

# A comprehensive quantification of global nitrous oxide sources and sinks

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91 Nitrous oxide (N<sub>2</sub>O), like carbon dioxide, is a long-lived greenhouse gas that accumulates in  
92 the atmosphere. The increase in atmospheric N<sub>2</sub>O concentrations over the past 150 years  
93 has contributed to stratospheric ozone depletion<sup>1</sup> and climate change<sup>2</sup>. Current national  
94 inventories do not provide a full picture of N<sub>2</sub>O emissions owing to their omission of  
95 natural sources and the limitations in methodology for attributing anthropogenic sources.  
96 In order to understand the steadily increasing atmospheric burden (about 2 percent per  
97 decade) and develop effective mitigation strategies, it is essential to improve quantification  
98 and attribution of natural and anthropogenic contributions and their uncertainties. Here  
99 we present a global N<sub>2</sub>O inventory that incorporates both natural and anthropogenic  
100 sources and accounts for the interaction between nitrogen additions and the biochemical  
101 processes that control N<sub>2</sub>O emissions. We use bottom-up (inventory; statistical  
102 extrapolation of flux measurements; process-based land and ocean modelling) and top-  
103 down (atmospheric inversion) approaches to provide a comprehensive quantification of  
104 global N<sub>2</sub>O sources and sinks resulting from 21 natural and human sectors between 1980  
105 and 2016. Global N<sub>2</sub>O emissions were 17.0 (minimum-maximum: 12.2–23.5) teragrams of  
106 nitrogen per year (bottom-up) and 16.9 (15.9–17.7) teragrams of nitrogen per year (top-  
107 down) between 2007 and 2016. Global human-induced emissions, which are dominated by  
108 nitrogen additions to croplands, increased by 30% over the past four decades to 7.3 (4.2–  
109 11.4) teragrams of nitrogen per year. This increase was mainly responsible for the growth  
110 in the atmospheric burden. Our findings point to growing N<sub>2</sub>O emissions in emerging  
111 economies—particularly Brazil, China and India. Analysis of process-based model  
112 estimates reveals an emerging N<sub>2</sub>O–climate feedback resulting from interactions between  
113 nitrogen additions and climate change. The recent growth in N<sub>2</sub>O emissions exceeds some

114 **of the highest projected emission scenarios<sup>3,4</sup>, underscoring the urgency to mitigate N<sub>2</sub>O**  
115 **emissions.**

116  
117 Nitrous oxide (N<sub>2</sub>O) is a long-lived stratospheric ozone-depleting substance and greenhouse gas  
118 (GHG) with a current atmospheric lifetime of 116±9 years (ref. <sup>1</sup>). The concentration of  
119 atmospheric N<sub>2</sub>O has increased by over 20% from 270 parts per billion (ppb) in 1750 to 331 ppb  
120 in 2018 (Extended Data Fig. 1), with the fastest growth observed in the past five decades<sup>5,6</sup>. Two  
121 key biochemical processes, nitrification and denitrification, control N<sub>2</sub>O production in both  
122 terrestrial and aquatic ecosystems, and are regulated by multiple environmental and biological  
123 factors, such as temperature, water, oxygen, acidity, substrate availability<sup>7</sup>, particularly nitrogen  
124 (N) fertilizer use and livestock manure management, and recycling<sup>8-10</sup>. In the coming decades,  
125 N<sub>2</sub>O emissions are expected to continue increasing due to the growing demand for food, feed,  
126 fiber and energy, and a rising source from waste generation and industrial processes<sup>4,11,12</sup>. Since  
127 1990, anthropogenic N<sub>2</sub>O emissions have been annually reported by Annex I Parties to the  
128 United Nations Framework Convention on Climate Change (UNFCCC). More recently, over 190  
129 national signatories to the Paris Agreement are now required to report biannually their national  
130 GHG inventory with sufficient detail and transparency to track progress towards their Nationally  
131 Determined Contributions. Yet, these inventories do not provide a full picture of N<sub>2</sub>O emissions  
132 due to their omission of natural sources, the limitations in methodology for attributing  
133 anthropogenic sources, and missing data for a number of key regions (e.g., South America,  
134 Africa)<sup>2,9,13</sup>. Moreover, we need a complete account of all human activities that accelerate the  
135 global N cycle and that interact with the biochemical processes controlling the fluxes of N<sub>2</sub>O in  
136 both terrestrial and aquatic ecosystems<sup>2,8</sup>. Here we present a comprehensive, consistent analysis

137 and synthesis of the global N<sub>2</sub>O budget across all sectors, including natural and anthropogenic  
138 sources and sinks, using both bottom-up (BU) and top-down (TD) methods and their cross-  
139 constraints. Our assessment enhances understanding of the global N cycle and will inform policy  
140 development for N<sub>2</sub>O mitigation, ideally helping to curb warming to levels consistent with the  
141 long-term goal of the Paris Agreement.

142 A reconciling framework (described in Extended Data Fig. 2) was utilized to take full  
143 advantage of BU and TD approaches in estimating and constraining sources and sinks of N<sub>2</sub>O.  
144 BU approaches include emission inventories, spatial extrapolation of field flux measurements,  
145 nutrient budget modeling, and process-based modeling for land and ocean fluxes. The TD  
146 approaches combine measurements of N<sub>2</sub>O mole fractions with atmospheric transport models in  
147 statistical optimization frameworks (inversions) to constrain the sources. Here we constructed a  
148 total of 43 flux estimates including 30 with BU approaches, five with TD approaches, and eight  
149 other estimates with observation and modeling approaches (see Methods; Extended Data Fig. 2).

150 With this extensive data and BU/TD framework, we establish the most comprehensive global  
151 and regional N<sub>2</sub>O budgets that include 18 sources and different versions of its chemical sink,  
152 which are further grouped into six categories (Fig. 1 and Table 1): 1) Natural sources (no  
153 anthropogenic effects) including a very small biogenic surface sink, 2) Perturbed fluxes from  
154 ecosystems induced by changes in climate, carbon dioxide (CO<sub>2</sub>) and land cover, 3) Direct  
155 emissions of N additions in the agricultural sector (Agriculture), 4) Other direct anthropogenic  
156 sources, which include fossil fuel and industry, waste and waste water, and biomass burning, 5)  
157 Indirect emissions from ecosystems that are either downwind or downstream from the initial  
158 release of reactive N into the environment, which include N<sub>2</sub>O release following transport and  
159 deposition of anthropogenic N via the atmosphere or water bodies as defined by the

160 Intergovernmental Panel on Climate Change (IPCC)<sup>14</sup>, and 6) The atmospheric chemical sink  
161 with one value derived from observations and the other (TD) from the inversion models. To  
162 quantify and attribute the regional N<sub>2</sub>O budget, we further partition the Earth's ice-free land into  
163 ten regions (Fig. 2 and Supplementary Fig. 1). With the construction of these budgets, we  
164 explore the relative temporal and spatial importance of multiple sources and sinks driving the  
165 atmospheric burden of N<sub>2</sub>O, their uncertainties, and interactions between anthropogenic forcing  
166 and natural fluxes of N<sub>2</sub>O as an emerging climate feedback.

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### 168 **The Global N<sub>2</sub>O Budget (2007–2016)**

169 The BU and TD approaches give consistent estimates of global total N<sub>2</sub>O emissions in the recent  
170 decade to well within their respective uncertainties, with values of 17.0 (min-max: 12.2–23.5) Tg  
171 N yr<sup>-1</sup> and 16.9 (15.9–17.7) Tg N yr<sup>-1</sup> for BU and TD sources, respectively. The global calculated  
172 atmospheric chemical sink (i.e., N<sub>2</sub>O losses via photolysis and reaction with O(<sup>1</sup>D) in the  
173 troposphere and stratosphere) is 13.5 (12.4–14.6) Tg N yr<sup>-1</sup>. The imbalance of sources and sinks  
174 of N<sub>2</sub>O derived from the averaged BU and TD estimates is 4.1 Tg N yr<sup>-1</sup>. This imbalance agrees  
175 well with the observed 2007–2016 increase in atmospheric abundance of 3.8–4.8 Tg N yr<sup>-1</sup> (see  
176 Methods). Natural sources from soils and oceans contributed 57% of total emissions (mean: 9.7;  
177 min-max: 8.0–12.0 Tg N yr<sup>-1</sup>) for the recent decade according to our BU estimate. We further  
178 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  
179 yr<sup>-1</sup> (see Methods).

180 Anthropogenic sources contributed on average 43% to the total N<sub>2</sub>O emission (mean: 7.3;  
181 min-max: 4.2–11.4 Tg N yr<sup>-1</sup>), in which direct and indirect emissions from N additions in  
182 agriculture and other sectors contributed ~52% and ~18%, respectively. Of the remaining

183 anthropogenic emissions, ~27% were from other direct anthropogenic sources including fossil  
184 fuel and industry (~13%), with ~3% from perturbed fluxes caused by climate/CO<sub>2</sub>/land cover  
185 change.

186

#### 187 **Four Decades of the Global N<sub>2</sub>O Budget**

188 The atmospheric N<sub>2</sub>O burden increased from 1462 Tg N in the 1980s to 1555 Tg N in the recent  
189 decade, with a possible uncertainty  $\pm 20$  Tg N. Our results (Table 1) demonstrate that global N<sub>2</sub>O  
190 emissions have also significantly increased, primarily driven by anthropogenic sources, with  
191 natural sources relatively steady throughout the study period. Our BU and TD global N<sub>2</sub>O  
192 emissions are comparable in magnitude during 1998–2016, but TD results imply a larger inter-  
193 annual variability (1.0 Tg N yr<sup>-1</sup>; Extended Data Fig. 3a). BU and TD approaches diverge in the  
194 magnitude of land versus ocean emissions, although they are consistent with respect to trends.  
195 Specifically, the BU land estimate during 1998–2016 was on average 1.8 Tg N yr<sup>-1</sup> higher than  
196 the TD estimate, but showed a slightly slower increasing rate of  $0.8 \pm 0.2$  Tg N yr<sup>-1</sup> per decade  
197 (95% confidence interval;  $P < 0.05$ ) compared to  $1.1 \pm 0.6$  Tg N yr<sup>-1</sup> per decade ( $P < 0.05$ ) from  
198 TD (Extended Data Fig. 3b). Since 2005, the difference in the magnitude of emissions between  
199 the two approaches has become smaller due to a large TD-inferred emission increase,  
200 particularly in South America, Africa, and East Asia (Extended Data Fig. 3d, f, i). Oceanic N<sub>2</sub>O  
201 emissions from BU [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>  
202 per decade ( $P < 0.05$ ), while the TD approach gave a higher but stable value of 5.1 (3.4–7.1) Tg  
203 N yr<sup>-1</sup> during 1998–2016 (Table 1).

