

# A comprehensive quantification of global nitrous oxide sources and sinks


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Nitrous oxide (N<sub>2</sub>O), like carbon dioxide, is a long-lived greenhouse gas that accumulates in the atmosphere. Over the past 150 years, increasing atmospheric N<sub>2</sub>O concentrations have contributed to stratospheric ozone depletion<sup>1</sup> and climate change<sup>2</sup>, with the current rate of increase estimated at 2 per cent per decade. Existing national inventories do not provide a full picture of N<sub>2</sub>O emissions, owing to their omission of natural sources and limitations in methodology for attributing anthropogenic sources. Here we present a global N<sub>2</sub>O inventory that incorporates both natural and anthropogenic sources and accounts for the interaction between nitrogen additions and the biochemical processes that control N<sub>2</sub>O emissions. We use bottom-up (inventory, statistical extrapolation of flux measurements, process-based land and ocean modelling) and top-down (atmospheric inversion) approaches to provide a comprehensive quantification of global N<sub>2</sub>O sources and sinks resulting from 21 natural and human sectors between 1980 and 2016. Global N<sub>2</sub>O emissions were 17.0 (minimum–maximum estimates: 12.2–23.5) teragrams of nitrogen per year (bottom-up) and 16.9 (15.9–17.7) teragrams of nitrogen per year (top-down) between 2007 and 2016. Global human-induced emissions, which are dominated by nitrogen additions to croplands, increased by 30% over the past four decades to 7.3 (4.2–11.4) teragrams of nitrogen per year. This increase was mainly responsible for the growth in the atmospheric burden. Our findings point to growing N<sub>2</sub>O emissions in emerging economies—particularly Brazil, China and India. Analysis of process-based model estimates reveals an emerging N<sub>2</sub>O–climate feedback resulting from interactions between nitrogen additions and climate change. The recent growth in N<sub>2</sub>O emissions exceeds some of the highest projected emission scenarios<sup>3,4</sup>, underscoring the urgency to mitigate N<sub>2</sub>O emissions.

Nitrous oxide (N<sub>2</sub>O) is a long-lived stratospheric ozone-depleting substance and greenhouse gas with a current atmospheric lifetime of  $116 \pm 9$  years<sup>1</sup>. The concentration of atmospheric N<sub>2</sub>O has increased by more than 20% from 270 parts per billion (ppb) in 1750 to 331 ppb in 2018 (Extended Data Fig. 1), with the fastest growth observed in the past five decades<sup>5,6</sup>. Two key biochemical processes—nitrification and denitrification—control N<sub>2</sub>O production in both terrestrial and aquatic ecosystems and are regulated by multiple environmental and biological factors including temperature, water and oxygen levels,

acidity, substrate availability<sup>7</sup> (which is linked to nitrogen fertilizer use and livestock manure management) and recycling<sup>8–10</sup>. In the coming decades, N<sub>2</sub>O emissions are expected to continue to increase as a result of the growing demand for food, feed, fibre and energy, and an increase in sources from waste generation and industrial processes<sup>4,11,12</sup>. Since 1990, anthropogenic N<sub>2</sub>O emissions have been reported annually by Annex I Parties to the United Nations Framework Convention on Climate Change (UNFCCC). More recently, over 190 national signatories to the Paris Agreement have been required to report biannually their

A list of affiliations appears at the end of the paper.

national greenhouse-gas inventory with sufficient detail and transparency to track progress towards their nationally determined contributions. However, these inventories do not provide a full picture of N<sub>2</sub>O emissions owing to their omission of natural sources, the limitations in methodology for attributing anthropogenic sources, and missing data for a number of key regions (for example, South America and Africa)<sup>2,9,13</sup>. Moreover, a complete account of all human activities that accelerate the global nitrogen cycle and that interact with the biochemical processes controlling the fluxes of N<sub>2</sub>O in both terrestrial and aquatic ecosystems is required<sup>2,8</sup>. Here we present a comprehensive, consistent analysis and synthesis of the global N<sub>2</sub>O budget across all sectors, including natural and anthropogenic sources and sinks, using both bottom-up and top-down methods and their cross-constraints. Our assessment enhances understanding of the global nitrogen cycle and will inform policy development for N<sub>2</sub>O mitigation, which could help to curb warming to levels consistent with the long-term goal of the Paris Agreement.

A reconciling framework (described in Extended Data Fig. 2) was used to take full advantage of bottom-up and top-down approaches for estimating and constraining sources and sinks of N<sub>2</sub>O. Bottom-up approaches include emission inventories, spatial extrapolation of field flux measurements, nutrient budget modelling and process-based modelling for land and ocean fluxes. The top-down approaches combine measurements of N<sub>2</sub>O mole fractions with atmospheric transport models in statistical optimization frameworks (inversions) to constrain the sources. Here we constructed a total of 43 flux estimates, including 30 using bottom-up approaches, 5 using top-down approaches, and 8 other estimates using observation and modelling approaches (Methods, Extended Data Fig. 2).

With this extensive data and bottom-up/top-down framework, we established comprehensive global and regional N<sub>2</sub>O budgets that include 18 sources and various different chemical sinks. These sources and sinks are further grouped into six categories (Fig. 1, Table 1): (1) natural sources (no anthropogenic effects) including a very small biogenic surface sink; (2) perturbed fluxes from ecosystems induced by changes in climate, carbon dioxide (CO<sub>2</sub>) and land cover; (3) direct emissions from nitrogen additions in the agricultural sector (agriculture); (4) other direct anthropogenic sources—including fossil fuel and industry, waste and waste water, and biomass burning; (5) indirect emissions from ecosystems that are either downwind or downstream from the initial release of reactive nitrogen into the environment—including N<sub>2</sub>O release after transport and deposition of anthropogenic nitrogen via the atmosphere or water bodies as defined by the Intergovernmental Panel on Climate Change (IPCC)<sup>14</sup>; and (6) the atmospheric chemical sink, for which one value is derived from observations and the other is derived from the inversion models. To quantify and attribute the regional N<sub>2</sub>O budget, we further partition the Earth's ice-free land into ten regions (Fig. 2, Supplementary Fig. 1). With the construction of these budgets, we explore the relative temporal and spatial importance of multiple sources and sinks that drive the atmospheric burden of N<sub>2</sub>O, their uncertainties, and interactions between anthropogenic forcing and natural fluxes of N<sub>2</sub>O as an emerging climate feedback.

### The global N<sub>2</sub>O budget (2007–2016)

The bottom-up and top-down approaches give consistent estimates of global total N<sub>2</sub>O emissions in the decade between 2007 and 2016 to well within their respective uncertainties, with values of 17.0 (minimum–maximum estimates: 12.2–23.5) Tg N yr<sup>-1</sup> and 16.9 (15.9–17.7) Tg N yr<sup>-1</sup> for bottom-up and top-down approaches, respectively. The global calculated atmospheric chemical sink (that is, N<sub>2</sub>O losses via photolysis and reaction with electronically excited atomic oxygen (O(<sup>1</sup>D)) in the troposphere and stratosphere) is 13.5 (12.4–14.6) Tg N yr<sup>-1</sup>. The imbalance of sources and sinks of N<sub>2</sub>O derived from the averaged bottom-up and top-down estimates is 4.1 Tg N yr<sup>-1</sup>. This imbalance agrees well

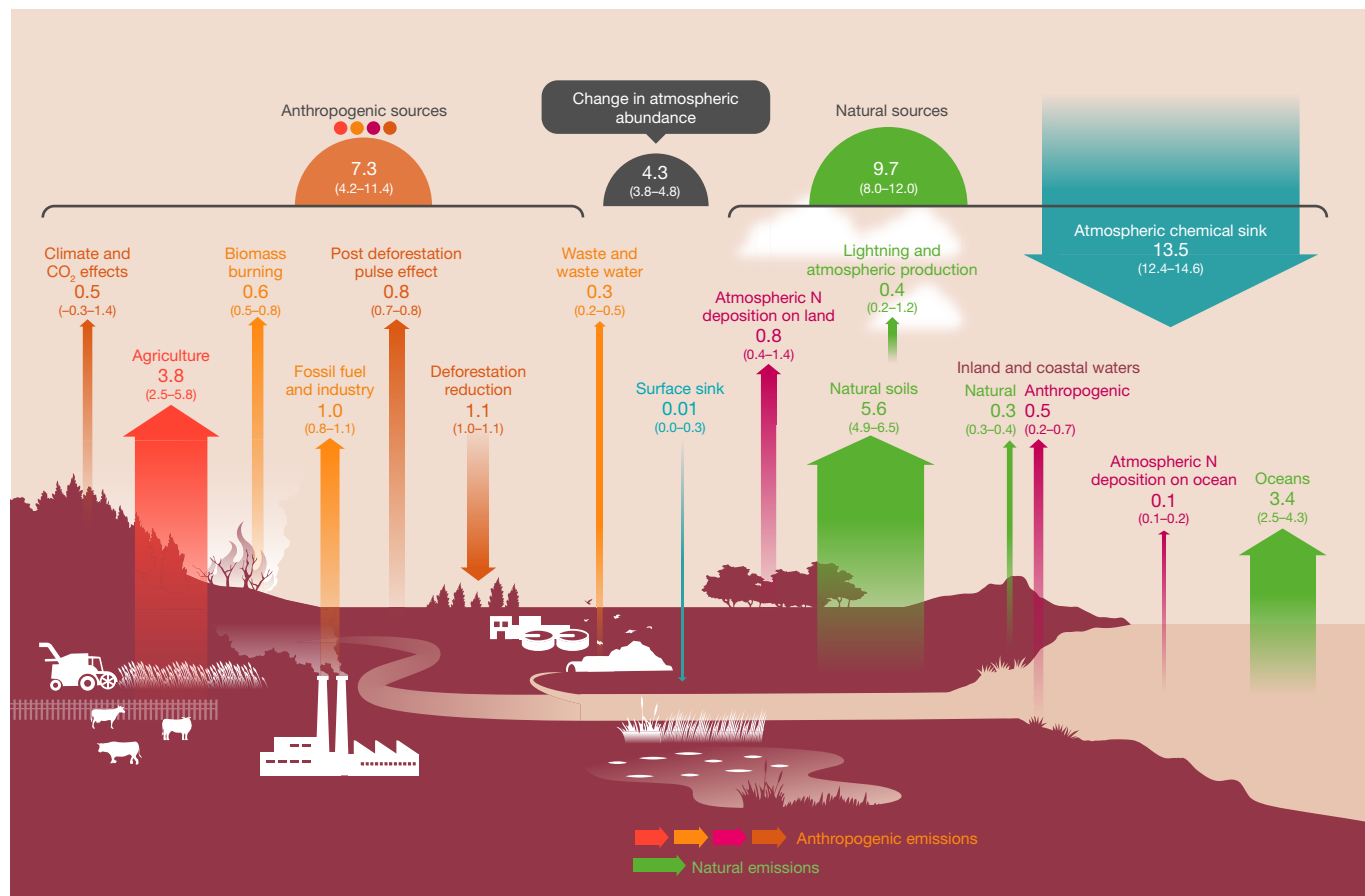
with the observed increase in atmospheric abundance of N<sub>2</sub>O between 2007 and 2016 of 3.8–4.8 Tg N yr<sup>-1</sup> (see Methods). Natural sources from soils and oceans contributed 57% of total emissions (mean: 9.7; min–max: 8.0–12.0 Tg N yr<sup>-1</sup>) during this time, according to our bottom-up estimate. We further estimate the natural soil flux at 5.6 (4.9–6.5) Tg N yr<sup>-1</sup> and the ocean flux at 3.4 (2.5–4.3) Tg N yr<sup>-1</sup> (see Methods).

Anthropogenic sources contributed, on average, 43% to the total N<sub>2</sub>O emission (mean: 7.3; min–max: 4.2–11.4 Tg N yr<sup>-1</sup>), of which direct and indirect emissions from nitrogen additions in agriculture and other sectors contributed around 52% and around 18%, respectively. Of the remaining anthropogenic emissions, about 27% were from other direct anthropogenic sources including fossil fuel and industry (around 13%), with about 3% from perturbed fluxes caused by changes in climate, CO<sub>2</sub> or land cover.

### Four decades of the global N<sub>2</sub>O budget

The atmospheric N<sub>2</sub>O burden increased from 1,462 Tg N in the 1980s to 1,555 Tg N in 2007–2016, with a possible uncertainty of ±20 Tg N. Our results (Table 1) show a substantial increase in global N<sub>2</sub>O emissions that is primarily driven by anthropogenic sources, as natural sources remained relatively steady throughout the study period. Global N<sub>2</sub>O emissions obtained from our bottom-up and top-down approaches are comparable in magnitude during 1998–2016, but top-down results imply a larger inter-annual variability (1.0 Tg N yr<sup>-1</sup>; Extended Data Fig. 3a). Bottom-up and top-down approaches diverge when estimating the magnitude of land emissions compared with ocean emissions, although they are consistent with respect to trends. Specifically, the bottom-up land estimate during 1998–2016 was on average 1.8 Tg N yr<sup>-1</sup> higher than the top-down estimate, but showed a slightly slower rate of increase of 0.8 ± 0.2 Tg N yr<sup>-1</sup> per decade (95% confidence interval;  $P < 0.05$ ) compared with 1.1 ± 0.6 Tg N yr<sup>-1</sup> per decade ( $P < 0.05$ ) from the top-down approach (Extended Data Fig. 3b). Since 2005, the difference in the magnitude of emissions between the two approaches has become smaller owing to a large increase in emission—particularly in South America, Africa and East Asia—that is inferred by the top-down approach (Extended Data Fig. 3d, f, i). Oceanic N<sub>2</sub>O emissions from the bottom-up approach (3.6 (2.7–4.5) Tg N yr<sup>-1</sup>) indicate a slight decline at a rate of 0.06 Tg N yr<sup>-1</sup> per decade ( $P < 0.05$ ), whereas the top-down approach gives a higher but stable value of 5.1 (3.4–7.1) Tg N yr<sup>-1</sup> during 1998–2016 (Table 1).

