Observatory using two Tekran® 2537 Model 2537A analyzers (Tekran Inc.,

165 Toronto, Canada), operating under Global Mercury Observation System (GMOS) standard operating procedures (Munthe et al., 2011; D'Amore et al., 2015). The ¼" unheated PFA sampling line was protected from UV radiation with a white opaque tube (polyvinyl chloride) to avoid photochemical reactions inside the line. The inlet, installed

2 m high on the stationary instrumented platform (6 m agl), was connected to a single PTFE filter-holder, composed of one 0.45 µm PES (polyethersulfone) filter (47 mm diameter). This analytical configuration has been

170 set up to measure GEM instead of total gaseous mercury (TGM), as well as to collect RM compounds for further analysis.

GEM then passed through the 9.5 m long sampling line and a second single PTFE filter-holder, composed of one 0.45 µm PTFE filter (47 mm diameter), into the Tekran® 2537, where it was collected on gold traps, before thermal desorption and quantification by cold vapor atomic fluorescence spectroscopy (CV-AFS). GEM was analyzed

175 every 15 min at a flow rate of 1.3 L min^{-1} (STP). The instrument detection limit is 0.1 ng m^{-3}. GEM concentrations are expressed in ng m^{-3} under standard conditions of 273.14 K and 1013.25 hPa. Calibration of the Tekran® 2537 was performed using both the internal Hg permeation sources (every week) and manual injections of saturated Hg vapor (every 3 months). The flow rate in the Tekran® 2537 as well as the internal mass flow meter itself was also frequently checked by an external and calibrated mass-flow meter. The Tekran® 2537 deployed at Maïdo

180 observatory experienced less than 5% and 1% shifts in the manual injection checking and mass-flow meter calibration, respectively. The room in which the instrument was placed was permanently air-conditioned to 22 - 23°C, thus allowing it to be permanently 2 °C to 3 °C above the outside temperatures, whatever the season, to avoid condensation in the sampling line. Raw GEM time series were quality controlled according to the guidelines proposed within the GMOS network, using a dedicated software developed at the Institute of Environmental

185 Geosciences in 2012 (see https://gmos.aeris.data.fr, last access: 25 May 2022). During this automated procedure, the raw dataset is compared against potential flags corresponding to more than 40 criteria that specifically refer to all operation phases related to the calculation of Hg concentrations and calibration (D'Amore et al., 2015), thus marking GEM readings as "valid", "warning", or "invalid". The quality-insured and quality-controlled dataset was then generated from the site manager under consideration of the previously flagged dataset, field notes,

190 logbooks, and site characteristics (Magand and Dommergue, 2022).

RM was sampled with a significantly lower frequency than GEM, with collection times varying from 5.8 to 14.1 days to allow for the concentration of enough RM mass on the dedicated 0.45 µm PES filter. It has been shown that PES membranes can collect RM (gaseous oxidized mercury + particle-bound mercury) quantitatively, similar to cation exchange membranes (Dunham-Cheatham et al., 2020; Gustin et al., 2015). After collection in the field,

195 PES filters were separately stored in Petri dishes, inside double-zipper bags, and kept frozen at -20 °C until analysis. After shipment to the laboratory, each PES filter was placed in a PTFE beaker. PES filters were digested in 16 mL of ERi 2.5% inverse aqua regia (solution of 97.5% vol H2O, 1.7% vol HNO3 [78% vol] and 0.8% vol HCl [83% vol]). The beakers were closed and placed on a heating plate (120 °C) for 12 hours before analysis, which was done with a Brooks Rand Model III cold vapor atomic fluorescence spectrometry detector (CVAFS).

200 The method detection limit was 5 pg Hg (Marusczak et al., 2017) and the average LOD was around 35 pg for the entire sampling and analytical process. Standard Reference Material was NIST 3133 and ORMC-5 from NRC. The standard measurement procedure included the CVAFS calibration (0 - 50 pg; NIST 3133) and a quality control sequence for 12 samples. The performance of the CVAFS was assessed based on ORMC-5, showing concentrations between 89 and 120 % of the expected value. The volume of air collected on PES filters was

205 calculated from the Tekran® 2537 flow rate at the inlet. RM concentrations are expressed in pg m^{-3} under standard conditions of 273.14 K and 1013.25 hPa.

2.3 Volatile organic compound (VOC) observations

We make use of VOC observations to characterize the origin of sampled air masses and the extent to which they were impacted by surface influences. This data was generated in the framework of the OCTAVE project (http://octave.aeronomie.be; last accessed 25 May 2022), which aimed to better understand the transport and role of VOCs in tropical regions (Verreyken et al., 2019, 2020, 2021; Rocco et al., 2020).

VOCs were measured by two high-sensitive proton-transfer-reaction quadrupole mass spectrometers (hs PTR-Quad-MS; Ionicon Analytik GmbH, Innsbruck, Austria) belonging to the Royal Belgian Institute for Space Aeronomy (BIRA-IASB) and the Laboratoire des Sciences du Climat et de l'Environnement (LSCE). The BIRA PTR-MS was deployed at Maïdo in multiple ion detection mode to generate a near-continuous 2-year data set of VOCs (from October, 19th, 2017 to November, 26th, 2019) (Verreyken et al., 2021). The LSCE PTR-MS measurements were recorded during the intensive field OCTAVE campaign at three different locations (Maïdo observatory, Le port coastal urban city, and the tropical Bélouve forest) (Rocco et al., 2020). Principal recorded VOCs were acetaldehyde (CH_3CHO), acetone (CH_3COCH_3), methanol (CH_3OH), formic acid (HCOOH), acetic acid (CH_3COOH), acetonitrile (CH_3CN), benzene (C_6H_6), toluene (C_7H_8), isoprene (C_5H_8), monoterpenes ($C_{10}H_{16}$), methyl vinyl ketone (MVK), methyl ethyl ketone (MEK) and dimethyl sulfide (DMS), a volatile organic sulfur compound. More technical and analytical information can be found in Verreyken et al. (2021, 2020) and Rocco et al. (2020).

2.4 Meteorological observations

We use observations on meteorological parameters (Temperature, relative humidity, wind speed, wind direction, and incoming solar radiation) as continuously taken at the Piton-Maïdo meteorological station, at ~2150 masl and around ~ 1 km from Maïdo Observatory. This meteorological station is permanently checked and validated by the French national meteorological service (Meteo France) and consists of a pyrocontrol probe, a Vaisala HMP110 humidity probe, an ultrasonic GILL WS2 sensor, and a K&Z CM5 pyranometer. Specific humidity was calculated from temperature and relative humidity observations but assuming a constant atmospheric pressure corresponding to the sampling altitude (2150 masl $\rightarrow$ 780 hPa).

2.5 Other ancillary data

Maïdo observatory is registered as an ICOS (Integrated Carbon Observation System), NDACC (Network for the Detection of Atmospheric Composition Change), and ACTRIS (Aerosols, Clouds and Trace gasses Research Infrastructure) atmospheric measurement site, and a regional GAW (Global Atmospheric Watch) station (WMO region I, Africa). In this framework, the atmospheric observatory continuously houses a suite of both in-situ and remote sensing instruments from which a list of continuous measurements can be found online (https://osur.univ-reunion.fr/observations/osu-r-stations/opar/, last access: 26 May 2022). Among them, greenhouse gases (CO, CO_2, CH_4) were specifically useful in the context of the present study.

Greenhouse gas measurements were performed with a PICARRO G2401 analyzer and following standardized ICOS protocols for measurement, processing, calibration, and quality control, described in detail elsewhere (Hazan

et al., 2016; Laurent, 2017; Heiskanen et al., 2022; Yver-Kwok et al., 2021). The analyzer was calibrated once a month with four cylinders of reference gases prepared and calibrated by the Flask and Calibration Laboratory (FCL) of ICOS in accordance with the WMO reference scales (CO2-X2019, CH4-X2004A, CO-X2014A). Two other reference gases were used for the quality control of the measurements at a rate of a daily injection for the short-term target gas, and one injection per month for the long-term target gas.

