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Deforestation as an anthropogenic driver of mercury pollution

Aryeh Feinberg[a*], *Martin Jiskra*[b*], *Pasquale Borrelli*[c], *Jagannath Biswakarma*[b,d], *and Noelle E. Selin*[a,e]

[a] Institute for Data, Systems, and Society, Massachusetts Institute of Technology, Cambridge, MA 02139, USA

[b] Environmental Geosciences, University of Basel, Basel 4056, Switzerland

[c] Department of Science, Roma Tre University, Rome 00146, Italy

[d] Department of Water Resources and Drinking Water, Eawag, Dübendorf 8600, Switzerland

[e] Department of Earth, Atmospheric, and Planetary Sciences, Massachusetts Institute of Technology, Cambridge, MA 02139, USA

*Correspondence to: arifeinberg@gmail.com (A.F.); martin.jiskra@gmail.com (M.J.)

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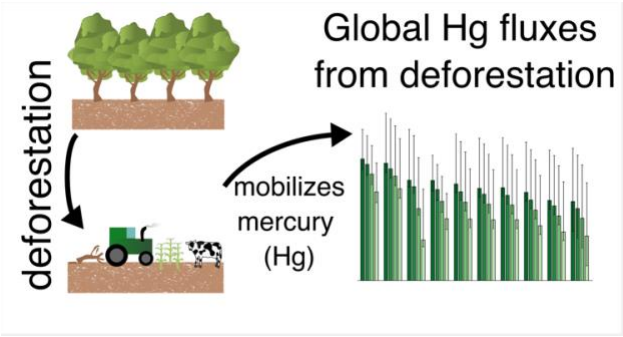
Abstract

Deforestation reduces the capacity of the terrestrial biosphere to take up the toxic pollutant mercury (Hg) and enhances the release of secondary Hg from soils. The consequences of deforestation for Hg cycling are not currently considered by anthropogenic emissions inventories or specifically addressed under the global Minamata Convention on Mercury. Using global Hg modeling constrained by field observations, we estimate that net Hg fluxes to the atmosphere due to deforestation are 217 Mg yr^{-1} (95% confidence interval, CI: 134–1650 Mg yr^{-1}) for 2015, approximately 10% of global primary anthropogenic emissions. If deforestation of the Amazon rainforest continues at business-as-usual rates, net Hg emissions from the region will increase by 153 Mg yr^{-1} by 2050 (CI: 97–418 Mg yr^{-1}), enhancing the transport and subsequent deposition of Hg to aquatic ecosystems. Substantial Hg emissions reductions are found for two potential cases of land use policies: conservation of the Amazon rainforest (92 Mg yr^{-1}, CI: 59 to 234 Mg yr^{-1}) and global reforestation (98 Mg yr^{-1}, CI: 64 to 449 Mg yr^{-1}). We conclude that deforestation-related emissions should be incorporated as an anthropogenic source in Hg inventories, and that land use policy could be leveraged to address global Hg pollution.

Synopsis

This study quantifies the impact of deforestation on the global Hg cycle, finding that deforestation increases Hg fluxes to air and water. Conservation and reforestation are important policy tools to mitigate these fluxes.

TOC Graphic



Main Text

Introduction

Humans are exposed to the organic form of mercury (Hg), methylmercury (MeHg), mainly through seafood consumption[1]. Methylmercury is a potent neurotoxin, impairing the neurodevelopment of fetuses and children and costing the global economy $20–117 billion annually according to some estimates[2,3]. Mercury is emitted to the atmosphere by: a) primary anthropogenic sources, including artisanal and small-scale gold mining (ASGM), fossil fuel combustion, and metal smelting; b) re-emissions of historical anthropogenic ("legacy") Hg from ocean and land; and c) geogenic sources[4]. Mercury spreads globally in the atmosphere due to its overall elemental lifetime against deposition of 4–6 months[5]. A global treaty, the Minamata Convention on Mercury, aims to protect human health and the environment from anthropogenic emissions and releases of Hg. The Convention targets primary anthropogenic emissions sources by phasing out Hg use and adopting best available technologies for pollution control[6]. Primary anthropogenic emissions account for only 30% of present-day total emissions, with legacy re-emissions from land and ocean accounting for 60% [7]. The future of Hg pollution will depend not only on reducing direct emissions through the Minamata Convention, but also on indirect anthropogenic influences on legacy Hg emissions and fate.

Terrestrial ecosystems, and especially forests, are important sinks of Hg from the atmosphere, taking up an estimated 2200–3600 Mg Hg per year[8], more than a third of total (anthropogenic, legacy, and geogenic) Hg emissions (7400 Mg yr^{-1})[9]. By taking up Hg, terrestrial ecosystems reduce the burden of Hg depositing in oceans and freshwater systems, where it can be more readily converted to MeHg and bioaccumulated in fish. Previous studies have drawn useful analogies between Hg and carbon cycling in terrestrial ecosystems[10,11]. Like carbon dioxide (CO_2), elemental mercury (Hg^0) is assimilated by foliage throughout the growing season[12]. Mercury is transported from the canopy to soil by foliage falling to the ground ("litterfall") and dry deposited Hg being washed off by precipitation ("throughfall"), which together are the major source (60–90%) of Hg in soils[8]. Anthropogenic land use and land cover changes (LULCC),

including deforestation, perturb both CO_2 and Hg fluxes to the atmosphere[13–15]. In the case of carbon, scientific assessments[14] have calculated the contribution of LULCC to total anthropogenic CO_2 emissions (13% of total), and land management practices are governed by Article 5 of the Paris Agreement[16]. For Hg, on the other hand, quantitative estimates of the overall importance of land cover change are limited. Only one previous study modeled the impact of future LULCC on atmospheric Hg cycling, focusing on the effects of climate-induced changes to vegetation[15]. No anthropogenic Hg emissions inventories have quantified the impacts of historical and future deforestation, and land management is not currently addressed by Hg policy efforts like the Minamata Convention.

Several processes mobilize Hg from terrestrial systems after deforestation. Along with removing a strong atmospheric sink of Hg[8], deforestation leads to more insolation reaching the soil, which increases volatilization of Hg from soils through enhanced microbial[17] or photochemical[18] reduction. Fire-mediated deforestation leads to direct emission of Hg from forest and soil biomass[19]. Soils in deforested areas are subject to accelerated erosion rates, enhancing Hg export to downstream ecosystems[17,20,21]. Direct measurement of deforestation-driven fluxes at larger scales is challenging given variations in the land sink due to trends in environmental conditions, necessitating the use of models to quantify these fluxes[22]. Models of terrestrial–atmosphere Hg fluxes, while still much more uncertain than analogous carbon cycle models, are improving due to a better process understanding and increasing availability of terrestrial measurements[8,12,23,24]. Thus, the time is ripe for assessing the relative importance of deforestation-driven fluxes in the Hg cycle.

Policies on local, national, and international scales will shape the future evolution of deforestation Hg fluxes. Deforestation due to agricultural land conversion threatens the Amazon rainforest[25,26], which currently contributes 29% of the global land sink for atmospheric Hg^0 (ref. [23]). At current deforestation rates, 40% of the Amazon rainforest could be lost by 2050, while enhanced environmental legislation (e.g., expansion of protected areas and enforcement) can reduce the deforested area to 15% (ref. [27]).

Reforestation and afforestation on the global scale are being studied as part of the solution to reach net zero greenhouse gas emissions in the future[28], though the efficacy of these measures has been debated[29]. In any case, the climate mitigation benefits of forestation would not be realized without accompanying aggressive CO_2 emissions reductions[29,30]. Similarly, forest conservation and reforestation policies may have potential benefits for Hg sequestration on land, yet the magnitude of impacts remain unquantified.

Here, we apply the GEOS-Chem Hg model[23] to calculate deforestation emission factors for Hg for different regions and evaluate them against available observations. We quantify the global atmospheric Hg fluxes in 2015 that result from deforestation. We study the impact of future Amazon deforestation policy scenarios[27] and potential global reforestation efforts[30] on the terrestrial Hg sink, to investigate the importance of land management policies for curbing Hg pollution.

Materials and Methods

Atmospheric Hg model (GEOS-Chem) description. In this study, we used the chemical-transport model GEOS-Chem v12.8.1 with Hg^0 dry deposition updates from Feinberg et al.[23]. The global model was run at 2.0° × 2.5° horizontal resolution and 47 vertical layers up to 80 km altitude. The model tracks emissions, transport, chemistry, and deposition of Hg in three chemical tracers: elemental mercury (Hg^0), oxidized mercury (Hg^{II}), and particulate-bound mercury (Hg^P). Atmospheric transport of Hg species is based on MERRA-2 reanalysis meteorological data[31]. The Hg chemical mechanism assumes that Br is the primary Hg^0 oxidant and uses offline monthly maps of previously-calculated oxidant concentrations to drive chemistry[32]. The aqueous photoreduction rate of Hg^{II} to Hg^0 is parametrized as a function of the organic aerosol concentration and the NO_2 photolysis rate[32].

The wet removal of oxidized Hg (Hg^{II} and Hg^P) from the atmosphere is calculated in online parametrizations considering large-scale and convective scavenging of gas and particulate species[33]. Dry deposition in GEOS-Chem applies a resistance-based approach[34], which determines the dry deposition velocities depending on meteorology

(e.g., temperature and windspeed), land surface parameters (e.g., land type and leaf area index, LAI), and compound-specific parameters (biological reactivity, f_0, and solubility, H^*). For Hg^0, f_0 was set to 0.2 within the Amazon rainforest and 3×10^{-5} elsewhere. These values of f_0 were found to yield the best agreement with available measurements of Hg^0 vegetation uptake[23], though we later tested the impacts of uncertainties in these parameters on the modeling results (Section S4). The solubility of Hg^0 is low ($H^* = 0.11$ M atm^{-1}), whereas gaseous Hg^{II} is assumed to be highly soluble ($H^* = 10^{14}$ M atm^{-1}) and biologically unreactive ($f_0 = 0$). Dry deposition of Hg^P is determined according to the aerosol deposition parametrization in GEOS-Chem[35]. Dry deposition is calculated separately over each land type within a grid cell (e.g., rainforest, grassland, cropland, etc.) and then an overall area-weighted average is calculated for the grid cell. GEOS-Chem accounts for 73 land types based on the Gibbs[36] land cover product. The LAI data for this study was taken from a reprocessed version of the Moderate Resolution Imaging Spectroradiometer (MODIS) satellite product[37].

Anthropogenic Hg emissions followed AMAP/UNEP estimates[38] for 2015. Biomass burning emissions were taken from the Global Fire Emissions Database (GFED) v4.1s[39]. Fixed concentrations of Hg^0 in the surface ocean based on the MITgcm 3-D ocean model[32] were used to calculate the Hg^0 air-sea exchange[40]. We adopted a new formulation[41] for the soil Hg^0 emissions parametrization (Supplementary Information, Section S1):

$$E_{\text{soil}} = aC^b R_g^c \qquad \text{(Eq. 1)}$$

where E_{soil} is the Hg^0 emissions from soil (units ng m^{-2} h^{-1}), C is the concentration of Hg in soils (units µg g^{-1}), R_g is solar radiation flux at the ground (units W m^{-2}), and a, b, and c, are coefficients (set to 71, 2.5, and 0.76, respectively). The coefficients of this parametrization were tuned to match available soil emissions measurements (Section S1). The soil concentration map of Hg (C) was calculated using the method of Selin et al.[42], deriving the spatial distribution of soil concentrations by first assuming a steady state balance between land emissions and deposition in the preindustrial and subsequently increasing soil concentrations according to the distribution of anthropogenic Hg

deposition. As in Selin et al.[42], the solar radiation at ground (R_g) is determined by considering attenuation of the solar radiation flux (R_S) by shading from the overhead canopy, parametrized by the LAI:

$$R_g = R_S \exp\left(-\frac{\alpha \mathrm{LAI}}{\cos\theta}\right) \qquad \text{(Eq. 2)}$$

where α = 0.5, assuming extinction from a random angular distribution of leaves[43], and θ is the solar zenith angle. Deforestation reduces the leaf area index (LAI) in impacted grid cells, increasing the solar radiation flux at the ground (Eq. 2) and consequently enhancing Hg^0 emissions from soils (Eq. 1). We have also updated GEOS-Chem to calculate soil emissions at the sub-grid scale for each land use category contained within the grid cell.

Reference (HIST) simulation. We ran a GEOS-Chem simulation for the land cover and LAI conditions of the year 2003 (HIST simulation), the first year where reprocessed LAI data is available. To highlight the role of land cover changes alone, meteorological conditions were kept constant by running all simulations with meteorology for 2014–2015. We considered the first year as spinup to equilibrate the new land cover conditions, and analyzed simulation differences for the meteorological year 2015.

