Global high-resolution emissions of soil NO$_x$, sea salt aerosols, and biogenic volatile organic compounds

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Background & Summary

Natural emissions of air pollutants from surface natural processes are an essential component of the Earth system, with strong impacts on air quality, climate, and ecosystems. In particular, soil emissions of nitrogen oxides (soil NO$_x$) contributed ~50% of global NO$_x$ emissions in preindustrial times and currently contribute ~15%, and are a major source of the NO$_x$ budget outside of cities$^1$. Sea salt aerosols (SSAs) are a key player in the climate and chemistry of the marine atmosphere, and dominate the top-of-atmosphere clear sky radiative effect over the oceans$^4$. They are also an important source of halogens, provide large surface area for heterogeneous reactions, and affect ozone, nitrogen, bromine chemistry and many other pollutants$^7$. Biogenic non-methane volatile organic compounds (BVOCs), among which the most abundant species are isoprene and monoterpenes, are the dominant contributor to the global VOCs flux into the atmosphere$^8$. BVOCs affect the production of near-surface ozone in urban and surrounding areas$^9$, alter the atmospheric oxidative capacity and methane lifetime on regional and global scales$^{10,11}$, and are important precursors of carbon dioxide$^{12,13}$.

Natural emissions are nonlinearly dependent on meteorological factors such as temperature, radiation, humidity, and winds$^{14,15}$. For soil NO$_x$ and BVOCs, the properties of soils (e.g., water content, organics content, microbes, and the amount of fertilizer applied) and vegetation (e.g., type, density and physiology) are also critical$^{16,17}$. Especially for the global domain, these natural emissions are typically estimated through parameterization, due to inadequate mechanistic knowledge about emission processes as well as concerns about the computational costs needed to fully resolve such processes. Parameterizations are typically nonlinear – meaning that...
the horizontal resolution of inputted meteorological and other variables has an important influence on the calculated emission totals and spatial distributions. Parameterizations are typically embedded in three-dimensional (3-D) chemical transport models (CTMs), climate-chemistry models and earth system models to calculate natural emissions online, and are thus sensitive to model resolution. Resolution-dependent emissions are a major factor affecting the accuracy of 3-D models 23–27.

Moreover, historical records of global high-resolution (≤ 50 km) natural emissions, shown in Tables 1–3, are relatively small, hindering the understanding of variations in global emission totals, spatial distributions and their air quality and climate impacts.

Here we use the Harvard-NASA Emissions Component 28 (HEMCO) to produce monthly global emissions of soil NOx, SSAs, and BVOCs at different resolutions. These emissions are calculated at 0.5° lat. × 0.625° lon. for 1980–2017 using the MERRA-2 assimilated meteorology and at three resolutions (0.25° lat. × 0.3125° lon., 2° lat. × 2.5° lon., and 4° lat. × 5° lon.) for 2014–2017 using GEOS-FP. The datasets will be continuously updated and published. The datasets can be used to study the effects of these natural emissions on air quality, climate, and the carbon cycle, as well as the effects of horizontal resolution on emissions estimates. The datasets can be downloaded freely through Peking University Atmospheric Chemistry & Modeling Group (http://www.phy.pku.edu.cn/~acm/acmProduct.php#NATURAL-EMISSION) and Figshare 29.

Table 1. Comparison with previous studies for soil NOx emissions.