Ozone (O_3) was measured with a UV photometric Thermo Scientific 49i analyzer with a detection limit of 1 ppb and a time resolution of 1 min. This instrument operates on the principle that O_3 molecules absorb UV light at a wavelength of 254 nm, and that the degree to which the UV light is absorbed is directly related to the O_3 concentration as described by the Beer-Lambert Law (Swinehart, 1962). Calibration was carried out every three months with an O_3 generator, validated by the French air quality monitoring agency.

2.6 FLEXPART-AROME transport modelling and Source-Receptor-Relationships (SRRs)

Here we use the model results obtained by Verreyken et al. (2021) concerning mesoscale air mass transport to Maïdo. Briefly, mesoscale transport to Maïdo was estimated with the help of FLEXPART-AROME (Verreyken et al., 2019; Brioude et al., 2013), which feeds the FLEXPART lagrangian particle dispersion model (Stohl et al., 2005; Pisso et al., 2019) with meteorological input from the high resolution (horizontal: 2.5 x 2.5 km^2) AROME regional climatological model (Seity et al., 2011). AROME is used by Meteo France as the operational mesoscale numerical weather prediction model for the Indian Ocean.

From 03.11.2017 to 26.11.2019, 20 000 initial air particles were launched every hour (on the hour: 8:00, 9:00, etc.) from Maïdo observatory (more specifically: homogeneously from a 20 m deep layer above the station) and followed for 24 hours into the past. Residence times of air parcels were calculated on a 0.025° x 0.025° grid (~2.5 km horizontal resolution) within a domain spanning 19.5 °S – 22.5 °S and 53.0 °E – 58.0 °E. The grid contained 15 vertical layers, 10 layers between 0 magl and 500 magl (50 m thickness), 3 layers between 500 magl and 2000 magl (500 m thickness), and two additional layers above (10 000 magl and 24 000 magl). More detailed information on this model run can be found in Verreyken et al. (2021).

Source-Receptor-Relationships (SRRs) describe the sensitivity of observed concentrations to surface emissions from regions of interest and are thus a practical tool to estimate quantitatively to what degree observed concentrations at Maïdo observatory are impacted by emissions from the island's surface. We calculated SRRs by dividing FLEXPART-AROME-derived air mass residence times by a constant minimal boundary layer height, as described in Seibert and Frank (2004). More details can be found in supplementary Sect. S6.

2.7 Data treatment and statistical tools

All data analysis was performed with R 3.6.0 (R Core Team, 2019) and using the "tidyverse" collection of R packages (Wickham et al., 2019). Most visualizations were done with the R package "ggplot2" (Wickham, 2016), while the site overview map (Fig.1) was generated with QGIS (QGIS Development Team, 2022), and the graphical abstract was created with Inkscape (Inkscape Project, 2022). Digitization of figures was done with WebPlotDigitizer (Rohatgi, 2021).

We worked with hourly averages whenever possible. Hourly averages were defined as follows: For GEM, CO, CO_2, CH_4, O_3, and VOCs, the hourly average at hour "h" corresponds to all data taken between "h" and "h+1", e.g. the hourly average for 9 AM corresponds to all observations between 9:00 and 9:59.

Relative humidity (RH) and temperature (T) measurements as obtained from Meteo France are reported at the round hour only. To make this as congruent as possible with the abovementioned hourly averaging of trace gases and VOCs, we assigned the average of the two reported values at "h" and "h+1" to measurements taken at hour "h". Continuing with the above example, the hourly average of RH (or T) for 9 AM corresponds to the average of the two RH (or T) observations reported at 9:00 and 10:00.

The used FLEXPART dispersion runs (see section 2.6) were launched instantaneously at every hour on the hour (e.g. 8:00, 9:00, 10:00). Here, we assigned to in-situ measurements taken between "h" and "h+1" the FLEXPART dispersion run arriving at "h+1", e.g., we assigned to the hourly average of 9 AM, which represents all data taken between 9:00 and 9:59:59, the backward dispersion run launched at 10:00. The same applies for all FLEXPART-derived SRRs.

3 Results and discussion
3.1 Overview: Hg observations at Maïdo
3.1.1 Gaseous Elemental Mercury (GEM)

For the 9 months of available observations (September 2017 – May 2018), mean GEM at Maïdo (see Figure 2a) was 0.85 ng m^{-3} (SD: 0.12 ng m^{-3}). This is quite low in comparison to reported atmospheric Hg concentrations at low-altitude SH background sites such as Amsterdam Island in the southern Indian Ocean (55 masl; GEM mean: ~1.05 ng m^{-3}; Angot et al., 2014; Slemr et al., 2015, 2020), Cape Point in South Africa (230 masl; ~1.0 ng m^{-3}; Slemr et al., 2015, 2020) and Darwin in northern Australia (25 masl; ~0.95 ng m^{-3}; Howard et al., 2017), but comparable to reported GEM concentrations from the more elevated Bariloche Argentina (800 masl; ~0.86 ng m^{-3}, Diéguez et al., 2019) and the high-altitude Chacaltaya observatory in Bolivia (5240 masl; total gaseous mercury: ~0.89 ng m^{-3} during the ENSO-neutral year 2014-2015, Koenig et al., 2021). Based on all data taken, no clear seasonality emerges in GEM at Maïdo, apart from somewhat increased concentrations between September and November (SON mean: 0.91 ng m^{-3}, SD: 0.12 ng m^{-3}). Higher GEM in SON might be related to the increased occurrence of biomass burning events in the SH from August to November (Edwards et al., 2006). In fact, it has been shown that biomass-burning-influenced air masses can get transported to Maïdo observatory, especially those originating from Africa and Madagascar (Verreyken et al., 2020).

GEM showed a marked diurnal variation, with a minimum at night (23:00 to 5:59 local time (LT); mean: 0.78 ng m^{-3}; SD: 0.11 ng m^{-3}), rising concentrations after dawn, and a peak around noon (from 12:00-13:59 LT; mean: 0.95 ng m^{-3}; SD: 0.08 ng m^{-3}), after which concentrations decrease again (Figure 2b). This contrasts with the relatively weak diurnal variation (diurnal range < 0.05 ng m^{-3}) at marine or coastal sites in the SH such as Amsterdam Island and Cape Point (Slemr et al., 2020; Angot et al., 2014; Slemr et al., 2015). On the other hand, Maïdo GEM diurnal variation with its midday peak is comparable to the summertime diurnal GEM variation at Dumont d'Urville and Concordia Station in Antarctica, where diurnal GEM variability has been attributed to

photo-reemission of GEM from the snowpack (Song et al., 2018; Angot et al., 2016a, b). The possible relationship between diurnal GEM cycles at Maïdo and photo-reemission is explored in section 3.3.

3.1.2 Reactive Mercury (RM)

Mean RM at Maïdo is quite low (10.6 pg m^{-3}; SD: 5.9; 35 samples in total) compared to reported mean RM concentrations at other mountain observatories (range: ~20 - 133 pg m^{-3}; Fu et al., 2016a; Marusczak et al., 2016; Nguyen et al., 2021; Swartzendruber et al., 2006; Luippold et al., 2020; Faïn et al., 2009). It must be noted that all these other observatories lie in the NH, and most of them measured RM (in the form of GOM and PBM separately) with a Tekran® speciation unit. The potassium chloride (KCl) denuder used in the Tekran® speciation unit has been proven to not collect all RM species quantitatively, causing RM measurements in ambient air to be biased low (Gustin et al., 2019, 2021; McClure et al., 2014; Lyman et al., 2010). Denuder-based RM observations at Pic du Midi mountain observatory in France were later corrected upwards by 30% (mean from 40 pg m^{-3} to 52 pg m^{-3}) after comparison to a PES-membrane-based measurement protocol near-identical to ours (Marusczak et al., 2017), which has been shown to sample RM more quantitatively (Gustin et al., 2021; Dunham-Cheatham et al., 2020). The difference between RM concentrations at Maïdo and these other mountain observatories may thus be even larger if the likely low bias of the earlier denuder-based measurements is accounted for.