On the basis of bottom-up approaches, anthropogenic N<sub>2</sub>O emissions increased from 5.6 (3.6–8.7) Tg N yr<sup>-1</sup> in the 1980s to 7.3 (4.2–11.4) Tg N yr<sup>-1</sup> in 2007–2016, at a rate of 0.6 ± 0.2 Tg N yr<sup>-1</sup> per decade ( $P < 0.05$ ). Up to 87% of this increase results from direct emission from agriculture (71%) and indirect emission from anthropogenic nitrogen additions into soils (16%). Direct soil emission from fertilizer application is the major source of increases in emission from agriculture, followed by a small but notable increase in emissions from livestock manure and aquaculture. Model-based estimates of direct soil emissions<sup>15–17</sup> show a faster increase than in the three inventories used in our study (see Methods; Extended Data Fig. 4a); this is largely attributed to the interactive effects between climate change and nitrogen additions, as well as spatio-temporal variability in environmental factors such as rainfall and temperature, that modulate the N<sub>2</sub>O yield from nitrification and denitrification. This result is in line with the increased emission factor deduced from the top-down estimates, in which the inversion-based soil emissions increased at a faster rate than suggested by the IPCC Tier 1 emission factor<sup>14</sup> (which assumes a linear response), especially after 2009 (ref. <sup>18</sup>). The remaining causes of the increase are attributed to other direct anthropogenic sources (6%) and perturbed fluxes from changes in climate, CO<sub>2</sub> or land cover (8%). The contribution from fossil fuel and industry emissions decreased rapidly between 1980 and 2000, largely due to the installation of emissions-abatement equipment in industrial facilities that produce nitric and adipic acid. However, after



**Fig. 1 | Global N<sub>2</sub>O budget for 2007–2016.** The coloured arrows represent N<sub>2</sub>O fluxes (in Tg N yr<sup>-1</sup> for 2007–2016) as follows: red, direct emissions from nitrogen additions in the agricultural sector (agriculture); orange, emissions from other direct anthropogenic sources; maroon, indirect emissions from anthropogenic nitrogen additions; brown, perturbed fluxes from changes in climate, CO<sub>2</sub> or land cover; green, emissions from natural sources. The anthropogenic and natural N<sub>2</sub>O sources are derived from bottom-up estimates. The blue arrows represent the surface sink and the observed atmospheric

chemical sink, of which about 1% occurs in the troposphere. The total budget (sources + sinks) does not exactly match the observed atmospheric accumulation, because each of the terms has been derived independently and we do not force top-down agreement by rescaling the terms. This imbalance readily falls within the overall uncertainty in closing the N<sub>2</sub>O budget, as reflected in each of the terms. The N<sub>2</sub>O sources and sinks are given in Tg N yr<sup>-1</sup>. Copyright the Global Carbon Project.

2000, such emissions began to increase slowly, owing to increasing fossil fuel combustion (Extended Data Fig. 5a, b).

Our analysis of process-based model estimates indicates that soil N<sub>2</sub>O emissions have accelerated substantially as a result of climate change since the early 1980s, and this has offset the reduction due to feedback with increased CO<sub>2</sub> concentration and climate (Extended Data Fig. 6a). Increased CO<sub>2</sub> concentrations enhance plant growth and thus increase nitrogen uptake, which in turn decreases soil N<sub>2</sub>O emissions<sup>16,19</sup>. Conversion of land from tropical mature forests, which have higher N<sub>2</sub>O emissions, to pastures and other unfertilized agricultural lands has considerably reduced global natural N<sub>2</sub>O emissions<sup>11,20,21</sup>. This decrease, however, has been partly offset by an increase in soil N<sub>2</sub>O emissions attributed to the temporary increase in emissions after deforestation (the post-deforestation pulse effect) and to background emissions from converted croplands or pastures<sup>21</sup> (see Methods; Extended Data Fig. 7).

From the ensemble of process-based land model emissions<sup>15,16</sup>, we estimate a global agricultural soil emission factor of 1.8% (1.3%–2.3%), which is considerably larger than the IPCC Tier 1 default for direct emission of 1%. This higher emission factor, derived from process-based models, suggests a strong interactive effect between nitrogen additions and other global environmental changes (Table 1, ‘Perturbed fluxes from climate, atmospheric CO<sub>2</sub> and land cover change’). Previous field experiments reported a better fit to local observations of soil N<sub>2</sub>O emissions when assuming a nonlinear response to fertilizer nitrogen inputs under varied

climate and soil conditions<sup>17,22</sup>. The nonlinear response is also likely to be associated with long-term nitrogen accumulation in agricultural soils from nitrogen fertilizer use and in aquatic systems from nitrogen loads (the legacy effect)<sup>18,23</sup>, which provides more substrate for microbial processes<sup>18,24</sup>. The increasing N<sub>2</sub>O emissions estimated by process-based models<sup>16</sup> also suggest that recent climate change—particularly warming—could have boosted soil nitrification and denitrification processes, contributing to the growing trend in N<sub>2</sub>O emissions together with increasing nitrogen additions to agricultural soils<sup>16,25–27</sup> (Extended Data Fig. 8).

### Regional N<sub>2</sub>O budgets (2007–2016)

Bottom-up approaches give estimates of N<sub>2</sub>O emissions in each of the five source categories, whereas top-down approaches provide only total emissions (Fig. 2). Bottom-up and top-down approaches indicate that Africa was the largest source of N<sub>2</sub>O in the last decade, followed by South America (Fig. 2). Bottom-up and top-down approaches agree well regarding the magnitudes and trends of N<sub>2</sub>O emissions from South Asia and Oceania (Extended Data Fig. 3j, l). For the remaining regions, bottom-up and top-down estimates are comparable in terms of trends but diverge when estimating the strengths of the sources. Clearly, much more work on regional N<sub>2</sub>O budgets is needed, particularly for South America and Africa where there are larger differences between bottom-up and top-down estimates and larger uncertainties in each

**Table 1 | The global N<sub>2</sub>O budget in the 1980s, 1990s, 2000s and 2007–2016**

		1980s			1990s			2000s			2007–2016		
<b>Anthropogenic sources</b>		<b>Mean</b>	<b>Min.</b>	<b>Max.</b>	<b>Mean</b>	<b>Min.</b>	<b>Max.</b>	<b>Mean</b>	<b>Min.</b>	<b>Max.</b>	<b>Mean</b>	<b>Min.</b>	<b>Max.</b>
Direct emissions from nitrogen additions in the agricultural sector (Agriculture)	Direct soil emissions	1.5	0.9	2.6	1.7	1.1	3.1	2.0	1.3	3.4	2.3	1.4	3.8
	Manure left on pasture	0.9	0.7	1.0	1.0	0.7	1.1	1.1	0.8	1.2	1.2	0.9	1.3
	Manure management	0.3	0.2	0.4	0.3	0.2	0.4	0.3	0.2	0.5	0.3	0.2	0.5
	Aquaculture	0.01	0.00	0.03	0.03	0.01	0.1	0.1	0.02	0.2	0.1	0.02	0.2
	<b>Subtotal</b>	<b>2.6</b>	<b>1.8</b>	<b>4.1</b>	<b>3.0</b>	<b>2.1</b>	<b>4.8</b>	<b>3.4</b>	<b>2.3</b>	<b>5.2</b>	<b>3.8</b>	<b>2.5</b>	<b>5.8</b>
Other direct anthropogenic sources	Fossil fuels and industry	0.9	0.8	1.1	0.9	0.9	1.0	0.9	0.8	1.0	1.0	0.8	1.1
	Waste and waste water	0.2	0.1	0.3	0.3	0.2	0.4	0.3	0.2	0.4	0.3	0.2	0.5
	Biomass burning	0.7	0.7	0.7	0.7	0.6	0.8	0.6	0.6	0.6	0.6	0.5	0.8
	<b>Subtotal</b>	<b>1.8</b>	<b>1.6</b>	<b>2.1</b>	<b>1.9</b>	<b>1.7</b>	<b>2.1</b>	<b>1.8</b>	<b>1.6</b>	<b>2.1</b>	<b>1.9</b>	<b>1.6</b>	<b>2.3</b>
Indirect emissions from anthropogenic nitrogen additions	Inland waters, estuaries, coastal zones	0.4	0.2	0.5	0.4	0.2	0.5	0.4	0.2	0.6	0.5	0.2	0.7
	Atmospheric nitrogen deposition on land	0.6	0.3	1.2	0.7	0.4	1.4	0.7	0.4	1.3	0.8	0.4	1.4
	Atmospheric nitrogen deposition on ocean	0.1	0.1	0.2	0.1	0.1	0.2	0.1	0.1	0.2	0.1	0.1	0.2
	<b>Subtotal</b>	<b>1.1</b>	<b>0.6</b>	<b>1.9</b>	<b>1.2</b>	<b>0.7</b>	<b>2.1</b>	<b>1.2</b>	<b>0.6</b>	<b>2.1</b>	<b>1.3</b>	<b>0.7</b>	<b>2.2</b>
Perturbed fluxes from climate/CO <sub>2</sub> /land cover change	CO <sub>2</sub> effect	-0.2	-0.3	0.0	-0.2	-0.4	0.0	-0.3	-0.5	0.1	-0.3	-0.6	0.1
	Climate effect	0.4	0.0	0.8	0.5	0.1	0.9	0.7	0.3	1.2	0.8	0.3	1.3
	Post-deforestation pulse effect	0.7	0.6	0.8	0.7	0.6	0.8	0.7	0.7	0.8	0.8	0.7	0.8
	Long-term effect of reduced mature forest area	-0.8	-0.8	-0.9	-0.9	-0.8	-1.0	-1.0	-0.9	-1.1	-1.1	-1.0	-1.1
	<b>Subtotal</b>	<b>0.1</b>	<b>-0.4</b>	<b>0.7</b>	<b>0.1</b>	<b>-0.5</b>	<b>0.7</b>	<b>0.2</b>	<b>-0.4</b>	<b>0.9</b>	<b>0.2</b>	<b>-0.6</b>	<b>1.1</b>
<b>Anthropogenic total</b>	<b>5.6</b>	<b>3.6</b>	<b>8.7</b>	<b>6.2</b>	<b>3.9</b>	<b>9.7</b>	<b>6.7</b>	<b>4.1</b>	<b>10.3</b>	<b>7.3</b>	<b>4.2</b>	<b>11.4</b>	
<b>Natural fluxes</b>													
Natural soils baseline		5.6	4.9	6.6	5.6	4.9	6.5	5.6	5.0	6.5	5.6	4.9	6.5
Ocean baseline		3.6	3.0	4.4	3.5	2.8	4.4	3.5	2.7	4.3	3.4	2.5	4.3
Natural (inland waters, estuaries, coastal zones)		0.3	0.3	0.4	0.3	0.3	0.4	0.3	0.3	0.4	0.3	0.3	0.4
Lightning and atmospheric production		0.4	0.2	1.2	0.4	0.2	1.2	0.4	0.2	1.2	0.4	0.2	1.2
Surface sink		-0.01	0.00	-0.3	-0.01	0.00	-0.3	-0.01	0.00	-0.3	-0.01	0.00	-0.3
<b>Natural total</b>		<b>9.9</b>	<b>8.5</b>	<b>12.2</b>	<b>9.8</b>	<b>8.3</b>	<b>12.1</b>	<b>9.8</b>	<b>8.2</b>	<b>12.0</b>	<b>9.7</b>	<b>8.0</b>	<b>12.0</b>
Bottom-up total source		15.5	12.1	20.9	15.9	12.2	21.7	16.4	12.3	22.4	17.0	12.2	23.5
Top-down ocean								5.1	3.1	7.2	5.1	3.4	7.1
Top-down land								10.8	9.3	12.5	11.8	10.6	13.8
Top-down total source								15.9	15.1	16.9	16.9	15.9	17.7
Top-down stratospheric sink								12.1	11.4	13.1	12.4	11.7	13.3
Observed atmospheric chemical sink <sup>a</sup>								13.3	12.2	14.4	13.5	12.4	14.6
Change in atmospheric abundance <sup>b</sup>								<b>3.7</b>	<b>3.2</b>	<b>4.2</b>	<b>4.3</b>	<b>3.8</b>	<b>4.8</b>
<b>Atmospheric burden</b>		<b>1,462</b>	<b>1,442</b>	<b>1,482</b>	<b>1,493</b>	<b>1,472</b>	<b>1,514</b>	<b>1,531</b>	<b>1,510</b>	<b>1,552</b>	<b>1,555</b>	<b>1,533</b>	<b>1,577</b>

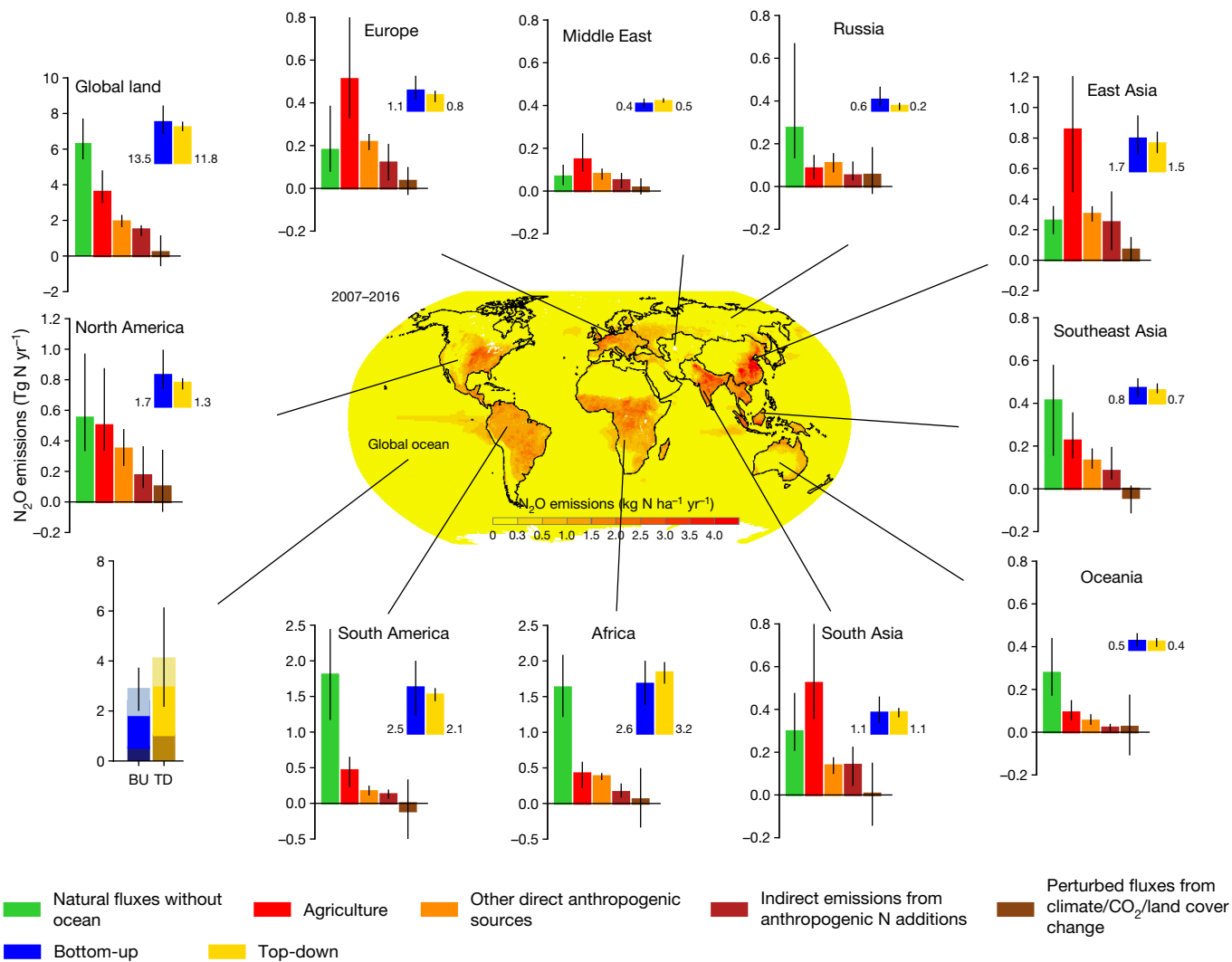
Bottom-up estimates include four categories of anthropogenic source and one category for natural sources and sinks. The sources and sinks of N<sub>2</sub>O are given in Tg N yr<sup>-1</sup>. The atmospheric burden is given in Tg N.