| Reference                  | Time         | Method                                      | Resolution          | Emission (Tg N/yr) |
|----------------------------|--------------|---------------------------------------------|---------------------|-------------------|
| Miyazaki, et al.           | 2005–2014    | Multi-constituent satellite data assimilation | 2.5° lat. × 2.5° lon. | 7.9               |
| Vinken, et al.             | 2005         | OML, GEOS-Chem                             | 2° lat. × 2.5° lon.  | 9–16.8            |
| Stavrakou, et al.          | 2007         | DOMINO, IMAGES CTM, MINLOSS setting        | 2° lat. × 2.5° lon.  | 9                 |
| Stavrakou, et al.          | 2007         | DOMINO, IMAGES CTM, MAXLOSS setting        | 2° lat. × 2.5° lon.  | 20.4              |
| Stavrakou, et al.          | 1997–2006    | GOME, SCIAMACHY, IMAGES CTM                | 2° lat. × 2.5° lon.  | 10–12             |
| Jaeglé, et al.             | 2000         | GOME, GEOS-Chem                            | 2° lat. × 2.5° lon.  | 7–10.8            |
| Müller and Stavrakou       | 1997         | GOME, IMAGES                               | 5° lat. × 5° lon.    | 10.9–12.1         |
| Heald, et al.              | 2000         | BNSNP                                       | 0.5° lat. × 0.667° lon. | 10                |
| Hudman, et al.             | 2006         | YL95                                        | 2° lat. × 2.5° lon.  | 6.2               |
| Hudman, et al.             | 2006         | BNSNP                                       | 2° lat. × 2.5° lon.  | 9                 |
| Steinkamp and Lawrence     | 1990–2000    | YL95EMAC                                    | –1°11′ lat. × 1°11′ lon. | 8.6               |
| Yan, et al.                | 2001         | Statistical model                           | 0.5° lat. × 0.5° lon. | 5                 |
| Ganz, et al.               | 2000         | YL95                                        | –3°75′ lat. × 3°75′ lon. | 8                 |
| Yenger and Levy            | 1990         | YL95                                        | 2° lat. × 2.5° lon.  | 3.3–7.7           |
| Müller                    | 1980         | Chemical transport model                    | 5° lat. × 5° lon.    | 4.1               |
| This Study                 | 1980–2017    | Updated BNSNP, MERRA-2                      | 0.5° lat. × 0.625° lon. | 9.5               |
| This Study                 | 2014–2017    | Updated BNSNP, GEOS-FP                      | 4° lat. × 5° lon.    | 7.1               |
| This Study                 | 2014–2017    | Updated BNSNP, GEOS-FP                      | 2° lat. × 2.5° lon.  | 7.5               |
| This Study                 | 2014–2017    | Updated BNSNP, GEOS-FP                      | 0.25° lat. × 0.3125° lon. | 8.8               |

Methods

HEMCO. The HEMCO 28 is a software package to compute pollutant emissions at user-defined resolutions. HEMCO can be run in a standalone mode or coupled to a 3-D model like GEOS-Chem. Here we use HEMCO version 2.1 at the standalone mode to calculate natural emissions based on different meteorological, ancillary variables, and nonlinear parameterizations.

Soil NOx emissions. Inside HEMCO, the algorithm for above-canopy soil NOx emissions (soil NOx) follows Hudman, et al. 2, with the efficiency of loss to canopy depending on vegetation type and density. Based on soil chamber and field measurements, soil NOx varies greatly with climate and edaphic conditions, and are most strongly correlated with N-availability, temperature, precipitation patterns, and fertilizer management practices 21,22. In the Hudman, et al. 2 algorithm, soil NOx emissions flux is a complex function of biological and meteorological drivers:

\[
S_{NOx} = A^{\prime}_{biome}(N_{avail}) \times f(T) \times g(\theta) \times P(\text{dry}, t) 
\]

\[
A^{\prime}_{biome} = A_w_{biome} + N_{avail} \times E 
\]

\[
N_{avail}(t) = N_{avail}(0) e^{-\frac{t}{\tau}} + F \times \tau \times \left( 1 - e^{-\frac{t}{\tau}} \right) 
\]
Table 2. Comparison with previous studies for sea salt emissions.

| Reference | Year | Method | Resolution | Emission (Tg/yr) |
|-----------|------|--------|------------|-----------------|
| Zhu et al. | 2011–2012 | Statistical model (Jaeglé et al.) in GEOS-Chem 12.3.0 | 4° lat. × 3° lon. | 3,140 |
| Grythe et al. | 1990–2005 | FLEXPART | 1° lat. × 1° lon. | 8,910 |
| Grythe et al. | 1980–2005 | FLEXPART fit to the observed | 1° lat. × 1° lon. | 9,000–10,800 |
| Sofiev et al. | 2001, 2008 | SILAM | 1° lat. × 1° lon. | 7,050 |
| Jaeglé et al. | 2008 | Statistical model (Gong) | 2° lat. × 2.5° lon. | 5,200 |
| Jaeglé et al. | 2008 | Statistical model (Jaeglé et al.) | 2° lat. × 2.5° lon. | 4,600 |
| This Study | 1980–2017 | Statistical model (Jaeglé et al.) in HEMCO, MERRA-2 | 0.5° lat. × 0.625° lon. | 3,560 |
| This Study | 2014–2017 | Statistical model (Jaeglé et al.) in HEMCO, GEOS-FP | 4° lat. × 5° lon. | 3,156 |
| This Study | 2014–2017 | Statistical model (Jaeglé et al.) in HEMCO, GEOS-FP | 2° lat. × 2.5° lon. | 3,239 |
| This Study | 2014–2017 | Statistical model (Jaeglé et al.) in HEMCO, GEOS-FP | 0.25° lat. × 0.3125° lon. | 3,860 |