204 Based on BU approaches, anthropogenic N<sub>2</sub>O emissions increased from 5.6 (3.6–8.7) Tg N yr<sup>-1</sup>  
205 in the 1980s to 7.3 (4.2–11.4) Tg N yr<sup>-1</sup> in the recent decade at a rate of  $0.6 \pm 0.2$  Tg N yr<sup>-1</sup> per

206 decade ( $P < 0.05$ ). Up to 87% of this increase is from direct emission from agriculture (71%) and  
207 indirect emission from anthropogenic N additions into soils (16%). Direct soil emission from  
208 fertilizer applications is the major source for agricultural emission increases, followed by a small  
209 but significant increase in emissions from livestock manure and aquaculture. The model-based  
210 estimates of direct soil emissions<sup>15-17</sup> exhibit a faster increase than the three inventories used in  
211 our study (see Methods; Extended Data Fig. 4a), which is largely attributed to the interactive  
212 effects between climate change and N additions as well as spatio-temporal variability in  
213 environmental factors such as rainfall and temperature that modulate the N<sub>2</sub>O yield from  
214 nitrification and denitrification. This result is in line with the elevated emission factor (EF)  
215 deduced from the TD estimates, in which the inversion-based soil emissions increased at a faster  
216 rate than suggested by the IPCC Tier 1 EF<sup>14</sup> (which assumes a linear response), especially after  
217 2009 (ref. <sup>18</sup>). The remaining causes of the increase are attributed to other direct anthropogenic  
218 sources (6%) and perturbed fluxes from climate/CO<sub>2</sub>/land cover change (8%). The part of fossil  
219 fuel and industry emissions decreased rapidly over 1980–2000 largely due to the installation of  
220 emissions abatement equipment in industrial facilities producing nitric and adipic acid. However,  
221 after 2000 such emissions began to increase slowly due to rising fossil fuel combustion  
222 (Extended Data Fig. 5a-b).

223 Our analysis of process-based model estimates indicates that soil N<sub>2</sub>O emissions accelerated  
224 substantially due to climate change since the early 1980s, which has offset the reduction due to  
225 elevated CO<sub>2</sub> concentration (Extended Data Fig. 6a). Elevated CO<sub>2</sub> enhances plant growth and  
226 thus increases N uptake, which in turn decreases soil N<sub>2</sub>O emissions<sup>16,19</sup>. Land conversion from  
227 tropical mature forests with higher N<sub>2</sub>O emissions to pastures and other unfertilized agricultural  
228 lands has significantly reduced global natural N<sub>2</sub>O emissions<sup>11,20,21</sup>. This decrease, however, was



229 partly offset by an increase in soil N<sub>2</sub>O emissions attributable to the temporary rise of emissions  
230 following deforestation (post-deforestation pulse effect) and background emissions from  
231 converted croplands or pastures<sup>21</sup> (see Methods; Extended Data Fig. 7).

232 From the ensemble of process-based land model emissions<sup>15,16</sup>, we estimate a global  
233 agricultural soil EF of 1.8% (1.3%–2.3%), which is significantly larger than the IPCC Tier-1  
234 default for direct emission of 1%. This higher EF, derived from process-based models, suggests a  
235 strong interactive effect between N additions and other global environmental changes (Table 1,  
236 Perturbed fluxes from climate, atmospheric CO<sub>2</sub>, and land cover change). Previous field  
237 experiments reported a better fit to local observations of soil N<sub>2</sub>O emissions when assuming a  
238 non-linear response to fertilizer N inputs under varied climate and soil conditions<sup>17,22</sup>. The non-  
239 linear response is likely also associated with long-term N accumulation in agricultural soils from  
240 N fertilizer use and in aquatic systems from N loads (the legacy effect)<sup>18,23</sup>, which provides more  
241 substrate for microbial processes<sup>18,24</sup>. The increasing N<sub>2</sub>O emissions estimated by process-based  
242 models<sup>16</sup> also suggest that recent climate change (particularly warming) may have boosted soil  
243 nitrification and denitrification processes, contributing to the growing trend in N<sub>2</sub>O emissions  
244 together with rising N additions to agricultural soils<sup>16,25-27</sup> (Extended Data Fig. 8).

245

### 246 **Regional N<sub>2</sub>O Budgets (2007–2016)**

247 BU approaches give estimates of N<sub>2</sub>O emissions in the five source categories, while TD  
248 approaches only provide total emissions (Fig. 2). BU and TD approaches indicate that Africa was  
249 the largest N<sub>2</sub>O source in the last decade, followed by South America (Fig. 2). BU and TD  
250 approaches agree well in the magnitudes and trends of N<sub>2</sub>O emissions from South Asia and  
251 Oceania (Extended Data Fig. 3j, l). For the remaining regions, BU and TD estimates are

252 comparable in their trends but diverge in their source strengths. Clearly, much more work on  
253 regional N<sub>2</sub>O budgets is needed, particularly for South America and Africa where we see larger  
254 differences between BU and TD estimates and larger uncertainty in each approach. Advancing  
255 the understanding and model representation of key processes responsible for N<sub>2</sub>O emissions from  
256 land and ocean are priorities for reducing uncertainties in BU estimates. Atmospheric  
257 observations in underrepresented regions of the world and better atmospheric transport models  
258 are essential for uncertainty reduction in TD estimates, while more accurate activity data and  
259 robust EFs are critical for GHG inventories (See Methods for additional discussion on  
260 uncertainty).

261 Based on the Global N<sub>2</sub>O Model Intercomparison Project (NMIP) estimates<sup>16</sup>, natural soil  
262 emissions (to different extents) dominated in tropical and sub-tropical regions. Soil N<sub>2</sub>O  
263 emissions in the tropics ( $0.1 \pm 0.04 \text{ g N m}^{-2} \text{ yr}^{-1}$ ) are about 50% higher than the global average,  
264 since many lowland, highly-weathered tropical soils have excess N relative to phosphorus<sup>20</sup>.  
265 Total anthropogenic emissions in the ten terrestrial regions were highest in East Asia (1.5;  
266 0.8–2.6 Tg N yr<sup>-1</sup>), followed by North America, Africa, and Europe. High direct agricultural N<sub>2</sub>O  
267 emissions can be attributed to large-scale synthetic N fertilizer applications in East Asia, Europe,  
268 South Asia, and North America, which together consume over 80% of the world's synthetic N  
269 fertilizers<sup>28</sup>. In contrast, direct agricultural emissions from Africa and South America are mainly  
270 induced by livestock manure that is deposited in pastures and rangelands<sup>28,29</sup>. East Asia  
271 contributed 71%–79% of global aquaculture N<sub>2</sub>O emissions; South Asia and Southeast Asia  
272 together contributed 10%–20% (refs. <sup>30,31</sup>). Indirect emissions play a moderate role in the total  
273 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

274 anthropogenic sources together contribute N<sub>2</sub>O emissions of approximately 0.2–0.4 Tg N yr<sup>-1</sup> in  
275 East Asia, Africa, North America, and Europe.

276 Both BU and TD estimates of ocean N<sub>2</sub>O emissions for northern, tropical, and southern ocean  
277 regions (90°–30°N, 30°N–30°S, and 30°–90°S, respectively) reveal that the tropical oceans  
278 contribute over 50% to the global oceanic source. In particular, the upwelling regions of the  
279 equatorial Pacific, Indian and tropical Atlantic (Fig. 3) provide significant sources of N<sub>2</sub>O<sup>32-34</sup>.  
280 BU estimates suggest the southern ocean is the second largest regional contributor with  
281 emissions about twice as high as from the northern oceans (53% tropical oceans, 31% southern  
282 oceans, 17% northern oceans), in line with their area, while the TD estimates suggest  
283 approximately equal contributions from the southern and northern oceans.

284

#### 285 **Four Decades of Anthropogenic N<sub>2</sub>O Emissions**

286 Trends in anthropogenic emissions varied among regions (Fig. 3). Fluxes from Europe and  
287 Russia decreased by a total of 0.6 (0.5–0.7) Tg N yr<sup>-1</sup> over the past 37 years (1980–2016). The  
288 decrease in Europe is associated with successful emissions abatement in industry as well as  
289 agricultural policies, while the decrease in Russia is associated with the collapse of the  
290 agricultural cooperative system after 1990. In contrast, fluxes from the remaining eight regions  
291 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%  
292 from Africa, 18% from South Asia, 13% from South America, only 6% from North America,  
293 and with remaining increases due to other regions.

294 The relative importance of each anthropogenic source to the total emission increase differs  
295 among regions. East Asia, South Asia, Africa, and South America show larger increases in total  
296 agricultural N<sub>2</sub>O emissions (direct and indirect) compared to the remaining six regions during

297 1980–2016 (Fig. 3). Southeast Asia, North America, and Middle East also show increasing direct  
298 N<sub>2</sub>O emissions but to smaller extent. Rising indirect emissions in these four regions (East Asia,  
299 South Asia, Africa, and South America) on average constitute 20% of total agricultural N<sub>2</sub>O  
300 emissions and are largely induced by the considerable increase in fertilizer N inputs to  
301 agricultural soils<sup>35,36</sup>. The most rapid increase in emissions from other direct anthropogenic  
302 sources was found in East Asia, primarily owing to the fast-growing industrial emissions. Africa  
303 and South Asia show a fast emission increase due to emissions from fossil fuel and industry and  
304 waste and waste water.

305 Our findings point to growing N<sub>2</sub>O emissions in emerging economies, particularly Brazil,  
306 China, and India. For example, we find here that the substantial increases in livestock manure  
307 left on pasture and in fertilizer use caused a ~120% increase in Brazilian agricultural N<sub>2</sub>O  
308 emissions during 1980–2016 (Extended Data Fig. 9). In addition to fertilizer applications, global  
309 livestock manure production has been growing steadily, in line with increased livestock  
310 numbers<sup>15,28</sup>. Rising demand for meat and dairy products has significantly increased global N<sub>2</sub>O  
311 emissions from livestock manure production and management associated with the expansion of  
312 pastures and grazing land<sup>37</sup>. Meanwhile, expansion of feed crop production to support the growth  
313 of livestock could further enhance global N<sub>2</sub>O emissions<sup>37,38</sup>. Likewise, increasing demand for  
314 fish has triggered a five-fold increase in global aquaculture production since the late 1980s<sup>39</sup>,  
315 with demand projected to increase further<sup>40</sup>, although this remains a small fraction (<1%) of total  
316 N<sub>2</sub>O emissions.

317 The acceleration of global N<sub>2</sub>O emissions resulting from anthropogenic sources is apparent in  
318 both BU and TD results and currently tracks the highest Representative Concentration Pathway  
319 (RCP8.5)<sup>4</sup> in the fifth assessment report (AR5) of IPCC<sup>2</sup> and exceeds all the Shared

320 Socioeconomic Pathways (SSPs)<sup>3</sup> in CMIP6 for the sixth assessment report (AR6) of IPCC (Fig.  
321 4). Observed atmospheric N<sub>2</sub>O concentrations are beginning to exceed predicted levels across all  
322 scenarios. Emissions need to be reduced to a level that is consistent with or below that in RCP2.6  
323 or SSP1-2.6 in order to limit warming well below the 2° C target of the Paris Agreement. Failure  
324 to include N<sub>2</sub>O within climate mitigation strategies will necessitate even greater abatement of  
325 CO<sub>2</sub> and CH<sub>4</sub>. Although N<sub>2</sub>O mitigation is difficult because N is the key-limiting nutrient in the  
326 agricultural production, this study demonstrates that effective mitigation actions have reduced  
327 emissions in some regions, such as Europe, through technological improvements in industry and  
328 improved N use efficiency in agriculture.