RM at Maïdo showed a more significant seasonal variation than GEM, with 14.9 pg m^{-3} (SD: 6.5) for September – November (SON), 8.2 pg m^{-3} (SD: 5.0) for December – March (DJFM), and 8.8 pg m^{-3} (SD: 3.1) for April – June (AMJ) (Figure 2c,d). These seasonal differences are statistically significant between SON and DJFM and between SON and AMJ ($p < 0.05$, Mann-Whitney U test), but statistically insignificant ($p = 0.3$) between DJFM and AMJ. Individual low RM observations (< 5 pg m^{-3}) may relate to elevated RM wet deposition, especially in DJFM, the wet season, which is characterized by elevated rainfall (Figure 2c) and increased relative humidity at Maïdo (mean: 86% in DJFM vs. 70% for the rest of the year).

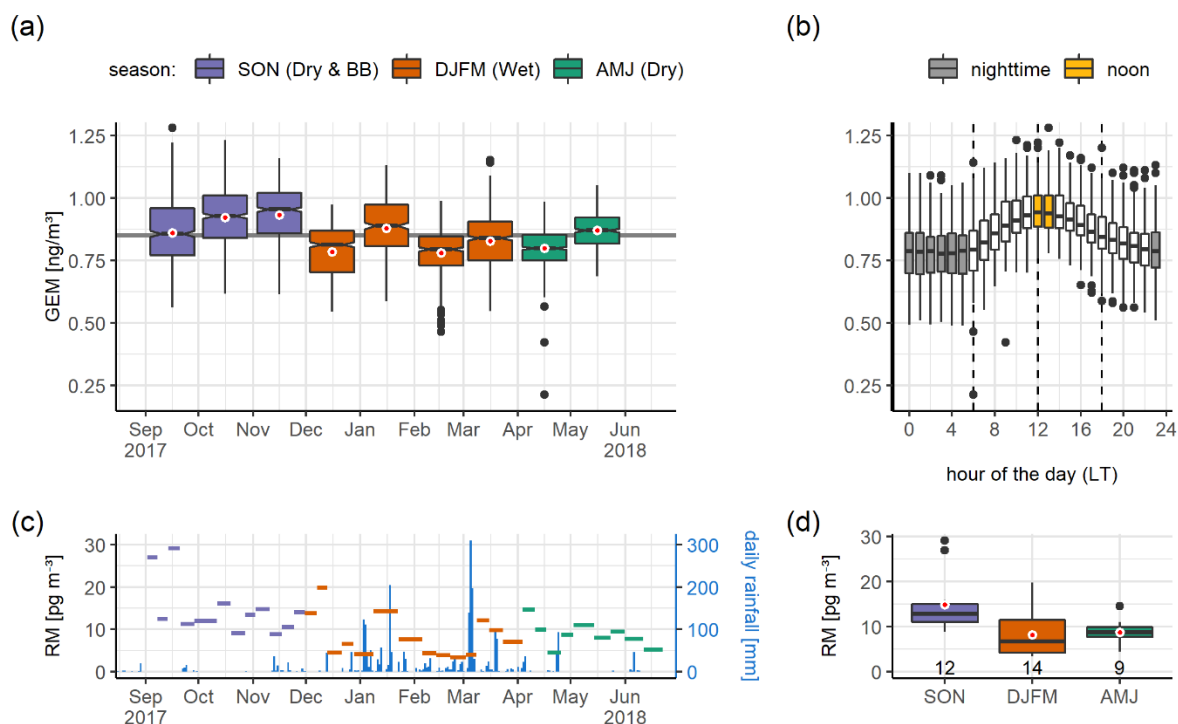



Figure 2. Overview of Hg observations

a) Overview of 9 months of hourly GEM measurements at Maïdo. Red dots show arithmetic means. The notches show the 95% confidence interval for the median. The grey horizontal line gives the arithmetic mean of the whole dataset, which is near-identical to the median (0.85 ng m^{-3} in both cases).

b) Diurnal GEM variation at Maïdo, characterized by a marked nighttime (23:00 – 5:59 local time (LT)) minimum and a noon (12:00 – 13:59 LT) peak.

c) Overview of RM measurements at Maïdo (a total of 35 weekly or biweekly samples). The length of each bar corresponds to the integration time of the respective RM observation. Total daily precipitation at Maïdo is given as blue bars (see right axis). Around 82% of all precipitation during the 9-month measurement period fell between 01.12.2017 and 31.03.2018 (DJFM, corresponding to the wet season).

d) Seasonal variation in RM sampled at Maïdo. Red dots show arithmetic means. The numbers indicate the number of available RM samples for the respective season.

3.2 Hg in the lower free troposphere (LFT) of the Southern Hemisphere (SH) tropical Indian Ocean
3.2.1 LFT GEM time series

A major goal of measuring Hg at Maïdo was getting insight into Hg in the LFT of the SH tropical Indian Ocean. While Maïdo is influenced by the boundary layer (BL) during the daytime, the observatory samples mostly air from the LFT during nighttime (see site description in section 2.1).

That said, we observed that nighttime observations at Maïdo are not always representative of the LFT, which is consistent with previous research at Maïdo observatory (Guilpart et al., 2017). Specifically, when humidity remained high at night (specific humidity > ~10 g kg^{-1}, RH > ~90%), nighttime observations at Maïdo most likely

365 remained importantly BL influenced. This appears to occur notably during cyclonic storms, during which the diurnal variability of GEM and specific humidity nearly disappears (see supplementary Sect. S2).

To estimate GEM in the LFT, we thus selected nighttime data and additionally removed all GEM observations sampled in air masses with a specific humidity higher than a seasonally variable threshold. As threshold, we used the monthly median nighttime specific humidity at coordinates for Maïdo and 800 hPa (~1950 masl) as obtained
370 from the ERA5 global reanalysis (Hersbach et al., 2020) (see supplementary Sect. S3).

Our estimate of GEM in the LFT (Figure 3; see data table in supplementary Sect. S4) shows a significant seasonal variation during the 9 months of observations, with remarkably low mean GEM concentrations of 0.66 ng m^{-3} (SD: 0.07 ng m^{-3}) in DJFM, corresponding to the wet season or austral summer. For each of the 9 months, monthly averaged LFT GEM was considerably lower (0.11 ng m^{-3} lower on average) than unfiltered Maïdo
375 GEM (i.e.: all data), with the difference being most pronounced in DJFM (0.25 ng m^{-3} lower in the LFT), likely due to the increased occurrence of cyclonic storms during that time of the year. Maïdo LFT GEM seasonality is nearly inverse to the TGM seasonality observed at the Chacaltaya mountain-top observatory (5240 masl) in the tropical Andes, to our knowledge the only other observatory in the SH where strongly free troposphere-influenced air is sampled with regularity (Aliaga et al., 2021) and Hg observations are available (Koenig et al.,
380 2021). This difference suggests that the seasonal variation of Maïdo LFT GEM is likely driven by other processes than TGM (GEM + RM) seasonality at Chacaltaya, which has been mainly attributed to biomass burning, vegetation uptake, and interhemispheric exchange in the upper troposphere (Koenig et al., 2021, 2022).