Emissions of sea salt aerosols. Parametrization of sea salt emissions in HEMCO is modified from Jaeglé et al. It considers two categories of SSAs based on their radii. The radius of accumulation mode sea salt aerosol (SALA) ranges from 0.01 to 0.5 μm, while that for coarse mode sea salt aerosol (SALC) ranges from 0.5 to 8 μm. Parametrization of sea salt aerosols emissions includes both a wind speed and a sea surface temperature (SST) dependence. The SSAs emission flux density function dE/dr80 is formulated as follows:

\[
f(T) \times g(\theta) = e^{0.103T} \times a\theta e^{-b \theta^2}
\]

where \( A'_{\text{biome}} \), representing the biome-dependent emission factors of N in the soil, is a function of \( N_{\text{avail}} \) and the A_w,biome coefficients. \( A_{w,}\text{biome} \) is the wet biome-dependent emission factors updated based on estimates from Steinkamp and Lawrence. \( E \) is the mean emission rate of fertilizer, and is treated identically to the natural pool of N. \( N_{\text{avail}} \), representing the sum of fertilizer N and deposited N, is the mass of available nitrogen in the soil. \( f \) is the fertilizer application rate and \( \tau \) is a decay lifetime, which is chosen as 4 months based on measurements within the top 10 cm of soil. Although atmospheric deposition also contributes to the available nitrogen in soils (about ~5% globally, based on Hudman et al.), this amount can only be calculated through 3-D model simulations and is thus not accounted for here.

\[
f(T) \times g(\theta) = e^{0.103T} \times a\theta e^{-b \theta^2}
\]

The temperature dependence of soil NOx is an exponential dependence on temperature between 0 °C and 30 °C (constant at \( T > 30 \) °C), where 0.103 is the weighted average of temperature dependencies for several biomes. The parameterization for soil moisture is a Poisson function scaling, where \( \theta \) (water-filled pore space) is defined as the ratio of the volumetric soil moisture content to the porosity.

\[
P(l_{\text{dry}}, t) = [13.01 \ln(l_{\text{dry}}) - 53.6] \times e^{-ct}
\]

\( A'_{\text{biome}} \), representing the pulsed soil NOx, which occur when very dry soil is wetted resulting in a reactivation of water-stressed bacteria. The parameterization, following Yan et al., is derived from four field studies relating pulsed emissions to the length of the antecedent dry period. The rate constant \( c \) reflects the rise/fall time of the pulse (\( c = 0.068 \) h⁻¹). The value of \( l_{\text{dry}} \) is the antecedent dry period in hours.

**Biogenic VOC emissions.** Inside HEMCO, BVOCs emissions are computed by the Model of Emissions of Gases and Aerosols from Nature version 2.1 (MEGAN2.1). MEGAN2.1 includes two major components: calculation of landscape average emission factors, and algorithms describing emission responses to variations in environmental conditions. The emissions (\( F_i \)) of species \( i \) is the product of the two components summed over all vegetation types:

\[
F_i = \gamma \sum_{j} e_i, j \chi_j
\]
The emission activity factor \( \gamma_j \) is assigned a value that results in \( F_i \) for vegetation type \( j \) at standard conditions (leaf temperature = 297 K; air temperature = 303 K; the photosynthetic photon flux density averaged over the past 24 h is equal to 200 \( \mu \)mol m\(^{-2}\)s\(^{-1}\) for sun leaves and 50 \( \mu \)mol m\(^{-2}\)s\(^{-1}\) for shade leaves), and \( \chi_j \) is the fractional grid box areal coverage for the same vegetation type. The emission factor accounts for the estimated in-canopy deposition flux so that \( F_i \) represents the net above-canopy flux.

The emission activity factor \( \gamma_j \) reflects the emission response to environmental drivers. The canopy environment coefficient (C) is assigned a value that results in \( \gamma_j = 1 \) for the standard conditions and is dependent on the canopy environment model being used. A detailed description of the model parameterizations for light \( (\gamma_{\text{L}}) \), temperature \( (\gamma_{\text{T}}) \), leaf age \( (\gamma_{\text{A}}) \), soil moisture \( (\gamma_{\text{S}}) \), leaf area index \( (L) \) and CO2 inhibition \( (\gamma_{\text{C}}) \) can be obtained from Guenther\(^4\).