Estimating historical global deforestation-driven Hg emissions. We calculated regional emissions factors (EFs) for deforestation through conducting perturbation experiments in GEOS-Chem. Emission factors were distinguished for the following regions based on biogeographic realms[44]: Palearctic, Nearctic, Afrotropic, Neotropic, Australasia & Oceania, Indomalaya, China, and the Amazon rainforest (mapped in Fig. S5). We separated China into its own region as soil Hg concentrations are higher than surrounding areas due to historical Hg emissions. The Amazon rainforest was separated from other Neotropic forests due to it having higher observed vegetation uptake fluxes and a different assigned f_0 parameter in the model dry deposition scheme. For each region, a simulation was conducted with perturbed land cover in the grid cells that experienced deforestation during 2000–2014 in the 0.25° × 0.25° resolution CMIP6 Land-Use Harmonization (LUH2) dataset[45]. As deforestation is mainly driven by agricultural expansion[46,47], we replaced forest land cover in these grid cells with the most common

agricultural land cover relevant to the region: "Crops and Town" (Afrotropic, Indomalaya, Palearctic, Australasia & Oceania, and China), "Corns and Beans Croplands" (Neotropic and Nearctic), and "Fields and Woody Savannah" (Amazon). For the new agricultural areas, the LAI was set to the average annual cycle for the existing agricultural grid cells within the region. Eight deforestation (DFR) simulations (1 for each region) were conducted for 2014–2015, comparing year 2015 fluxes to the HIST simulation. To calculate the net emissions factor (EF) from deforestation, we calculated changes to the land-air exchange over the deforested grid cells:

$$\mathrm{EF} = \frac{(E_{\mathrm{DFR}} - D_{\mathrm{DFR}}) - (E_{\mathrm{HIST}} - D_{\mathrm{HIST}})}{A_{\mathrm{DFR}}} \qquad \text{(Eq. 3)}$$

where *E* refers to Hg emissions, *D* refers to Hg deposition, and *A* refers to the area that is deforested in the simulation. The emissions factor represents the net emissions of Hg released by a deforested area annually, in units Mg m^{-2} yr^{-1}, capturing both the impact of increased soil Hg^0 emissions and reduced forest Hg^0 uptake. The assumption of linearity of the net emissions to deforested area holds in simulations conducted in the Amazon with differing spatial distributions of deforestation (Fig. S4), supporting an emissions factor approach to deforestation. We compared calculated emissions factors with existing estimates from observational studies[18,21,24,48–64] for total deforestation EFs and the component of EFs due to soil Hg^0 emissions (Supplementary Information, Section S2). Based on our literature review (SI Spreadsheet), observational data is available for three of the tested regions (Amazon, China, and Nearctic).

We applied the regional emissions factor to historical land use data from the LUH2 dataset to calculate emissions from deforestation. We defined gross deforested areas from the LUH2 dataset by summing the areas with transitions from primary or secondary forest to a non-forest land type. This approach does not consider LULCC fluxes due to harvesting of a forest without complete deforestation or the regrowth of vegetation after clearing, due to a lack of corresponding observations for Hg to constrain these parameters. Likewise, the emissions factors were assumed to be constant annually, so a deforested area continues to have the same total emissions for each year over the

considered time horizon. In reality, deforested areas could have a recovery timescale as vegetation regrows, which is accounted for in carbon LULCC fluxes[65]; for Hg, the response timescales during regrowth are largely unknown. To account for these uncertainties, we produced global and country-level estimates of Hg emissions in 2015 due to deforestation by summing deforestation over different time horizons: 15 years (2000–2014), 30 years (1985–2014), 45 years (1970–2014), and 60 years (1955–2014). The 45-year (1970–2014) accumulated results are presented in the main text, with the others presented in Fig. S6.

Future Amazon deforestation scenarios. We employed deforestation scenarios from Soares-Filho et al.[27], who developed a model for predicting the extent of deforestation within the Amazon based on environmental policies and highway construction. They presented two scenarios for 2050, encompassing a range of future deforestation trajectories. In the Business as Usual (BAU) scenario, recent deforestation trends continue into the future, assuming that compliance with conservation laws remains low and no new areas will be protected. On the other hand, the Governance (GOV) scenario assumes that the expansion of environmental legislation and increased enforcement of protected areas will lead to a reduction in the deforestation rate. Compared to the Amazon forest area in 2003 (5.3 million km^2), in 2050 the BAU scenario projects 3.2 million km^2 remaining and GOV projects 4.5 million km^2 remaining[27]. We focused our analysis on comparing the forest coverage in the years 2003 and 2050.

We translated these scenarios into required inputs for the calculations in GEOS-Chem (spatially gridded land use categories, LAI, and biomass burning emissions). The Soares-Filho et al.[27] dataset assigns 1 km^2 pixels within the Amazon basin as being forested, deforested, or agricultural areas for every year between 2003 and 2050. These annual datasets were regridded to 0.25° × 0.25° resolution, the native resolution of land use and LAI maps in GEOS-Chem. We calculated the relative change in forested area in the scenarios for every 0.25° × 0.25° grid cell. The rainforest land use category in deforested grid cells was correspondingly reduced by this factor, with the lost land area added to the land use category for "Fields and Woody Savanna". The LAI annual cycle for existing

Fields and Woody Savanna grid cells within the Amazon basin was spatially averaged over 2003 and assigned to the deforested areas. Annual average LAI maps for the Amazon scenarios used in GEOS-Chem are shown in Fig. S9. For these simulations, we assumed that conversion of forest to agricultural land within the Amazon is fire-mediated[66]. Gridded biomass burning emissions were calculated by multiplying the newly deforested areas for each year by mean fire Hg emissions (380 μg m^{-2} yr^{-1}) from two observational studies in the Amazon[19,67]. An additional 50% of the emissions (190 μg m^{-2} yr^{-1}) are released to the atmosphere within the first year as post-burn Hg^0 emissions from soils[18]. To account for seasonal differences in meteorology and realistic timing for forest clearing and burning[66], we assumed that deforestation occurs at the start of June and deforestation biomass burning emissions occur in August and September .

The BAU and GOV scenarios do not account for any land-climate feedbacks[27], wherein deforestation of the rainforest can lead to reduced moisture recycling and widespread *savannization* (conversion of rainforest to savanna)[68]. As an upper bound for this process, we considered an extreme scenario (SAV) where the Amazon rainforest is fully converted to savanna[69]. The impact of this scenario on Hg^0 deposition was previously quantified[23], but here we reran the SAV simulation in GEOS-Chem to account for updates in the soil Hg^0 emissions parametrization. Fluxes for the Amazon region were calculated by averaging over the area covered by the Soares-Filho et al.[27] deforestation projections (shown in Fig. S8).

Potential reforestation scenario. We applied a reforestation scenario (RFR) in GEOS-Chem based on the Global Reforestation Potential map[30,70], which considers the binary potential of every 1 km^2 grid cell to be converted from non-forest (<25% tree cover in 2000–2009) to forest (>25% tree cover). The reforestation potential dataset does not include areas that are native non-forest land cover types (e.g., grasslands) or cropland areas. The reforestation potential was regridded to 0.25° × 0.25° resolution. For every grid cell where reforestation can occur, we identified the corresponding biome in the Ecoregions2017 dataset[44] to determine the type of native forest vegetation that would occur. If the corresponding biome of the grid cell was not a forest (e.g., coastal grid

cells), the most common forest type in the 8 neighboring grid cells was selected. The added forest was assumed to have a LAI annual cycle equal to the 2003 spatial average for all grid cells in the corresponding biome and biogeographic realm (LAI_{biome}). For grid cells that were not a forest land type in 2003, we converted the reforested area fraction (f_{rfr}) from the original land type to the new forest land type. Only grid cells where LAI_{biome} is larger than the original land type LAI (LAI_{old}) were reforested. Since the land map used in GEOS-Chem is at coarser resolution (0.25° × 0.25°) than the reforestation potential dataset (1 km × 1 km), the reforested grid cell may already be a forest land type in GEOS-Chem. In this case, we assumed that the grid cell LAI (LAI_{new}) will become denser due to the new reforested area:

$$\mathrm{LAI_{new}} = \mathrm{LAI_{old}} + f_{\mathrm{rfr}} \cdot \mathrm{LAI_{biome}} \quad \text{(Eq. 4)}$$

The resultant average LAI map in the RFR scenario is shown in Fig. S10.

Uncertainty analysis. We employed offline Python-based models for Hg^0 dry deposition and soil Hg^0 emissions to estimate uncertainties in the simulated terrestrial-atmosphere Hg fluxes from GEOS-Chem. These models are made publicly available for further reuse (see Code and Data Availability). We focused on offline modeling of the Hg^0 dry deposition and soil emissions as these processes contribute the overwhelming majority (>98%) of the flux response to deforestation. The offline models contain the stand-alone GEOS-Chem code for calculation of dry deposition velocities and soil emissions across the horizontal model grid, but do not calculate atmospheric transport or chemical transformations. Dry deposition fluxes of Hg^0 were calculated by multiplying the deposition velocities by previously computed monthly Hg^0 concentration fields from the online simulations. The offline models were run for the year 2015 using monthly average diurnal cycles (12 × 24 h = 288 timesteps) of meteorological parameters, land surface parameters, and Hg^0 concentration fields. At this time resolution, the offline models showed sufficient accuracy compared to full online GEOS-Chem simulations, with maximum errors compared to online predictions of 1% for annual mean soil emissions and 5% for Hg^0 deposition. Given this level of accuracy and reduced computational expense, the offline models are appropriate for estimating the parametric uncertainties in

atmosphere-terrestrial fluxes of the online GEOS-Chem model. We considered the contributions of deposition parameters (f_0), soil emission parametrizations, the assumption for LAI for replaced land types, and biomass burning emission factors (for the Amazon simulations) to the overall uncertainty in fluxes. Uncertainty bounds of these parameters are tabulated in Table S4. Latin Hypercube sampling[71] was used to sample 100 parameter combinations. We conducted 100 simulations in the offline emissions and deposition models for each studied scenario, calculating 95% confidence intervals from the 2.5th and 97.5th percentile values in the offline calculated fluxes.

Results and Discussion

Global estimate of deforestation-driven Hg fluxes. To calculate net deforestation emissions, we computed the difference in the net terrestrial-atmosphere exchange (emissions from a grid cell minus deposition to a grid cell) before and after deforestation (Eq. 3). For our global estimate of deforestation-driven emissions, we did not consider immediate biomass burning emissions of Hg due to fire-mediated forest clearing nor enhanced erosion fluxes, instead focusing on the impact on net Hg fluxes to the atmosphere in the years after the clearing event. The major impacts to Hg fluxes arise through enhanced soil Hg^0 emissions and decreased Hg^0 dry deposition due to reduced canopy coverage, which can continue many years after the initial deforestation event[18,61]. Using perturbation simulations in GEOS-Chem for 8 global land regions, we calculated regional emission factors (EFs) representing net fluxes to the atmosphere per unit deforested area (in units Mg Hg m^{-2} yr^{-1}).

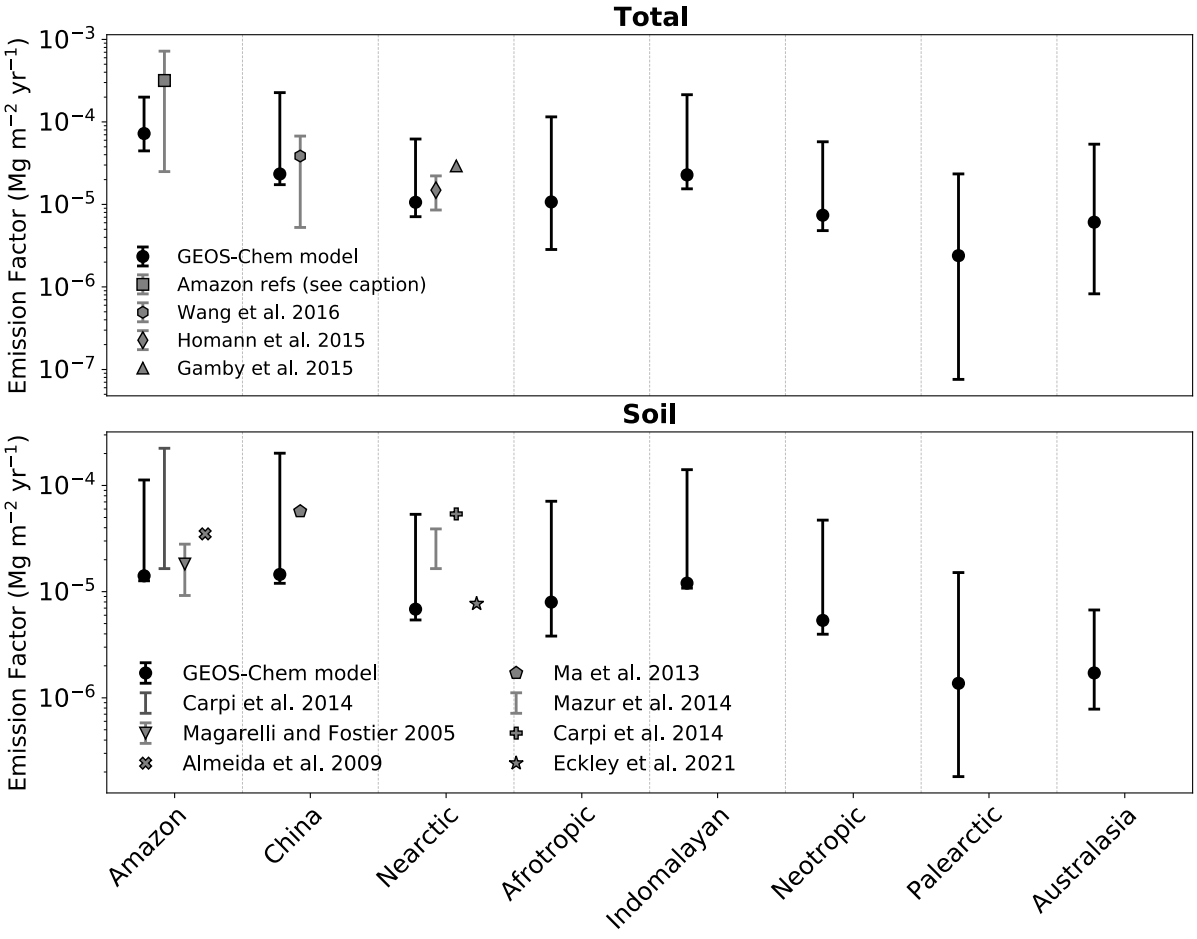


Figure 1. Comparison between modeled and observation-derived net emission factors (EFs) for deforestation in different regions. The upper panel shows total EFs and the lower panel shows the soil Hg^0 emissions component of deforestation EFs. Modeled circles show the best estimate (online simulations), while error bars show the 95% confidence interval due to model parameter uncertainties (calculated in offline simulations, Section S4). Observation estimates are from refs.[18,21,24,48–64], with the Amazon Total EF estimate based on measurements in Fig. S3. Observed error bars refer to uncertainty ranges when multiple plots were measured within a study (further information about these calculations can be found in Section S2 and the SI Spreadsheet).