Based on the same filtering procedure as for LFT GEM, we estimated LFT CO, CH$_4$, and O$_3$ (see Figure 3), although no O$_3$ observations were available from September to December 2017. In contrast to GEM, the
385 application of the LFT filter had little effect on CO and O$_3$ concentrations, indicating that concentration differences between BL and LFT are much less pronounced for these compounds than for GEM. The strong CO enhancement between September and November is likely due to increased biomass burning in the SH during that time of the year (Edwards et al., 2006; Callewaert et al., 2022). While it is likely that LFT GEM concentrations in SON are biomass burning-influenced, the seasonal variations of LFT GEM and LFT CO are clearly different, especially
390 after February 2018 when CO remains low while GEM increases, again suggesting that the seasonal variation of Maïdo LFT GEM is likely dominated by other drivers than biomass burning. Seasonal variation in transport pathways and long-range transport of anthropogenic emissions could conceivably impact Maïdo LFT GEM seasonality. While long-range transport pathways were very similar for SON (Sept. – Nov. 2017) and AMJ (Apr. – Jun. 2018), some differences could be seen in DJFM (Dec. 2017 – Mar. 2018), during which a fraction of
395 northerly air masses even originated north of the equator (within the last 10 days before arrival at Maïdo; see supplementary Sect.1). However, given that Hg concentrations tend to be significantly higher in the NH than in the SH (Horowitz et al., 2017; Shah et al., 2021), it appears unlikely that the low LFT GEM concentrations in DJFM are linked to these northerly air masses. Intriguingly, the seasonality of LFT GEM, with its minimum between December and March, is similar to the seasonality of LFT CH$_4$, which, in the remote tropical Indian
400 Ocean, is mainly driven by oxidation loss through reaction with OH (Khalil and Rasmussen, 1983). This could indicate that OH-related Hg redox chemistry (see introduction) plays an important role in driving LFT GEM seasonality.

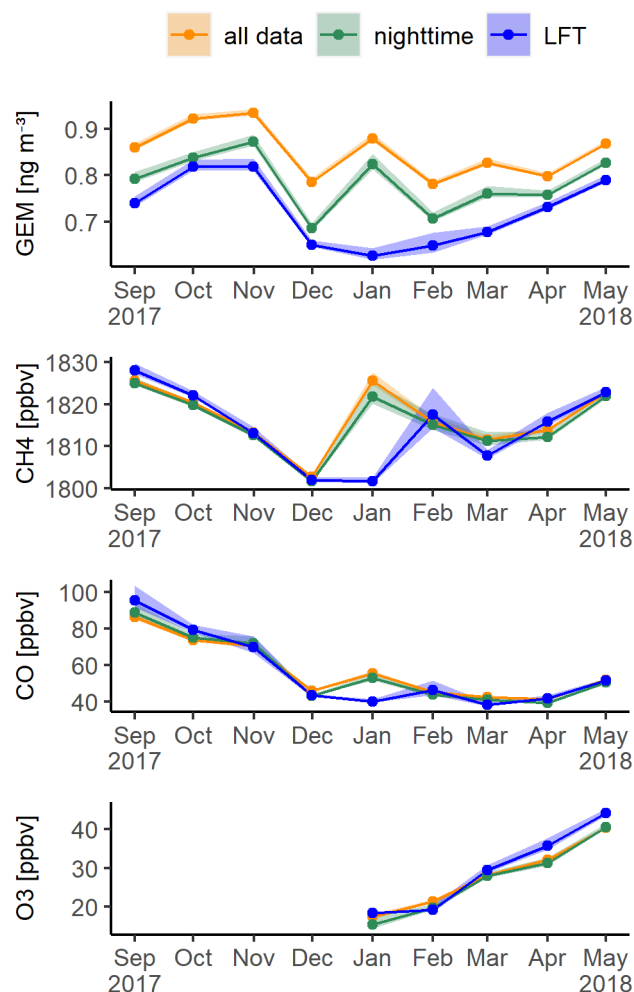



Figure 3. Estimated concentrations of atmospheric GEM, CH₄, CO, and O₃ in the lower free troposphere (LFT) of the SH tropical Indian Ocean, as inferred from dry nighttime air masses arriving at Maïdo. LFT time series (blue lines) were obtained by selecting only nighttime data (23:00 LT – 5:59 LT, green lines) from all observations (orange lines), followed by the application of a specific humidity filter. The shaded area encloses the mean ± 2 times the standard error.

3.2.2 Maïdo LFT GEM seasonality in the context of atmospheric Hg models

GEOS-Chem modeling results predict an important seasonality of atmospheric Hg in both the NH and the SH (Horowitz et al., 2017; Shah et al., 2021; Feinberg et al., 2022). In contrast, while such a Hg seasonality has been observed in the NH, only a weak Hg seasonality has been observed at SH background sites such as Amsterdam Island and Cape Point (Slemr et al., 2015, 2020). It has been proposed that atmospheric Hg seasonality is less pronounced at SH monitoring stations as a consequence of the lower land cover in the SH (19%) compared to the NH (39%) and consequently a lesser importance of vegetation GEM uptake as a driver for Hg seasonality (Feinberg et al., 2022; Jiskra et al., 2018; Zhang and Zhang, 2022).

Adding another piece to this puzzle, we observed an important seasonal variation of GEM at Maïdo, but only after isolating the LFT from BL influences (Figure 3, section 3.2.1). This suggests that Hg seasonality, seemingly weak in the SH BL, might be much more pronounced in the SH free troposphere.

To illustrate how this LFT seasonal variation compares to modeled Hg seasonality in the SH, and how it contrasts with the reported lack of observed Hg seasonality in the BL at other SH background sites, we digitized the GEOS-Chem modeling results from Horowitz et al. (2017) and Shah et al. (2021), which differ importantly in the used atmospheric Hg redox chemistry schemes (see introduction). As can be seen in Figure 4, Maïdo LFT GEM is lower than atmospheric Hg observed at other SH background sites (Amsterdam Island and Cape Point; note that at these sites TGM ~ GEM), which itself is notably lower than modeled Hg in Shah et al. (2021). Maïdo LFT GEM is also somewhat lower than modeled Hg in Horowitz et al. (2017). As for seasonality, the minimum of Maïdo LFT GEM lies somewhere in between the modeled Hg minimum of Horowitz et al. (2017) and Shah et al. (2021), albeit somewhat closer to Horowitz et al. (2017).

Important caveats to the comparison with GEOS-Chem results have to be mentioned: 1) Both Horowitz et al. (2017) and Shah et al. (2021) evaluated Hg seasonality not for the LFT but for the lowest model layer (~ 0 – 60 magl), which lies entirely within the BL, 2) Modeled Hg corresponds to different time periods than Maïdo GEM observations, and 3) the digitized modeled Hg seasonalities correspond to model results evaluated for the geographical coordinates of Amsterdam Island (37.8 °S, 77.57 °E), Cape Point (34.35 °S, 18.48 °E), and Bariloche (41.13 °S, 71.42 °W; only in the case of Shah et al., 2021), not Maïdo (~21.08 °S, 55.38 °E).

The goal of this comparison is thus not to evaluate which chemistry scheme performs better, but to illustrate the added benefit from relatively continuous Hg observations in the free troposphere. Modeled seasonality of atmospheric Hg is evidently sensitive to the used chemistry scheme (Figure 4). In consequence, if Hg seasonality is observed in the free troposphere, as in the case of Maïdo, it could be used to evaluate and improve our understanding of atmospheric Hg chemistry. Inversely, the observed discrepancies between modeled and observed seasonalities in the SH MBL may indicate a model misrepresentation of more BL-specific processes, such as surface emissions, deposition velocities, or more BL-specific Hg chemistry.