Our BVOCs emission dataset includes isoprene (ISOP, the most abundant species), acetone (ACET), acetaldehyde (ALD\(_2\)), ethene (C\(_2\)H\(_4\)), ethanol (EOH), propene (PRPE), lumped monoterpenes (MTPA, sum of \( \alpha \) pinene, \( \beta \) pinene, sabinene and carene), other monoterpenes (MTPO, sum of myrcene, ocimene and other monoterpenes), limonene (LIMO), and sesquiterpenes (SESQ, sum of farnesene, \( \beta \) caryophyllene and other sesquiterpenes).

**LAI data for calculation of soil NO\(_x\) and BVOCs emissions.** Vegetation composition is principal information needed to estimate BVOCs and soil NO\(_x\) emissions\(^4,15\). The density of vegetation is represented in the parameterizations by leaf area index (LAI), which is defined as the amount of leaf area per unit surface of the ground (m\(^2\)m\(^{-2}\)). We use monthly MODIS-derived LAI with gap filling and smoothing described by Yuan, et al.\(^{40}\) (here after referred to as Yuan LAI). For 2005–2017, we use year-specific Yuan LAI data. For years prior to 2005, we use the LAI values in 2005 due to lack of year-specific data. This would introduce certain uncertainty for these earlier years. We did a test to fix LAI to a certain year, and the effect on global emissions is relatively small (within 5%). Therefore, our extrapolation of LAI data before 2005 does not significantly affect the emissions time series.

**Data Records**

Our datasets contain 4 data records for monthly global gridded emissions. Each record contains monthly emission data for soil NO\(_x\), 2 SSAs species (SALA and SALC), and 10 BVOCs species (ISOP, ACET, ALD\(_2\), C\(_2\)H\(_4\), EOH, PRPE, MTPA, MTPO, LIMO, and SESQ). Our data were constructed in nc file format which can be read by many tools like IDL, MatLab, and so on. Of these,
One is from 2014 to 2017 based on GEOS-FP at 2° lat. ALD2, C2H4, EOH, PRPE, MTPA, MTPO, LIMO, and SESQ). The total BVOCs exhibits strong seasonality and 1990s than in later years (with a difference by about 10%), and by a modest seasonality (within 15%).

Ocean. The temporal variation of global total SSAs emissions is characterized by lower values in the 1980s and the North Atlantic in January and over the Indian Ocean in July. Emissions are also strong over the Southern seasons, and Figure 2e shows the temporal profile of the respective global total emissions. SSAs are the largest over

The parameterized nonlinear relationships between emissions and controlling factors means that the horizontal resolution of inputted meteorological and other variables has important influences on the calculated emission magnitudes and spatial distributions.

Table 5 presents the global annual total emissions of soil NOx, SSAs, and BVOCs derived based on GEOS-FP at different resolutions (4° lat. × 5° lon., 2° lat. × 2.5° lon., and 0.25° lat. × 0.625° lon.) over 2014–2017. The resolution dependence of emission magnitude is evident especially for soil NOx and SSAs, that is, a higher resolution inputted data instead of hourly data would reduce the global ISOP emission total by 7%.

Table 4. Global annual total emissions of soil NOx, SSAs, and BVOCs (with standard deviation) over 1980–2017 derived based on MERRA-2 at 0.5° lat. × 0.625° lon.

| Species                              | 1980–2017 mean (Tg/yr) | Rel. contribution (%) | Maximum (Tg/yr) | Minimum (Tg/yr) |
|--------------------------------------|------------------------|-----------------------|-----------------|-----------------|
| Soil NOx (TgN/year)                  | 9.5 ± 0.4              | 100%                  | 10.5            | 8.5             |
| Sea salt aerosols (Tg/year)          |                        |                       |                 |                 |
| Accumulation mode                    | 57.9 ± 2.6             | 1.6%                  | 62.3            | 52.0            |
| Coarse mode                          | 3,502 ± 157            | 98.4%                 | 3,771           | 3,130           |
| Biogenic VOCs (TgC/year)             |                        |                       |                 |                 |
| Isoprene                             | 345 ± 17               | 61.2%                 | 381             | 315             |
| Acetone                              | 247 ± 0.9              | 4.4%                  | 266             | 23.1            |
| Acetaldehyde                        | 9.2 ± 0.4              | 1.6%                  | 9.9             | 8.7             |
| Ethene                               | 19.1 ± 0.8             | 3.4%                  | 20.7            | 17.7            |
| Ethanol                              | 8.8 ± 0.3              | 1.6%                  | 9.5             | 8.3             |
| Propene                              | 16.9 ± 0.6             | 3.0%                  | 18.1            | 15.9            |
| Lumped monoterpenes                  | 75.7 ± 3.1             | 13.5%                 | 82.0            | 70.4            |
| Other monoterpenes                   | 36.0 ± 1.5             | 6.4%                  | 39.3            | 33.3            |
| Limonene                             | 8.2 ± 0.3              | 1.5%                  | 8.8             | 7.7             |
| Sesquiterpenes                       | 19.1 ± 1.2             | 3.4%                  | 21.6            | 16.9            |