329 There are a number of mitigation options in the agriculture sector available for immediate  
330 deployment, including increased N use efficiency in (i) animal production through tuning of feed  
331 rations to reduce N excretion, and (ii) in crop production through precision delivery of N  
332 fertilizers, split applications and better timing to match N applications to crop demand,  
333 conservation tillage, prevention of waterlogging, and the use of nitrification inhibitors<sup>43,44</sup>.  
334 Success stories include the stabilization or reduction of N<sub>2</sub>O emissions through improving N use  
335 efficiency in the United States and Europe, while maintaining or even increasing crop yields<sup>44,45</sup>.  
336 There is every reason to expect that additional implementation of more sustainable practices and  
337 emerging technologies will lead to further reductions in these regions. For example, N<sub>2</sub>O  
338 emissions from European agricultural soils decreased by 21% between 1990 and 2010, a decline  
339 attributable to the implementation of the Nitrates Directive (an agricultural policy favoring  
340 optimization and reduction of fertilizer use as well as water protection legislation)<sup>46</sup>. For regions  
341 where emissions are growing, an immediate opportunity lies in the reduction of excess fertilizer  
342 use along with the implementation of more sustainable agricultural practices that together have

343 been shown to increase crop yields, reduce N<sub>2</sub>O emissions, increase water quality, and increase  
344 farm income<sup>47</sup>. In addition, N<sub>2</sub>O emissions can be efficiently abated in the chemical  
345 industry<sup>11,43,48,49</sup>, as has been achieved successfully in nitric acid plants in the European Union  
346 where industrial N<sub>2</sub>O emissions dropped from 11% to 3% of total emissions between 2007 and  
347 2012 (ref. <sup>46</sup>). Additional available strategies to reduce N<sub>2</sub>O emissions include promoting lower  
348 meat consumption in some parts of the world<sup>9</sup> and reducing food waste<sup>11</sup>.

349 We present the most comprehensive global N<sub>2</sub>O budget to date, with a detailed sectorial and  
350 regional attribution of sources and sinks. Each of the past four decades had higher global N<sub>2</sub>O  
351 emissions than the previous one, and in all, agricultural activities dominated the growth in  
352 emissions. Total industrial emissions have been quite stable with increased emissions from the  
353 fossil fuel sector offset to some extent by the decline in emissions in other industrial sectors as a  
354 result of successful abatement policies. We also highlight a number of complex interactions  
355 between N<sub>2</sub>O fluxes and human-driven changes whose impact on the global atmospheric N<sub>2</sub>O  
356 growth rate was previously unknown. Those interactions include the effects of climate change,  
357 increasing atmospheric CO<sub>2</sub>, and deforestation. Cumulatively, these exert a relatively small  
358 effect on the overall N<sub>2</sub>O growth, however, individual flux components, such as the growing  
359 positive climate-N<sub>2</sub>O feedback, are significant. These fluxes are not currently included in the  
360 national GHG reporting. We further find that Brazil, China, and India dominate the regional  
361 contributions to the increase in global N<sub>2</sub>O emissions over the most recent decade. Our extensive  
362 database and modelling capability fill current gaps in national and regional emissions  
363 inventories. Future research is needed to further constrain complex biogeochemical interactions  
364 between natural/anthropogenic fluxes and global environmental changes, which could lead to  
365 significant feedbacks in the future. Reducing excess N applications to croplands and adopting

366 precision fertilizer application methods provide the largest immediate opportunities for N<sub>2</sub>O  
367 emissions abatement.

368

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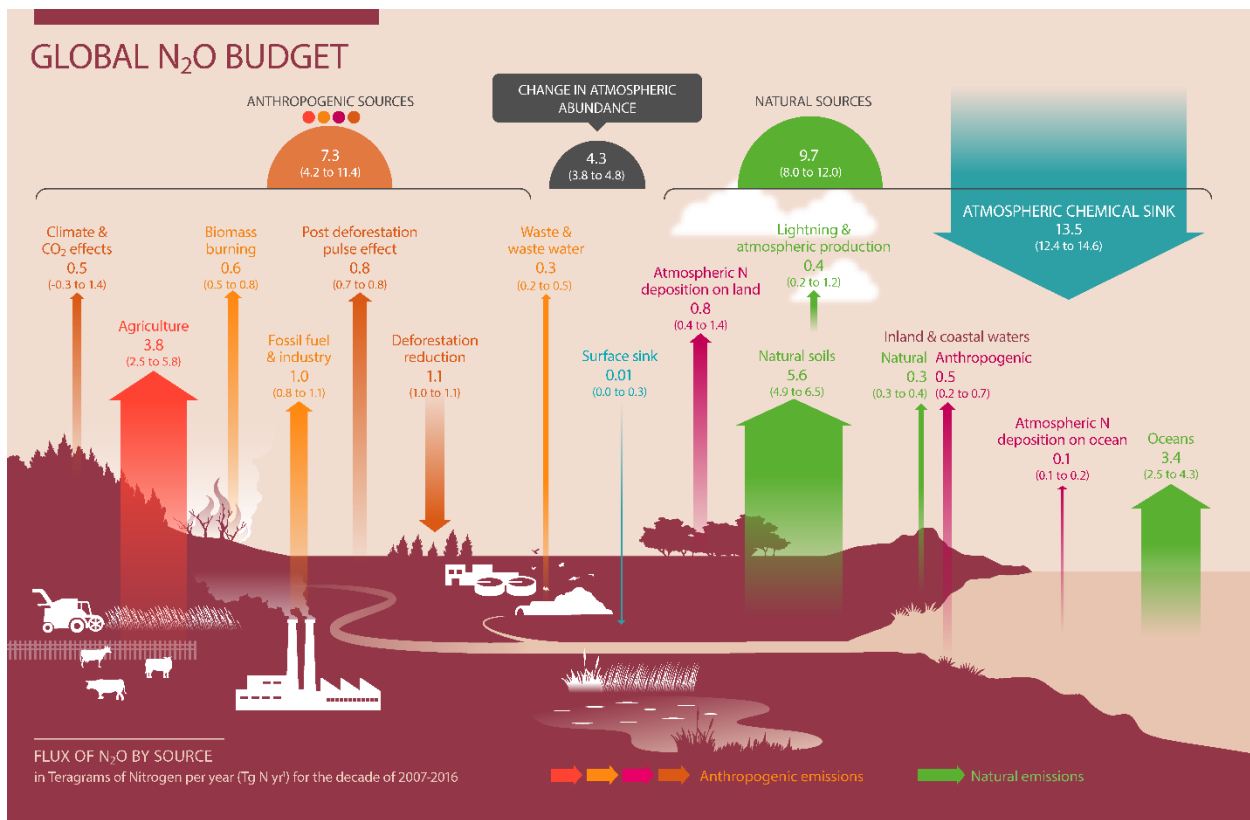
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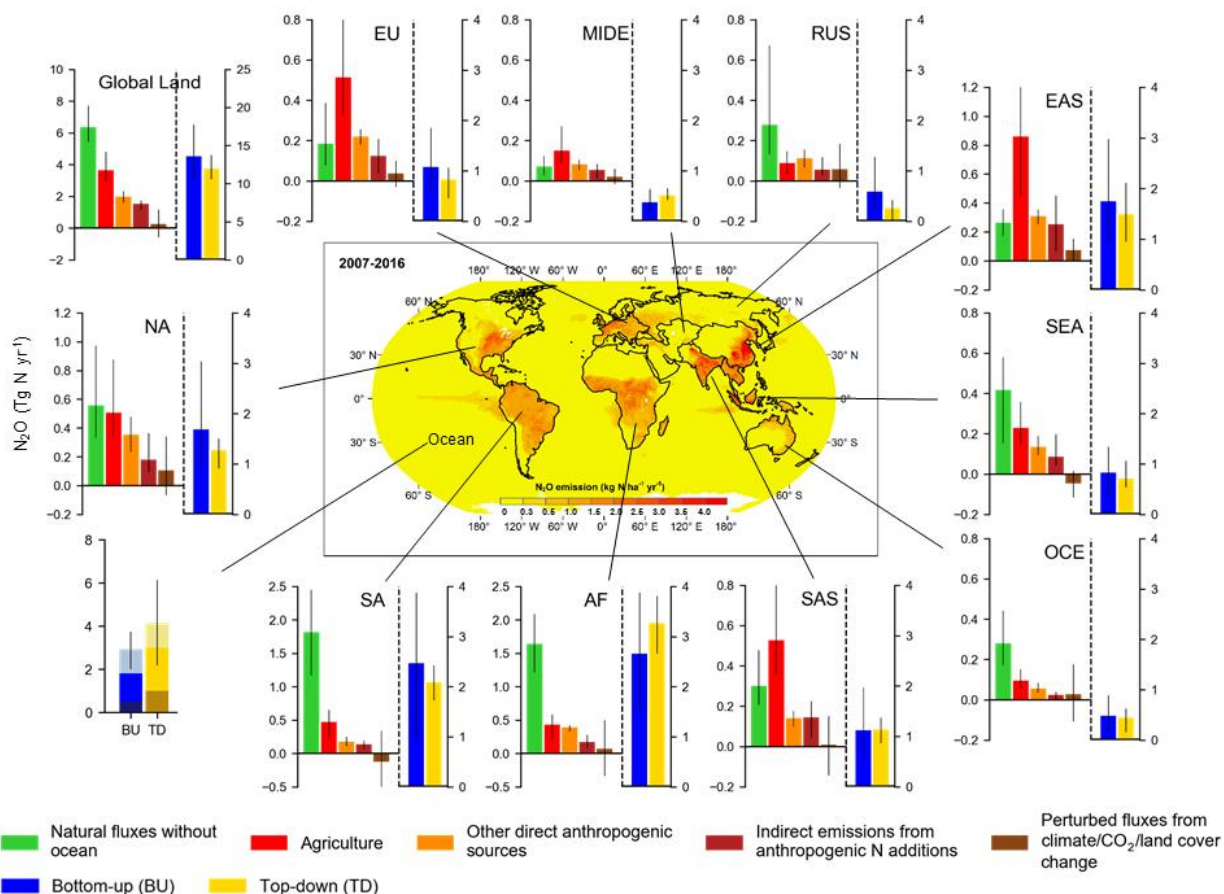
496 **Table 1** The global N<sub>2</sub>O budget in the 1980s, 1990s, 2000s, and 2007–2016.