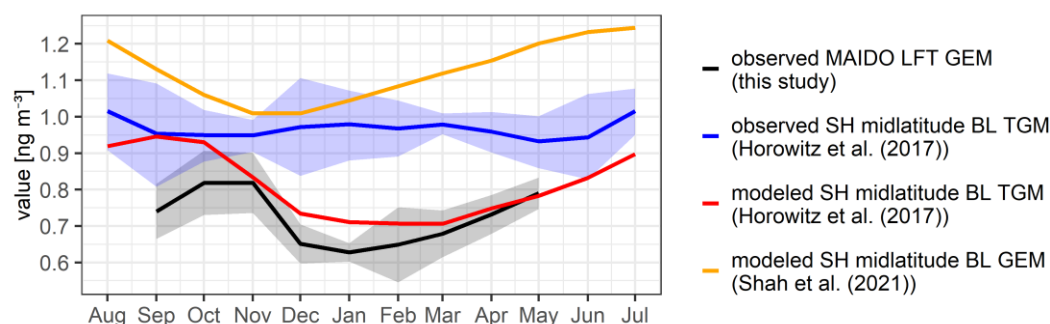



Figure 4. Comparison between observed and modelled Hg seasonalities in the SH. The abbreviations LFT and BL correspond to "lower free troposphere" and "boundary layer", respectively. Observed TGM seasonality as digitized from Horowitz et al. (2017) corresponds to observations from Amsterdam Island and Cape Point. The shaded areas give the mean ± 1 standard deviation.

3.2.3 An upper bound on RM concentrations in the LFT

While LFT and BL influences on Maïdo vary diurnally (see site description in section 2.1), our RM observations correspond to integration times of ~6-14 days. In consequence, RM concentrations reported here correspond to a mix of air masses from the LFT, the PBL, and the MBL. Given the low time resolution of our RM observations, it was not possible to isolate RM in the LFT by only considering dry nighttime air masses, as we did for hourly sampled GEM (see section 3.2.1).

We hence used a different approach to constrain RM in the LFT: We combined FLEXPART-AROME results with ERA5 reanalysis data (Hersbach et al., 2020) to estimate, on a seasonal basis, the fraction of sampled air masses coming from the PBL, the MBL, the cloud-free LFT, and from clouds (see supplementary Sect. S5). We then estimated RM in the (cloud-free) LFT with the mixing equation below (Equation 1). Clouds were addressed explicitly because of the elevated water solubility of atmospheric RM, which can be efficiently scavenged into cloud droplets and rain (Nair et al., 2013).

$$RM_{LFT(cloud-free)} = \frac{RM_{observed} - f_{MBL} * RM_{MBL} - f_{PBL} * RM_{PBL} - f_{clouds} * RM_{clouds}}{f_{LFT(cloud-free)}} \qquad \text{Equation 1}$$

Where f_{MBL}, f_{PBL}, $f_{LFT(cloud-free)}$, and f_{clouds} are the estimated fractions of air coming from the MBL, PBL, cloud-free LFT and clouds, respectively.

To estimate RM in the cloud-free LFT ($RM_{LFT(cloud-free)}$), assumptions about mean RM concentrations in the MBL (RM_{MBL}), the PBL (RM_{PBL}), and clouds (RM_{clouds}), all not known from Maïdo observations alone, had to be made. As for the RM_{MBL}, recent observations at Amsterdam Island (~2700 km south-east of Maïdo) with the same measurement protocol as ours (see methods) have revealed MBL RM concentrations of ~4 pg m^{-3} (Jeroen Sonke, Beatriz F. Araujo, Olivier Magand, Aurelien Dommergue, personal communication, unpublished data). However, higher RM concentrations of ~13 pg m^{-3} have recently been derived for MBL air arriving at the Australian coast (~9200 km east of Maïdo), based on observations with cation-exchange membranes (Miller et al., 2021). Here, we only aim to estimate the upper bound of RM in the (cloud-free) LFT at Maïdo, which is obtained when the lower bound of concentrations is assumed for RM_{MBL}, RM_{PBL} and RM_{clouds} (see Equation 1). With this goal in mind, we used 0 pg m^{-3} for RM_{clouds}, assuming complete scavenging of RM in clouds, and we used 4 pg m^{-3} for both RM_{PBL} and RM_{MBL}, based on observations at Amsterdam Island.

Our results suggest that RM in the cloud-free LFT might be about twice as high as the overall average at Maïdo, i.e., up to 30 pg m^{-3} (SD: 7) in SON, 14 pg m^{-3} (SD: 10) in DJFM, and 15 pg m^{-3} (SD: 6) in AMJ, with an average of 20 pg m^{-3} (SD: 13) over the whole measurement period. It is notable that even this upper limit is still at the lower end of RM concentrations reported from NH mountain sites (~20 – 133 pg m^{-3}, see section 3.1.2). This might be partly explained by the difference in GEM concentrations between Maïdo (mean 0.85 ng m^{-3}) and these NH measurement sites (mean ~1.5 ng m^{-3}), considering that the chemical production rate of RM in the atmosphere depends directly on GEM concentrations. However, any interpretation has to be approached with care, as our estimate of RM in the LFT is subject to important uncertainties arising from the low time resolution of our RM observations, the use of FLEXPART-AROME to estimate air mass mixing (see supplementary Sect. S5), and several assumptions.

3.3 Exploring GEM diurnal variation

3.3.1 Conditions leading to marked GEM diurnal cycles

As described above, GEM at Maïdo exhibits a marked diurnal variation, with a minimum at night when the observatory samples mostly air from the LFT, rising concentrations after dawn, and a peak around noon, after which concentrations decrease again (Figure 2b).

To investigate possible drivers for this diurnal variation and to determine which conditions affect the amplitude of GEM diurnal cycles, we grouped days into 1) days with a strong GEM diurnal variation (Group 1: difference between noon and nighttime > 0.16 ng m^{-3}; 55 days in total), and 2) days with a weak diurnal GEM variation (Group 2: difference between noon and nighttime < 0.08 ng m^{-3}; 21 days in total). Days belonging to neither of these two groups were excluded from this analysis.

As can be seen in Figure 5, Group 1 diurnal cycles are characterized by remarkably low mean GEM concentrations at night (arithmetic mean: 0.72 ng m^{-3} between 23:00 LT and 5:59 LT, SD: 0.09 ng m^{-3}) and, by definition, by a large GEM difference between nighttime and noon (0.26 ng m^{-3} difference on average). On the other hand, Group 2 diurnal cycles are characterized by comparatively high GEM concentrations at night (on average 0.87 ng m^{-3}, SD: 0.11 ng m^{-3}) and, by definition, by a low GEM difference between nighttime and noon (0.04 ng m^{-3} difference on average).

We find large differences in meteorological conditions corresponding to the two groups. While marked GEM diurnal cycles (Group 1) are associated with mostly sunny days and dry nights, atypically weak diurnal cycles (Group 2) are associated with cloudy days and humid nights (Figure 5). This suggests that Maïdo receives predominantly LFT air at night for days assigned to Group 1, while this is not the case for days assigned to Group 2.

For data belonging to Group 1, mean diurnal GEM variation anticorrelates significantly (Pearson: r = - 0.98, p << 0.01) with the diurnal variation of carbon dioxide (CO$_2$; see Figure 5), a long-lived (Archer et al., 2009) greenhouse gas importantly taken up by vegetation, especially during daytime (Black and Clanton, 1973). Indeed, diurnal CO$_2$ variation at Maïdo is mostly driven by vegetation gas exchange (Callewaert et al., 2022). Group 1 mean diurnal GEM variation also correlates significantly with the diurnal variation of isoprene (r = 0.95, p << 0.01), a short-lived (~1 hour during daytime) VOC mostly emitted by terrestrial vegetation under sunlight and heat stress (Guenther et al., 1993; Pacifico et al., 2009). At Maïdo, observed isoprene mainly originates from the vegetated mountain slopes and the densely vegetated "Cirque de Mafate" (see site description in section 2.1) relatively close to the observatory (Verreyken et al., 2021). These strong relationships suggest that diurnal cycles of GEM may be related to similar drivers and regions of influence as the diurnal cycles of isoprene and CO$_2$. In other words, GEM diurnal variation appears to be linked to the island's vegetated surface under sunlight.