Table 4 presents the global annual total emissions of soil NOx, SSAs, and BVOCs over 1980–2017 derived from MERRA-2 at 0.5° lat. × 0.625° lon. Averaged over all years, global total soil NOx emissions amounts to 9.5 TgN/yr. Global total SSAs reaches 3,560 Tg/yr, as contributed by SALC (98.4%) and SALA (1.6%). Global total BVOCs emission reaches 563 TgC/yr, as contributed by ISOP (61.2%), ACET (4.4%), ALD 2 (1.6%), C2H4 (3.4%), EOH (1.6%), PRPE (3.0%), MTPA (13.5%), MTPO (6.4%), LIMO (1.5%), and SESQ (3.4%).

Figure 1a–d shows the MERRA-2 based spatial distribution of soil NOx emissions at 0.5° lat. × 0.625° lon. in January, April, July, and October averaged over 1980–2017. High values of soil NOx emissions move between the two hemispheres as a result of the seasonal variation of temperature. Spatially, the highest emissions occur over regions with intensive agricultural activities, e.g., the Ganges River Basin of India and the North China Plain. Figure 1e further shows the temporal profile of global monthly total emissions (blue line) and annual total emissions (red line), which indicates the strong seasonality (with a July to January ratio of 2.5) and interannual variability (with maximum values in the early 2000s).

Figure 2a–d represents the spatial distribution of total SSAs (sum of SALA and SALC) emissions in different seasons, and Fig. 2e shows the temporal profile of the respective global total emissions. SSAs are the largest over the North Atlantic in January and over the Indian Ocean in July. Emissions are also strong over the Southern Ocean. The temporal variation of global total SSAs emissions is characterized by lower values in the 1980s and 1990s than in later years (with a difference by about 10%), and by a modest seasonality (within 15%).

Figure 3 shows the spatial and temporal distributions of total BVOCs emissions (sum of ISOP, ACET, ALD2, C2H4, EOH, PRPE, MTPA, MTPO, LIMO, and SESQ). The total BVOCs emissions exhibits strong seasonality and cross-hemispheric seasonal migration (Fig. 3a–d) because of changes in radiation and temperature. The highest emissions occur over the Amazon, Southeast Asia, Southeast United States, and Central Africa. The global total emission also exhibits a large seasonality, with a July to January ratio of 1.3, due to variation of LAI, especially in the Northern Hemisphere. The interannual variation is modest (within 20%) (Fig. 3e).

The parameterized nonlinear relationships between emissions and controlling factors means that the horizontal resolution of inputted meteorological and other variables has important influences on the calculated emission magnitudes and spatial distributions.

Table 5 presents the global annual total emissions of soil NOx, SSAs, and BVOCs derived based on GEOS-FP × 0.625° lon. [Available in Hongjian and Jintai29, File 'MERRA-2_0.5×0625_monthly_1980–2017.nc']

One is from 2014 to 2017 based on GEOS-FP at 0.25° lat. [Available in Hongjian and Jintai29, File 'GEOS-FP_025×03125_monthly_2014–2017.nc']

One is from 2014 to 2017 based on GEOS-FP at 2° lat. × 2.5° lon. [Available in Hongjian and Jintai29, File GEOS-FP_2×25_monthly_2014–2017.nc]

One is from 2014 to 2017 based on GEOS-FP at 4° lat. × 5° lon. [Available in Hongjian and Jintai29, File GEOS-FP_4×5_monthly_2014–2017.nc]
Figure 4 shows the temporal profile of monthly global total emissions of soil NO\textsubscript{x} (a), sea salt (b), and biogenic VOCs (c) over 2014–2017 derived based on GEOS-FP at 4°lat. × 5° lon. (gray line), 2° lat. × 2.5° lon. (blue line), and 0.25° lat. × 0.3125° lon. (red line). Although the total emissions of soil NO\textsubscript{x} and sea salt increase with the horizontal resolution, the interannual and seasonal variability and trends are similar at different horizontal resolutions.