		the 1980s			the 1990s			the 2000s			2007-2016		
<b>Anthropogenic sources</b>		mean	min	max	mean	min	max	mean	min	max	mean	min	max
Direct emissions of N 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
	sub-total	2.6	1.8	4.1	3.0	2.1	4.8	3.4	2.3	5.2	3.8	2.5	5.8
Other direct anthropogenic sources	Fossil fuel 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
	sub-total	1.8	1.6	2.1	1.9	1.7	2.1	1.8	1.6	2.1	1.9	1.6	2.3
Indirect emissions from anthropogenic N 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 N 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 N 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
	sub-total	1.1	0.6	1.9	1.2	0.7	2.1	1.2	0.6	2.1	1.3	0.7	2.2
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
	sub-total	0.1	-0.4	0.7	0.1	-0.5	0.7	0.2	-0.4	0.9	0.2	-0.6	1.1
<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>
<b>Bottom-up total source</b>		<b>15.5</b>	<b>12.1</b>	<b>20.9</b>	<b>15.9</b>	<b>12.2</b>	<b>21.7</b>	<b>16.4</b>	<b>12.3</b>	<b>22.4</b>	<b>17.0</b>	<b>12.2</b>	<b>23.5</b>
<i>Top-down Ocean</i>								5.1	3.1	7.2	5.1	3.4	7.1
<i>Top-down Land</i>								10.8	9.3	12.5	11.8	10.6	13.8
<b>Top-down total source</b>								<b>15.9</b>	<b>15.1</b>	<b>16.9</b>	<b>16.9</b>	<b>15.9</b>	<b>17.7</b>
<i>Top-down Statospheric sink</i>								12.1	11.4	13.1	12.4	11.7	13.3
Observed atmospheric chemical sink*								13.3	12.2	14.4	13.5	12.4	14.6
<b>Change in atmospheric abundance**</b>								<b>3.7</b>	<b>3.2</b>	<b>4.2</b>	<b>4.3</b>	<b>3.8</b>	<b>4.8</b>
Atmospheric burden		1462	1442	1482	1493	1472	1514	1531	1510	1552	1555	1533	1577

497 Note: BU estimates include four categories of anthropogenic sources (red for agriculture, orange for  
 498 other direct anthropogenic sources, maroon for indirect emissions from anthropogenic N additions, and  
 499 brown for perturbed fluxes from climate/CO<sub>2</sub>/land cover change) and one category for natural sources  
 500 and sinks (green). The sources and sinks of N<sub>2</sub>O are given in Tg N yr<sup>-1</sup>. The atmospheric burden is given  
 501 in Tg N. \*calculated from satellite observations with a photolysis model (about 1% of this sink  
 502 occurs in the troposphere). \*\*Calculated from the combined NOAA and AGAGE record of surface N<sub>2</sub>O,  
 503 and adopting the uncertainty of the IPCC AR5 (Chapter 6)<sup>2</sup>. Detailed information on calculating each  
 504 sub-category is shown in Supplementary Tables 1–13.

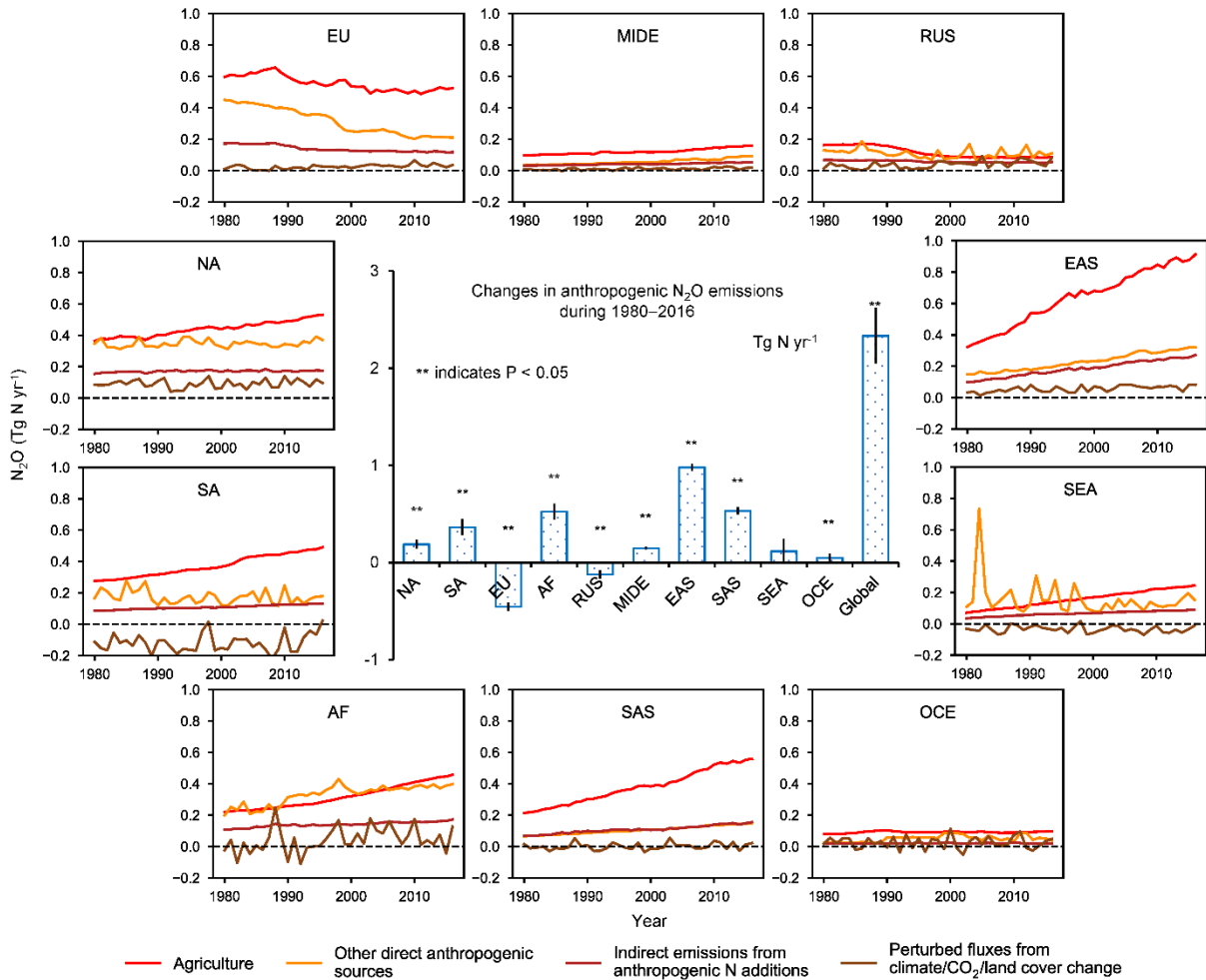


505  
 506 **Fig. 1 Global N<sub>2</sub>O budget for the recent decade (2007–2016).** The red arrow represents direct  
 507 emissions of N additions in the agricultural sector (Agriculture). The orange arrows represent emissions  
 508 from other direct anthropogenic sources. The maroon arrows represent indirect emissions from  
 509 anthropogenic N additions. The brown arrows represent perturbed fluxes from climate/CO<sub>2</sub>/land cover  
 510 change effects. The green arrows represent natural source. The anthropogenic and natural N<sub>2</sub>O sources  
 511 are derived from BU estimates. The blue arrows represent surface sink and observed atmospheric  
 512 chemical sink of which about 1% occurs in the troposphere. The total budget (sources + sinks) does not  
 513 exactly match the observed atmospheric accumulation, because each of the terms has been derived  
 514 independently and we do not force top-down agreement by rescaling the terms. This imbalance readily  
 515 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  
 516 sources and sinks are given in Tg N yr<sup>-1</sup>.  
 517



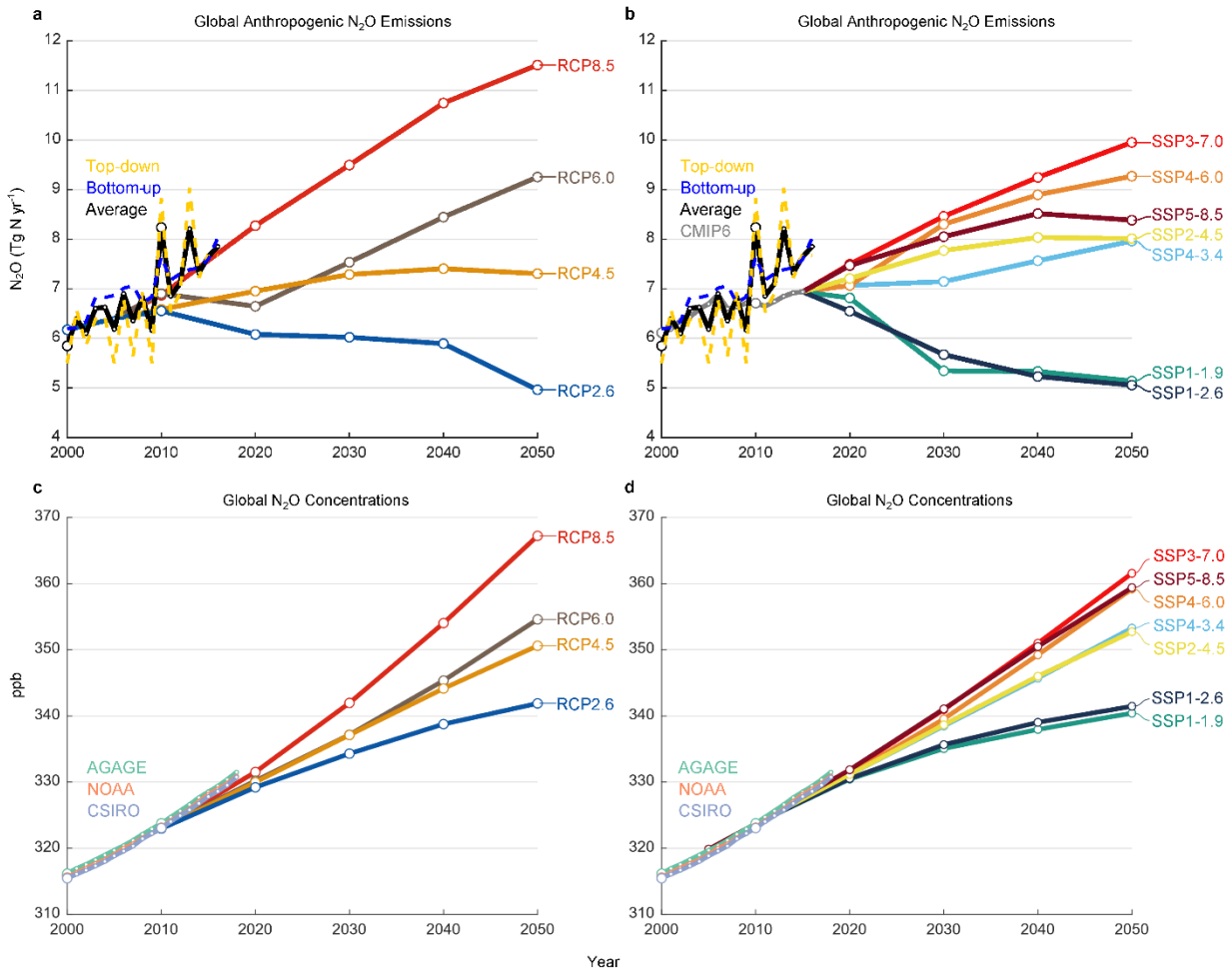
518  
 519 **Fig. 2 Regional N<sub>2</sub>O sources in the recent decade (2007–2016) over 11 regions.** *The Earth's*  
 520 *ice-free land is partitioned into ten regions: North America (NA), South America (SA), Europe (EU),*  
 521 *Middle East (MIDE), Africa (AF), Russia (RUS), East Asia (EAS), South Asia (SAS), Southeast Asia*  
 522 *(SEA), and Oceania (OCE). In each subplot from left to right: emissions from five sub-sectors using BU*  
 523 *approaches: natural fluxes without ocean (green), direct emissions of N additions in the agricultural*  
 524 *sector (Agriculture, red), other direct anthropogenic sources (orange), indirect emissions from*  
 525 *anthropogenic N additions (maroon), and perturbed fluxes from climate/CO<sub>2</sub>/land cover change (brown);*  
 526 *the sum of these five categories by BU approaches (blue), and the estimates by TD approaches (gold). BU*  
 527 *and TD estimates of ocean emissions are shown at the bottom left (from bottom to top: 30°–90°N,*  
 528 *30°S–30°N, and 90°–30°S). Error bars indicate the spread between the minimum and the maximum*  
 529 *values. The center map shows the spatial distribution of 10-year average N<sub>2</sub>O emissions from land and*  
 530 *ocean based on the land and ocean models. Per capita N<sub>2</sub>O emission (kg N capita<sup>-1</sup> yr<sup>-1</sup>) during*  
 531 *2007–2016 is shown in Supplementary Fig. 2.*  
 532