Finally, Group 1 GEM diurnal variation correlates (r = 0.87, p << 0.01) with that of dimethyl sulfide (DMS), a relatively short-lived (~1 - 2 days; Chen et al., 2018; Kloster et al., 2006) VOC that is mostly emitted by marine phytoplankton (Stefels et al., 2007) and is frequently used as a tracer for marine air masses. Even though DMS concentrations at Maïdo are relatively low, the diurnal variation of Maïdo DMS is most likely related to marine

influences (Verreyken et al., 2021). It is noteworthy that GEM concentrations increase immediately after sunrise (~6 LT – 7 LT) and quickly decrease in the afternoon, while DMS concentrations start rising later in the day (at around 9 LT), and do not decline until the evening (~18 LT, Figure 5). This suggests that diurnal GEM variation is not in phase with the diurnal variation of MBL influences.

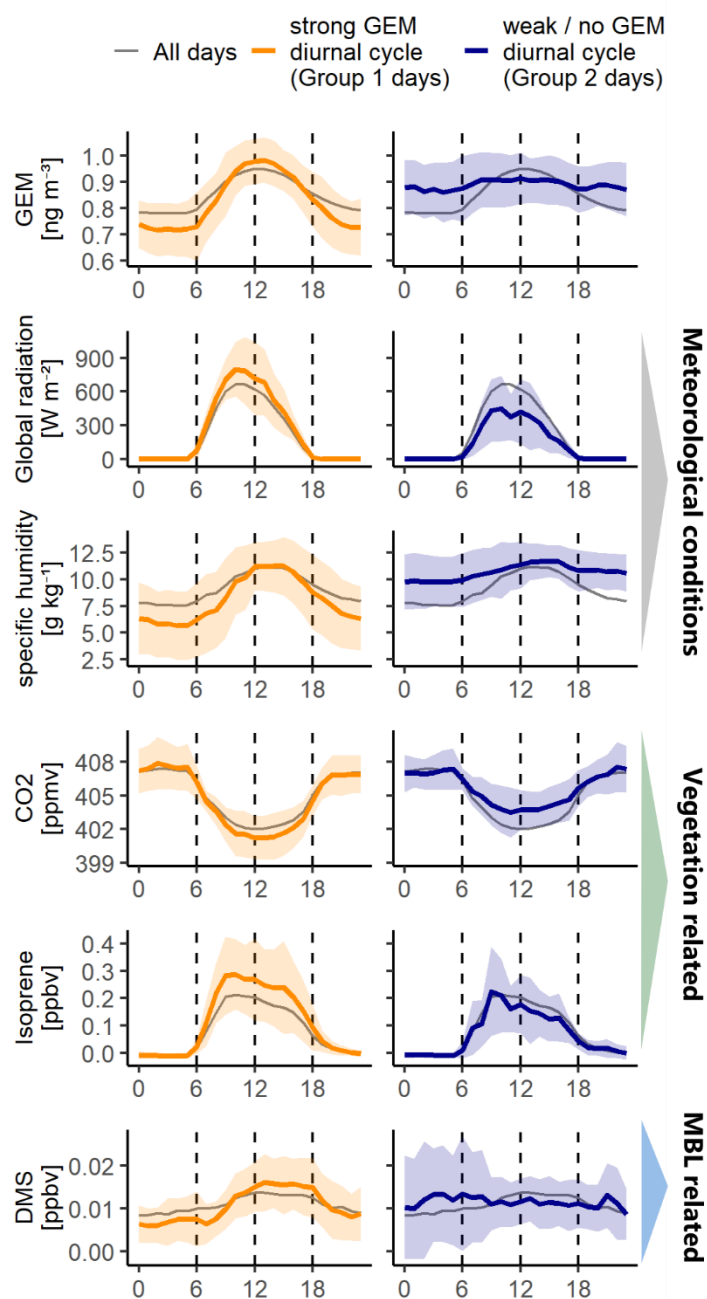



Figure 5. Mean diurnal variation (arithmetic average) of GEM, solar radiation, specific humidity, CO₂, isoprene, and DMS. Days have been grouped into two main groups in function of the amplitude of diurnal GEM variation (the difference between nighttime and noon). The left column corresponds to all data on days with an especially marked GEM diurnal variation (Group 1), the right column corresponds to all data on days with no or only a weak diurnal GEM variation (Group 2). The thin grey line shows the average diurnal cycle for all days (for which GEM observations are also available). The shaded area corresponds to the mean ± 1 standard deviation.

3.3.2 Constraining the role of mixing processes

Above, we showed that very pronounced diurnal GEM cycles (Group 1) are related to the sampling of LFT air at night and to a strong diurnal variation of DMS, which suggests an important diurnal variation in the fraction of air coming from the MBL.

Considering the reported importance of mixing processes at Maïdo (Lesouëf et al., 2013; Guilpart et al., 2017), we first explore the hypothesis that GEM diurnal cycles are purely driven by a diurnal variation in the sampled mix between LFT and MBL air masses. In this baseline hypothesis, we assume that there are no influences from the island's surface, even though the abovementioned similarities between GEM, CO_2, and isoprene diurnal variations suggest otherwise (see Figure 5).

To test this hypothesis, we built a two-box mixing model where we assume that Maïdo GEM depends only on GEM concentrations in the MBL, GEM concentrations in the LFT, as well as the diurnal variation in the mixing between LFT and MBL air (see supplementary Sect. S6). We estimated the latter with the help of FLEXPART-AROME (see supplementary Sect. S5). For simplicity, we only focus on those days that show a strong diurnal GEM variation (Group 1 in Figure 5; 55 days in total, of which FLEXPART output is available for 40), which generally correspond to sunny days with few clouds and low nighttime humidity. This choice allows us to minimize the difficult-to-parametrize effect of clouds and provides confidence that air masses sampled at night come indeed predominantly from the LFT (see section 3.2.1).

To represent LFT GEM concentrations in the two-box mixing model, we use the LFT GEM time series derived in section 3.2.1 (see also Figure 3). MBL GEM concentrations at Réunion Island are more difficult to constrain from Maïdo observations alone, as pure MBL air is rarely sampled. We thus assumed MBL GEM concentrations to be within $0.9 - 1.1$ ng m^{-3} for the two-box mixing model, based on previously reported GEM concentrations in the SH MBL (Slemr et al., 2015, 2020). Considering the reported absence of clear GEM diurnal variation and the weak seasonality in the MBL of the SH (Slemr et al., 2015), we also assumed MBL concentrations to be constant throughout the day and year.

While the two-box mixing model suggests an important influence of mixing processes on GEM diurnal variation, we find that the modeled and observed diurnal GEM variations do not agree in timing (Figure 6). A variation in mixing between LFT and MBL air as the sole driver of GEM diurnal variation cannot explain the early morning rise in GEM and would place the GEM peak in the afternoon, around 2 - 4 hours later than observed. It must be said that the modeled diurnal GEM variation in the two-box mixing model depends directly on the FLEXPART-AROME-based estimate of the mixing between LFT air and MBL air, which could conceivably be biased. To exclude such a bias, we compared the estimated LFT/MBL mixing to the observed diurnal DMS variation as a

proxy for marine influences, finding that diurnally varying MBL influences appear to be captured adequately (see
supplementary Sect. S5).

We thus consider that mixing processes between MBL and LFT air, even though likely an important contributor,
cannot adequately explain GEM diurnal variability alone. In the following section, we explore the potential role
of the island's vegetated surface under sunlight.