The calculated EFs are on the order of 10^{-6} to 10^{-4} Mg Hg m^{-2} yr^{-1} depending on the region (Fig. 1; Table S3), with the Amazon rainforest showing the highest EF (7×10^{-5} Mg Hg m^{-2} yr^{-1}; 95% confidence interval, CI: 4×10^{-5} to 2×10^{-4} Mg Hg m^{-2} yr^{-1}). This is to be expected from litterfall and throughfall measurements in the Amazon, which show some of the highest levels of Hg^0 vegetation uptake observed globally[13], as well as Hg^0 soil flux measurements from deforested areas in the Amazon, which show higher levels

of emissions compared to deforested North American soils[18]. The variation of simulated EFs between regions depend on the factors that affect dry deposition (vegetation type and LAI) and soil emission fluxes (LAI, soil Hg concentrations, and solar radiation). We compiled available estimates of deforestation EFs from previous observational studies[18,21,24,48–64] and compared these to our modeled values (Fig. 1). Our EFs overlap with available factors derived from observations, for the three regions where data are available. The modeled error ranges appear well-calibrated as they cover a similar range as the variability between observation-derived fluxes in the same region (Fig. 1). Fig. 1 also highlights that no observations of the impact of deforestation on Hg cycling are currently available from the Afrotropic and Indomalayan regions, where deforestation is widespread.

We multiplied the regional EFs by the deforested area from the CMIP6 Land-Use Harmonization (LUH2) dataset[45] to calculate the net Hg fluxes to the atmosphere from deforestation. Given the uncertain timescale for recovery in Hg sink capacity after deforestation, we assumed that a deforested area has constant annual emissions over a considered time horizon. Previous LULCC studies for carbon suggest that forests recover their original biomass within 75 years after deforestation[65], so we employed time horizons between 15–60 years (Fig. S6) to calculate 2015 deforestation-driven emissions. In Fig. 2a, country-level deforestation emissions are shown based on a 45-year time horizon (emissions released from areas deforested between 1970 and 2014). Net emissions occurring in 2015 considering this 45-year deforestation time horizon are 217 Mg yr^{-1} globally (CI: 134–1650 Mg yr^{-1}). Countries with substantial (>10 Mg yr^{-1}) deforestation-driven emissions include Brazil (43 Mg yr^{-1}), Indonesia (35 Mg yr^{-1}), China (16 Mg yr^{-1}), Colombia (14 Mg yr^{-1}), India (13 Mg yr^{-1}), Philippines (11 Mg yr^{-1}), and Myanmar (11 Mg yr^{-1}). To put these emissions into context, Fig. 2b compares the deforestation emissions with 2015 primary anthropogenic emissions inventory from AMAP/UNEP[9,38]. Deforestation Hg emissions are minor (<5%) compared to primary anthropogenic emissions for most countries. However, for 32 countries, all located in the tropics, deforestation emissions are greater than 30% of primary emissions. For Brazil, which is the fifth highest emitter of primary Hg[9,38], deforestation emissions (43 Mg yr^{-1})

are only 40% smaller than the 2015 emissions from primary anthropogenic sources (71 Mg yr^{-1}). Deforestation emissions even exceed primary emissions in some countries, including Madagascar (deforestation emissions are 2.4× larger), Paraguay (2.3×), Liberia (2.0×), and Bangladesh (1.8×). Currently, Hg emissions inventories[9] only consider primary anthropogenic emissions (2222 Mg yr^{-1} in 2015), overlooking deforestation as a significant source of anthropogenic Hg to the atmosphere (217 Mg yr^{-1}). The relative importance of deforestation as an anthropogenic driver of Hg pollution could increase over the next decades, with primary anthropogenic emissions of Hg projected to halve to 1020 Mg yr^{-1} by 2035 under Minamata policies and reductions in fossil fuel use[72]. Therefore, assessing the potential impacts of land use policy scenarios will be crucial for predicting future Hg cycling, as primary anthropogenic emissions decline in the future.

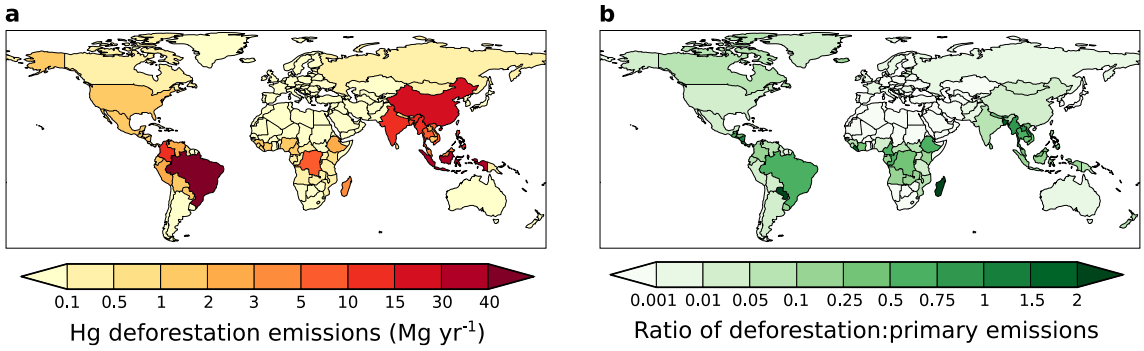


Figure 2. Country-level annual deforestation emissions of Hg in 2015. (*a*) Deforestation-driven net emissions of Hg by country, assuming that deforested areas from the previous 45 years (1970–2014) contribute to emissions. (*b*) Ratio of deforestation emissions to primary anthropogenic emissions[9,38] by country.

Amazon conservation policy impacts on Hg cycling. The Amazon is one of the regions with the highest Hg fluxes from deforestation (Fig. 2) and land policy choices will determine how this evolves in the future. Under historical forest coverage from 2003 (HIST simulation), the Amazon rainforest stands out as a strong global sink of Hg (Fig. 3a), with net input from the atmosphere to the rainforest totaling 332 Mg yr^{-1} (CI: 179–463 Mg yr^{-1}). We studied the evolution of the Amazon Hg sink in two deforestation scenarios[27] for 2050: a business-as-usual scenario (BAU), which extrapolates historical deforestation tendencies into the future, and a governance scenario (GOV), which

assumes expanded conservation of the rainforest in the future. In the BAU scenario, widespread deforestation, mainly in eastern Amazonia, reduces the net Hg inputs to soils (Fig. 3b). While the Amazon region overall remains a net Hg sink in BAU, the removed vegetation leads to decreased Hg^0 deposition in the Amazon (change from HIST: -105 Mg yr^{-1}; CI: -53 to -152 Mg yr^{-1}) and enhanced Hg^0 emissions from soils (+35 Mg yr^{-1} ; CI: 28–275 Mg yr^{-1}). For the Amazon policy scenarios, we have also considered the impact that fire-mediated forest clearing[66,73] has on biomass burning emissions of Hg, which are 15 Mg yr^{-1} (CI: 10–17 Mg yr^{-1}) larger in BAU than HIST. The BAU scenario shows atmospheric Hg^0 concentrations increasing up to 0.3 ng m^{-3} (+50%) within the Amazon region (Fig. S11); this would be a detectable change in Hg^0, comparable to the 0.5 ng m^{-3} (-30%) decrease between 1995–2015 in North American Hg^0 observations[74]. The additional Hg fluxes from deforested areas can be transported over long distances in the atmosphere and lead to more Hg deposition over oceans and remaining intact forest areas (Fig. 3b). In the GOV scenario, deforestation is slowed by the conservation measures, leading to smaller perturbations in the dry deposition flux from HIST (-47 Mg yr^{-1} ; CI: -25 to -68 Mg yr^{-1}) and the soil emission flux (+16 Mg yr^{-1}; CI: 12–126 Mg yr^{-1}) (Fig. 3b). In GOV, burning emissions from deforestation are 1 Mg yr^{-1} lower than in HIST, due to lower annual rates of deforestation in the 2050 GOV scenario compared to the HIST case representing 2003. Globally, the weakened rainforest sink of Hg yields higher deposition of Hg to oceans compared to the reference simulation (BAU – HIST = +108 Mg yr^{-1}; GOV – HIST = +44 Mg yr^{-1}).

Deforestation can be exacerbated through climate feedbacks, which are not considered in these policy scenarios. For example, BAU projects that 40% of the Amazon will be deforested by 2050[27], which could trigger a tipping point with widespread transition of the rainforest to a savannah biome under diminished regional moisture recycling[68]. To evaluate this, we also re-ran an upper limit scenario from our previous work[23] where the entire rainforest is converted to savannah (SAV). In this case, a strong decline in Hg^0 dry deposition (-359 Mg yr^{-1}; CI: -210 to -503 Mg yr^{-1}) and an increase in Hg^0 soil emissions (+89 Mg yr^{-1}; CI: 68 to 652 Mg yr^{-1}) drive enhanced inputs of Hg to the ocean (343 Mg yr^{-1}) (Fig. 3b).

This change in the fate of atmospheric Hg (deposition to ocean instead of land) affects both the spatial distribution and bioavailability of Hg pollution. When sequestered in soils, Hg has an estimated residence time on the order of hundreds of years, whereas in the surface ocean Hg is recycled to the atmosphere within months to years[7,11]. Deforestation thus increases the mobility of Hg by transferring Hg from locally-sequestered reservoirs to the global pool. Human health risks are driven by exposure to the more toxic form of the element, MeHg, which is produced through methylation in the environment[2,75]. Deforestation shifts Hg inputs from land to the ocean, where Hg can more readily be methylated and bioaccumulate to dangerous levels in commercial fish. Methylation and bioaccumulation of Hg can also occur in forested soils, but MeHg levels in aquatic ecosystems are generally much higher (overall global ocean average = 15%)[76] than in Amazonian soils (1–5%)[48,77] . In addition, the long length of aquatic food chains leads to high levels of MeHg in commonly consumed fish species at higher trophic levels (e.g., tuna, cod, and swordfish)[75].

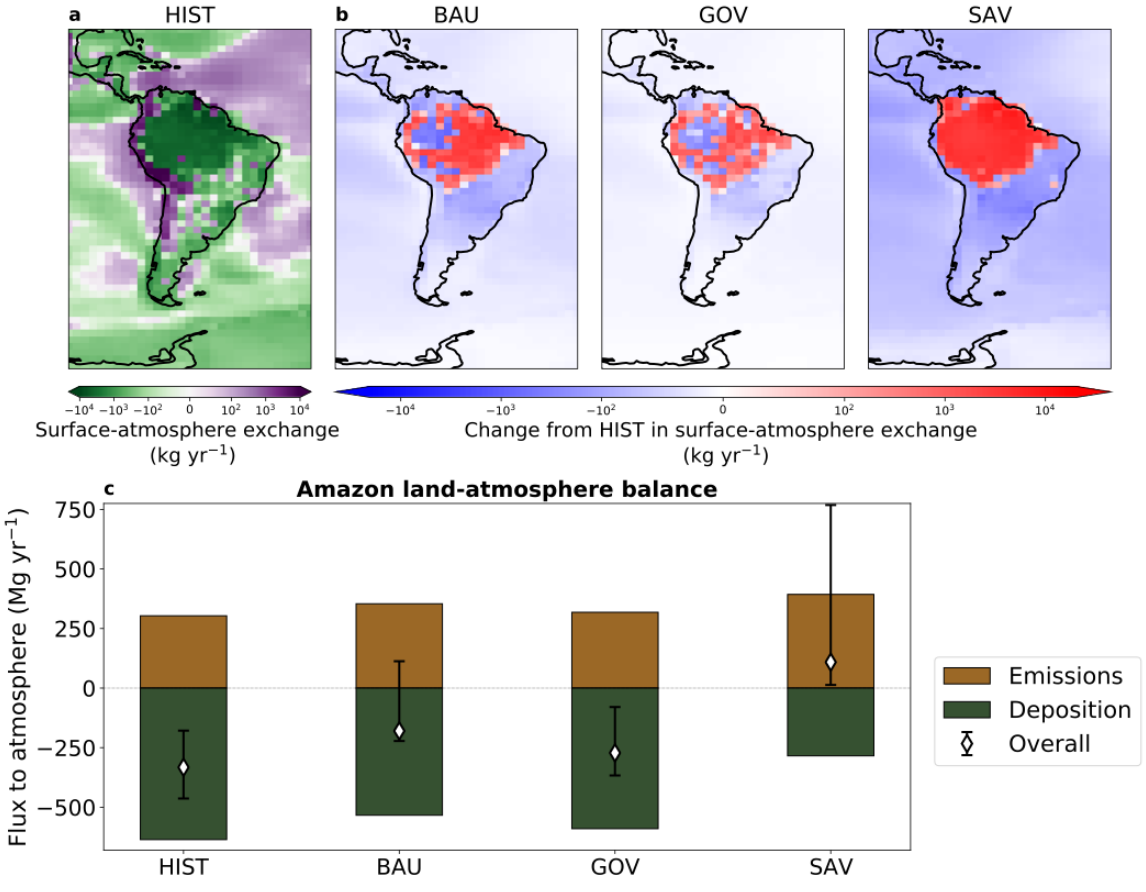


Figure 3. Impacts of Amazon deforestation scenarios on surface-atmosphere Hg exchange. (*a*) The simulated surface-atmosphere exchange (net deposition is negative and net emission is positive) of Hg in the reference simulation (HIST). (*b*) Changes in exchange fluxes from HIST are shown for the deforestation scenarios: Business-as-usual (BAU), Governance (GOV), and Savannization (SAV); negative values refer to increased net fluxes to the surface compared to HIST and positive values refer to increased net fluxes to the atmosphere. (*c*) Total simulated fluxes of Hg emissions and deposition are calculated for the Amazon region in each scenario. White diamonds illustrate the net flux of Hg to the atmosphere (= emissions – deposition) and error bars refer to the 95% confidence interval based on model parameter uncertainties.