<sup>a</sup>Calculated from satellite observations with a photolysis model (about 1% of this sink occurs in the troposphere).

<sup>b</sup>Calculated from the combined NOAA and AGAGE record of surface N<sub>2</sub>O, and adopting the uncertainty of the IPCC Assessment Report 5 (Chapter 6)<sup>2</sup>. Detailed information on calculating each sub-category is shown in Supplementary Tables 1–13.

approach. Advancing the understanding and model representation of key processes responsible for N<sub>2</sub>O emissions from land and ocean are priorities to reduce uncertainties in bottom-up estimates. Atmospheric observations in underrepresented regions of the world and better atmospheric transport models are essential to reduce uncertainty in top-down estimates, whereas more accurate activity data and robust emission factors are critical for greenhouse-gas inventories (see Methods for additional discussion on uncertainty).

According to estimates from the Global N<sub>2</sub>O Model Intercomparison Project<sup>16</sup>, natural soil emissions dominate (to different extents) in tropical and sub-tropical regions. Soil N<sub>2</sub>O emissions in the tropics (0.1 ± 0.04 g N m<sup>-2</sup> yr<sup>-1</sup>) are about 50% higher than the global average, because many lowland, highly weathered tropical soils have excess nitrogen relative to phosphorus<sup>20</sup>. Total anthropogenic emissions in the 10 terrestrial regions (Fig. 2) were highest in East Asia (1.5 (0.8–2.6) Tg N yr<sup>-1</sup>), followed by North America, Africa and Europe. High direct



**Fig. 2 | Regional N<sub>2</sub>O sources in the decade 2007–2016.** The Earth’s ice-free land is partitioned into ten regions: North America, South America, Europe, Middle East, Africa, Russia, East Asia, South Asia, Southeast Asia and Oceania. Each subplot shows the emissions from five sub-sectors using bottom-up approaches, followed by the sum of these five categories using bottom-up approaches (blue) and the estimates from top-down approaches (yellow). Bottom-up and top-down estimates of ocean emissions are shown at the

bottom left (from bottom to top, lighter to darker, the contributions from the 30°–90° N, 30° S–30° N and 90°–30° S regions). Error bars indicate the spread between the minimum and the maximum values. The centre map shows the spatial distribution of 10-year average N<sub>2</sub>O emissions from land and ocean based on the land and ocean models. Per capita N<sub>2</sub>O emission (kg N per capita per year) during 2007–2016 is shown in Supplementary Fig. 2. The map was created using ESRI ArcMap 10.4.1.

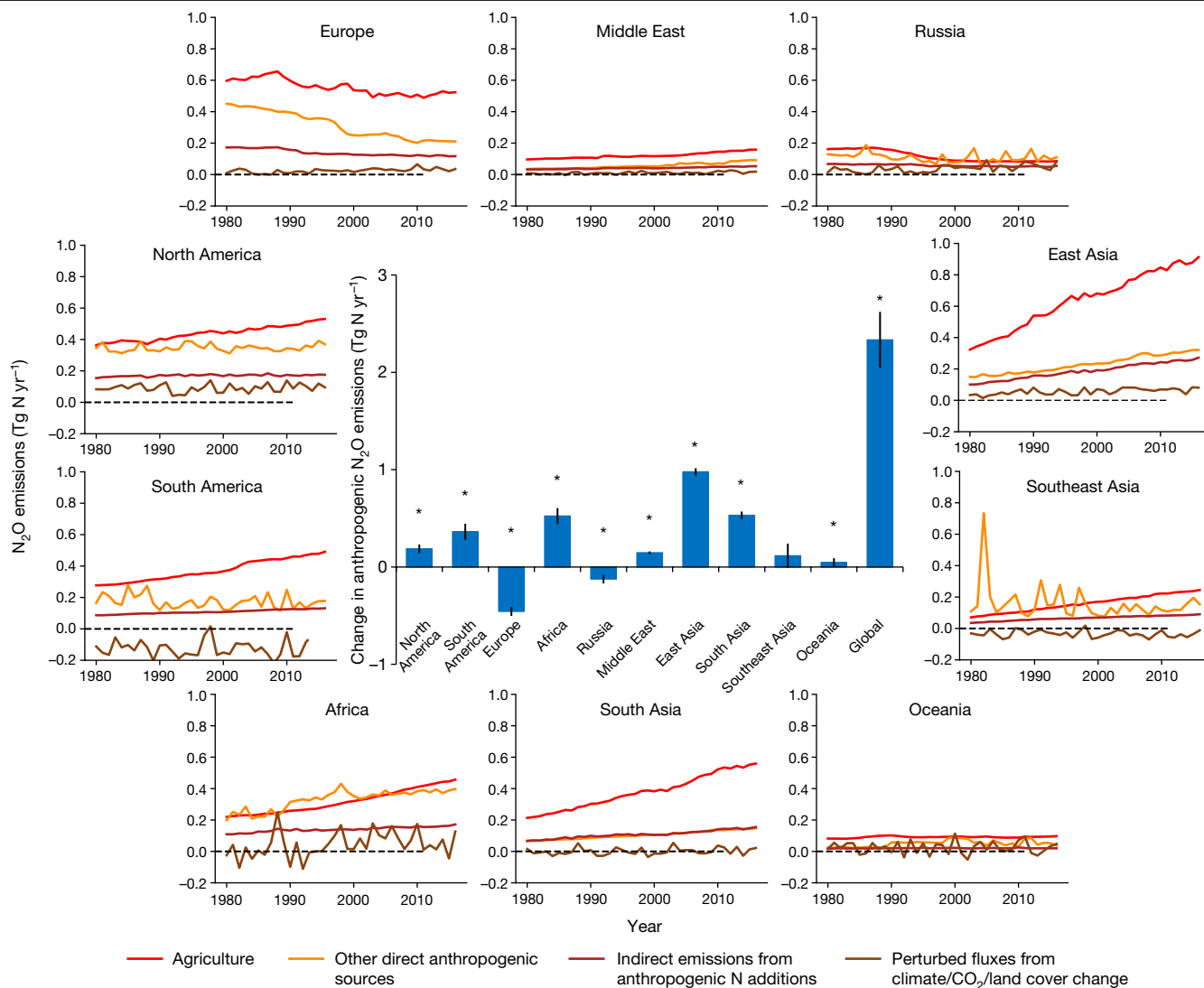
agricultural N<sub>2</sub>O emissions can be attributed to the large-scale application of synthetic nitrogen fertilizers in East Asia, Europe, South Asia and North America, which together consume over 80% of the world’s synthetic nitrogen fertilizers<sup>28</sup>. By contrast, direct agricultural emissions from Africa and South America mainly arise from livestock manure that is deposited in pastures and rangelands<sup>28,29</sup>. East Asia contributed 71%–79% of global aquaculture N<sub>2</sub>O emissions; South Asia and Southeast Asia together contributed 10%–20% (refs.<sup>30,31</sup>). Indirect emissions have a moderate role in the total N<sub>2</sub>O budget, with the highest emission in East Asia (0.3 (0.1–0.5) Tg N yr<sup>-1</sup>). Other direct anthropogenic sources together contribute N<sub>2</sub>O emissions of approximately 0.2–0.4 Tg N yr<sup>-1</sup> in each of East Asia, Africa, North America and Europe.

Both bottom-up and top-down estimates of ocean N<sub>2</sub>O emissions for northern, tropical and southern ocean regions (90° N–30° N, 30° N–30° S and 30° S–90° S, respectively) reveal that the tropical oceans contribute over 50% to the global oceanic N<sub>2</sub>O source. In particular, the upwelling regions of the equatorial Pacific, Indian and tropical Atlantic (Fig. 3) provide considerable sources of N<sub>2</sub>O<sup>32–34</sup>. Bottom-up estimates suggest that the southern ocean region is the second largest

contributor, with emissions around twice as high as those from the northern oceans (53% tropical oceans, 31% southern oceans, 17% northern oceans), in line with their respective areas. Top-down estimates, however, suggest approximately equal contributions from the southern and northern ocean regions.

### Four decades of anthropogenic N<sub>2</sub>O emissions

Trends in anthropogenic emissions were found to vary among regions (Fig. 3). Fluxes from Europe and Russia decreased by a total of 0.6 (0.5–0.7) Tg N yr<sup>-1</sup> over the 37 years from 1980 to 2016. The decrease in Europe is associated with successful emissions abatement in industry as well as agricultural policies, whereas the decrease in Russia is associated with the collapse of the agricultural cooperative system after 1990. By contrast, fluxes from the remaining eight regions increased by a total of 2.9 (2.4–3.4) Tg N yr<sup>-1</sup> (Fig. 3), of which 34% came from East Asia, 18% from Africa, 18% from South Asia, 13% from South America and 6% from North America, with the remaining increase attributed to the three other regions.



**Fig. 3 | Ensembles of regional anthropogenic N<sub>2</sub>O emissions over the period 1980–2016.** The bar chart in the centre shows the accumulated changes in regional and global N<sub>2</sub>O emissions during the study period of 1980–2016. Error bars indicate the 95% confidence interval for the average of accumulated changes. The Mann–Kendall test was performed to examine a monotonic increasing or decreasing trend in the estimated ensemble N<sub>2</sub>O emissions

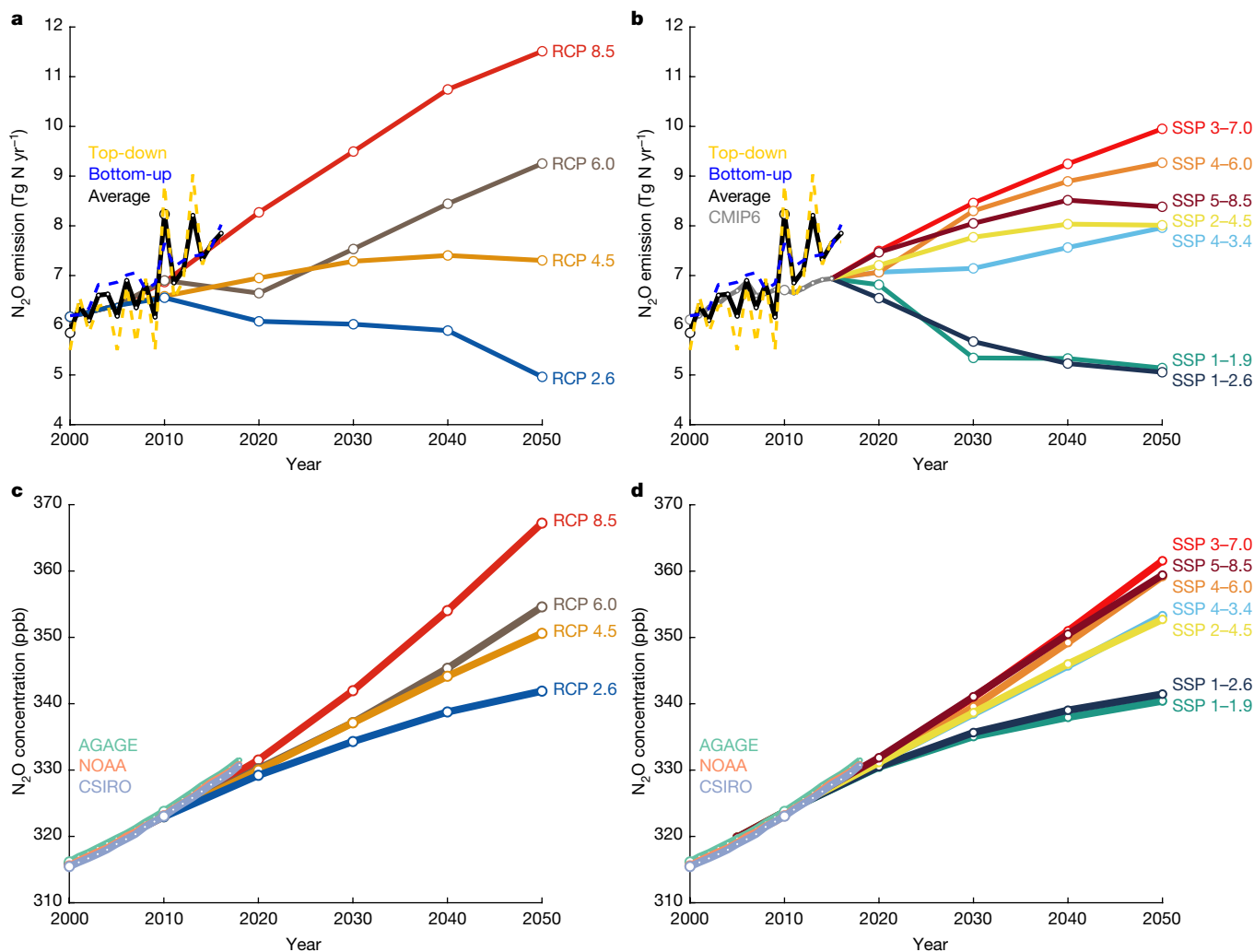
globally and for each region over the period 1980–2016. The accumulated changes were calculated from the linear regressed annual change rate (Tg N yr<sup>-2</sup>) multiplied by 37 years. All regions except Southeast Asia show a significant increasing or decreasing trend in the estimated ensemble N<sub>2</sub>O emissions during the study period. \**P* < 0.05.