533



534  
 535 **Fig. 3 Ensembles of regional anthropogenic  $N_2O$  emissions over the 1980–2016 period.** *The*  
 536 *bar chart in the center shows the accumulated changes in regional and global  $N_2O$  emissions during the*  
 537 *study period. Error bars indicate the 95% confidence interval for the average of accumulated changes.*  
 538 *The Mann-Kendall test was performed to examine a monotonic increasing or decreasing trend in the*  
 539 *estimated ensemble  $N_2O$  emissions for each region and the globe during 1980–2016. The accumulated*  
 540 *changes were calculated from the linear regressed annual change rate ( $Tg\ N\ yr^{-2}$ ) multiplied by 37 years.*  
 541 *All regions except SEA show a significant increasing or decreasing trend in the estimated ensemble  $N_2O$*   
 542 *emissions during the study period (indicated by \*\* for each bar).*

543  
 544



545  
 546 **Fig. 4 Historical and projected global anthropogenic  $N_2O$  emissions and concentrations.**  
 547 *Global anthropogenic  $N_2O$  emissions (a, b) and concentrations (c, d) compared to the four*  
 548 *representative concentration pathways (RCPs) in the IPCC AR5 (a, c, ref. <sup>2</sup>) and the new marker*  
 549 *scenarios based on the Shared Socioeconomic Pathways (SSPs) used in CMIP6 (b, d, ref. <sup>41</sup>).*  
 550 *The historical data is represented as the mean of the BU and TD estimates of anthropogenic  $N_2O$*   
 551 *emissions, while the atmospheric concentration uses the three observation networks available,*  
 552 *AGAGE, NOAA, and CSIRO. TD anthropogenic emissions were calculated by subtracting BU-*  
 553 *derived natural fluxes. To aid the comparison, the four RCPs were shifted down so that the 2005*  
 554 *value is equal to the 2000–2009 average of the mean of TD and BU estimates. The SSPs are*  
 555 *harmonized<sup>3</sup> to match the historical emissions used in CMIP6<sup>42</sup> and Extended Data Fig. 10*  
 556 *shows the unharmonized data.*

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558

559

560 **Methods**

561 **Terminology.** This study provides an estimation of the global N<sub>2</sub>O budget considering all  
562 possible sources and all global change processes that can perturb the budget. A total of 18  
563 sources and three sinks of N<sub>2</sub>O are identified and grouped into six categories (Figure 1, Table 1):  
564 1) Natural fluxes in absence of climate change and anthropogenic disturbances including Soil  
565 emissions, Surface sink, Ocean emissions, Lightning and atmospheric production, and Natural  
566 emission from inland waters, estuaries, coastal zones (inland and coastal waters), 2) Perturbed  
567 fluxes from climate/CO<sub>2</sub>/land cover change including CO<sub>2</sub> effect, Climate effect, Post-  
568 deforestation pulse effect, and Long-term effect of reduced mature forest area, 3) Direct  
569 emissions of N additions in the agricultural sector (Agriculture) including emissions from direct  
570 application of synthetic N fertilizers and manure (henceforth Direct soil emissions), Manure left  
571 on pasture, Manure management, and Aquaculture, 4) Indirect emissions from anthropogenic N  
572 additions including atmospheric N deposition (NDEP) on land, atmospheric NDEP on ocean, and  
573 effects of anthropogenic loads of reactive N in inland waters, estuaries, coastal zones, 5) Other  
574 direct anthropogenic sources including Fossil fuel and industry, Waste and waste water, and  
575 Biomass burning, and 6) Two estimates of stratospheric sinks obtained from atmospheric  
576 chemistry transport models and observations, and one tropospheric sink (Table 1, Extended Data  
577 Fig. 2).

578 For the purpose of compiling national GHG inventories for country reporting to the climate  
579 convention, our anthropogenic N<sub>2</sub>O emission categories are aligned with those used in UNFCCC  
580 reporting and IPCC 2006 methodologies (Supplementary Table 14). We also provide the detailed  
581 comparison of our methodology and quantification with the IPCC AR5 (see Supplementary  
582 Section 4; Supplementary Table 15).

583 **Data synthesis.** We consider global N<sub>2</sub>O emission from land and ocean consisting of natural  
584 fluxes and anthropogenic emissions based on BU and TD approaches, however, the TD approach  
585 cannot separate natural and anthropogenic sources.

586 ‘Natural soil baseline’ emissions were obtained from six terrestrial biosphere models  
587 (NMIP<sup>16</sup>, Supplementary Tables 16–17) and provided here reflect a situation without  
588 consideration of land use change (e.g., deforestation) and without consideration of indirect  
589 anthropogenic effects via global change (i.e., climate, elevated CO<sub>2</sub>, and atmospheric N  
590 deposition). BU oceanic N<sub>2</sub>O emissions were based on an inter-comparison of five global ocean  
591 biogeochemistry models (Supplementary Table 18). The natural emission from ‘Inland water,  
592 estuaries, coastal zones’ includes coastal upwelling<sup>50</sup> and inland and coastal waters that were  
593 obtained from Yao et al.<sup>36</sup>, Maavara et al.<sup>35</sup>, and Lauerwald et al.<sup>51</sup>. Since the data (rivers,  
594 reservoirs, and estuaries) provided by Maavara et al. and Lauerwald et al. are for the year 2000,  
595 we assume that these values are constant during 1980–2016. Yao et al.<sup>36</sup> provided annual  
596 riverine N<sub>2</sub>O emissions using DLEM during the same period. Here, we averaged estimates from  
597 Yao et al. with that from Maavara et al.<sup>35</sup>. In addition, we estimated N<sub>2</sub>O emissions from global  
598 and regional reservoirs in the 2000s, and averaged their estimates with that from Maavara et al.<sup>35</sup>  
599 to represent emissions from reservoirs during 1980–2016. The estimate for global and regional  
600 estuaries and lakes is still based on the long-term averaged values provided by Maavara et al.<sup>35</sup>  
601 and Lauerwald et al.<sup>51</sup>, respectively. We considered the riverine emissions in the year 1900 as  
602 equivalent to the natural emission for the DLEM estimate assuming that the N load from land  
603 was negligible in that period<sup>52</sup>. We quantified the contribution of natural sources to total  
604 emission from reservoirs, lakes, and estuaries at 44% (36%–52%), with consideration of all N  
605 inputs (i.e., inorganic, organic, dissolved, particulate forms). We combined the estimate from



606 lightning with that from atmospheric production into an integrated category ‘Lightning and  
607 atmospheric production’. We make the simplification of considering the category ‘Lightning and  
608 atmospheric production’ as purely natural, however, atmospheric production is affected to some  
609 extent by anthropogenic activities through enhancing the concentrations of the reactive species  
610  $\text{NH}_2$  and  $\text{NO}_2$ . This category is in any case very small and the anthropogenic enhancement effect  
611 is uncertain. Lightning produces  $\text{NO}_x$ , the median estimate of which is  $5 \text{ Tg N yr}^{-1}$  (ref. <sup>53</sup>). We  
612 assumed an EF of 1% (ref. <sup>54</sup>) and a global estimate of  $0.05 (0.02\text{--}0.09) \text{ Tg N yr}^{-1}$  from lightning.  
613 Atmospheric production of  $\text{N}_2\text{O}$  results from the reaction of  $\text{NH}_2$  with  $\text{NO}_2$  (refs. <sup>55,56</sup>), N with  
614  $\text{NO}_2$ , and oxidation of  $\text{N}_2$  by  $\text{O}(^1\text{D})$ <sup>57</sup>, all of which constitute an estimated source of  $0.3 (0.2\text{--}1.1)$   
615  $\text{Tg N yr}^{-1}$ . The estimate of ‘Surface sink’ was obtained from Schlesinger<sup>58</sup> and Syakila et al.<sup>59</sup>.

616 The anthropogenic sources include four sub-sectors:

617 **(a) Agriculture.** It consists of four components: ‘Direct soil emissions’, ‘Manure left on  
618 pasture’, ‘Manure management’, and ‘Aquaculture’. Data for ‘Direct soil emissions’ were  
619 obtained as the ensemble mean of  $\text{N}_2\text{O}$  emissions from an average of three inventories (EDGAR  
620 v4.3.2, FAOSTAT, and GAINS), the SRNM/DLEM models, and the NMIP/DLEM models. The  
621 statistical model SRNM only covers cropland  $\text{N}_2\text{O}$  emissions, the same as the NMIP. Thus, we  
622 add the DLEM-based estimate of pasture  $\text{N}_2\text{O}$  emissions into the two estimates in cropland to  
623 represent direct agricultural soil emissions (i.e., SRNM/DLEM or NMIP/DLEM). The ‘Manure  
624 left on pasture’ and ‘Manure management’ emissions are the ensemble mean of EDGAR v4.3.2,  
625 FAOSTAT, and GAINS databases. Global N flows (i.e., fish feed intake, fish harvest, and waste)  
626 in freshwater and marine aquaculture were obtained from Beusen et al.<sup>30</sup> and Bouwman et al.<sup>60,61</sup>  
627 based on a nutrient budget model for the period 1980–2016. We then calculated global  
628 aquaculture  $\text{N}_2\text{O}$  emissions through considering 1.8% loss of N waste in aquaculture, the same

629 EF used in Hu et al.<sup>62</sup> and Macleod et al.<sup>31</sup>. The uncertainty range of the EF is from 0.5% (ref. <sup>14</sup>)  
630 to 5% (ref. <sup>63</sup>), the same range used in the UNEP report<sup>9</sup>. The ‘Aquaculture’ emission for the  
631 period 2007–2016 was a synthesis data from Hu et al.<sup>62</sup> in 2009, the FAO Report<sup>31</sup> in 2013, and  
632 our calculations. The estimate of aquaculture N<sub>2</sub>O emission prior to 2009 was from our  
633 calculations only.