Deforestation policy substantially impacts the soil mass balance of Hg in the Amazon region, illustrated by our modeling simulations (Fig. 3c) and available field observations (Fig. S3). If agricultural expansion continues as in BAU, the net Amazon sink of atmospheric Hg is weakened by 153 Mg yr^{-1} (CI: 97–418 Mg yr^{-1}) (Fig. 3c). The reduction of forest Hg^0 uptake contributes two-thirds of the net flux response in the BAU scenario, while increases in emissions contribute the remaining third. Under the more moderate GOV scenario, the Amazon Hg sink (272 Mg yr^{-1}; CI: 79–367 Mg yr^{-1}) is better

preserved, though still 18% (CI: 14–65%) smaller than HIST. Stricter conservation policies in GOV yield an additional 92 Mg yr^{-1} (CI: 59–234 Mg yr^{-1}) of Hg sequestered in the Amazon compared to BAU. The SAV scenario illustrates that additional climate feedbacks could flip the Amazon from a net Hg sink to a source (+109 Mg yr^{-1}; CI: 13–768 Mg yr^{-1}). These Hg projections parallel recent findings on Amazon carbon cycling, which have demonstrated that climate change and deforestation are turning the Amazon into a CO_2 source[25]. In addition to atmosphere-terrestrial exchange fluxes, soil erosion of Hg can also be altered due to deforestation. We applied a soil erosion model GloSEM[78,79] to evaluate the impact of deforestation on erosion in the Amazon basin (Supplementary Information Section S6). In terms of Hg flux magnitudes, perturbations to erosion are smaller (<15%) than changes to the atmosphere-terrestrial exchange fluxes (Section S6), which is supported by field studies[64]. Nevertheless, deforestation also enhances Hg erosion in both scenarios (BAU: +33%; GOV: +14%), accelerating the transfer of terrestrial Hg to aquatic ecosystems.

Quantifying the Hg mitigation potential of reforestation. Reforestation has been identified as a potential mitigation approach for climate change, by strengthening the terrestrial CO_2 sink[30,80]. To investigate the concurrent strengthening of the terrestrial Hg sink and the impacts on Hg cycling, we considered a global reforestation scenario (RFR) based on the Global Reforestation Potential Map[30,70], which identified areas suitable for reforestation worldwide (i.e., not including croplands or areas where forests are not native). Figure 4 maps the impacts of reforestation on Hg surface-atmosphere exchange, comparing to the reference HIST simulation. The spatial distribution of reforestation impacts depends both on the areal extent of reforestation as well as the reforested vegetation type. Net deposition of Hg increases over reforested areas (blue areas in Fig. 4), while net deposition declines over the ocean as well as land areas with existing forests (red areas in Fig. 4). Globally, RFR enhances uptake of Hg on land by 98 Mg yr^{-1} (CI: 64–449 Mg yr^{-1}) compared to HIST, thereby reducing Hg deposition to oceans. Reforestation could thus take up approximately 5% of the anthropogenic Hg emission flux (~2200 Mg yr^{-1})[9]. In addition to the targeted benefits for biodiversity and climate change mitigation[30], reforestation could moderately reduce levels of Hg in marine

ecosystems, and thus commercial fish. Nevertheless, the magnitude of reforestation impact (5% of primary emissions) illustrates that reforestation is not a substitute for implementing extensive cuts to primary Hg emissions, like in the CO_2 context[29].

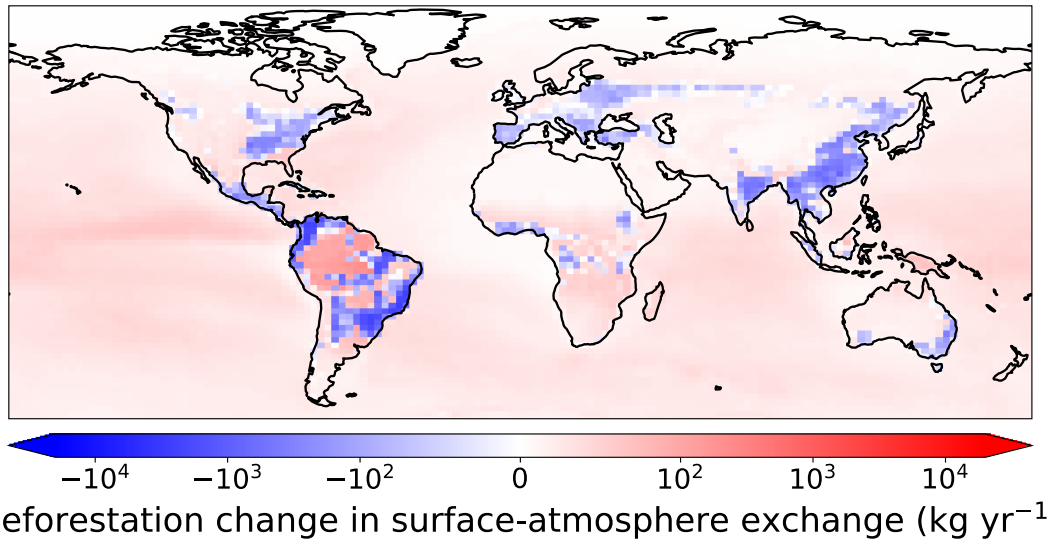


Figure 4. Enhanced land sink of Hg with reforestation. The impact of the potential reforestation (RFR) scenario on surface-atmosphere exchange. The differences from the reference (HIST) simulation are shown, with negative values referring to increased net fluxes to the surface and positive values referring to decreased net fluxes to the surface.

Potential reforestation opportunities for Hg are dominated by the Amazon and Atlantic forest regions in South America (71 Mg yr^{-1}, 72% of total land sink impact) (Fig. 4). The potential reforestation impact on atmospheric fluxes in Northern extratropical areas alone (-29 Mg yr^{-1}) would not compensate for increased Hg emissions due to deforestation in the Amazon (BAU: +153 Mg yr^{-1}; GOV: +61 Mg yr^{-1}). Overall, more information would be needed to compare the potentials of reforestation and conservation policies on a global scale, as the deforestation policy scenarios focused only on a specific region (the Amazon); future research could study conservation impacts in other tropical regions with high Hg deforestation emissions (Fig. 2) (e.g., in Africa and Southeast Asia). Our simulated fluxes consider the uptake of Hg upon maturation of forest stands, as reforested areas are assumed to have LAI of existent corresponding biomes. Further experimental

research would be required to understand the transient response of Hg uptake during the growth of forests.

Limitations of modeling approach. The current work provides an initial assessment of the global emissions of Hg from deforestation, which can spur future investigation into the impact of LULCC on Hg. Other LULCC processes (e.g., wood harvest and agricultural practices) may also affect Hg fluxes but have not been considered within this study. As well, due to the early stage of Hg research, there is not yet the same level of information for Hg that is commonly included in LULCC assessments for carbon, including temporal information on the release of Hg from soils and Hg uptake rates during regrowth of vegetation[22]. Although we have assembled a dataset of available deforestation flux measurements covering multiple regions (SI Spreadsheet), there continues to be a lack of measurements in relevant regions (e.g., Afrotropic and Indomalayan) to constrain the response of Hg fluxes to deforestation, contributing uncertainty to this work. As information from field measurements becomes more available, it will be possible for future modeling studies to analyze smaller sub-regions differentiated by ecosystem types, improving the accuracy of deforestation-driven emissions. In the current work, the parametrization of Hg^0 soil emissions is based on solar radiation and soil Hg concentration, which is the current state of the art for global models and in agreement with available flux measurements (Fig. 1). Field observations[81,82] have investigated the role of other environmental parameters including precipitation, soil moisture, soil chemistry, soil physics, and microbial interactions, along with anthropogenic factors such as emissions from directly contaminated soils[83] that would not be captured at the resolution of the global modeling approach. Regional models of Hg^0 soil emissions include a wider array of these parameters[24], but further research would be required to produce a tuned parametrization of this complexity at the global scale. The development of terrestrial Hg cycles and LULCC processes within Earth system models[84] will be vital to investigate the evolution of the Hg land sink over time and the effect on environmental Hg risks.

Implications for global Hg policy. Land use policy has been largely unexplored as a lever to mitigate Hg pollution. On the global scale, the estimated deforestation-driven Hg emissions in 2015 (217 Mg yr^{-1}; CI: 134–1650 Mg yr^{-1}) correspond to 10% of the global primary anthropogenic emissions[9] (2222 Mg yr^{-1}) (Fig. 5a). Therefore, though cutting primary anthropogenic emissions remains a priority, deforestation fluxes should not be overlooked in assessments of Hg pollution, especially for countries in the tropics (Fig. 2b). The potential of Amazon conservation and global reforestation to reduce net Hg emissions in the future is substantial compared to previously quantified policies aimed at tackling primary anthropogenic emissions (Fig. 5b). Potential emissions reductions from Amazon conservation (92 Mg yr^{-1}) and global reforestation (98 Mg yr^{-1}) are within the range of impacts of past policy and future policy scenarios aimed at reducing Hg from specific anthropogenic sources or due to national climate and air pollution policies (5–262 Mg yr^{-1})[85–90]. Emissions reductions from land use policies are different from primary emissions reductions in that their efficacy depends on whether the storage of Hg in soils is over a long-term period. Similar to CO_2, the potential benefits of enhanced Hg uptake on land can be reversed by human or natural disturbances, e.g., by climate change increasing the frequency of wildfires — which re-emit Hg and carbon from terrestrial ecosystems — and droughts — which reduce Hg and CO_2 uptake by plants[30,91]. Thus, mitigation of Hg pollution by conserving and increasing forest area can only be realized with concurrent efforts to sustainably manage land areas and preventing severe climate change. The potential of sustainable land use to mitigate Hg pollution could enable collaborations between the Minamata Convention and other global policy efforts to reduce deforestation, e.g., the 2021 Glasgow Declaration[92]. Ultimately, mitigation of global Hg pollution depends not only on reducing primary anthropogenic emissions, but also reducing anthropogenic activities like deforestation that re-mobilize legacy Hg.

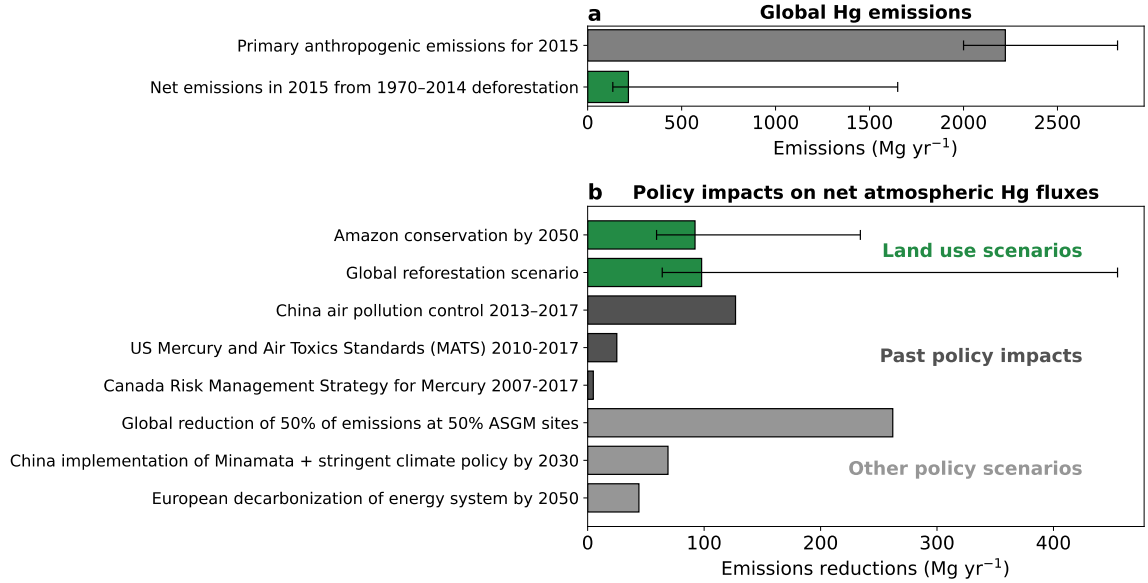


Figure 5. Potential of land use policies to reduce net Hg fluxes to the atmosphere. (*a*) Comparing global 2015 emissions from primary anthropogenic emissions[9,38] and deforestation-driven emissions, assuming a 45-year time horizon (1970–2014 deforested areas). (*b*) Net Hg emissions reductions from land use policies (this study) are compared to primary anthropogenic emissions policies, whose impacts have been quantified in the literature[85–90]. ASGM refers to artisanal and small-scale gold mining. For land use scenarios, "Amazon conservation by 2050" refers to the net emissions reductions in the 2050 governance (GOV) from the business-as-usual (BAU) simulations and "Global reforestation scenario" compares the net emissions reductions in the reforestation scenario (RFR) compared to the reference simulation (HIST). Error bars for this study refer to the 95% confidence interval based on model parameter uncertainties.

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Code and Data Availability

Model code, analysis scripts, and simulation data supporting the results of this study are published in Zenodo (https://doi.org/10.5281/zenodo.8364698) under a CC BY 4.0 license (https://creativecommons.org/licenses/by/4.0/).