The relative importance of each anthropogenic source to the total increase in emission differs among regions. East Asia, South Asia, Africa and South America show larger increases in total agricultural N<sub>2</sub>O emissions (direct and indirect) compared with the remaining six regions during 1980–2016 (Fig. 3). Southeast Asia, North America and the Middle East also show increasing direct N<sub>2</sub>O emissions, but to a smaller extent. Increasing indirect emissions in East Asia, South Asia, Africa and South America on average constitute 20% of total agricultural N<sub>2</sub>O emissions and largely result from the considerable increase in fertilizer nitrogen inputs to agricultural soils<sup>35,36</sup>. The fastest increase in emissions from other direct anthropogenic sources was found in East Asia, and is primarily due to rapidly increasing industrial emissions. Africa and South Asia also show a rapid increase in emissions, arising from fossil fuels and industry, and from waste and waste water.

Our findings point to growing N<sub>2</sub>O emissions in emerging economies—particularly Brazil, China and India. For example, we find here that the substantial increases in livestock manure left on pasture and in fertilizer use caused an increase of approximately 120% in Brazilian

agricultural N<sub>2</sub>O emissions during 1980–2016 (Extended Data Fig. 9). In addition to fertilizer applications, the production of global livestock manure has been growing steadily, in line with increased livestock numbers<sup>15,28</sup>. Growing demand for meat and dairy products has substantially increased global N<sub>2</sub>O emissions from livestock manure production and management associated with the expansion of pastures and grazing land<sup>37</sup>. Meanwhile, expansion of feed crop production to support the growth of livestock could further enhance global N<sub>2</sub>O emissions<sup>37,38</sup>. Likewise, increasing demand for fish has resulted in a fivefold increase in global N<sub>2</sub>O production from aquaculture since the late 1980s<sup>39</sup>, and demand is projected to increase further<sup>40</sup>; however, this remains a small fraction (less than 1%) of total N<sub>2</sub>O emissions.

The acceleration of global N<sub>2</sub>O emissions resulting from anthropogenic sources is apparent from both bottom-up and top-down estimates. It currently tracks the highest Representative Concentration Pathway<sup>4</sup> (RCP) in the fifth assessment report of the IPCC<sup>2</sup>, RCP 8.5, and exceeds all the Shared Socioeconomic Pathways (SSPs)<sup>3</sup> in the Coupled Model Intercomparison Project Phase 6 (CMIP6) for the sixth



**Fig. 4 | Historical and projected global anthropogenic  $N_2O$  emissions and concentrations. a–d**, Global anthropogenic  $N_2O$  emissions (a, b) and concentrations (c, d) compared to the four RCPs in the IPCC assessment report 5 (a, c; ref. <sup>2</sup>) and the new marker scenarios based on the SSPs used in CMIP6 (b, d; ref. <sup>48</sup>). The historical emissions data are represented as the mean of the bottom-up and top-down estimates of anthropogenic  $N_2O$  emissions, whereas the historical atmospheric concentration data are from the three available

observation networks: AGAGE, NOAA, and CSIRO. Top-down anthropogenic emissions were calculated by subtracting natural fluxes derived from bottom-up approaches. To aid the comparison, the four RCPs were shifted down so that the 2005 value is equal to the 2000–2009 average of the mean of top-down and bottom-up estimates. The SSPs are harmonized<sup>3</sup> to match the historical emissions used in CMIP6<sup>49</sup>; Extended Data Fig. 10 shows the unharmonized data.

assessment report of the IPCC (Fig. 4). Observed atmospheric  $N_2O$  concentrations are beginning to exceed predicted levels across all scenarios. Emissions need to be reduced to a level that is consistent with or below that of RCP 2.6 or SSP 1–2.6 in order to limit warming to well below the 2 °C target of the Paris Agreement. Failure to include  $N_2O$  within climate mitigation strategies will necessitate even greater abatement of  $CO_2$  and methane. Although  $N_2O$  mitigation is difficult because nitrogen is the key limiting nutrient in agricultural production, this study demonstrates that effective mitigation actions have reduced emissions in some regions—such as Europe—through technological improvements in industry and improved efficiency of nitrogen use in agriculture.

There are numerous mitigation options in the agriculture sector that are available for immediate deployment, including increasing the efficiency of nitrogen use both in animal production (through the tuning of feed rations to reduce nitrogen excretion) and in crop production (through precision delivery of nitrogen fertilizers, split applications and better timing to match nitrogen applications to crop

demand, conservation tillage, prevention of waterlogging, and the use of nitrification inhibitors<sup>41,42</sup>). Success stories include the stabilization or reduction of  $N_2O$  emissions through improving nitrogen use efficiency in the United States and Europe, while maintaining or even increasing crop yields<sup>42,43</sup>. There is every reason to expect that additional implementation of more sustainable practices and emerging technologies will lead to further reduction of emissions in these regions. For example,  $N_2O$  emissions from European agricultural soils decreased by 21% between 1990 and 2010—a decline attributed to the implementation of the Nitrates Directive (an agricultural policy that favours optimization and reduction of fertilizer use as well as water protection legislation)<sup>44</sup>. For regions in which emissions are growing, an immediate opportunity lies in the reduction of excess fertilizer use along with the implementation of more sustainable agricultural practices; together, these strategies have been shown to increase crop yields, reduce  $N_2O$  emissions, increase water quality and increase farm income<sup>45</sup>. In addition,  $N_2O$  emissions can be efficiently abated in the chemical industry<sup>11,41,46,47</sup>. This has been achieved successfully in nitric

acid plants in the European Union, where industrial N<sub>2</sub>O emissions decreased from 11% to 3% of total emissions between 2007 and 2012 (ref. 44). Additional strategies available to reduce N<sub>2</sub>O emissions include promoting lower meat consumption in some parts of the world<sup>9</sup> and reducing food waste<sup>11</sup>.

We present the most comprehensive, to our knowledge, global N<sub>2</sub>O budget so far, with a detailed sectorial and regional attribution of sources and sinks. Each of the past four decades has had higher global N<sub>2</sub>O emissions than the last, and overall, agricultural activities have dominated the growth in emissions. Total industrial emissions have been quite stable, with increased emissions from the fossil fuel sector offset to some extent by the decline in emissions in other industrial sectors as a result of successful abatement policies. We also highlight a number of complex interactions between N<sub>2</sub>O fluxes and human-driven changes, the effect of which on the global atmospheric N<sub>2</sub>O growth rate was previously unknown. These interactions include the effects of climate change, increasing atmospheric CO<sub>2</sub> and deforestation. Cumulatively, these exert a relatively small effect on the overall increase in N<sub>2</sub>O concentrations; however, individual flux components—such as the increasing positive climate–N<sub>2</sub>O feedback—are considerable. These fluxes are not currently included when reporting national greenhouse gas emissions. We further find that Brazil, China and India dominate the regional contributions to the increase in global N<sub>2</sub>O emissions over the most recent decade. Our extensive database and modelling capability fill current gaps in national and regional emissions inventories. Future research is needed to further constrain complex biogeochemical interactions between natural and anthropogenic fluxes and global environmental changes, which could lead to considerable feedback in the future. Reducing excess nitrogen applications to croplands and adopting precision fertilizer application methods provide the greatest immediate opportunities for the abatement of N<sub>2</sub>O emissions.

## Online content

Any methods, additional references, Nature Research reporting summaries, source data, extended data, supplementary information, acknowledgements, peer review information; details of author contributions and competing interests; and statements of data and code availability are available at <https://doi.org/10.1038/s41586-020-2780-0>.

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## Methods

### Terminology

This study provides an estimation of the global N<sub>2</sub>O budget considering all possible sources and all global change processes that can perturb the budget. A total of 18 sources and three sinks of N<sub>2</sub>O are identified and grouped into six categories (Fig. 1, Table 1): (1) natural fluxes in the absence of climate change and anthropogenic disturbances including soil emissions, surface sink, ocean emissions, lightning and atmospheric production, and natural emission from inland waters, estuaries, coastal zones (inland and coastal waters); (2) perturbed fluxes from climate/CO<sub>2</sub>/land cover change including the effect of CO<sub>2</sub>, the effect of climate, the post-deforestation pulse effect, and the long-term effect of reduced mature forest area; (3) direct emissions from nitrogen additions in the agricultural sector ('agriculture') including emissions from direct application of synthetic nitrogen fertilizers and manure (henceforth 'direct soil emissions'), manure left on pasture, manure management and aquaculture; (4) indirect emissions from anthropogenic nitrogen additions including atmospheric nitrogen deposition (NDEP) on land, atmospheric NDEP on ocean, and effects of anthropogenic loads of reactive nitrogen in inland waters, estuaries and coastal zones; (5) other direct anthropogenic sources including fossil fuel and industry, waste and waste water, and biomass burning; and (6) two estimates of stratospheric sinks obtained from atmospheric chemistry transport models and observations, and one tropospheric sink (Table 1, Extended Data Fig. 2).

For the purpose of compiling national greenhouse-gas inventories for reporting data for each country to the climate convention, our anthropogenic N<sub>2</sub>O emission categories are aligned with those used in UNFCCC reporting and IPCC 2006 methodologies (Supplementary Table 14). We also provide a detailed comparison of our methodology and quantification with that of the IPCC assessment report 5 (see Supplementary Information section 4, Supplementary Table 15).

### Data synthesis

We consider global N<sub>2</sub>O emission from land and ocean consisting of natural fluxes and anthropogenic emissions estimated from bottom-up and top-down approaches; however, the top-down approach cannot separate natural and anthropogenic sources.

'Natural soil baseline' emissions were obtained from six terrestrial biosphere models (Global N<sub>2</sub>O Model Intercomparison Project (NMIP)<sup>16</sup>, Supplementary Tables 16, 17) and reflect a situation without consideration of land use change (for example, deforestation) and without consideration of indirect anthropogenic effects via global change (that is, climate, increased CO<sub>2</sub> and atmospheric nitrogen deposition). Bottom-up oceanic N<sub>2</sub>O emissions were based on an inter-comparison of five global ocean biogeochemistry models (Supplementary Table 18). The natural emission from 'Inland water, estuaries, coastal zones' includes coastal upwelling<sup>50</sup> and inland and coastal waters that were obtained from ref. <sup>36</sup>, ref. <sup>35</sup> and ref. <sup>51</sup>. Because the data (rivers, reservoirs, and estuaries) provided in ref. <sup>35</sup> and ref. <sup>51</sup> are for the year 2000, we assume that these values are constant during 1980–2016. Ref. <sup>36</sup> provided annual riverine N<sub>2</sub>O emissions using the DLEM model during the same period. Here, we averaged estimates from ref. <sup>36</sup> with those from ref. <sup>35</sup>. In addition, we estimated N<sub>2</sub>O emissions from global and regional reservoirs in the 2000s, and averaged their estimates with those from ref. <sup>35</sup> to represent emissions from reservoirs during 1980–2016. The estimate for global and regional estuaries and lakes is still based on the long-term averaged values provided by ref. <sup>35</sup> and ref. <sup>51</sup>, respectively. We considered the riverine emissions in the year 1900 as equivalent to the natural emission for the DLEM estimate assuming that the nitrogen load from land was negligible in that period<sup>52</sup>. We quantified the contribution of natural sources to total emission from reservoirs, lakes and estuaries at 44% (36%–52%), with consideration of all nitrogen inputs (that is, inorganic, organic, dissolved and particulate

forms). We combined the estimate from lightning with that from atmospheric production into an integrated category denoted 'Lightning and atmospheric production'. We make a simplification by considering the category 'Lightning and atmospheric production' as purely natural; however, atmospheric production is affected to some extent by anthropogenic activities through enhancing the concentrations of the reactive species NH<sub>2</sub> and NO<sub>2</sub>. This category is in any case very small and the anthropogenic enhancement effect is uncertain. Lightning produces NO<sub>x</sub>, the median estimate of which is 5 Tg N yr<sup>-1</sup> (ref. <sup>53</sup>). We assumed an emission factor of 1% (ref. <sup>54</sup>) and a global estimate of 0.05 (0.02–0.09) Tg N yr<sup>-1</sup> from lightning. Atmospheric production of N<sub>2</sub>O results from the reaction of NH<sub>2</sub> with NO<sub>2</sub> (refs. <sup>55,56</sup>), N with NO<sub>2</sub>, and from the oxidation of N<sub>2</sub> by O(<sup>1</sup>D)<sup>57</sup>, all of which constitute an estimated source of 0.3 (0.2–1.1) Tg N yr<sup>-1</sup>. The estimate of the 'Surface sink' was obtained from ref. <sup>58</sup> and ref. <sup>59</sup>.