Figure 5 shows the 2014–2017 average spatial distributions of annual emissions of soil NO\textsubscript{x}, SSAs (SALA + SALC), and total BVOCs (summed over all species) estimated at different resolutions based on GEOS-FP. Figures 6–8 further show the respective spatial distributions of emission differences and percentage differences from 0.25° lat. × 0.3125° lon. to coarser resolutions. As the resolution increases, fine scale patterns of emissions become much more evident, which has important implications for air quality simulations. For soil NO\textsubscript{x}, northern India and North China, which are major source regions, exhibit the largest resolution dependence.
for absolute emission differences (Fig. 6a,b). The percentage difference is most evident along the coasts where a fine resolution (0.25° lat. × 0.3125° lon.) resolves the land-ocean contrast much better than coarser resolutions do (Fig. 6c,d). For sea salt emissions, the major source regions at high latitudes of both hemispheres exhibit a large resolution dependence (Fig. 7). For BVOCs, tropical regions have the largest resolution dependence in terms of absolute difference (Fig. 8a,b), while the coastal and low-emission regions exhibit the largest resolution dependence in terms of percentage difference (Fig. 8c,d).

Figure 9 further shows the resolution dependence of calculated regional annual emission totals over eight major regions. Compared to results for global total emissions in Table 5, the resolution dependence of emission magnitude in some regions is more evident. For Southeast Asia, soil NOx emissions total at 0.25° lat. × 0.3125° lon. is higher than that at 4° lat. × 5° lon. by 38%. Similar results are shown for Europe (38% higher) and Australia (37% higher). For sea salt, emissions for North Hemisphere Africa and Southeast Asia increase by 38% and 30%,
respectively, from 4° lat. × 5° lon. to 0.25° lat. × 0.3125° lon. The resolution dependence of regional emissions is smaller for BVOCs (within 10% for all regions) than for soil NOx and SSAs.

Table and figure data are available from Peking University Atmospheric Chemistry & Modeling Group, including global and regional monthly totals and spatial distributions from 1980 to 2017 derived based on MERRA-2 at 0.5° lat. × 0.625° lon., as well as respective results from 2014 to 2017 derived based on GEOS-FP at 0.25° lat. × 0.3125° lon., 2° lat. × 2.5° lon., and 4° lat. × 5° lon.

**Technical Validation**

**Uncertainty.** A major source of uncertainty in our calculated emission data is the use of parameterization as an approximate of the complex processes involved in the emissions of these species. Parameterization is also sensitive to errors in the inputted meteorological and ancillary data.
The parameterization of soil NO\textsubscript{x} emissions includes a continuous dependence on soil moisture and temperature, a representation of biogeochemistry that induces pulsing of the emissions following dry spells, and a detailed spatial and temporal representation of N-inputs both from chemical/manure fertilizer and atmospheric N-deposition (not included here). Our sensitivity test for 2017 at GEOS-FP 4° lat. \times 5° lon. shows that a 1 °C increase in temperature would lead to 5.2% increase in the calculated global total emission, a 10% increase in soil moisture would lead to 15.8% decrease in emission, and a 10% increase in LAI would lead to 1% decrease in emission. Sensitivity tests for other resolutions show similar results.

For sea salt emissions, the strong power law relationship with wind speed and the polynomial relationship with SST mean that errors in wind speed and SST have a significant impact on calculated sea salt emission. Based on our sensitivity test for 2017 at GEOS-FP 4° lat. \times 5° lon., a 10% increase in wind speed would lead to 38.4% increase in the calculated global emission total, and a 1 °C increase in SST would lead to 6.7% increase in emission.