Associated Content

Supporting Information. Further supporting information can be found in the following files:

Soil emissions parametrization; Observations of deforestation fluxes; Global deforestation-driven emissions estimates; Parameters for uncertainty analysis; Scenario maps; Erosion calculations; Atmospheric concentration impacts (PDF)

Tabulated dataset of literature Hg deforestation flux measurements (XLSX)

Author Contributions

All authors conceived the study. M.J., J.B., and A.F. compiled Hg field data through literature review. A.F. and P.B performed the simulations. All authors contributed to the data analysis. A.F. wrote the draft of the paper with contributions and revisions from all authors.

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Supplementary Information (SI) for

Deforestation as an anthropogenic driver of mercury pollution

Aryeh Feinberg[a]*, *Martin Jiskra*[b]*, *Pasquale Borrelli*[c], *Jagannath Biswakarma*[b,d], *and Noelle E. Selin*[a,e]

[a] Institute for Data, Systems, and Society, Massachusetts Institute of Technology, Cambridge, MA 02139, USA

[b] Environmental Geosciences, University of Basel, Basel 4056, Switzerland

[c] Department of Science, Roma Tre University, Rome 00146, Italy

[d] Department of Water Resources and Drinking Water, Eawag, Dübendorf 8600, Switzerland

[e] Department of Earth, Atmospheric, and Planetary Sciences, Massachusetts Institute of Technology, Cambridge, MA 02139, USA

*Correspondence to: arifeinberg@gmail.com (A.F.); martin.jiskra@gmail.com (M.J.)

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Section S1. Soil emissions parameterization

We improved the model's parametrization of Hg^0 soil emissions by adopting a new formulation for the parametrization, suggested by Khan et al.[1]:

$$E_{\text{soil}} = aC^b R_g^c \qquad \text{(Eq. S1)}$$

where E_{soil} are soil emissions (ng m^{-2} h^{-1}), C is the concentration of Hg in soils (µg g^{-1}), R_g is the solar radiation flux at the ground (W m^{-2}), and a, b, and c are coefficients.

As in Selin et al.[2], the solar radiation atwi ground (R_g) is determined by considering attenuation of the solar radiation flux (R_S) by shading from the overhead canopy, parametrized by the leaf area index (LAI):

$$R_g = R_S \exp\left(-\frac{\alpha \text{LAI}}{\cos\theta}\right) \qquad \text{(Eq. S2)}$$

where α = 0.5, assuming extinction from a random angular distribution of leaves[3] and θ is the solar zenith angle.

We compiled several relevant observational constraints for the parametrization in Tables S1 and S2. Observational studies from the Amazon region suggest that deforestation has a large impact on soil emissions due to removal of canopy shading, showing factors of 1.8×, 6.7×, and >31× more emissions in forested compared to deforested land plots (Table S1). Observational studies from other regions find a similarly high sensitivity of soil emissions to the presence of forest: open fields in China showed 6–10 times higher Hg emissions than forests[4] and logging in the US flipped the surface-air Hg^0 flux from net deposition to net emissions (-2.2 µg m^{-2} yr^{-1} to +5.5 µg m^{-2} yr^{-1}) [5]. For extratropical grassland soil emissions, we use the compiled median values from Zhu et al.[6] and Agnan et al.[7]

We conducted a parameter sweep of a, b, and c, calculating globally-gridded soil emissions using annual solar radiation data (Fig. S1). Sensitivity simulations showed that the ratio of deforested to forested soil emissions in the Amazon (median value 6.7) can tune the exponent for the radiation term (c in Eq. S1), i.e., the response of emissions to canopy shading. The exponent for the soil concentration term (b) was tuned with the ratio of deforested Amazon soil emissions (Table S1) to extratropical grassland soil emissions from the Northern Hemisphere from two review studies[6,7] (overall Amazon to extratropical ratio of 5.3). Lastly, after these coefficients are tuned, the prefactor a is adjusted so that predicted annual mean emissions match the observed median magnitudes of Amazon deforested soil emissions (23 µg m^{-2} yr^{-1}) and extratropical grassland emissions (4.3 µg m^{-2} yr^{-1}).

We recognize the uncertainties in the observed data used to tune this parametrization, and thus we constructed 100 alternative parametrizations that fit within observed data bounds (Table S5). These parametrizations were applied in offline uncertainty analyses to assess 95% confidence intervals in the fluxes driven by deforestation (Section S4).

Table S1. Literature review of available Hg^0 soil emission flux measurements from the Amazon region, differentiated by land cover type.

Reference	Location	Site	Deforested Hg^0 flux ($\mu g\ m^{-2}\ yr^{-1}$)	Forested Hg^0 flux ($\mu g\ m^{-2}\ yr^{-1}$)	Flux ratio (deforest:forest)
Magarelli and Fostier[8]	Negro River Basin, Brazil	#1	27 ± 9	0.6 ± 1.5	
		#2	19	–1.0 ± 0.8	
		#3	9.8 ± 0.7		
		Mean	18	–0.2	> 31[a]
Almeida et al.[9]	Rondônia, Brazil	#1	79 ± 110	44 ± 18	1.8
Carpi et al.[10]	Acre, Brazil	#1	19 ± 2	2.9 ± 0.8	6.7
		#2	230[b]		
	Median		**23**	**1.8**	**6.7**

[a]lower limit calculated assuming the forested flux is equal to site #1, as site #2 shows negative overall flux; deforested flux assumed as mean.

[b]this site was 2-months post-fire and soil temperatures were still elevated; this flux is excluded from ratio calculations

Table S2. Observational constraints used to tune the soil emissions parametrization.

Constraint	Value	Reference	Coefficient constrained
Amazon deforested soil emissions ($\mu g\ m^{-2}\ yr^{-1}$)	23	Table S1	*a*
Extratropical grassland soil emissions ($\mu g\ m^{-2}\ yr^{-1}$)	4.3[†]	Zhu et al.[6]; Agnan et al.[7]	*a*
Ratio of Amazon to extratropical soil emissions	5.3	(23:4.3)	*b*
Ratio of deforested to forested Amazon soil emissions	6.7	Table S1	*c*

[†]average of grassland median Hg^0 fluxes from the two independent review studies

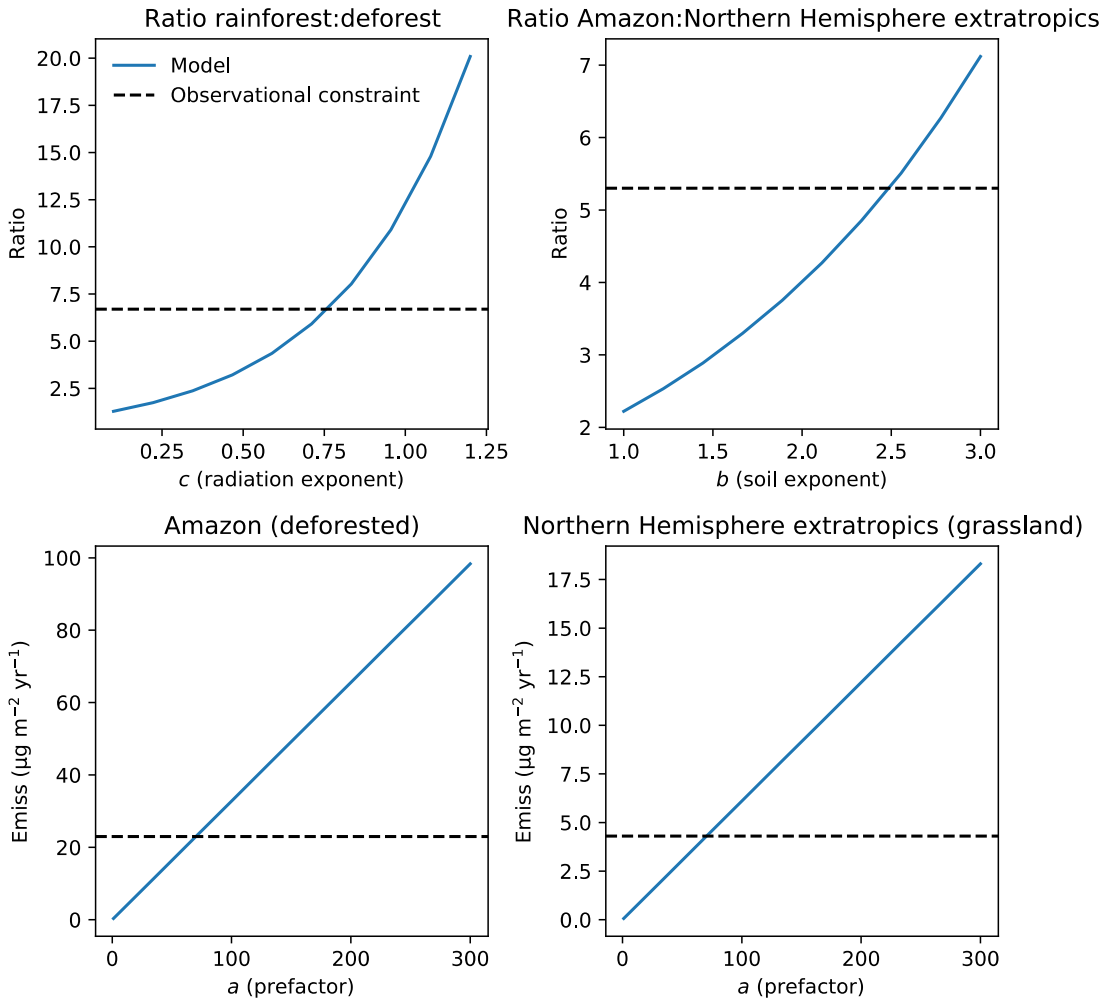


Figure S1. Parameter tuning (Eq. S1) to match observational constraints from Table S2.

The tuning procedure is illustrated in Fig. S1, yielding best matches for a = 71, b = 2.5, and c = 0.76. We compare the gridded annual mean soil emissions from the previous soil emission parametrization (GEOS-Chem v12.8) and the current study (Eq. S1) in Fig. S2. Global annual mean soil Hg^0 emissions in the new parametrizations (954 Mg yr^{-1}) is similar to the predictions from two GEOS-Chem studies[11,12] using the previous parametrization: 860 ± 440 Mg yr^{-1} and 910 Mg yr^{-1}. The spatial distribution of emissions (Fig. S2) shows a decrease in vegetated regions (e.g., the Amazon and Congo rainforests) and an increase in regions with high soil Hg concentrations (e.g., eastern China).

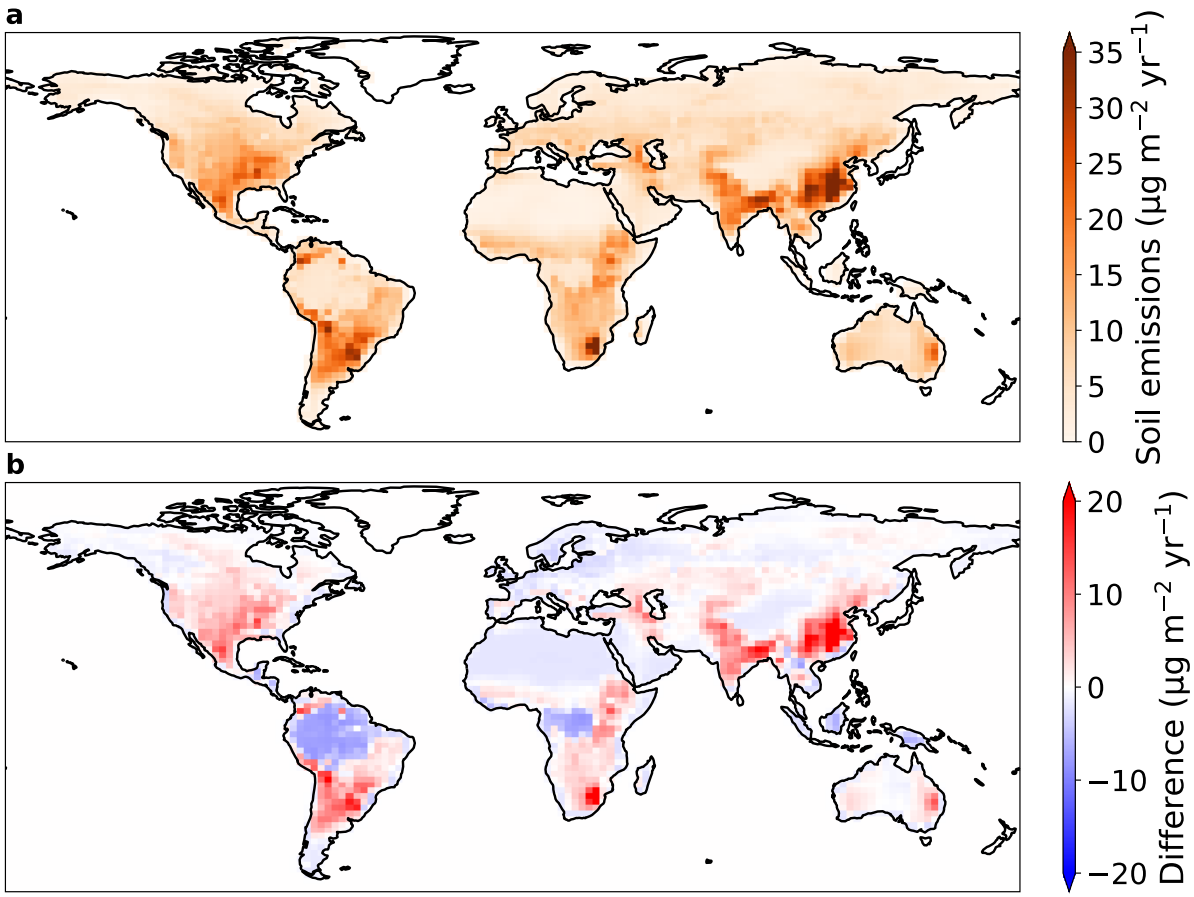


Figure S2. (*a*) Annual mean soil emissions of Hg^0 with the new parametrization. (*b*) Difference between new and old (GEOS-Chem v12.8) soil emissions parametrizations (new minus old).