The anthropogenic sources include four sub-sectors:

(a) Agriculture. This consists of four components: 'Direct soil emissions', 'Manure left on pasture', 'Manure management' and 'Aquaculture'. Data for 'Direct soil emissions' were obtained as the ensemble mean of N<sub>2</sub>O emissions from an average of three inventories (EDGAR v4.3.2, FAOSTAT and GAINS), the SRNM/DLEM models and the NMIP/DLEM models. The statistical model SRNM covers only cropland N<sub>2</sub>O emissions, the same as the NMIP. Thus, we add the DLEM-based estimate of pasture N<sub>2</sub>O emissions into the two estimates in cropland to represent direct agricultural soil emissions (that is, SRNM/DLEM or NMIP/DLEM). The 'Manure left on pasture' and 'Manure management' emissions are the ensemble mean of the values from the EDGAR v4.3.2, FAOSTAT and GAINS databases. Global nitrogen flows (that is, fish feed intake, fish harvest and waste) in freshwater and marine aquaculture were obtained from ref. <sup>30</sup> and refs. <sup>60,61</sup> based on a nutrient budget model for the period 1980–2016. We then calculated global aquaculture N<sub>2</sub>O emissions through considering 1.8% loss of nitrogen waste in aquaculture, the same emission factor used in ref. <sup>62</sup> and ref. <sup>31</sup>. The uncertainty range of the emission factor is from 0.5% (ref. <sup>14</sup>) to 5% (ref. <sup>63</sup>), the same range used in the UNEP report<sup>9</sup>. The 'Aquaculture' emission for the period 2007–2016 was estimated through synthesizing multiple sources of data from ref. <sup>62</sup> in 2009, the FAO report<sup>31</sup> in 2013 and our calculations. The estimate of aquaculture N<sub>2</sub>O emission before 2009 was from our calculations only.

The estimated direct emissions from agriculture have increased from 2.6 (1.8–4.1) Tg N yr<sup>-1</sup> in the 1980s to 3.8 (2.5–5.8) Tg N yr<sup>-1</sup> over the recent decade (2007–2016, Table 1). Specifically, direct soil emission from the application of fertilizers is the major source and increased at a rate of 0.27 ± 0.01 Tg N yr<sup>-1</sup> per decade ( $P < 0.05$ ; Table 1). Compared with the three global inventories (FAOSTAT, EDGAR v4.3.2, and GAINS), the estimates from process-based models (NMIP/DLEM<sup>15,16</sup>) and a statistical model (SRNM)/DLEM<sup>15,17</sup> exhibited a faster increase (Extended Data Fig. 4a). Over the past four decades, we also found a small but significant increase in emissions from livestock manure (that is, manure left on pasture and manure management) at a rate of 0.1 ± 0.01 Tg N yr<sup>-1</sup> per decade ( $P < 0.05$ ; Extended Data Fig. 4b–c). Meanwhile, global aquaculture N<sub>2</sub>O emissions increased tenfold, however, this flux remains the smallest term in the global budget (Extended Data Fig. 4d).

(b) Other direct anthropogenic sources. This includes 'Fossil fuel and industry', 'Waste and waste water', and 'Biomass burning'. Both 'Fossil fuel and industry' and 'Waste and waste water' are the ensemble means of the values from EDGAR v4.3.2 and GAINS databases. The 'Biomass burning' emission is the ensemble mean of values from FAOSTAT, DLEM and GFED4s databases.

Emissions from a combination of fossil fuel and industry, waste and waste water, and biomass burning increased from 1.8 (1.6–2.1) Tg N yr<sup>-1</sup> in the 1980s to 1.9 (1.6–2.3) Tg N yr<sup>-1</sup> over the period 2007–2016 (Table 1). The waste and waste water emission showed a continuous increase at a rate of 0.04 ± 0.01 Tg N yr<sup>-1</sup> per decade ( $P < 0.05$ ) (Extended Data Fig. 5c). Emissions from biomass burning, estimated on the basis of three data

sources (DLEM, GFED4s, and FAOSTAT), slightly decreased at a rate of  $-0.03 \pm 0.04 \text{ Tg N yr}^{-1}$  per decade ( $P = 0.3$ ) since the 1980s (Extended Data Fig. 5d). This contribution is largely affected by climate and land use change<sup>64,65</sup>. Of the three data sources, the DLEM estimate exhibited substantial inter-annual variability, especially during 1980–2000 when extreme fire events were detected in 1982, 1987, 1991, 1994 and 1998. The occurrences of these extreme fires were associated with El Niño/Southern Oscillation (ENSO) events, especially in Indonesia (for example, the Great Fire of Borneo in 1982)<sup>66</sup>. Since 1997, N<sub>2</sub>O emissions from fires estimated by DLEM, GFED4s and FAOSTAT were consistent in terms of inter-annual variability. All three estimates showed a decreasing trend, agreeing well with the satellite-observed decrease of the global burned area<sup>64,65</sup>.

(c) Indirect emissions from anthropogenic nitrogen additions. Data were obtained from various sources and considered as nitrogen deposition on land and ocean ('Nitrogen deposition on land' and 'Nitrogen deposition on ocean'), as well as the nitrogen leaching and runoff from upstream ('Inland and coastal waters'). The emission from 'Nitrogen deposition on ocean' was provided in ref.<sup>67</sup>, whereas emission from 'Nitrogen deposition on land' was the ensemble mean of an average of three inventories: FAOSTAT/EDGAR v4.3.2, GAINS/EDGAR v4.3.2 and NMIP. FAOSTAT and GAINS documented the sector 'Indirect agricultural N<sub>2</sub>O emissions' by separating estimates from nitrogen leaching or nitrogen deposition, whereas EDGAR v4.3.2 did not. Here, we treated 'Indirect agricultural N<sub>2</sub>O emissions' from EDGAR v4.3.2 as 'Inland and coastal waters' emissions for data synthesis. Only EDGAR v4.3.2 provided an estimate of indirect emission from non-agricultural sectors, whereas both FAOSTAT and GAINS—following the IPCC guidelines—provided NH<sub>3</sub>/NO<sub>y</sub> volatilization from agricultural sectors. Here, we sum FAOSTAT or GAINS data with EDGAR v4.3.2 data (that is, FAOSTAT/EDGAR v4.3.2 or GAINS/EDGAR v4.3.2) to represent nitrogen-deposition-induced soil emissions from both agricultural and non-agricultural sectors. The N<sub>2</sub>O emissions from 'Inland and coastal waters' consist of emissions from rivers, reservoirs, lakes, estuaries and coastal zone, and is the ensemble mean of an average of three inventories (EDGAR v4.3.2, FAOSTAT, GAINS), and the mean of process-based models. The anthropogenic emission estimated in ref.<sup>36</sup> considered annual nitrogen inputs and other environmental factors (that is, climate, increased CO<sub>2</sub> and land cover change). For long-term average in rivers, reservoirs, estuaries and lakes, we applied a mean of 56% (based on the ratio of anthropogenic to total nitrogen additions from land) to calculate anthropogenic emissions. Seagrass, mangrove, salt-marsh and intertidal N<sub>2</sub>O emissions were undated and obtained from ref.<sup>68</sup>. Coastal waters with low disturbance generally either have low N<sub>2</sub>O emissions or act as a sink for N<sub>2</sub>O<sup>69,70</sup>. Here, coastal zone emissions were treated as anthropogenic emissions owing to intensive human disturbances<sup>71</sup>.

N<sub>2</sub>O emissions after transport of anthropogenic nitrogen additions via the atmosphere and via water bodies increased from 1.1 (0.6–1.9) Tg N yr<sup>-1</sup> in the 1980s to 1.3 (0.7–2.2) Tg N yr<sup>-1</sup> during 2007–2016 (Table 1). The N<sub>2</sub>O emissions from inland and coastal waters increased at a rate of  $0.03 \pm 0.00 \text{ Tg N yr}^{-1}$  per decade ( $P < 0.05$ ). Such an increase was reported by all three inventories (FAOSTAT, GAINS and EDGAR v4.3.2) with FAOSTAT giving the largest estimate. By contrast, the DLEM-based estimate presented a divergent trend: first increasing from 1980–1998 and then slightly decreasing thereafter (Extended Data Fig. 6a). Emissions from atmospheric nitrogen deposition on oceans were relatively constant with a value of 0.1 (0.1–0.2) Tg N yr<sup>-1</sup>, whereas a large increase in emissions was found from atmospheric nitrogen deposition on land, with  $0.06 \pm 0.01 \text{ Tg N yr}^{-1}$  per decade ( $P < 0.05$ ) reported in the three estimates (FAOSTAT/EDGAR v4.3.2, GAINS/EDGAR v4.3.2 and NMIP). The FAOSTAT agricultural source—together with the EDGAR v4.3.2 industrial source—is consistent with NMIP estimates regarding the magnitude of N<sub>2</sub>O emissions, with the latter estimating a slightly slower increase from 2010 to 2016 (Extended Data Fig. 6b).

(d) Perturbed fluxes from climate/CO<sub>2</sub>/land cover change. Perturbed N<sub>2</sub>O fluxes represent the sum of the effects of climate, increased atmospheric CO<sub>2</sub> and land cover change. The estimate of climate and CO<sub>2</sub> effects on emissions was based on NMIP. The effect of land cover change on N<sub>2</sub>O dynamics includes the reduction due to 'Long-term effect of reduced mature forest area' and the emissions due to 'Post-deforestation pulse effect'. The two estimates were based on the book-keeping approach and the DLEM model simulation. The book-keeping method is developed in ref.<sup>72</sup> for accounting for carbon flows due to land use. In this study, an observation dataset consisting of 18 tropical sites was collected to follow the book-keeping logic. The dataset covers N<sub>2</sub>O emissions from a reference mature forest and their nearby converted pastures aged between 1 and 60 years. The average tropical forest N<sub>2</sub>O emission rate of 1.974 kg N<sub>2</sub>O-N ha<sup>-1</sup> yr<sup>-1</sup> was adopted as the baseline<sup>73</sup>. Two logarithmic response curves of soil N<sub>2</sub>O emissions (normalized to the baseline) after deforestation were developed:  $y = -0.31 \ln(x) + 1.53$  ( $R^2 = 0.30$ ) and  $y = -0.454 \ln(x) + 2.21$  ( $R^2 = 0.09$ ). The first logarithmic function uses data collected by a review analysis<sup>74</sup>, and the second is based upon this but further considers observations from ref.<sup>21</sup> and ref.<sup>75</sup>. In the first function,  $x$  indicates pasture age in years after deforestation, and  $y$  (unitless; 0–1) indicates the ratio of pasture N<sub>2</sub>O emission over the N<sub>2</sub>O emission from the nearby reference mature forest. In the second function,  $x$  indicates secondary forest age and  $y$  indicates the ratio of secondary forest N<sub>2</sub>O emission over that of a reference mature forest. This form of the response functions can effectively reproduce the short-lived increase in soil N<sub>2</sub>O emissions after initial forest clearing and the gradually declining emission rates of converted crops and/or pastures<sup>21,76</sup>. Using these two curves and the baseline, we kept track of the N<sub>2</sub>O reduction of tropical forests and the post-deforestation crop/pasture N<sub>2</sub>O emissions at an annual timescale. This book-keeping method was applied to the two deforestation area datasets (Supplementary Information section 2.8), so we could investigate not only the difference caused by the two sets of land use data but also the difference between this empirical method and the process-based model. For land conversion from natural vegetation to croplands or pastures, DLEM uses a similar strategy to that used in ref.<sup>72</sup> and ref.<sup>77</sup> to simulate its influences on carbon and nitrogen cycles. Moreover, through using the sites of field observation from ref.<sup>20</sup> and ref.<sup>75</sup>, we estimated N<sub>2</sub>O emission from secondary tropical forests based on the algorithm:  $y = 0.0084x + 0.2401$  ( $R^2 = 0.44$ ).  $x$  indicates secondary forest age and  $y$  indicates the ratio of secondary forest N<sub>2</sub>O emission over that of a reference mature forest. The difference between primary forests and secondary forests were subtracted from natural soil emissions simulated by six terrestrial biosphere models in NMIP.

We calculated the ensemble of oceanic N<sub>2</sub>O emission based on the bottom-up approach (five ocean biogeochemical models; Supplementary Table 18) and the top-down approach (five estimates from four inversion models; Supplementary Table 19). The atmospheric burden and its rate of change during 1980–2016 were derived from mean maritime surface mixing ratios of N<sub>2</sub>O (refs.<sup>78,79</sup>) with a conversion factor of 4.79 Tg N ppb<sup>-1</sup> (ref.<sup>80</sup>). Combining uncertainties in measuring the mean surface mixing ratios<sup>78</sup> and that of converting surface mixing ratios to a global mean abundance<sup>80</sup>, we estimate an uncertainty in the burden of  $\pm 1.4\%$ . Annual change in atmospheric abundance is calculated from the combined NOAA and AGAGE record of surface N<sub>2</sub>O and uncertainty is taken from the IPCC assessment report 5 (ref.<sup>3</sup>). There is an agreement between stratospheric loss from atmospheric chemistry transport models (top-down modelled chemical sink<sup>18,81</sup>) and satellite observations from a photolysis model (observed photochemical sink<sup>1</sup>), differing by only about 1 Tg N yr<sup>-1</sup>. The satellite-based lifetime,  $116 \pm 9 \text{ yr}$ , gives an overall uncertainty in the annual loss of  $\pm 8\%$ . The tropospheric loss of N<sub>2</sub>O from reaction with O(<sup>1</sup>D) is included in the observed atmospheric chemical sink (Table 1) and is small (around 1% of the stratospheric sink), with an estimated range of 0.1 to 0.2 Tg N yr<sup>-1</sup>.

## Comparison with the IPCC guidelines

The IPCC has provided guidance to quantify N<sub>2</sub>O emissions, which is widely used in emission inventories for reporting to the UNFCCC. Over time the recommended approaches have changed, which is critical for estimating emissions from agricultural soils, the largest emission source. Previous global N<sub>2</sub>O assessments<sup>52,82,83</sup> based on the IPCC 1996 guidelines<sup>84</sup> attributed about 6.3 Tg N yr<sup>-1</sup> to the agricultural sector, including both direct and indirect emissions. This estimate is notably larger than our results (Fig. 1, Table 1) derived from multiple methods, and is also larger than the most recent estimates from global inventories (EDGAR v4.3.2, FAOSTAT and GAINS) that are based on the IPCC 2006 guidelines<sup>14</sup>. The main reason is that indirect emissions from leaching and groundwater were overestimated in previous studies<sup>85</sup>. Correspondingly, projections of atmospheric N<sub>2</sub>O concentrations that are based on these overestimated emissions<sup>82</sup> led to biased estimates. For example, in ref.<sup>82</sup>, atmospheric N<sub>2</sub>O concentrations were expected to be 340–350 ppb in the year 2020, instead of 333 ppb<sup>5</sup> as observed. The 2019 Refinement to the 2006 IPCC Guidelines for National Greenhouse Gas Inventories has recently been published<sup>86</sup> and adopts the same approach for nitrogen application on soils, but considers the effects of different climate regimes. The new guidelines, which are based on a wealth of new scientific literature, proposed much smaller emissions from grazing animals, by a factor of 5–7. Our preliminary calculations indicate that global soil emissions based on these new guidelines may decrease by 20%–25%. Integrating estimates that rely on the IPCC methodology with estimates from process-based models provides for a more balanced assessment in this paper. We also added information from assessments<sup>87,88</sup> that derived agricultural emissions as the difference between atmospheric terms and other emissions such as combustion, industry and nature, and they gave comparable magnitudes (4.3–5.8 Tg N yr<sup>-1</sup>) to our bottom-up results.