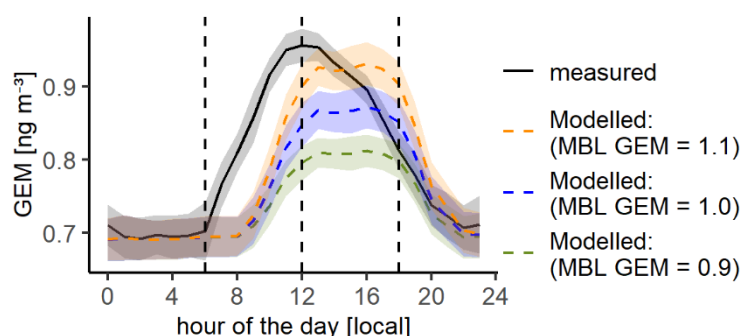



**Figure 6. Assessment of the role of mixing between LFT and MBL as a driver for diurnal GEM cycles. The solid line
shows the observed mean diurnal GEM cycle for those days characterized by an especially marked diurnal GEM
variation (Group 1). The dashed lines give the corresponding modelled diurnal GEM cycles if only the diurnal
variation in the sampled mix of LFT and MBL air is considered, under the assumption of no surface influence from
the island. Modeled diurnal cycles based on three different MBL concentrations (1.1 ng m^{-3}, 1.0 ng m^{-3} and 0.9 ng m^{-3})
are shown. Shaded areas give the arithmetic mean ± 2 times the standard error.**

3.3.3 Radiation-driven surface emissions as potential driver for GEM diurnal variation

We found that diurnal GEM variation at Maïdo depends on solar radiation, and that marked diurnal cycles of GEM
at Maïdo relate importantly to the diurnal cycles of isoprene and CO_2, which have been attributed to the island's
vegetated surface. This suggests that GEM diurnal cycles are related to vegetated surfaces under sunlight.

Previous work has shown that solar radiation, especially in the ultraviolet range, can cause photo-reemission of
Hg from surfaces, such as snow, and lead to diurnal GEM cycles characterized by a midday peak (Song et al.,
2018; Angot et al., 2016a). While terrestrial vegetation is globally a net sink of atmospheric mercury (Zhou and
Obrist, 2021), fluxes between vegetation and the atmosphere are bidirectional (Agnan et al., 2016; Luo et al.,
2016), and re-emission also occurs from vegetated surfaces and soils (Yuan et al., 2019; Yu et al., 2020; Converse
et al., 2010; Osterwalder et al., 2017). Not only RM deposited on leaf surfaces can get photo-reduced and re-
emitted as GEM, but also Hg from within the leaf tissue (Yuan et al., 2019).

Considering all this, we propose the hypothesis that Maïdo GEM diurnal variation is, in addition to mixing between
LFT and MBL air, driven by net daytime photo re-emission of GEM from the island's vegetated surfaces (i.e.
vegetation + soil), especially from the vegetated mountain slopes close to the observatory.

We addressed this hypothesis by including a term expressing the (net) photo-reemission of GEM from vegetated surfaces into the mixing model from the previous section. We parametrized the impact of photo-reemission on observed GEM as the product of 1) the FLEXPART-AROME-derived SRRs between Maïdo observatory and vegetated surfaces, 2) total solar radiation as measured at Maïdo, and 3) a constant radiation-dependent surface emission term (see supplementary Sect. S6 for details). As in the previous section concerning the influence of mixing processes, and for the same reasons, we only focus on those days that exhibit a strong diurnal cycle (Group 1 in Figure 5). While we assume direct proportionality between net GEM reemission and solar radiation (see supplementary Sect. S6), the magnitude of this relationship (in other words: the slope) is not known. We estimate this slope in an inverse modeling approach, i.e., we determine the most likely slope by computing the model for a wide range of values and evaluating the root mean square error (RMSE) between modeled and observed mean diurnal GEM variation.

We find that (Group 1) diurnal GEM cycles could be well explained (RMSE = 0.022 ng m^{-3}) by net surface GEM emission of 0.032 ng m^{-2} h^{-1} per watt (more precisely W m^{-2}) of incoming total solar radiation (Figure 7), corresponding to a net emission of 21 ng m^{-2} h^{-1} at the mean diurnal solar radiation maximum at Maïdo (~660 W m^{-2}), and to an average net emission flux of 13.5 ng m^{-2} h^{-1} during hours of daylight. Extrapolated to the whole year, this flux would imply a net daytime surface emission of ~59 µg m^{-2} y^{-1}. Our flux estimates depend on a variety of parameters used for the inverse modeling, such as the assumed MBL GEM concentration, and the used parametrizations to estimate air mass mixing (see supplementary Sect. S5) and SRRs (see supplementary Sect. S6). To assess uncertainties, we recalculated in a Monte Carlo approach (Janssen, 2013; Metropolis and Ulam, 1949) the average net daytime emission flux for a wide range of parameters, obtaining a 95% confidence interval of 8 – 22 ng m^{-2} h^{-1} (see supplementary Sect. S7). It should be noted that our flux estimate corresponds to the full ecosystem, i.e. the sum of vegetation and soil fluxes, with their relative contributions being unknown. In addition, our estimate assumes that GEM concentrations at Maïdo are not strongly affected by anthropogenic emissions downslope, similar to what has been reported for CO and CO$_2$ (mean anthropogenic contribution at noon < 7 ppbv and < 0.2 ppm, respectively; Callewaert et al., 2022).

To our knowledge, no previous studies have derived differentiated day- and nighttime GEM fluxes for mostly pristine tropical mountain forests or shrublands comparable to those on Réunion Island. Observed daytime Hg emission fluxes from background sites in other terrestrial environments (i.e. predominantly low-altitude and extra-tropical) were generally below ~ 3.5 ng m^{-2} h^{-1} (median ~ 0.8 ng m^{-2} h^{-1}), significantly lower than found here (Agnan et al., 2016). Mean daytime fluxes of up to ~6 ng m^{-2} h^{-1} (depending on the season) have been reported for temperate mountain meadows in the US (Converse et al., 2010) and Tibet (Sun et al., 2020). Daytime fluxes above ~10 ng m^{-2} h^{-1}, similar to what we derived here, were observed in some tropical environments, e.g. for an open field soil in Amazonia (Almeida et al., 2009), and a naturally preserved but anthropogenically influenced (TGM > 5 ng m^{-3}) forest soil (soil Hg: ~0.13 mg$_{Hg}$kg$_{soil}^{-1}$) in tropical China (Fu et al., 2012). It is well-reported that daytime emissions from terrestrial surfaces correlate with soil Hg concentrations and solar radiation (Agnan et al., 2016). Soil Hg concentrations on Réunion Island are high (median: 0.16 mg kg^{-1}; range: 0.03-0.81 mg kg^{-1}; Dœlsch et al., 2006) compared to European and other tropical topsoils (median ~0.02 and ~0.06 mg kg^{-1}, respectively; Panagos et al., 2021; Lim et al., 2020). This, alongside the strong incident solar radiation around Maïdo (see Fig.5), might explain the large daytime emission flux found here. In any case, given that studies in directly comparable

environments are scarce, it is not yet possible to say whether a large daytime emission flux is specific to mountain forests and shrublands on Réunion Island, or characteristic of tropical mountain environments in general.

Our results suggest that vegetated surfaces around and downslope of Maïdo are a net source of GEM to the air during hours of daylight, but not necessarily during the entire 24-hour period. Maïdo observatory is mostly insensitive to surface-atmosphere interactions during the night when sampled air masses come predominantly from the LFT. Even though little can be said about nighttime atmosphere-surface fluxes based on Maïdo observations alone, it is likely that net Hg deposition occurs at night to vegetated surfaces around and downslope of Maïdo to sustain net daytime GEM emission. It is thought that vegetation GEM uptake happens mostly through plant stomata (Zhou and Obrist, 2021), whose opening varies as a function of daytime, heat stress, and evaporative loss (Roelfsema and Hedrich, 2005). While stomata tend to open most widely during the day, they often remain at least partly open during the night, especially in the case of tropical vegetation (Wang et al., 2021). This could allow for nighttime stomatal GEM uptake, in addition to possible non-stomatal uptake pathways (Converse et al., 2010). In fact, significant nighttime GEM uptake by vegetation has been reported before (Kurz et al., 2020; Fu et al., 2016b; Jiskra et al., 2018; Yu et al., 2018). Adding to GEM dry deposition, katabatic winds could bring comparatively high RM concentrations in the LFT (see section 3.2.3) to the mountain vegetation around and downslope of Maïdo and lead to elevated nighttime RM deposition on leaf surfaces, which could then be readily reduced and reemitted at daytime under the strong tropical sun.