Section S2. Observational constraints on deforestation Hg fluxes

There are several available sources of information that can be used to validate the deforestation emission factors (EF) calculated by GEOS-Chem (Fig. 1, SI Spreadsheet):

1) *Soil Hg concentration measurements of paired forest-deforested sites*:
Previous studies have measured the concentrations of Hg soils at deforested sites (C_d) and nearby forest (C_f) plots. For this analysis, we assume that the difference in these soil concentrations is due to mainly the change in atmospheric exchange, which is supported by the magnitude of modeled erosion fluxes (Section S6) and available measurements[5]. We use the following equation to convert the difference in these concentrations to a deforestation emission factor of Hg in Mg m^{-2} yr^{-1}:

$$\text{Total EF} = \frac{(C_d - C_f) \times \rho \times h}{t_d} \qquad \text{(S3)}$$

where ρ is the density of the soil, h is the depth of the soil layer, and t_d is the time since deforestation. In the US (Nearctic), there have been studies in Ohio[13] and Oregon[14] with measurements of Hg in deforested and forested soils, which we use to calculate deforestation EFs for the Nearctic. For the Amazon, more measurements are available (24 pairs of soil plots)[8–10,15–25]. We compiled a literature database of studies that compared Hg concentrations in deforested Amazonian soils with nearby forest plots (Fig. S3; SI Spreadsheet). Deforested sites show a consistent decrease compared to paired forested sites (p-value < 0.001; Wilcoxon signed-rank test), with the median decrease being 25 ng g^{-1} (10^{th}–90^{th} percentile: 2–58 ng g^{-1}). To calculate a deforestation EF for the Amazon, we apply this concentration decrease in Eq. S3 and assume an average Amazon soil density of 1.25 ng g^{-1}, a surface soil layer of 10 cm, and that deforested soils in the literature studies were measured 10 years after deforestation.

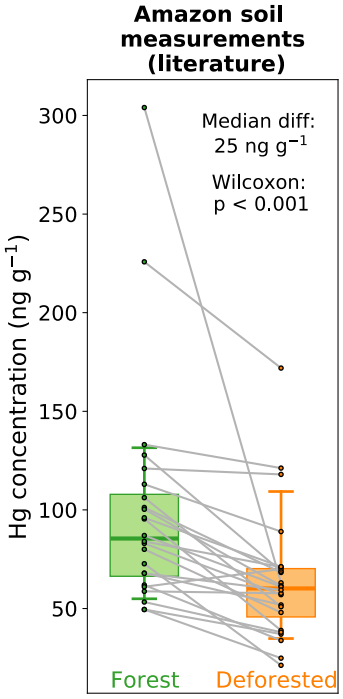


Figure S3. Measured Hg concentrations in forest (green) and deforested (orange) soils (0–20 cm depth) from the literature (n = 24)[8–10,15–25]. Box plots show the median values (solid lines), interquartile range (shaded), and 10th and 90th percentiles (whiskers). Gray lines connect paired sites from the same study. Listed p-value (<0.001) refers to the Wilcoxon signed-rank test of the null hypothesis that paired forest and deforested sites come from the same distribution.

2) *Terrestrial-atmosphere exchange models validated by Hg observations*:
An estimate for the deforestation EF over China is available from the Wang et al.[26] modeling study. We use their area-averaged mean fluxes over forest and agricultural land cover to calculate a deforestation emission factor:

$$\text{Total EF} = (E_d - D_d) - (E_f - D_f) \quad \text{(S4)}$$

where E_d and E_f are the terrestrial emission fluxes (Mg m^{-2} yr^{-1}) from Chinese agricultural land and forest, and D_d and D_f are the deposition fluxes (Mg m^{-2} yr^{-1}) to Chinese agricultural land and forest. Although this EF estimate is model-based, the Wang et al.[26] model was validated extensively with available terrestrial-atmosphere exchange measurements from China.

3) *Dynamic flux chamber measurements of forested and deforested soils*:
Additional studies investigating the impact of deforestation on atmospheric fluxes quantified the response of soil emissions using dynamic flux chamber measurements[5,8–10,27,28]. We compare these measurements to the soil-only EF modeled by GEOS-Chem. The soil emission factors measured by the studies is calculated as the difference between soil emissions (Mg m^{-2} yr^{-1}) over deforested and forested soils:

$$\text{Soil EF} = E_d - E_f \quad \text{(S5)}$$

The comparison between GEOS-Chem simulated deforestation EFs and observation-derived values is summarized in Fig. 1. Observations are only available from three regions (Amazon, China and Nearctic). We found further references investigating the impact of deforestation on Hg for the Palearctic region[29,30], yet these focused on measuring Hg concentrations in aquatic media and methylation potential rather than soil concentrations or atmospheric exchange. Australian soil measurements[31,32] have been made before and after vegetation burning events, but do not cover the longer term soil Hg response to deforestation.

The modeled EF estimates and their uncertainties overlap with observation-derived EFs for all 3 regions. If anything, the modeled best estimate used in online simulations is conservative compared to

available observations, showing generally lower EFs (Fig. 1). However, it is unclear whether the sparse observations available are representative of the overall region. The modeled EF uncertainty estimates cover 1–2 orders of magnitude, emphasizing the current uncertainties in the response of Hg fluxes to deforestation. Figure 1 also reveals the regions where no observations of the impact of deforestation on Hg cycling are currently available. Specifically, the Afrotropic and Indomalayan domains would be priorities for future measurement campaigns, given the current impact of deforestation in those regions (Fig. 2). It remains unknown whether Southeast Asian and African rainforests show similarly high levels of Hg in litterfall as the Amazon rainforest[33].

Section S3. Global deforestation-driven emissions estimates

We use perturbation simulations in which a set area within each region is deforested to calculate each deforestation EF. In the EF approach, we assume that 1) land-air fluxes respond linearly to deforested area and 2) spatial variability in the deforestation response within regions can be ignored. We explored the validity these assumptions using the four Amazon deforestation scenario simulations conducted in this work (Fig. S4). In the Amazon simulations — the reference simulation with 2003 forest cover (HIST), governance scenario for 2050 (GOV), business-as-usual for 2050 (BAU), and savannization (SAV) — different areas (both in spatial pattern and extent) were deforested in the Amazon region. The total fluxes from the Amazon basin for Hg^0 dry deposition, soil Hg^0 emissions, and the overall land-air balance of Hg all respond linearly ($R^2 > 0.98$) to the magnitude of the deforested area. Therefore, the approach of calculating deforestation EFs and scaling these with deforested areas would likely not be highly sensitive to the spatial distribution and amount of deforestation. Therefore, we conducted 7 other idealized deforestation simulations for the other land regions (Fig. S5).

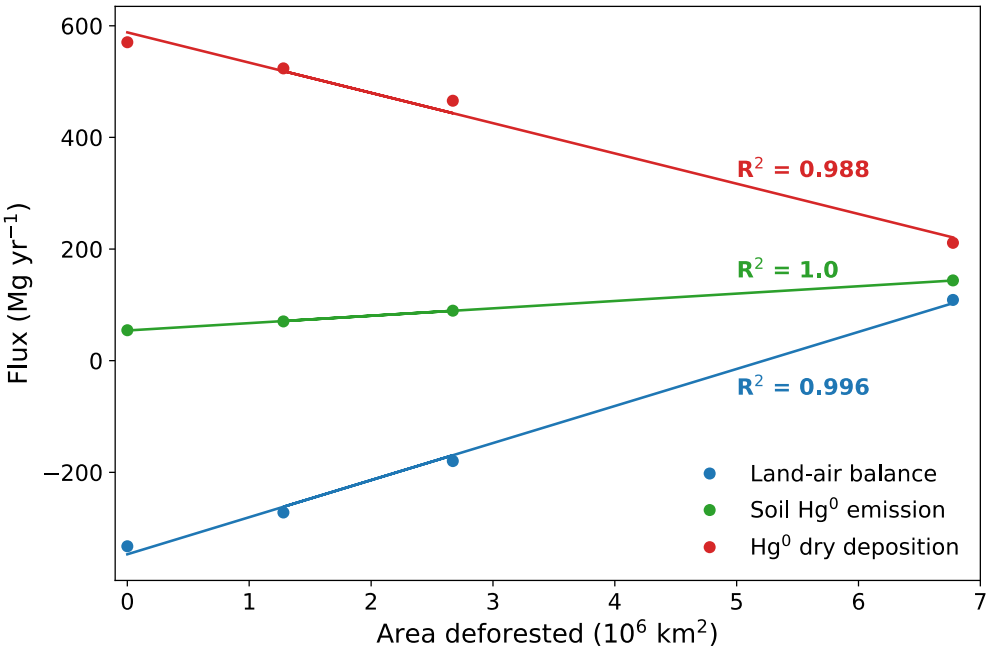


Figure S4. Relationship between land-air fluxes and the area deforested in GEOS-Chem simulations for the Amazon rainforest. Fluxes are averaged over the Amazon rainforest domain and listed R^2 values refer to linear models.

Additional data related to the calculation of historical deforestation-driven emissions of Hg are presented in this section. The maps defining the regions used in this study is shown in Fig. S5. Table S3 tabulates the results from the perturbation simulations for the different regions and the resultant emission factors. Fig. S6 explores the impact of choosing different time horizons for the deforestation area on the calculated Hg emissions globally and by country. Fig. S7 shows the map of Hg deforestation-driven emissions, assuming a 45 year time horizon (deforestation area of 1970–2014 from the LUH2 dataset[34]).

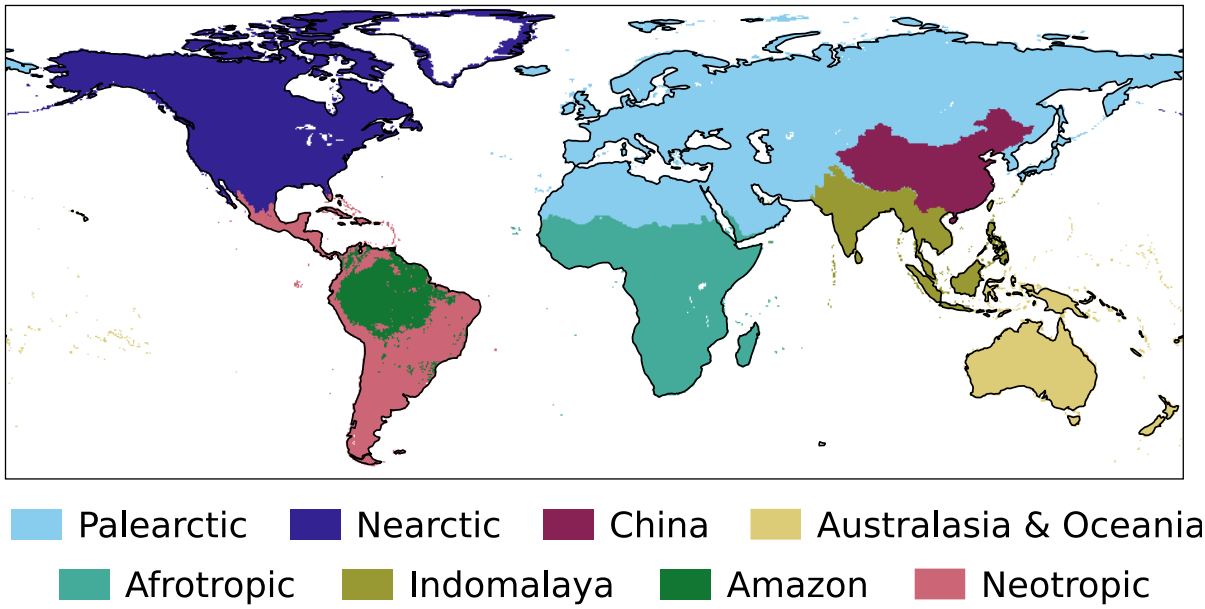


Figure S5. Definition of regions used to calculate the deforestation emission factors.

Table S3. Results from the deforestation perturbation simulations in GEOS-Chem for determining the response of land-air fluxes to deforesting a specified area. Emissions factors are listed with the 95% confidence interval calculated in offline simulations assessing the uncertainties due to model parameters (Section S4).