## Uncertainty

Current data analysis and synthesis of long-term N<sub>2</sub>O fluxes are based on a wide variety of top-down and bottom-up methods. Top-down approaches, consisting of four inversion frameworks<sup>89–92</sup>, provide a wide range of estimates largely because of systematic errors in the modelled atmospheric transport and stratospheric loss of N<sub>2</sub>O. In addition, the emissions from top-down analyses are dependent on the magnitude and distribution of the prior flux estimates to an extent that is strongly determined by the number of atmospheric N<sub>2</sub>O measurements<sup>18</sup>. Inversions are generally not well constrained (and thus rely heavily on a priori estimates) in Africa, Southeast Asia, southern South America, and over the oceans, owing to the paucity of observations in these regions. The improvement of atmospheric transport models, more accurate priors, and more atmospheric N<sub>2</sub>O measurements would reduce uncertainty in further top-down estimates, particularly for ocean and regional emissions.

Bottom-up approaches are subject to uncertainties in various sources from land<sup>16</sup> and oceans<sup>32</sup>. For process-based models (for example, NMIP and ocean biogeochemical models), the uncertainty is associated with differences in model configuration as well as process parameterization<sup>16,32</sup>. The uncertainty of estimates from NMIP could be reduced in several ways<sup>16</sup>. First, the six models in NMIP exhibited different spatial and temporal patterns of N<sub>2</sub>O emissions even though they used the same forcings. Although these models have considered essential biogeochemical processes in soils (for example, biological nitrogen fixation, nitrification/denitrification, mineralization/immobilization, etc.)<sup>93</sup>, some missing processes such as freeze–thaw cycles and ecosystem disturbances should be included in terrestrial biosphere models to reduce uncertainties. Second, the quality of input datasets—specifically the amount and timing of nitrogen application, and spatial and temporal changes in distribution of natural vegetation and agricultural land—is critical for accurately simulating soil N<sub>2</sub>O emissions. Third,

national and global N<sub>2</sub>O flux measurement networks<sup>17</sup> could be used to validate model performance and to constrain large-scale model simulations. Data assimilation techniques could be used to improve model accuracy.

Current remaining uncertainty in global ocean model estimates of N<sub>2</sub>O emission includes the contribution of N<sub>2</sub>O flux derived from the tropical oceanic low oxygen zones (for example, the eastern Equatorial Pacific, the northern Indian ocean) relative to the global ocean. These low oxygen zones are predominantly influenced by high yield N<sub>2</sub>O formation processes (for example, denitrification and enhanced nitrification). Regional observation-based assessments have also suggested that these regions may produce more N<sub>2</sub>O than is simulated by the models<sup>32</sup>. The current generation of global ocean biogeochemistry models are not sufficiently accurate to represent the high N<sub>2</sub>O production processes in low-oxygen zones and their associated variability (see refs.<sup>34,94,95</sup> for more detail). Thus, precisely representing the local ocean circulation and associated biogeochemical fluxes of these regions could further reduce the uncertainty in estimates of global and regional oceanic N<sub>2</sub>O emissions.

Regardless of the tier approach used, greenhouse-gas inventories for agriculture suffer from high uncertainty in the underlying agriculture and rural data and statistics used as input, including statistics on fertilizer use, livestock manure availability, storage and applications, and nutrient, crop and soils management. For instance, animal waste management is an uncertain aspect, because much of the manure is either not used, or is used as a fuel or building material, or may be discharged directly to surface water<sup>96</sup>, with important repercussions for the calculated emissions. Furthermore, greenhouse-gas inventories using default emission factors show large uncertainties at local to global scales, especially for agricultural N<sub>2</sub>O emissions, due to the poorly captured dependence of emission factors on spatial diversity in climate, management, and soil physical and biochemical conditions<sup>2,22</sup>. It is well known, for example from the IPCC guidelines, that higher-tier greenhouse-gas inventories may provide more reasonable estimates by using the alternative emission factors that are disaggregated by environmental factors and management-related factors<sup>86</sup>. A large range of emission factors have been used to estimate aquaculture N<sub>2</sub>O emissions<sup>31,39,62,87</sup>, and long-term estimates of nitrogen flows in freshwater and marine aquaculture are scarce<sup>30</sup>. Uncertainty also remains in several N<sub>2</sub>O sources that have not yet been fully understood or quantified. To date, robust estimates of N<sub>2</sub>O emissions from global peatland degradation are still lacking, although we have accounted for N<sub>2</sub>O emissions due to the drainage of organic soils (histosols) obtained from FAOSTAT and GAINS databases<sup>28,41</sup>. Recent evidence shows that permafrost thawing<sup>97</sup> and the freeze–thaw cycle<sup>98</sup> contribute to increasing N<sub>2</sub>O emissions; however, are not well established in the current estimates of the global N<sub>2</sub>O budget.

## Statistics

The Mann–Kendall test in R-3.4.4 was used to assess the significance of trends in annual N<sub>2</sub>O emissions from each sub-sector based on the bottom-up approach.

## Data availability

The relevant datasets of this study are archived in the box site of the International Center for Climate and Global Change Research at Auburn University (<https://auburn.box.com/>). Researchers that are interested in using the results made available in the repository are encouraged to contact the original data providers.

## Code availability

The relevant codes used in this study are archived in the box site of the International Center for Climate and Global Change Research at Auburn University (<https://auburn.box.com/>).

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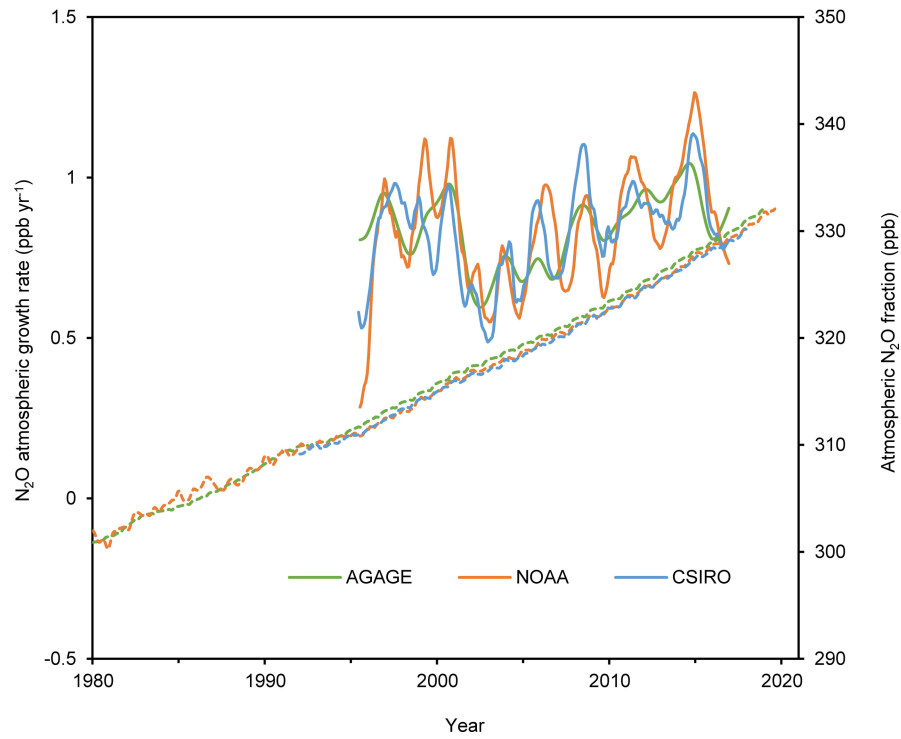
**Additional information**

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**Correspondence and requests for materials** should be addressed to H.T.

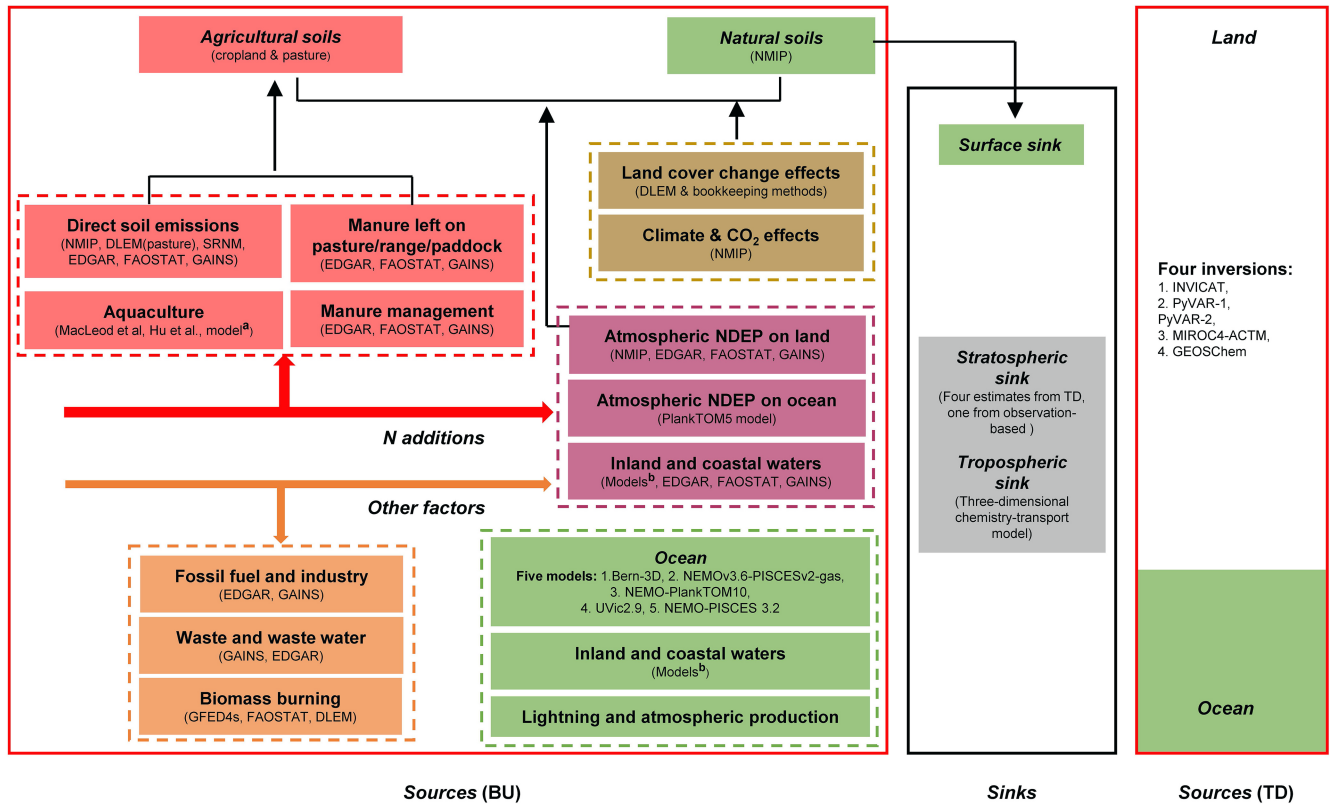
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**Extended Data Fig. 1 | Global mean growth rates and atmospheric concentration of N<sub>2</sub>O.** Global mean growth rates (solid lines, during 1995–2017) and atmospheric N<sub>2</sub>O concentration (dashed lines, during 1980–2017) are from the AGAGE<sup>6</sup> (green), NOAA<sup>5</sup> (orange) and CSIRO (blue) networks.

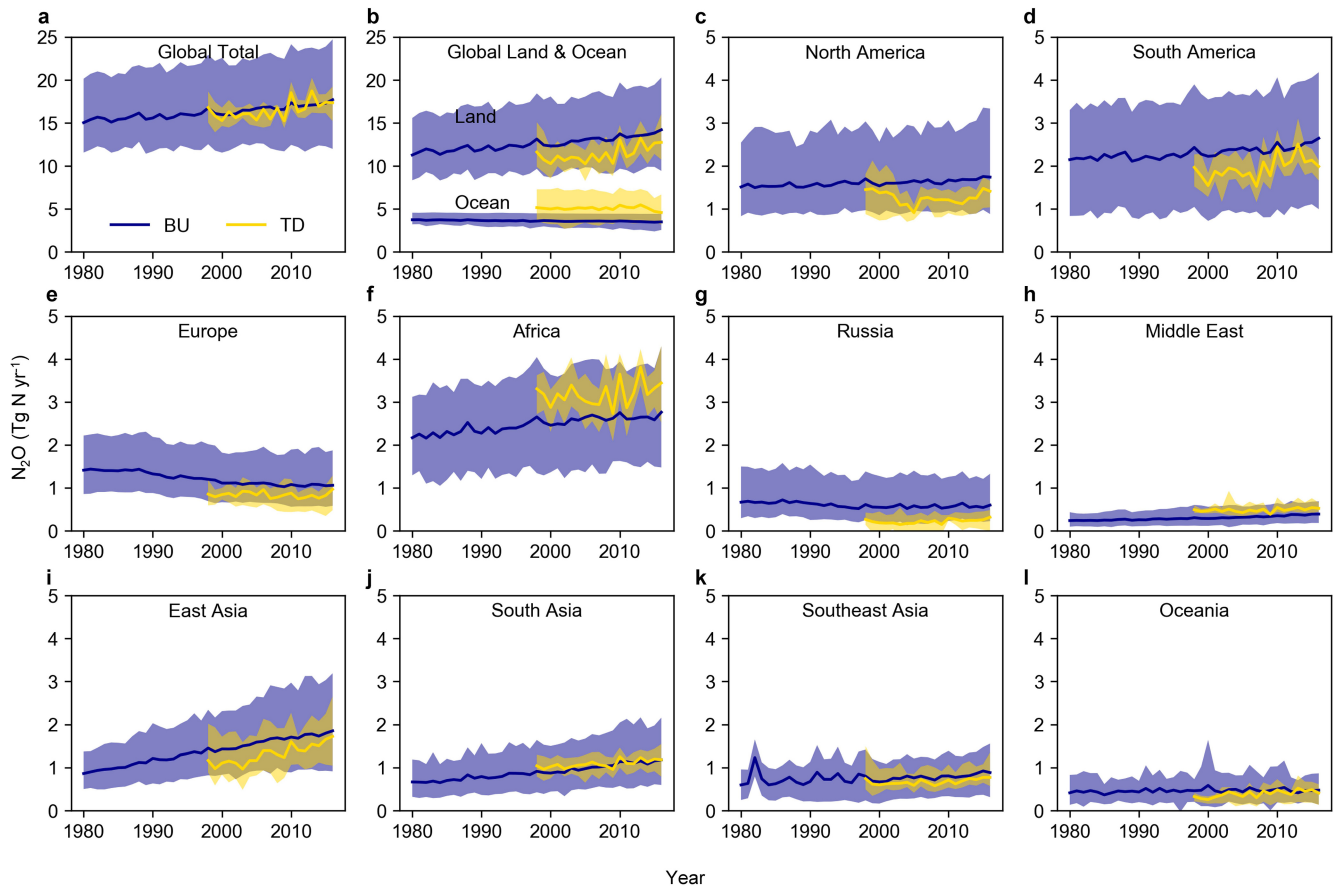
Global mean growth rates were calculated with annual time steps and are shown as 12-month moving averages. Growth rates are not calculated before 1995 owing to insufficient data and higher uncertainties on the measurements.