### Table 5.

Global annual total emissions of soil NO\textsubscript{x}, SSAs, and BVOCs over 2014–2017 derived based on GEOS-FP at three resolutions. The percentage values represent the relative changes from emissions at 4° lat. \times 5° lon.

| Species                        | 4° lat. \times 5° lon. | 2° lat. \times 2.5° lon. | 0.25° lat. \times 0.3125° lon. |
|--------------------------------|------------------------|--------------------------|-------------------------------|
| Soil NO\textsubscript{x} (TgN/year) | 7.1                    | 7.5 (+5.6%)              | 8.8 (+23.9%)                  |
| Accumulation mode              | 51.3                   | 52.6 (+2.5%)             | 62.7 (+22.2%)                 |
| Coarse mode                    | 3,105                  | 3,186 (+2.6%)            | 3,797 (+22.3%)                |
| Sea salt aerosols (Tg/year)    |                        |                          |                               |
| Isoprene                       | 330                    | 333 (+0.9%)              | 341 (+3.3%)                   |
| Acetone                        | 27.0                   | 26.2 (-3.0%)             | 24.8 (-8.1%)                  |
| Acetaldehyde                   | 9.8                    | 9.5 (-3.1%)              | 9.3 (-5.1%)                   |
| Ethene                         | 20.4                   | 19.9 (-2.5%)             | 18.9 (-7.4%)                  |
| Ethanol                        | 9.4                    | 9.1 (-3.2%)              | 8.9 (-5.3%)                   |
| Propene                        | 18.5                   | 18.0 (-2.7%)             | 17.0 (-8.1%)                  |
| Lumped monoterpenes            | 80.1                   | 78.6 (-1.9%)             | 75.7 (-5.5%)                  |
| Other monoterpenes             | 37.8                   | 37.1 (-1.9%)             | 35.8 (-5.3%)                  |
| Limonene                       | 8.1                    | 8.2 (+1.2%)              | 8.3 (+2.5%)                   |
| Sesquiterpenes                 | 20.6                   | 20.0 (-2.9%)             | 19.2 (-6.8%)                  |

Table 5

| Biogenic VOCs (TgC/year) | 4° lat. × 5° lon. | 2° lat. × 2.5° lon. | 0.25° lat. × 0.3125° lon. |
|--------------------------|-------------------|---------------------|---------------------------|
| Isoprene                 | 330 (+0.9%)       | 333 (+0.9%)         | 341 (+3.3%)               |
| Acetone                  | 27.0 (-3.0%)      | 26.2 (-3.0%)        | 24.8 (-8.1%)              |
| 2° lat. × 2.5° lon.      |                   |                     |                           |
| Acetaldehyde             | 9.8 (-3.1%)       | 9.5 (-3.1%)         | 9.3 (-5.1%)               |
| Ethene                   | 20.4 (-2.5%)      | 19.9 (-2.5%)        | 18.9 (-7.4%)              |
| Ethanol                  | 9.4 (-3.2%)       | 9.1 (-3.2%)         | 8.9 (-5.3%)               |
| Propene                  | 18.5 (-2.7%)      | 18.0 (-2.7%)        | 17.0 (-8.1%)              |
| Lumped monoterpenes      | 80.1 (-1.9%)      | 78.6 (-1.9%)        | 75.7 (-5.5%)              |
| Other monoterpenes       | 37.8 (-1.9%)      | 37.1 (-1.9%)        | 35.8 (-5.3%)              |
| Limonene                 | 8.1 (+1.2%)       | 8.2 (+1.2%)         | 8.3 (+2.5%)               |
| Sesquiterpenes           | 20.6 (-2.9%)      | 20.0 (-2.9%)        | 19.2 (-6.8%)              |

Fig. 4

Temporal profiles of monthly global total emissions of soil NO\textsubscript{x} (a), sea salt (b) and BVOCs (c) over 2014–2017 derived based on GEOS-FP at different resolutions.
emission. This is consistent with the evident dependence of calculated emissions to the horizontal resolution of inputted meteorological data. By comparison, an increase in the shape parameter by 10% (from constant 30 to 33) would lead to a 0.1% increase in the calculated emissions.

The parameterization of BVOCs emissions involves meteorological (temperature, solar radiation, humidity, wind speed and soil moisture), land cover data (LAI and PFT fractions) and the PFT-specific average emission factor at standard conditions. According to Guenther, et al., uncertainties associated with the global annual emissions of several compounds (isoprene, acetone and acetaldehyde) are about a factor of two while estimates of uncertainties in land cover and meteorological variables are also important. Wang, et al., showed that an average bias of about 2 °C in temperature is associated with an error in isoprene emissions by ~23% in the Pearl River Delta of China. The error in LAI and its extrapolation to 1980–2004 leads to an additional uncertainty in calculated emissions. Our sensitivity test for 2017 at GEOS-FP 4° lat. × 5° lon. shows that a 1 °C increase in temperature would lead to a 12.9% increase in the calculated global total BVOCs, and a 10% increase in LAI would lead to a 4.6% increase in emission.