634 The estimated direct emissions from agriculture have increased from 2.6 (1.8–4.1) Tg N yr<sup>-1</sup>  
635 in the 1980s to 3.8 (2.5–5.8) Tg N yr<sup>-1</sup> over the recent decade (2007–2016, Table 1).  
636 Specifically, direct soil emission from the application of fertilizers is the major source and  
637 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  
638 three global inventories (FAOSTAT, EDGAR v4.3.2, and GAINS), the estimates from process-  
639 based models (NMIP/DLEM<sup>15,16</sup>) and a statistical model (SRNM)/DLEM<sup>15,17</sup> exhibited a faster  
640 increase (Extended Data Fig. 4a). Over the past four decades, we also found a small but  
641 significant increase in emissions from livestock manure (i.e., manure left on pasture and manure  
642 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).  
643 Meanwhile, global aquaculture N<sub>2</sub>O emissions increased 10-fold, however, this flux remains the  
644 smallest term in the global budget (Extended Data Fig. 4d).

645 **(b) Other direct anthropogenic sources.** It includes ‘Fossil fuel and industry’, ‘Waste and  
646 waste water’, and ‘Biomass burning’. Both ‘Fossil fuel and industry’ and ‘Waste and waste  
647 water’ are the ensemble means of EDGAR v4.3.2 and GAINS databases. The ‘Biomass burning’  
648 emission is the ensemble mean of FAOSTAT, DLEM, and GFED4s databases.

649 Emissions from a combination of fossil fuel and industry, waste and waste water, and biomass  
650 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  
651 period of 2007–2016 (Table 1). The waste and waste water emission showed a continuous

652 increase at a rate of  $0.04 \pm 0.01 \text{ Tg N yr}^{-1}$  per decade ( $P < 0.05$ ) (Extended Data Fig. 5c).  
653 Emissions from biomass burning, estimated based on three data sources (DLEM, GFED4s, and  
654 FAOSTAT), slightly decreased at a rate of  $-0.03 \pm 0.04 \text{ Tg N yr}^{-1}$  per decade ( $P = 0.3$ ) since  
655 the 1980s (Extended Data Fig. 5d). This item is largely affected by climate and land use  
656 change<sup>64,65</sup>. Of the three data sources, the DLEM estimate exhibited significant inter-annual  
657 variability, especially during 1980–2000 when extreme fire events were detected in 1982, 1987,  
658 1991, 1994, and 1998. The occurrences of these extreme fires were associated with El Niño-  
659 Southern Oscillation (ENSO) events, especially in Indonesia (e.g., ‘Great Fire of Borneo’ in  
660 1982)<sup>66</sup>. Since 1997,  $\text{N}_2\text{O}$  emissions from fires estimated by DLEM, GFED4s, and FAOSTAT  
661 were consistent in the inter-annual variability. All the three estimates showed a decreasing trend,  
662 agreeing well with satellite-observed decrease of global burned area<sup>64,65</sup>.

663 **(c) Indirect emissions from anthropogenic N additions.** Data were obtained from various  
664 sources and considered N deposition on land and ocean (‘N deposition on land’ and ‘N  
665 deposition on ocean’), as well as the N leaching and runoff from upstream (‘Inland and coastal  
666 waters’). The emission from ‘N deposition on ocean’ was provided by Suntharalingam et al.<sup>67</sup>,  
667 while emission from ‘N deposition on land’ was the ensemble mean of an average of three  
668 inventories: FAOSTAT/EDGAR v4.3.2, GAINS/EDGAR v4.3.2, and NMIP. FAOSTAT and  
669 GAINS documented the sector ‘Indirect agricultural  $\text{N}_2\text{O}$  emissions’ by separating estimates  
670 from N leaching or N deposition, while EDGAR v4.3.2 did not. Here, we treated ‘Indirect  
671 agricultural  $\text{N}_2\text{O}$  emissions’ from EDGAR v4.3.2 as ‘Inland and coastal waters’ emissions for  
672 data synthesis. Only EDGAR v4.3.2 provided an estimate of indirect emission from non-  
673 agricultural sectors, while both FAOSTAT and GAINS, following the IPCC guidelines, provided  
674  $\text{NH}_x/\text{NO}_y$  volatilization from agricultural sectors. Here, we sum FAOSTAT or GAINS with

675 EDGAR v4.3.2 (i.e., FAOSTAT/EDGAR v4.3.2 or GAINS/EDGAR v4.3.2) to represent N  
676 deposition induced soil emissions from both agricultural and non-agricultural sectors. The N<sub>2</sub>O  
677 emissions from ‘Inland and coastal waters’ consist of rivers, reservoirs, lakes, estuaries, and  
678 coastal zone, which is the ensemble mean of an average of three inventories (EDGAR v4.3.2,  
679 FAOSTAT, GAINS), and the mean of process-based models. The anthropogenic emission  
680 estimated by Yao et al.<sup>36</sup> considered annual N inputs and other environmental factors (i.e.,  
681 climate, elevated CO<sub>2</sub>, and land cover change). For long-term average in rivers, reservoirs,  
682 estuaries and lakes, we applied a mean of 56% (based on the ratio of anthropogenic to total N  
683 additions from land) to calculate anthropogenic emissions. Seagrass, mangrove, saltmarsh and  
684 intertidal N<sub>2</sub>O emissions were undated from Murray et al<sup>68</sup>. Coastal waters with low disturbance  
685 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  
686 emissions were treated as anthropogenic emissions due to intensive human disturbances<sup>71</sup>.

687 N<sub>2</sub>O emissions following transport of anthropogenic N additions via atmosphere and water  
688 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  
689 2007–2016 (Table 1). The N<sub>2</sub>O emissions from inland and coastal waters increased at a rate of  
690 0.03±0.00 Tg N yr<sup>-1</sup> per decade (P < 0.05). Such an increase was reported by all the three  
691 inventories (FAOSTAT, GAINS, and EDGAR v4.3.2) with FAOSTAT giving the largest  
692 estimate. In contrast, the DLEM-based estimate presented a divergent trend: first increasing from  
693 1980–1998 and then slightly decreasing thereafter (Extended Data Fig. 6a). Emissions from  
694 atmospheric N deposition on oceans were relatively constant with a value of 0.1 (0.1–0.2) Tg N  
695 yr<sup>-1</sup>, while a large increase in emissions was found from atmospheric N deposition on land, with  
696 0.06±0.01 Tg N yr<sup>-1</sup> per decade (P < 0.05) reported in the three estimates (FAOSTAT/EDGAR  
697 v4.3.2, GAINS/EDGAR v4.3.2, and NMIP). The FAOSTAT agricultural source, together with

698 the EDGAR v4.3.2 industrial source, is consistent with NMIP estimates in the magnitude of N<sub>2</sub>O  
699 emissions, with the latter estimating a slightly slower increase from 2010 to 2016 (Extended  
700 Data Fig. 6b).

701 **(d) Perturbed fluxes from climate/CO<sub>2</sub>/land cover change.** Perturbed N<sub>2</sub>O fluxes represent the  
702 sum of the effects of climate, elevated atmospheric CO<sub>2</sub>, and land cover change. The estimate of  
703 climate and CO<sub>2</sub> effects on emissions was based on NMIP. The effect of land cover change on  
704 N<sub>2</sub>O dynamics includes the reduction due to ‘Long-term effect of reduced mature forest area’  
705 and the emissions due to ‘Post-deforestation pulse effect’. The two estimates were based on the  
706 book-keeping approach and the DLEM model simulation. The book-keeping method is  
707 developed by Houghton et al.<sup>72</sup> for accounting for carbon flows due to land use. In this study, an  
708 observation dataset consisting of 18 tropical sites was collected to follow the book-keeping logic.  
709 The dataset covers N<sub>2</sub>O emissions from a reference mature forest and their nearby converted  
710 pastures aged between one and 60 years. The average tropical forest N<sub>2</sub>O emission rate of 1.974  
711 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  
712 emissions (normalized to the baseline) after deforestation were developed:  $y = -0.31 \ln(x) +$   
713  $1.53$  ( $R^2 = 0.30$ ) and  $y = -0.454 \ln(x) + 2.21$  ( $R^2 = 0.09$ ). The first logarithmic function  
714 uses data collected by a review analysis<sup>74</sup>, based upon which the second one further considers  
715 observations from Verchot et al.<sup>21</sup> and Keller and Reiners<sup>75</sup>. In the first function,  $x$  (unit: year)  
716 indicates pasture age in years after deforestation and  $y$  (unitless; 0–1) indicates the ratio of  
717 pasture N<sub>2</sub>O emission over the N<sub>2</sub>O emission from the nearby reference mature forest. In the  
718 second function,  $x$  (unit: year) indicates secondary forest age and  $y$  (unitless; 0–1) indicates the  
719 ratio of secondary forest N<sub>2</sub>O emission over that of a reference mature forest. This form of the  
720 response functions can effectively reproduce the short-lived increase in soil N<sub>2</sub>O emissions after

721 initial forest clearing and the gradually declining emission rates of converted crops/pastures<sup>21,76</sup>.  
722 Using these two curves and the baseline, we kept track of the N<sub>2</sub>O reduction of tropical forests  
723 and the post-deforestation crop/pasture N<sub>2</sub>O emissions at an annual time-scale. This book-  
724 keeping method was applied to the two deforestation area datasets (Supplementary Text 2.8), so  
725 we could investigate not only the difference caused by the two sets of land use data but also the  
726 difference between this empirical method and the process-based model. For land conversion  
727 from natural vegetation to croplands or pastures, DLEM uses a similar strategy to Houghton et  
728 al.<sup>72</sup> and McGuire et al.<sup>77</sup> to simulate its influences on carbon and N cycles. Moreover, through  
729 using the sites of field observation from Davidson et al.<sup>20</sup> and Keller and Reiners<sup>75</sup>, we estimated  
730 N<sub>2</sub>O emission from secondary tropical forests based on the algorithm:  $y = 0.0084x + 0.2401$  ( $R^2$   
731  $= 0.44$ ).  $x$  (unit: year) indicates secondary forest age and  $y$  (unitless; 0–1) indicates the ratio of  
732 secondary forest N<sub>2</sub>O emission over that of a reference mature forest. The difference between  
733 primary forests and secondary forests were subtracted from natural soil emissions simulated by  
734 six terrestrial biosphere models in NMIP.