**Extended Data Fig. 2 | The methodology for data synthesis of the global N<sub>2</sub>O budget.** BU and TD represent bottom-up and top-down methods, respectively. The colour codes are the same as that used in Table 1 and Figs. 1–3. We use both approaches, including 22 bottom-up and five top-down estimates of N<sub>2</sub>O fluxes from land and oceans. For sources estimated by the bottom-up approach, we include six process-based terrestrial biosphere modelling studies<sup>16</sup>; five process-based ocean biogeochemical models<sup>99</sup>; one nutrient budget model<sup>30,60,61</sup>; five inland water modelling studies<sup>35,36,50,51,68</sup>; one statistical model SRNM based on spatial extrapolation of field measurements<sup>17</sup>; and four greenhouse-gas inventories: EDGAR v4.3.2<sup>100</sup>, FAOSTAT<sup>101</sup>, GAINS<sup>41</sup>, and

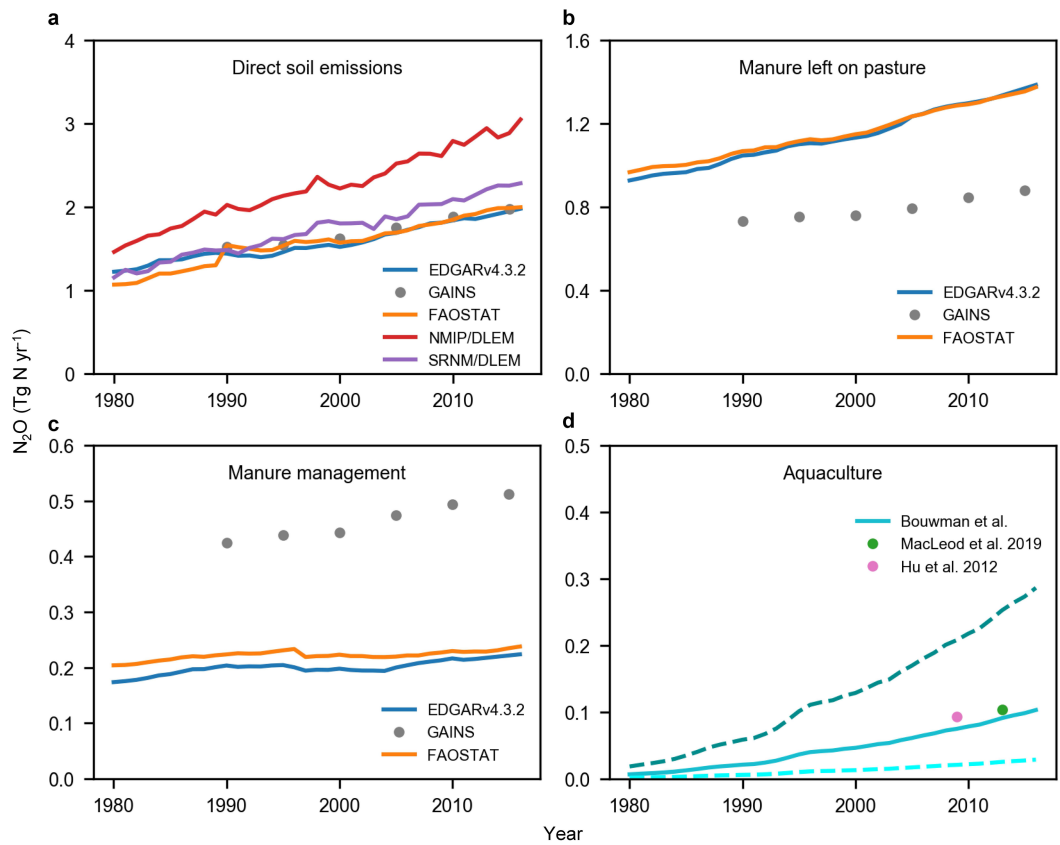
GFED4s<sup>102</sup>. In addition, previous studies regarding estimates of surface sink<sup>58,73</sup>, lightning<sup>53,54</sup>, atmospheric production<sup>56,57,103</sup>, aquaculture<sup>31,62</sup> and model-based tropospheric sink<sup>81</sup> and observed stratospheric sink<sup>1</sup> are included in the current synthesis. <sup>a</sup>Ref. <sup>31</sup> and ref. <sup>62</sup> provide global aquaculture N<sub>2</sub>O emissions in 2013 and in 2009, respectively; and the nutrient budget model<sup>30,60,61</sup> provides nitrogen flows in global freshwater and marine aquaculture over the period 1980–2016. <sup>b</sup>Model-based estimates of N<sub>2</sub>O emissions from inland and coastal waters include rivers and reservoirs<sup>35,36</sup>, lakes<sup>51</sup>, estuaries<sup>35</sup>, coastal zones (that is, seagrasses, mangroves, saltmarsh and intertidal saltmarsh)<sup>68</sup> and coastal upwelling<sup>50</sup>.





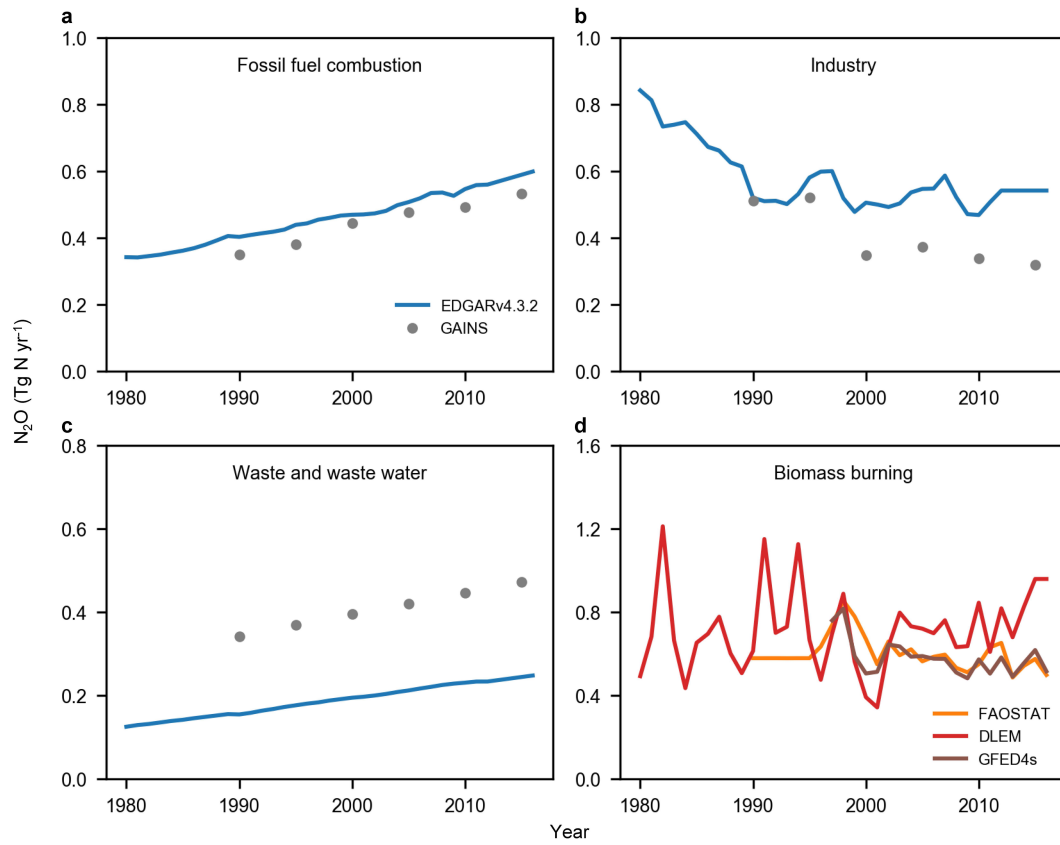
**Extended Data Fig. 3 | Comparison of annual total  $N_2O$  emissions at global and regional scales estimated by bottom-up and top-down approaches.** The blue lines represent the mean  $N_2O$  emission from bottom-up methods and

the shaded areas show minimum and maximum estimates; the gold lines represent the mean  $N_2O$  emission from top-down methods and the shaded areas show minimum and maximum estimates.



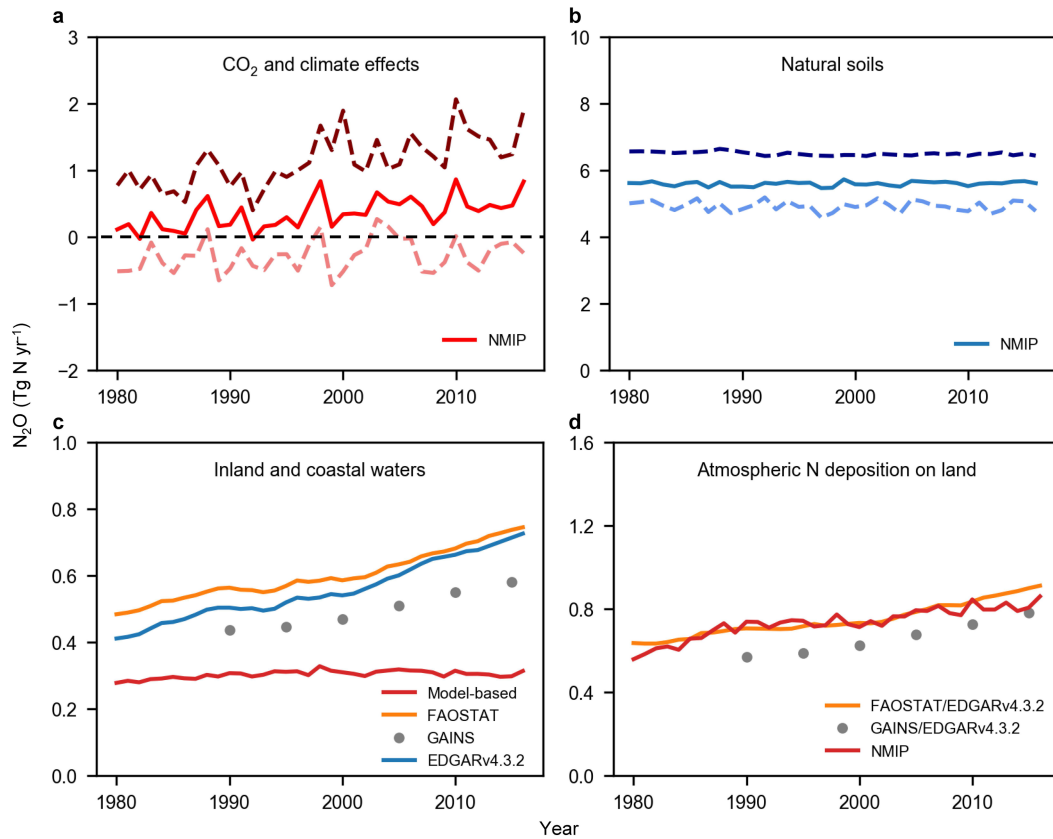
**Extended Data Fig. 4 | Global agricultural  $N_2O$  emissions.** **a**, Direct emission from agricultural soils associated with mineral fertilizer, manure and crop residue inputs, and cultivation of organic soils based on EDGAR v4.3.2, GAINS, FAOSTAT, NMIP/DLEM and SRNM/DLEM estimates. NMIP/DLEM or SRNM/DLEM indicates the combination of  $N_2O$  emission estimated by NMIP or SRNM from croplands with  $N_2O$  emission from intensively managed grassland (pasture) by estimated by DLEM. **b**, Direct emission from the global total area under permanent meadows and pasture, due to manure nitrogen deposition

(left on pasture) based on EDGAR v4.3.2, FAOSTAT and GAINS estimates. **c**, Emission from manure management based on FAOSTAT, GAINS and EDGAR v4.3.2. **d**, Aquaculture  $N_2O$  emission based on a nutrient budget model<sup>30</sup>, ref.<sup>31</sup> and ref.<sup>62</sup>; the solid line represents the 'best estimate' that is the product of emission factor (1.8%) and nitrogen waste from aquaculture provided by the nutrient budget model; the dashed lines represent the minimum and maximum values.