Comparison with existing emission estimates. Comparisons with existing emission estimates are mainly for our results derived based on MERRA-2, which contain much longer data records than those based on GEOS-FP.

As shown in Table 1, global total above-canopy soil NOx emissions are estimated at 3.3–10 TgN/yr in previous bottom-up studies and at 7.9–16.8 TgN/yr for satellite-based top-down estimates. The Yienger and Levy II43 bottom-up algorithm used in many CTM simulations results in global soil NOx emissions of 3.3–7.7 TgN/yr depending on parameters used. Updating upon Yienger and Levy II43, Steinkamp and Lawrence30 use a new biome type land-cover map and improved emission factors, resulting in an estimate of 8.6 TgN/yr. Hudman, et al., further includes a more physical parameterization that takes into account the pulsing, soil moisture and temperature dependence. Based on Hudman, et al., parameterization and MERRA-2 meteorological data, our calculated soil NOx emissions are 9.5 TgN/yr averaged over 1980–2017, within the (wide) range of values in previous bottom-up and top-down estimates.
Fig. 6 Spatial distributions of annual soil NO$_x$ emissions differences between 0.25° lat. × 0.3125° lon. and coarser resolutions over 2014–2017 based on GEOS-FP. Case_0.25 represents emissions at 0.25° lat. × 0.3125° lon. Case_4 represents emissions re-gridded from 4° lat. × 5° lon. to 0.25° lat. × 0.3125° lon. Case_2 represents emissions re-gridded from 2° lat. × 2.5° lon. to 0.25° lat. × 0.3125° lon. Percentage differences are calculated as $2 \times \frac{(\text{Case}_{0.25} - \text{Case}_4)}{(\text{Case}_{0.25} + \text{Case}_4)} \times 100\%$ and $2 \times \frac{(\text{Case}_{0.25} - \text{Case}_2)}{(\text{Case}_{0.25} + \text{Case}_2)} \times 100\%$.

Fig. 7 Similar to Fig. 6 but for total sea salt emissions.
Table 2 shows that our global sea salt aerosols emission total (3,560 Tg/yr, based on MERRA-2 for 1980–2017) is in lower end of previous estimates (3,140–10,800 Tg/yr), but is in the middle of the range presented in the IPCC Fifth Assessment Synthesis Report (1,400–6,800 Tg/yr). It is reduced from the estimate (4,300 Tg, based on GEOS-4 winds for 2003) by Jaeglé, et al., by 20%. The decrease is due to the difference in meteorological field data (especially for winds) and some recent updates of sea salt simulation (http://wiki.seas.harvard.edu/geos-chem/index.php/Sea_salt_aerosols#Recent_Updates_to_sea_salt_simulation). In particular, Jaeglé, et al., included one accumulation bin (0.01–0.5 μm) and two coarse mode bins (0.5–4 μm; 4–10 μm), whereas we use one accumulation bin (0.1–0.5 μm) and one coarse bin (0.5–8 μm) here.

The existing estimates of total BVOCs range from 200 to 1,000 TgC/yr depending on the meteorological and vegetation datasets used, and our estimate (563 TgC/yr averaged over 1980–2017 based on MERRA-2) is within this range. Table 3 further compares our estimate of global isoprene emission total with others. Our global isoprene emission total (330–345 TgC/yr) is at the lower end of previous bottom-up estimates (303–529 TgC/yr).
TgC/yr), although it is consistent with those used in various versions of GEOS-Chem (http://wiki.seas.harvard.edu/geos-chem/index.php/Benchmark/GEOS-Chem_12.5.0#GEOS-Chem_Classic_1-month_benchmark). Our isoprene emission total is larger than a recent top-down estimate based on formaldehyde measurements from the Ozone Monitoring Instrument (240 TgC/yr for 2005–2013)17, but lower than an earlier top-down estimate (566 TgC/yr for 1996–1997)46. The tropical ecosystems are a crucial factor affecting these estimates17.

Code availability
The official HEMCO code is a collection of FORTRAN-90 routines that are freely available at http://wiki.geoschem.org/HEMCO. The official HEMCO code with a few necessary changes (based on HEMCO v2.1) to run at the standalone mode are available at Peking University Atmospheric Chemistry & Modeling Group (http://www.phy.pku.edu.cn/~acm/acmProduct.php#NATURAL-EMISSION) and Figshare29.

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Competing interests
The authors declare no competing interests.

Additional information
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