735 We calculated the ensemble of oceanic N<sub>2</sub>O emission based on the BU approach (five ocean  
736 biogeochemical models; Supplementary Table 18) and the TD approach (five estimates from  
737 four inversion models; Supplementary Table 19), respectively. The atmospheric burden and its  
738 rate of change during 1980–2016 were derived from mean maritime surface mixing ratios of  
739 N<sub>2</sub>O (refs. <sup>78,79</sup>) with a conversion factor of 4.79 Tg N/ppb (ref. <sup>80</sup>). Combining uncertainties in  
740 measuring the mean surface mixing ratios<sup>78</sup> and that of converting surface mixing ratios to a  
741 global mean abundance<sup>80</sup>, we estimate a  $\pm 1.4\%$  uncertainty in the burden. Annual change in  
742 atmospheric abundance is calculated from the combined NOAA and AGAGE record of surface  
743 N<sub>2</sub>O and uncertainty is taken from the IPCC AR5 (ref. <sup>2</sup>). There shows an agreement of the

744 stratospheric loss from atmospheric chemistry transport models (TD modeled chemical sink<sup>18,81</sup>)  
745 and from satellite observations with a photolysis model (observed photochemical sink<sup>1</sup>), which  
746 differ only by  $\sim 1$  Tg N yr<sup>-1</sup>. The satellite-based lifetime,  $116 \pm 9$  years, gives an overall  
747 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  
748 included in observed atmospheric chemical sink (Table 1) and is small ( $\sim 1\%$  of the stratospheric  
749 sink) with an estimated range of 0.1 to 0.2 Tg N yr<sup>-1</sup>.

750 **Comparison with the IPCC guidelines.** The IPCC has provided guidance to quantify N<sub>2</sub>O  
751 emissions, which is widely used in emission inventories for reporting to the UNFCCC. Over time  
752 the recommended approaches have changed, which is critical for estimating emissions from  
753 agricultural soils, the largest emission source. Previous global N<sub>2</sub>O assessments<sup>52,82,83</sup> based on  
754 the IPCC 1996 guidelines<sup>84</sup> attributed about 6.3 Tg N yr<sup>-1</sup> to the agricultural sector, including  
755 both direct and indirect emissions. This estimate is significantly larger than our results (Fig. 1;  
756 Table 1) derived from multiple methods, and is also larger than the most recent estimates from  
757 global inventories (EDGAR v4.3.2, FAOSTAT, and GAINS) that are based on the IPCC 2006  
758 guidelines<sup>14</sup>. The main reason is that indirect emissions from leaching and groundwater were  
759 overestimated in previous studies<sup>85</sup>. Correspondingly, projections of atmospheric N<sub>2</sub>O  
760 concentrations based on these overestimated emissions<sup>82</sup> led to biased estimates. For example,  
761 Mosier and Kroeze<sup>82</sup> expected atmospheric N<sub>2</sub>O concentrations to be 340–350 ppb in the year  
762 2020, instead of 333 ppb<sup>5</sup> as observed. Recently, the 2019 Refinement to the 2006 IPCC  
763 Guidelines for National Greenhouse Gas Inventories has been published. It adopts the same  
764 approach for N application on soils, but considers impacts of different climate regimes. The new  
765 guidelines, based on a wealth of new scientific literature, proposed much smaller emissions from  
766 grazing animals by a factor of 5–7. Preliminary calculations we have made indicate that global

767 soil emissions based on these new guidelines may decrease by 20%–25%. Integrating estimates  
768 relying on the IPCC methodology with estimates by process-based models provides for a more  
769 balanced assessment in this paper. We also added information from assessments<sup>86,87</sup> that derived  
770 agricultural emissions as the difference between atmospheric terms and other emissions like  
771 combustion, industry and nature, and they gave comparable magnitudes (4.3–5.8 Tg N yr<sup>-1</sup>) to  
772 our bottom-up results.

773 **Uncertainty.** Current data analysis and synthesis of long-term N<sub>2</sub>O fluxes are based on a wide  
774 variety of TD and BU methods. TD approaches, consisting of four inversion frameworks<sup>88-91</sup>,  
775 provide a wide range of estimates largely due to systematic errors in the modelled atmospheric  
776 transport and stratospheric loss of N<sub>2</sub>O. In addition, the emissions from TD analyses are  
777 dependent on the magnitude and distribution of the prior flux estimates to an extent that is  
778 strongly determined by the number of atmospheric N<sub>2</sub>O measurements<sup>18</sup>. Inversions are  
779 generally not well constrained (and thus rely heavily on a priori estimates) in Africa, Southeast  
780 Asia, southern South America, and over the oceans, owing to the paucity of observations in these  
781 regions. The improvement of atmospheric transport models, more accurate priors, and more  
782 atmospheric N<sub>2</sub>O measurements would reduce uncertainty in further TD estimates, particularly  
783 for ocean and regional emissions.

784 BU approaches are subject to uncertainties in various sources from land<sup>16</sup> and oceans<sup>32</sup>. For  
785 process-based models (e.g. NMIP and ocean biogeochemical models), the uncertainty is  
786 associated with differences in model configuration as well as process parameterization<sup>16,32</sup>. The  
787 uncertainty of estimates from NMIP could be reduced in multiple ways<sup>16</sup>. First, the six models in  
788 NMIP exhibited different spatial and temporal patterns of N<sub>2</sub>O emissions even though they used  
789 the same forcings. Although these models have considered essential biogeochemical processes in



790 soils (e.g., biological N fixation, nitrification/denitrification, mineralization/immobilization,  
791 etc.)<sup>92</sup>, some missing processes such as freeze-thaw cycles and ecosystem disturbances should be  
792 included in terrestrial biosphere models to reduce uncertainties. Second, the quality of input  
793 datasets, specifically the amount and timing of N application, and spatial and temporal changes  
794 in distribution of natural vegetation and agricultural land, is critical for accurately simulating soil  
795 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  
796 validate model performance and constrain large-scale model simulations. Data assimilation  
797 techniques could be utilized to improve model accuracy.

798 Current remaining uncertainty in global ocean model estimates of N<sub>2</sub>O emission includes the  
799 contribution of N<sub>2</sub>O flux derived from the tropical oceanic low oxygen zones (e.g., the Eastern  
800 Equatorial Pacific, the northern Indian ocean) relative to the global ocean. These low oxygen  
801 zones are predominantly influenced by high yield N<sub>2</sub>O formation processes (e.g., denitrification  
802 and enhanced nitrification). Regional observation-based assessments have also suggested that  
803 these regions may produce more N<sub>2</sub>O than is simulated by the models<sup>32</sup>. The current generation  
804 of global ocean biogeochemistry models are not sufficiently accurate to represent the high N<sub>2</sub>O  
805 production processes in low-oxygen zones, and their associated variability (see refs. <sup>34,93,94</sup> for  
806 more detail). Thus, precisely representing the local ocean circulation and associated  
807 biogeochemical fluxes of these regions could further reduce the uncertainty in estimates of  
808 global and regional oceanic N<sub>2</sub>O emissions.

809 Regardless of the tier approach used, GHG inventories for agriculture suffer from high  
810 uncertainty in the underlying agriculture and rural data and statistics used as input, including  
811 statistics on fertilizer use, livestock manure availability, storage and applications, and nutrient,  
812 crop and soils management. For instance, animal waste management is an uncertain aspect, since

813 much of the manure is either not used, or employed as a fuel or building material, or may be  
814 discharged directly to surface water<sup>95,96</sup>, with important repercussions for the calculated  
815 emissions. Furthermore, GHG inventories using default EFs show large uncertainties at local to  
816 global scales, especially for agricultural N<sub>2</sub>O emissions, due to the poorly captured dependence  
817 of EFs on spatial diversity in climate, management, and soil physical and biochemical  
818 conditions<sup>2,22</sup>. It is well known, for example from the IPCC guidelines, that higher-tier GHG  
819 inventories may provide more reasonable estimates by using the alternative EFs that are  
820 disaggregated by environmental factors and management-related factors<sup>97</sup>. A large range of EFs  
821 have been used to estimate aquaculture N<sub>2</sub>O emissions<sup>31,39,62,86</sup> and long-term estimates of N  
822 flows in freshwater and marine aquaculture are scarce<sup>30</sup>. Uncertainty also remains in several N<sub>2</sub>O  
823 sources that have not yet been fully understood or quantified. To date, robust estimates of N<sub>2</sub>O  
824 emissions from global peatland degradation are still lacking, although we have accounted for  
825 N<sub>2</sub>O emissions due to the drainage of organic soils (histosols) obtained from FAOSTAT and  
826 GAINS databases<sup>28,43</sup>. Recent evidence shows that permafrost thawing<sup>98</sup> and the freeze-thaw  
827 cycle<sup>99</sup> contribute to increasing N<sub>2</sub>O emissions, which, however, have not been well established  
828 in the current estimates of the global N<sub>2</sub>O budget.

829 **Statistics.** Through using the Mann-Kendall test in R-3.4.4, we checked the significance of  
830 trends in annual N<sub>2</sub>O emissions from each sub-sector based on the BU approach.

## 831 **References**

832

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973  
974 **Data availability**

975 The relevant datasets of this study are archived in the box site of International Center for Climate  
976 and Global Change Research at Auburn University (<https://auburn.box.com/>). Source data for  
977 Figs. 1–4, Table 1, Extended Figs. 1–10 and Supplementary Information are provided with the  
978 paper. Additional description on data availability for atmospheric N<sub>2</sub>O observations from  
979 NOAA, AGAGE and CSIRO networks is provided in the Supplementary Information. The data  
980 presented here are made available in the belief that their dissemination will lead to greater  
981 understanding and new scientific insights on the global and regional N<sub>2</sub>O budgets and changes to  
982 it, and helping to reduce the uncertainties. As data are the result of initial processing to fit to the  
983 purpose of this publication, typically a wealth of underlying information is with the original data  
984 providers. Researchers interested to use results made available in the repository are encouraged,  
985 as good practice, to take advantage of underlying information by contacting the original data  
986 providers. If such a contact develops into a more intensive scientific discussion, further  
987 involvement including co-authorship should be considered.