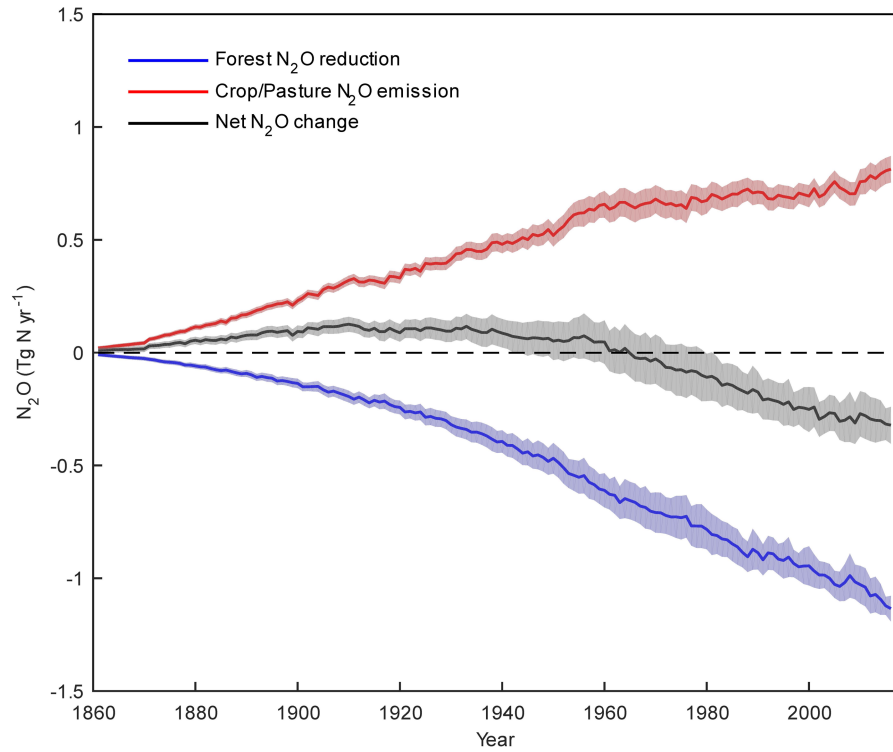
**Extended Data Fig. 5 | Global  $N_2O$  emission from other direct anthropogenic sources.** **a**, Emission from fossil fuel combustion based on EDGAR v4.3.2 and GAINS estimates. **b**, Emission from industry based on EDGAR

v4.3.2 and GAINS estimates. **c**, Emission from waste and waste water based on EDGAR v4.3.2 and GAINS estimates. **d**, Emission from biomass burning based on FAOSTAT, DLEM, and GFED4s estimates.



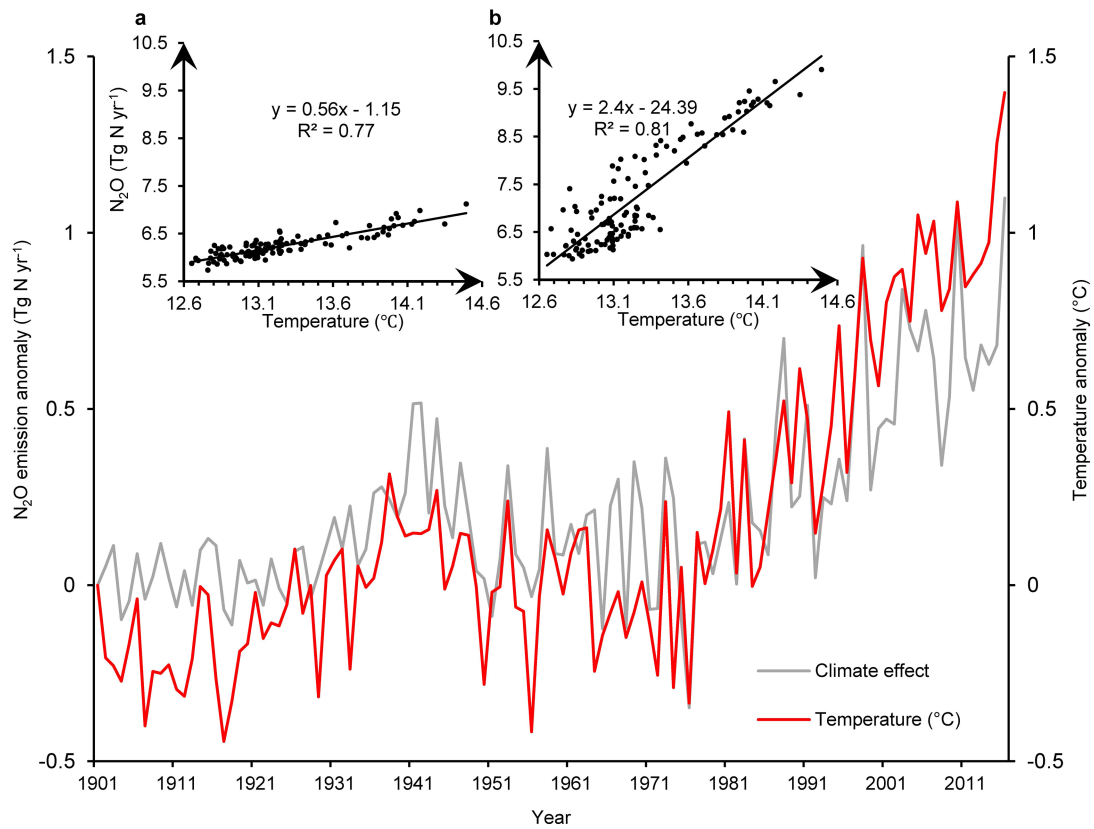
**Extended Data Fig. 6 | Global N<sub>2</sub>O emissions from natural soils, inland and coastal waters and due to change in climate, atmospheric CO<sub>2</sub> and nitrogen deposition.** **a**, Changes in global soil N<sub>2</sub>O fluxes due to changing CO<sub>2</sub> and climate. **b**, Global natural soil N<sub>2</sub>O emissions without consideration of land use change (for example, deforestation) and without consideration of indirect anthropogenic effects via global change (that is, climate, increased CO<sub>2</sub> and atmospheric nitrogen deposition). The estimates are based on NMIP estimates during 1980–2016 including six process-based land biosphere models. Here, we also subtracted the difference between including and not including emissions from secondary forests (that grow back after pasture or cropland abandonment) as part of natural soil emissions based on NMIP estimates. The

solid lines represent the ensemble and dashed lines show the minimum and maximum values. **c**, Global anthropogenic N<sub>2</sub>O emission from inland waters, estuaries, coastal zones based on models (model-based), FAOSTAT, GAINS and EDGAR v4.3.2 estimates. **d**, Emission due to atmospheric nitrogen deposition on land based on NMIP, FAOSTAT/EDGAR v4.3.2 and GAINS/EDGAR v4.3.2. FAOSTAT/EDGAR v4.3.2 or GAINS/EDGAR v4.3.2 indicates the combination of agricultural source estimates from FAOSTAT or GAINS with non-agricultural source estimates from EDGAR v4.3.2. A process-based model DLEM<sup>36</sup> and a mechanistic stochastic model<sup>35,51</sup> were used to estimate N<sub>2</sub>O emission from inland waters and estuaries, whereas site-level emission rates of N<sub>2</sub>O were upscaled to estimate global N<sub>2</sub>O fluxes from the global seagrass area<sup>68</sup>.



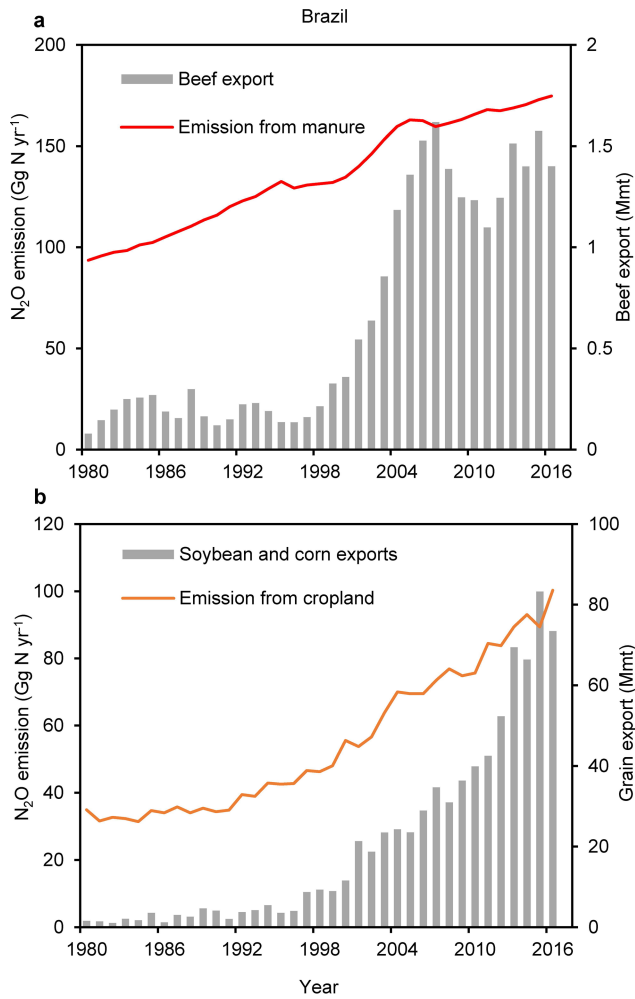
**Extended Data Fig. 7 | Global N<sub>2</sub>O dynamics due to land cover changes.** The blue line represents the mean forest N<sub>2</sub>O reduction caused by the long-term effect of reduced mature forest area (that is, deforestation) and shaded areas show minimum and maximum estimates; the red line represents the mean N<sub>2</sub>O emission from the post-deforestation pulse effect (that is, crop/pasture N<sub>2</sub>O

emissions from legacy nitrogen of previous forest soil, not accounting for new fertilizer nitrogen added to these crop/pasture lands) and shaded areas show minimum and maximum estimates; the grey line represents the mean net deforestation emission of N<sub>2</sub>O and shaded areas show minimum and maximum estimates.

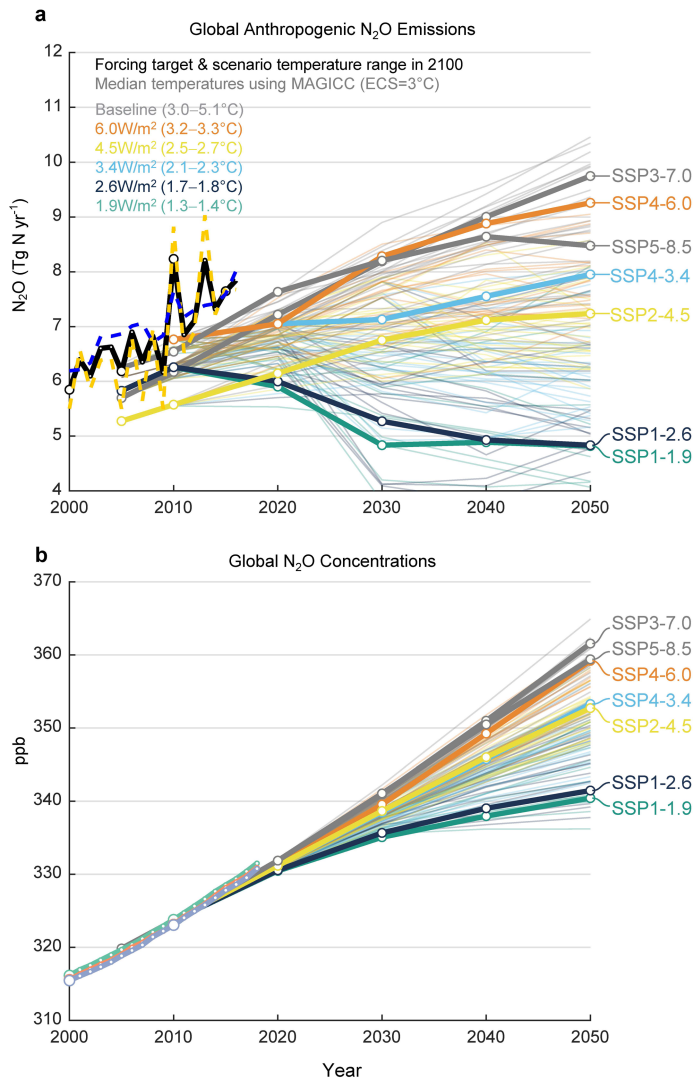


**Extended Data Fig. 8 | Global simulated N<sub>2</sub>O emission anomaly due to climate effect and global annual land surface temperature anomaly during 1901–2016.** Global N<sub>2</sub>O emission anomalies are the ensemble of six process-based land biosphere models in NMIP. The temperature data were obtained from the CRU-NCEP v8 climate dataset (<https://vesg.ipsl.upmc.fr>). **a.** The correlation between average global annual land surface temperature and simulated N<sub>2</sub>O

emissions (that is, the result of SE6 experiment in NMIP<sup>16</sup>) considering annual changes in climate but keeping all other factors (that is, nitrogen fertilizer, manure, NDEP, increased CO<sub>2</sub> and land cover change) at the level of 1860. **b.** The correlation between average global annual land surface temperature and simulated N<sub>2</sub>O emissions (that is, the result of SE1 experiment in NMIP<sup>16</sup>) considering annual changes in all factors during 1860–2016.



**Extended Data Fig. 9 | Direct soil emissions and agricultural product trades in Brazil.** **a**, The red line shows the ensemble direct  $N_2O$  emissions from livestock manure based on EDGAR v4.3.2, GAINS and FAOSTAT, the sum of ‘manure left on pasture’ and ‘manure management’. The grey columns show the amount of beef exported by Brazil. **b**, Orange line shows the ensemble direct  $N_2O$  emissions from croplands due to nitrogen fertilization based on NMIP and SRNM. The grey columns show the amount of soybeans and corn exported by Brazil. Data regarding beef and cereal product exports were adapted from the ABIEC (beef) and FAOSTAT (soybean and corn) databases.  $Mmt\ yr^{-1}$  represents millions of metric tons per year.



**Extended Data Fig. 10 | A comparison of anthropogenic N<sub>2</sub>O emissions and atmospheric N<sub>2</sub>O concentrations in the unharmonized SSPs.** An extension of Fig. 4, in which the emission and concentration data are the same as in Fig. 4. **a**, Global anthropogenic N<sub>2</sub>O emissions; **b**, Global N<sub>2</sub>O concentrations. The unharmonized emissions from the Integrated Assessment Models (IAMs)<sup>104</sup> show a large variation due to different input data and model assumptions. Comparison with Fig. 4b, d illustrates the modifications to the IAM scenario data for use in CMIP6. All baseline scenarios (SSP 3–7.0 and SSP 5–8.5; without climate policy applied) are shown in grey regardless of the radiative forcing level they reach in 2100.