Realm	Area deforested (km^2)	Change in emissions ($Mg\ yr^{-1}$)	Change in deposition ($Mg\ yr^{-1}$)	Change in net emissions ($Mg\ yr^{-1}$)	Emissions factor ($Mg\ m^{-2}\ yr^{-1}$) [95% confidence interval]
Afrotropic	3 644 969	29.1	-10.0	39.1	1.1×10^{-5} [2.8×10^{-6} to 1.2×10^{-4}]
Neotropic	2 422 577	13.0	-4.9	17.9	7.4×10^{-6} [4.8×10^{-6} to 5.7×10^{-5}]
Indomalaya	2 626 474	31.6	-28.3	59.9	2.3×10^{-5} [1.5×10^{-5} to 2.1×10^{-4}]
Palearctic	4 221 663	5.8	-4.3	10.1	2.4×10^{-6} [7.6×10^{-8} to 2.3×10^{-5}]
Nearctic	4 606 898	31.6	-17.4	48.9	1.1×10^{-5} [7.1×10^{-6} to 6.2×10^{-5}]
Australasia	1 088 250	1.9	-4.8	6.6	6.1×10^{-6} [8.3×10^{-7} to 5.4×10^{-5}]
China	1 141 180	16.6	-10.1	26.7	2.3×10^{-5} [1.7×10^{-5} to 2.3×10^{-4}]
Amazon	6 775 429	96.2	-394.0	490.2	7.2×10^{-5} [4.5×10^{-5} to 2.0×10^{-4}]

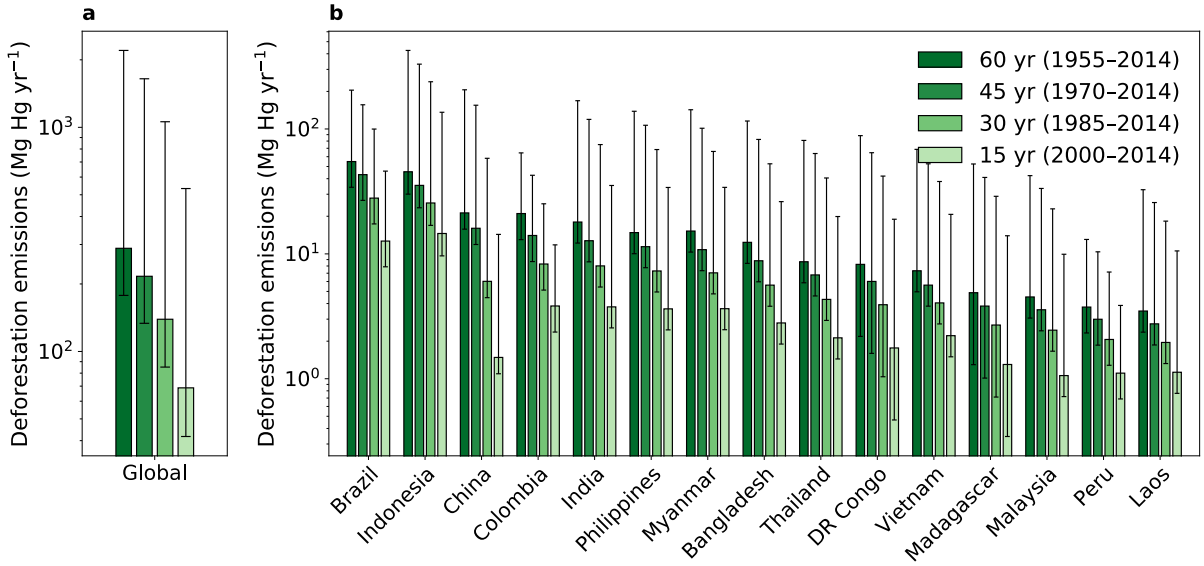


Figure S6. (*a*) Global and (*b*) country-level deforestation emissions of Hg for the top 15 emitting countries. Results are summarized accumulating deforested area over different time horizons (15 years, 30 years, 45 years, and 60 years) before 2015. Error bars refer to the 95% confidence interval based on the uncertainty in model parameters (Section S4).

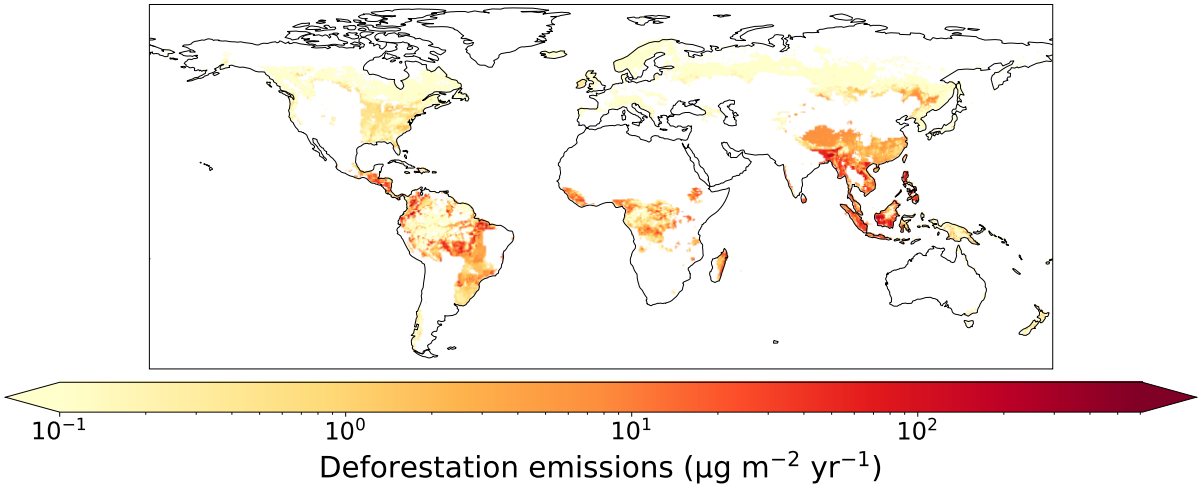


Figure S7. Map of net emissions of Hg from deforestation calculated over a 45 year time horizon before 2015 (1970–2014), using deforested area from the LUH2 dataset[34].

Section S4. Model uncertainty analysis

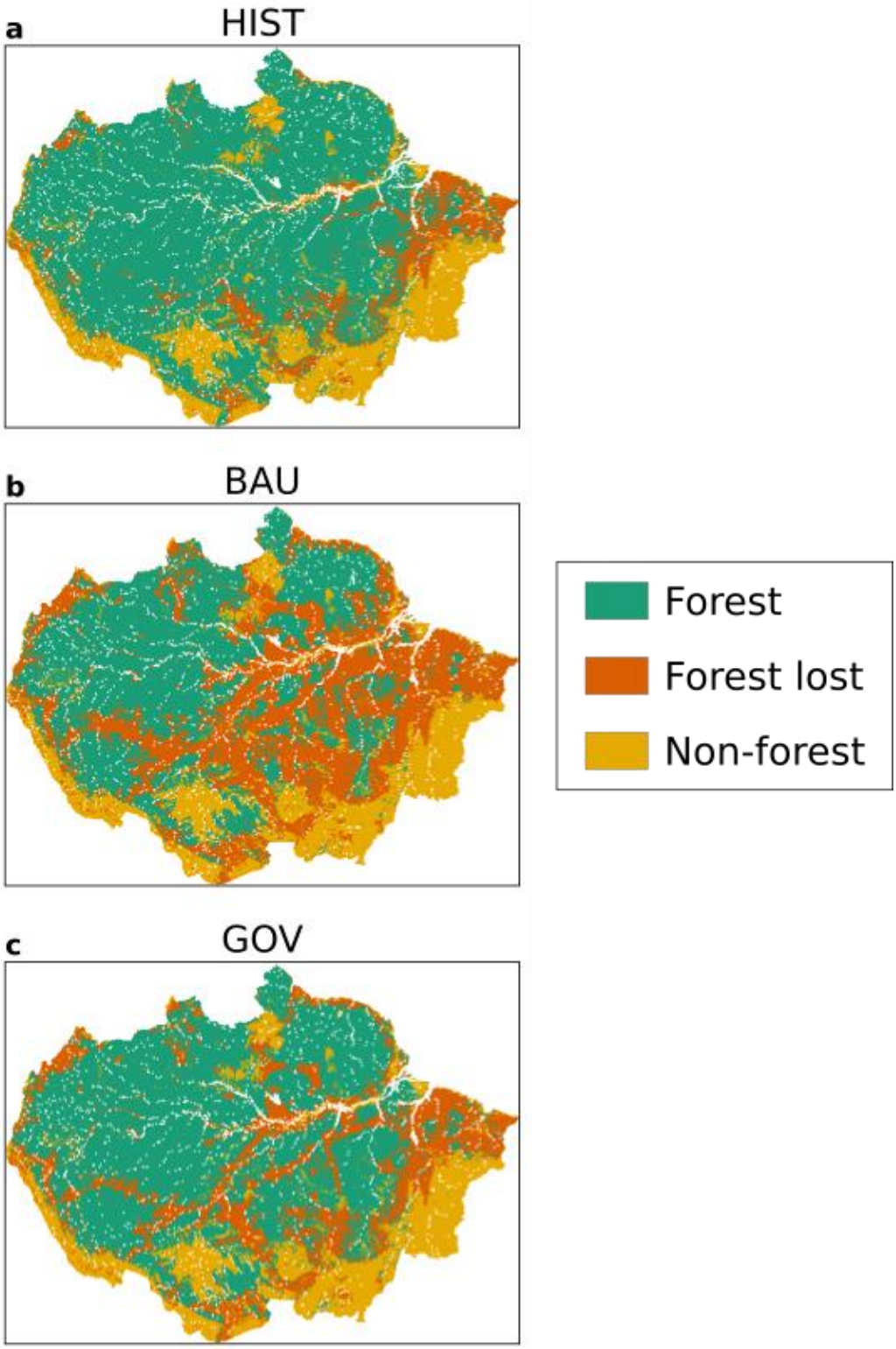
Table S4. Parameter uncertainty bounds applied in the uncertainty analysis.

Parameter	Min	Max	Units	Distribution	Comment
Soil emission parametrization	1	100	-	Uniform	Integer representing one of 100 reasonable parametrizations calculated within the range of observed uncertainties (Table S5)
Percentile of replaced LAI when building scenarios	10	90	-	Uniform	e.g., deforested Amazon area is assigned 10th percentile LAI of HIST savanna, instead of mean for default estimate
Dry deposition Hg^0 reactivity (f_0) Amazon rainforest	10^{-2}	0.5	-	Loguniform	Based on Feinberg et al.[33], within range of available vegetation uptake measurements
Dry deposition Hg^0 reactivity (f_0) other rainforests	10^{-5}	0.2	-	Loguniform	Based on Feinberg et al.[33]; no available measurements from other rainforests, leading to wider f_0 uncertainty
Dry deposition Hg^0 reactivity (f_0) elsewhere	10^{-5}	5×10^{-5}	-	Uniform	Based on Feinberg et al.[33], within range of available vegetation uptake measurements
Biomass burning emission factor for Amazon	350	615	μg m^{-2}	Uniform	Estimated range in literature[10,35,36]

Table S5. Bounds of observed parameters used to calculate 100 reasonable soil emission parametrizations, which are then applied in the uncertainty analysis (Table S4).

Parameter	Min	Max	Units	Comment
Ratio of deforested to forested Amazon soil emissions	1.8	31	-	Range from Table S1
Ratio of Amazon to extratropical soil emissions	3.5	8	-	Assume 50% error from Table S2
Extratropical grassland soil emissions	3.5	11.4	μg m^{-2} yr^{-1}	Grasslands and background soil range from literature reviews[6,7]
Deforested Amazon soil emissions	9.8	79	μg m^{-2} yr^{-1}	Range from Table S1

206 **Section S5. Scenarios for Amazon deforestation and global reforestation**



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208 **Figure S8.** Map of the Amazon basin showing the area of forest, forest loss and rangeland and
209 agriculture in (*a*) HIST; and projections for 2050 in (*b*) Business as Usual (BAU) and (*c*) Governance
210 (GOV) scenarios (replotted from Soares-Filho et al.[37] data).

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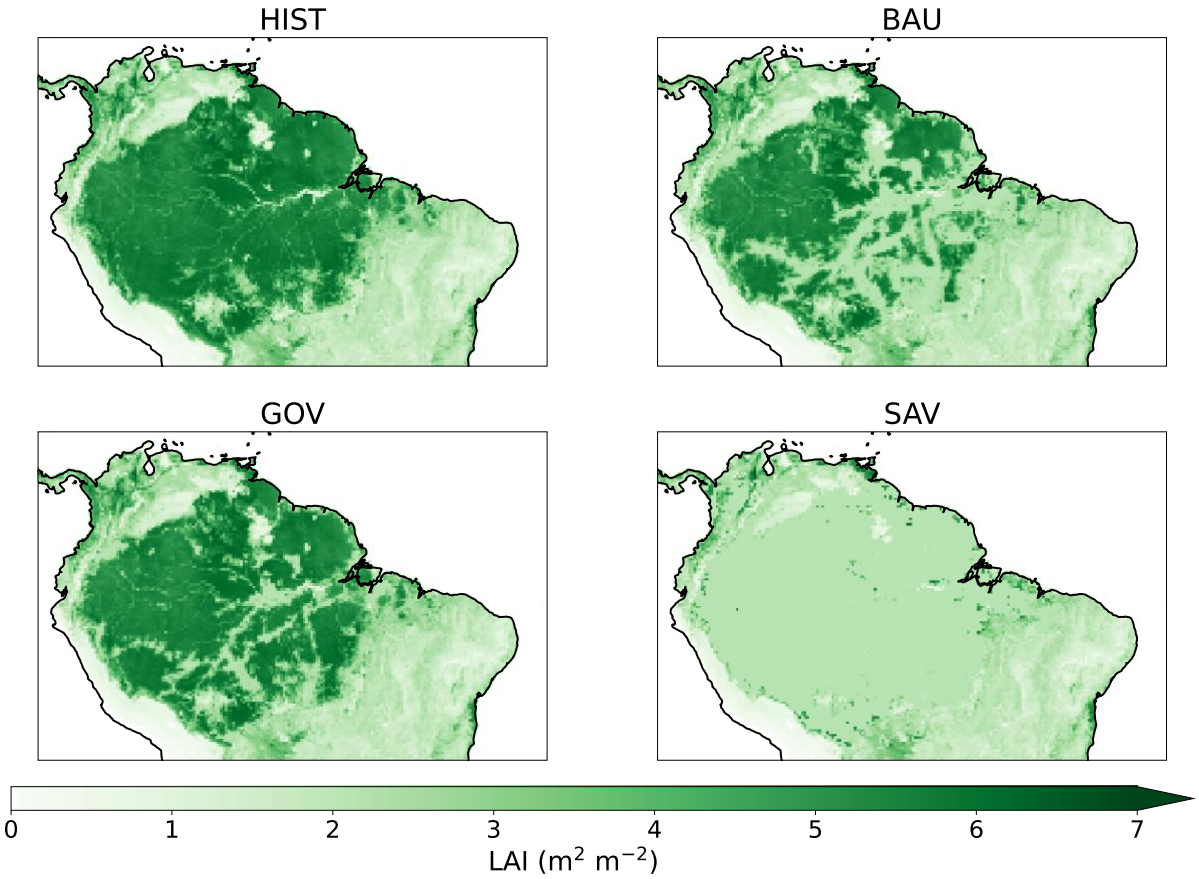

Figure S9. Annual mean leaf area index (LAI) maps for the Amazon deforestation scenarios at 0.25° × 0.25° resolution. The simulations names refer to the following scenarios: reference (HIST), Business-as-usual (BAU), Governance (GOV), and Savannization (SAV).