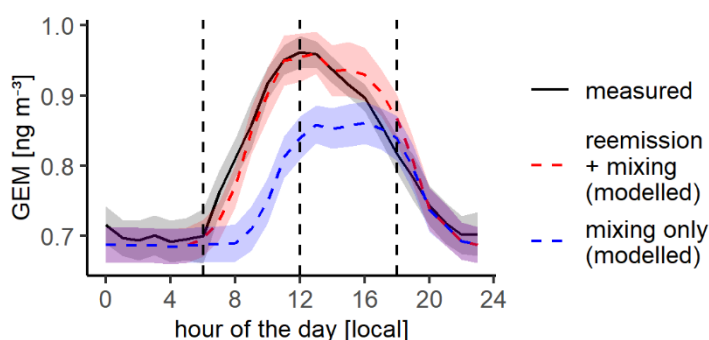



Figure 7. Assessment of the potential role of surface GEM reemission as a driver for GEM diurnal cycles. The solid line shows the observed mean diurnal GEM cycle for those days characterized by an especially marked diurnal GEM variation (Group 1). The dashed lines give the corresponding modelled mean diurnal GEM cycles if (1) only mixing processes between MBL and LFT are considered (dashed blue line; assumed MBL GEM = 1.0 ng m^{-3}) and (2) parametrized photo-reemission from the island's vegetated surface is added to the mixing model (dashed red line; assumed surface flux is a result of inverse modelling). Shaded areas show the arithmetic mean ± 2 times the standard error.

4   Conclusions & perspectives

We presented 9 months of observations of GEM (sampled every 15 minutes) and RM (integrated over ~ 6 - 14 days) at Maïdo mountain observatory (2160 masl) on Réunion Island in the Southern Hemisphere (SH) tropical

Indian Ocean. Due to mesoscale circulation characteristics, Maïdo is influenced by the boundary layer (BL) during the daytime, but samples mostly air from the lower free troposphere (LFT) during nighttime. Based on nighttime observations in especially dry airmasses, we estimated GEM in the LFT of the tropical Indian Ocean to be, on average, significantly lower than unfiltered Maïdo GEM observations (0.74 ng m^{-3} vs. 0.85 ng m^{-3}, respectively). Maïdo LFT GEM showed an important seasonal variation, with the highest concentrations from October to November 2017 and lowest concentrations from December 2017 to March 2018, corresponding to austral summer. Such a marked seasonal variation stands in contrast to the reported weak Hg seasonality at background sites in the SH marine boundary layer (MBL) but is congruent with the significant and photochemistry-dependent Hg seasonality reported in modeling studies. This suggests that Hg observations in the SH LFT may be particularly useful to constrain Hg chemistry and that continuous Hg monitoring at SH mountain sites could prove highly beneficial for the community.

Mean RM at Maïdo, sampled on polyether sulfone (PES) membranes, was ~10.6 pg m^{-3}, significantly lower than reported RM concentrations for mountain sites in the NH. While we estimate that RM in the (cloud-free) LFT at Maïdo may be about twice as high (~20 pg/m^3), we were limited by the low time resolution of RM observations (sample integration time of ~ 6 - 14 days). This limitation did not allow us to capture the likely significant diurnal RM variation at Maïdo and to rigorously separate the LFT from boundary layer influences. In future studies on mountain observatories, it would be advisable to measure RM with a higher time resolution that permits resolving diurnal variations, or at least capturing day- and nighttime differences in RM concentrations.

GEM at Maïdo exhibits marked diurnal cycling, with a nighttime minimum and a noon maximum. GEM diurnal cycling is significantly more pronounced on sunny days than on cloudy days and disappears altogether during large-scale cyclonic storms. Marked GEM diurnal cycles are significantly correlated with isoprene, emitted from vegetation under sunlight, and anti-correlated with CO_2, which is taken up by vegetation during photosynthesis. This suggests net GEM emission from the island's mostly vegetated surface during the day. GEM diurnal cycles at Maïdo could be well explained by significant GEM photo-reemission from the island's vegetated surfaces (i.e. vegetation + soil) during daylight hours, combined with important MBL influences in the afternoon. Through inverse modeling, we estimated that vegetated surfaces around and downslope of Maïdo could emit on average 13.5 ng m^{-2} h^{-1} (95% CI: 8 – 22 ng m^{-2} h^{-1}) of GEM during daylight hours. To maintain this net daytime emission, significant net nighttime Hg deposition from the atmosphere to these vegetated surfaces is to be expected, likely through deposition of both GEM and RM. While these results are subject to considerable uncertainties, they suggest important and diurnally variable bidirectional fluxes between vegetated surfaces and the atmosphere, at least for tropical evergreen mountain forests and shrublands such as those found on Réunion Island. Future measurement campaigns on Réunion Island could explore Hg concentrations and isotopic signatures below the forest canopy, in soil, in rainfall, litterfall, and throughfall, to rigorously constrain these bidirectional Hg fluxes and to further investigate the potentially important role of nighttime GEM uptake by vegetation.

Data availability:

Maïdo L2 GEM data (https://doi.org/10.25326/352) are freely available (Magand and Dommergue, 2022) at https://gmos.aeris-data.fr/ from GMOS-FR data portal coordinated by IGE (Institut des Géosciences de l'Environnement—Grenoble, France; technical PI: Olivier Magand) and maintained by the French national center for Atmospheric data and services AERIS (last access: 11 August 2022).

Monthly averaged Maïdo L2 GEM data for different data selections (all data, nighttime only, LFT) is given in supplementary Sect. S4.

Maïdo RM data is given in supplementary Sect. S8.

Maïdo (O)VOC data as acquired with a hs-PTR-MS instrument in the framework of the OCTAVE project are freely available under https://doi.org/10.18758/71021061 (Amelynck et al., 2021).

The hourly means of CO_2, CH_4, and CO concentrations, and a set of meteorological parameters, are available on the Carbon Portal (https://www.icos-cp.eu/) and under https://doi.org/10.18160/1QQG-6RP6 (De Mazière et al., 2022)

Author contributions

AMK: Formal Analysis, Methodology, Software, Validation, Visualization, Writing – original draft preparation, Writing – review & editing; **OM:** Conceptualization, Funding acquisition, Data curation, Investigation, Methodology, Project administration, Resources, Supervision, Validation, Writing – original draft preparation, Writing – review & editing; **BV:** Methodology, Resources, Writing – review & editing; **JB:** Funding acquisition, Methodology, Resources, Writing – review & editing; **CA:** Funding acquisition, Resources, Writing – review & editing; **NS:** Funding acquisition, Resources, Writing – review & editing; **AC:** Resources, Writing – review & editing; **BFA:** Data curation, Investigation, Methodology, Writing – review & editing; **MR:** Resources, Writing – review & editing; **MKS:** Resources, Writing – review & editing; **JPC:** Funding acquisition, Resources, Writing – review & editing; **JES:** Conceptualization, Data Curation, Funding acquisition, Investigation, Methodology, Resources, Supervision, Writing – original draft preparation, Writing – review & editing; **AD:** Conceptualization, Funding acquisition, Methodology, Project administration, Resources, Supervision, Writing – original draft preparation, Writing – review & editing;

Competing interests

The authors declare that they have no conflict of interest.

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