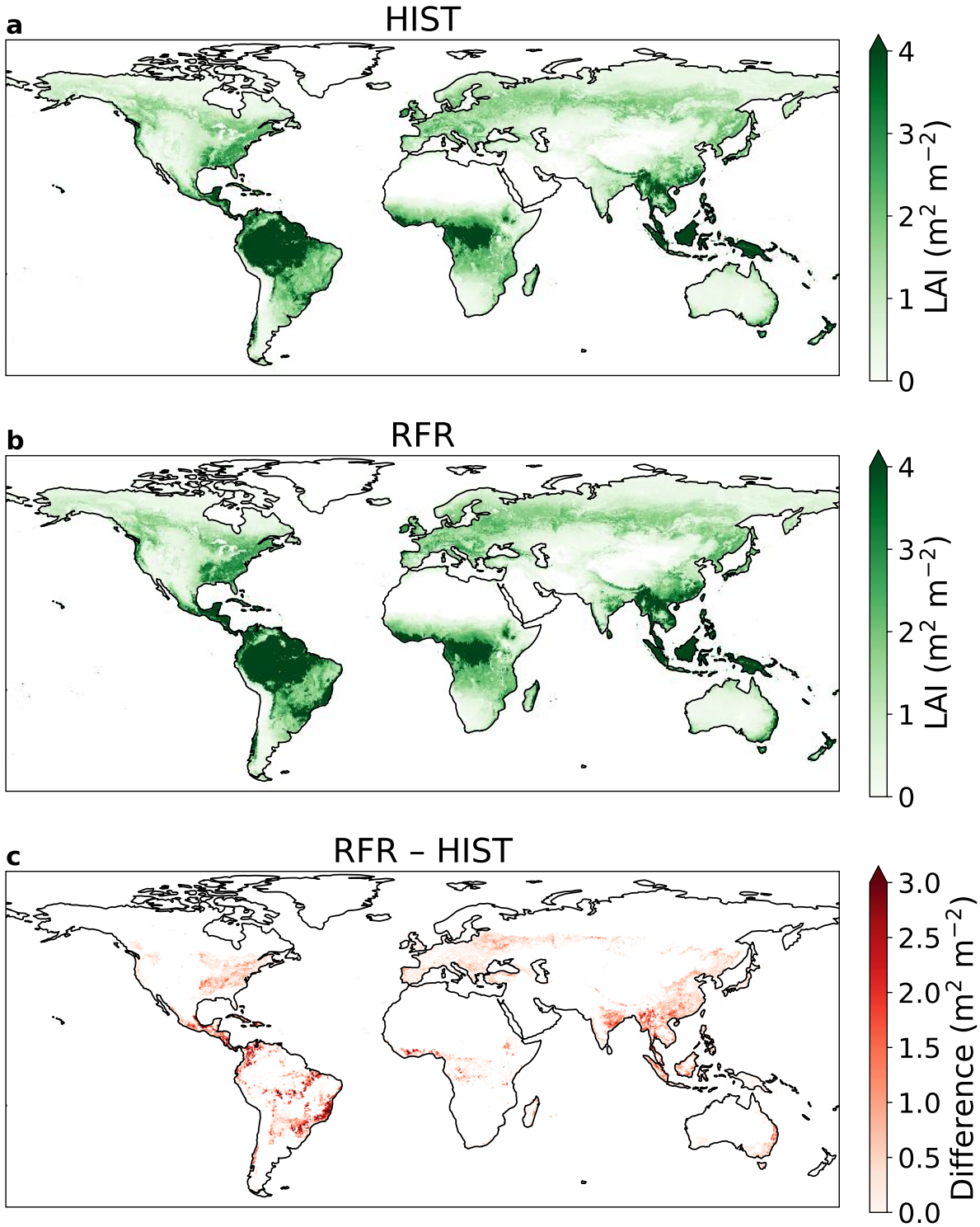

Figure S10. Annual mean leaf area index (LAI) maps at 0.25 × 0.25° resolution for: (*a*) the reference (HIST) scenario (*b*) Reforestation scenario (RFR) (*c*) Difference between RFR and HIST.

Section S6. Impact of Amazon deforestation on erosion

Previous field studies[15,38] have suggested that erosion of Hg is increased after deforestation in the Amazon, measuring enhanced runoff of Hg in deforested catchments. We estimated the change in soil displacement by water erosion (soil erosion) in the Amazon deforestation scenarios using the RUSLE-based[39] modeling platform Global Soil Erosion Modeling (GloSEM)[40,41]. As a detachment-limited soil erosion prediction model, GloSEM estimates soil erosion (expressed as a mass of soil lost per unit area and time, Mg ha^{-1} yr^{-1}) due to inter-rill and rill erosion processes by multiplication of six contributing

factors. The modeling scheme follows the same principle of most RUSLE-type models or more complex catchment-scale process-based models, with a driving force (erosivity of the climate, R), a resistance term (erodibility of the soil, K) and other factors representing the farming choice, i.e., topographical conformation of the field (LS), cropping system (C), and soil conservation practices (P).

Our approach for calculating soil erosion in the Amazon scenarios is similar to the GloSEM parametrization adopted by Borrelli et al.[40,41] to estimate human-induced soil erosion change between 2001 and 2070 at a global scale. The horizontal resolution of the native soil erosion modeling is 250 × 250 m. The calculation of erosivity (R), erodiblity (K), topographical conformation of the field (LS), and soil conservation practices (P) factors are described in Borrelli et al.[40,41]. We acknowledge that the calculation of erosion model factors for the Amazon rainforest may be associated with higher uncertainties than other regions due to the lower density in meteorological stations[42] and soil sampling sites[43]. For this study, we adapted the computation of the land cover and management factor (C-factor), which measures the combined effect of vegetation cover and cropping system variables on the soil erosion process. We parametrize the C-factor according to two layers of information: 1) the spatial dimension of land use classes according to the deforestation scenarios from Soares-Filho et al.[37] (described below); 2) the vegetation condition in each land use class using the MODIS MOD44B Vegetation Continuous Fields product (VCF) (~250m spatial resolution) as a proxy to quantify (i) surface vegetation cover, (ii) tree cover, and (iii) bare soil. As we focus our analysis on comparing the forest coverage in the years 2003 and 2050, the baseline vegetation condition is given by the average VCF values over the years 2000, 2001 and 2002. The C-factor for noncropland areas (C_{nc}) is estimated in two steps. First, a preliminary C-factor (C_p) not considering tree cover is calculated as:

$$C_p = C_{min} + \left((C_{max} - C_{min})\,\mathrm{NVS}\right) \quad \text{(S6)}$$

where the C_{min} (0.01) and C_{max} (0.15) express the potential range in C-factor values for dense to sparse grassland cover. NVS (non-vegetated surface) is spatially defined using the MODIS MOD44B VCF data normalized to a range from 0 to 1 and describes the percentage of ground covered by any vegetation type. For the NVS, the C-factor is set to 0.5. Within the next step, the final land cover and management C-factor for non-croplands (C_{nc}) is computed including the tree coverage (TC) defined using the MODIS MOD44B VCF normalized to range from 0 to 1:

$$C_{nc} = C_{p\,min} + \left(\left(C_{p\,max} - C_{p\,min}\right)\mathrm{TC}\right) \quad \text{(S7)}$$

where the $C_{p\,min}$ and $C_{p\,max}$ values are set to 0.0001 (100% canopy cover) and 0.009 (sparse forest vegetation).

While the deforestation scenarios proposed by Soares-Filho et al.[37] provide a spatial quantification of the forest losses between 2003 and 2050, the annual shares of conversion from forest to grassland or cropland are separate from the annual projection of the Land-Use Harmonization (LUH2) data[34], which provides fractional land-use patterns (850-2100) at 0.25° × 0.25° resolution. The downscaling of the LUH2 fractional cropland and grassland data from 0.25° × 0.25° resolution to the 250 m × 250 m resolution of the erosion model is performed through a probabilistic land use allocation scheme based on classification rules applied to auxiliary information (i.e., a crop suitability index, more detail in Borrelli et al.[40]). Finally, the C-factor of the cropland is defined at sub-national administrative level (Global Administrative Unit Levels) based on the Food and Agriculture Organization's (FAO) FAOSTAT database, which allowed to statistically describe typical crop rotations in each region. The C-factor of the croplands ranges from 0.131 (Northern Suriname) to 0.332 (Northeast Brazil).

Following the assumption of Lugato et al.[44] for eroded carbon, we assume that 30% of the eroded soil flux is not redeposited on land and enters riverine systems. The fraction of eroded Hg which enters aquatic systems is uncertain, depending on hillslopes dynamics and flow patterns that are not explicitly modeled by the RUSLE-based framework, as well as whether Hg would be selectively eroded relative to carbon. We recognize that this assumption introduces uncertainty into our calculations, and assume that the fraction of eroded soil which enters riverine systems can vary between 5–47%, the range reported by Van Oost et al.[45] We calculate the eroded flux of Hg from land by multiplying the soil flux by the median Hg concentration in Amazon forested soils from a literature review (86 ng g^{-1}; see SI Spreadsheet).

For each Amazon scenario, we tabulate the Hg erosion fluxes in Table S6. Erosion in the HIST scenario represents a flux of 64 Mg yr^{-1} (uncertainty range: 11–100 Mg yr^{-1}). Erosion is enhanced in the deforestation scenarios, ranging from +14% increase in GOV to a 96% increase in the extreme SAV scenario. The absolute magnitudes of erosion flux changes are smaller than the perturbations in the land-air flux, driven by changes in Hg^0 soil emissions and dry deposition (Table S6). Overall, perturbations to the erosion flux are approximately 14% of the perturbations to the land-air flux due to deforestation. A previous field study[5] has also suggested that the majority of flux changes after deforestation occurs through atmospheric exchange (97%) rather than erosion to riverine systems. Therefore, the land-air changes to the fluxes play the larger role in the impact of deforestation on the mass balance of Hg in soils. Nevertheless, changes to erosion will affect downstream Hg concentrations and the methylation potential after deforestation[5,29], which would be important to consider when assessing the impact of deforestation on local ecosystems.

Table S6. Soil erosion fluxes for the Amazon basin calculated by the erosion model GloSEM. The simulations names refer to the following scenarios: reference (HIST), Business-as-usual (BAU), Governance (GOV), and Savannization (SAV).

Scenario	**HIST**	**BAU**	**GOV**	**SAV**
Soil loss (Mt yr^{-1})	2467	3276	2816	4834
30% of soil loss (Mt yr^{-1})[a]	740	983	845	1450
[5%–47%]	[123–1159]	[164–1540]	[141–1323]	[242–2272]
Hg erosion (Mg yr^{-1})	64	85	73	125
[uncertainty range]	[11–100]	[14–132]	[12–114]	[21–195]
Change from HIST (Mg yr^{-1})	-	21	9	61
(relative change)		(+33%)	(+14%)	(+96%)
Land-air flux change from HIST (Mg yr^{-1})	-	153	61	441

[a] This is the flux assumed to be entering riverine systems

Section S7. Impacts on atmospheric Hg concentrations

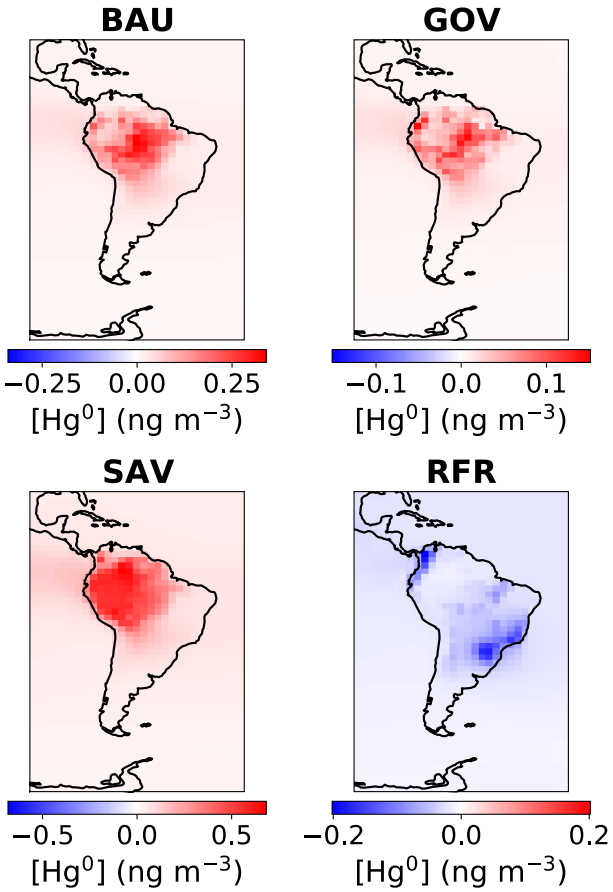

Figure S11. Annual mean differences in simulated atmospheric Hg^0 concentration at the surface between scenarios — Business-as-usual (BAU), Governance (GOV), Savannization (SAV), and global reforestation (RFR) — and the HIST reference simulation.

Supplementary References

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