988

### 989 **Code availability**

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

992

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### 1033 **Author contributions**

1034 Author contributions. H.T., R.L.T., J.G.C. and R.B.J. designed and coordinated the study. H.T.,  
1035 R.X., J.G.C., R.L.T., W.W., P.S., E.A.D., P.C., R.B.J., G.J.M., M.J.P., N.P., S.P., P.R., H.S.,  
1036 F.N.T., S.Z., F.Z., B.F. and G.P. conducted data analysis, synthesis and wrote the paper. R.L.T.  
1037 led atmospheric inversions teaming with M.P.C., T.M., D.B.M., P.K.P., K.C.W., and C.W.; H.T.  
1038 led land biosphere modeling teaming with P.C., H.S., S.Z., A.A., F.J., J.C., S.R.S.D., A.I., W.L.,  
1039 S.L., S.O., N.V., E.A.D., S.D. and W. Li; P.S. led ocean biogeochemical modeling teaming with  
1040 G.B., L.B., S.B., E.T.B., F.J. and A.L.; P.R. led inland water and coastal modeling and synthesis  
1041 teaming with B.D.E., G.G.L., R.L., T.M., P.A.R., H.T. and Y.Y.; A.F.B., J.W., M.M. provided  
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1043 burning. F.Z. provided cropland N<sub>2</sub>O flux data from a statistical model and field observations.  
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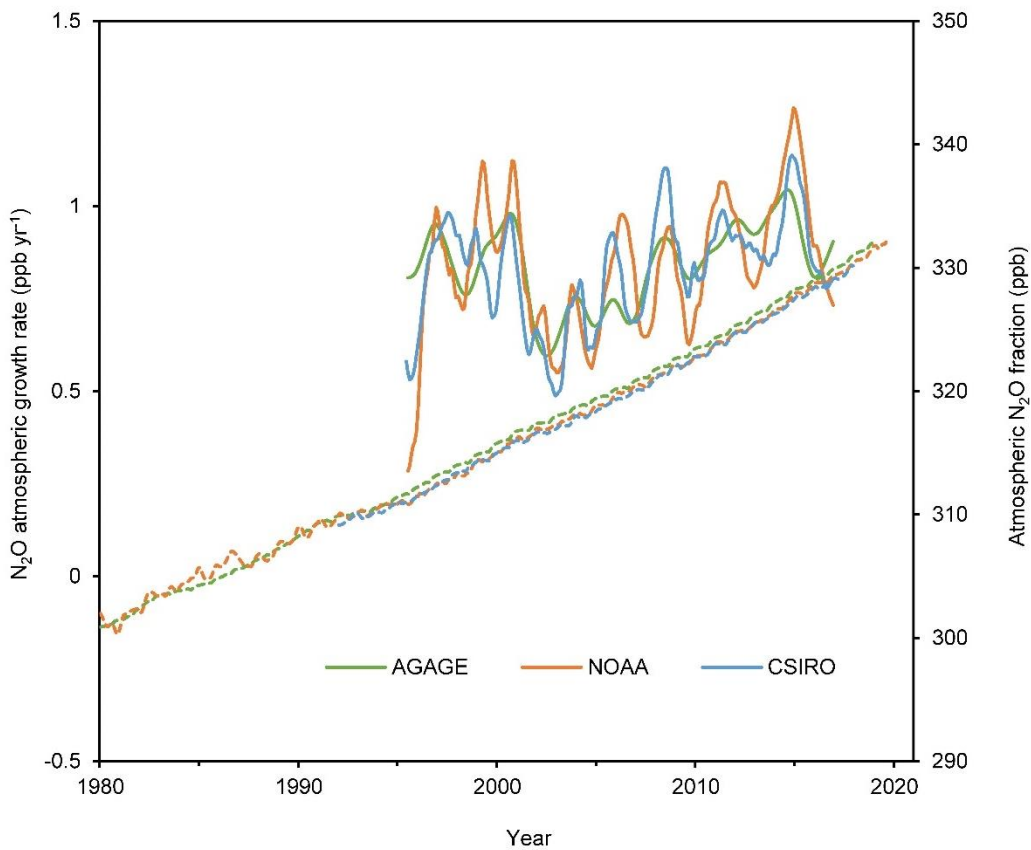
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1051  
1052 **Additional information**

1053  
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1055  
1056 **Correspondence and requests for materials should be addressed to H.T.**

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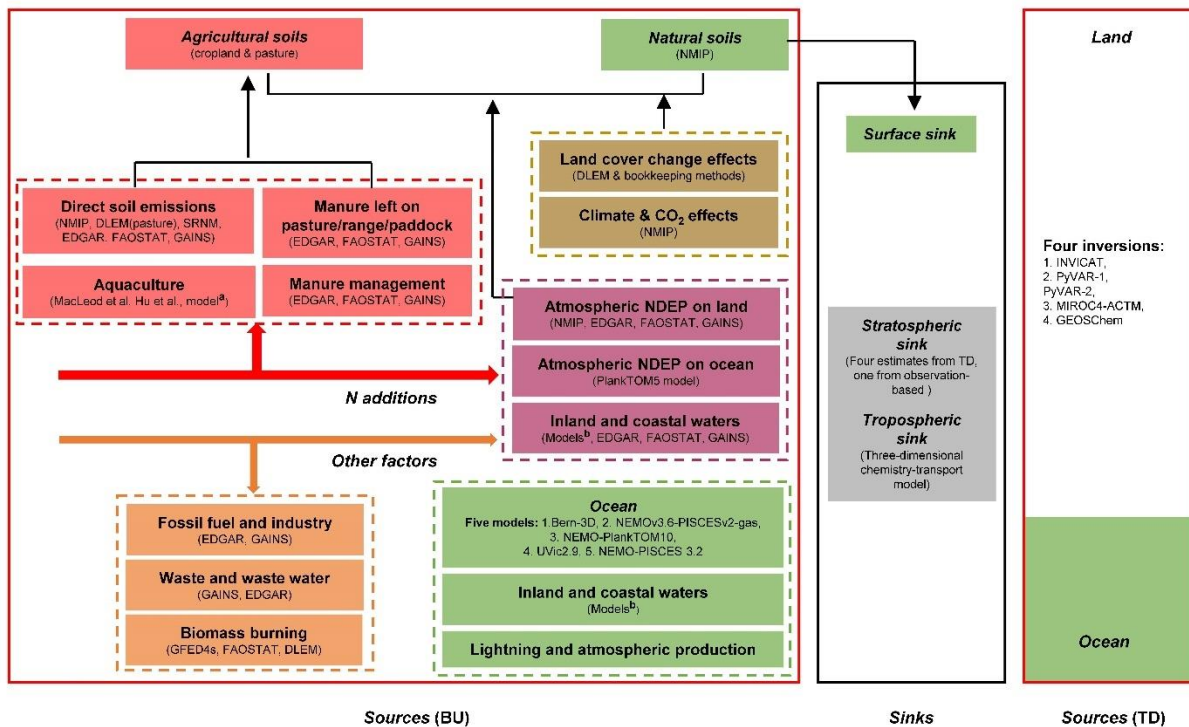
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1059 **Extended Data Fig. 1 Global mean growth rates and atmospheric concentration of N<sub>2</sub>O.**

1060 Global mean growth rates (solid lines, during 1995–2017) and atmospheric N<sub>2</sub>O concentration  
 1061 (dashed lines, during 1980–2017) are from the AGAGE<sup>6</sup> (green), NOAA<sup>5</sup> (orange), and CSIRO  
 1062 (blue) networks. Global mean growth rates were calculated with annual time steps and are shown  
 1063 as 12-month moving averages. Growth rates are not calculated prior to 1995 due to insufficient  
 1064 data and higher uncertainties on the measurements.

1065



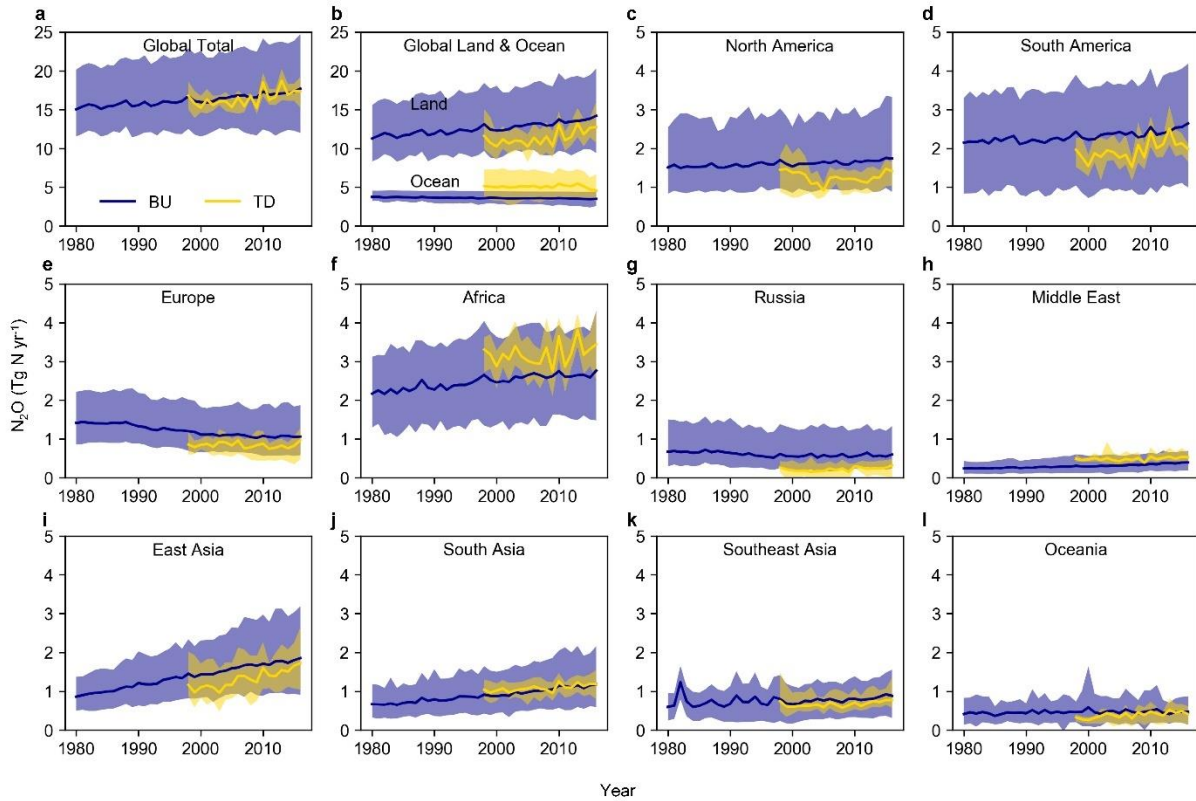


1066

1067 **Extended Data Fig. 2 The methodology for data synthesis of global N<sub>2</sub>O budget.** BU and TD  
 1068 represent bottom-up and top-down methods, respectively. The color codes are the same as that  
 1069 used in Table 1 and Figs. 1–3. We utilize both approaches, including 22 BU and five TD  
 1070 estimates of N<sub>2</sub>O fluxes from land and oceans. For sources estimated by BU, we include six  
 1071 process-based terrestrial biosphere modeling studies<sup>16</sup>; five process-based ocean biogeochemical  
 1072 models<sup>100</sup>; one nutrient budget model<sup>30,60,61</sup>; five inland water modeling studies<sup>35,36,50,51,68</sup>; one  
 1073 statistical model SRNM based on spatial extrapolation of field measurements<sup>17</sup>; and four GHG  
 1074 inventories: EDGAR v4.3.2<sup>101</sup>, FAOSTAT<sup>102</sup>, GAINS<sup>43</sup>, and GFED4s<sup>103</sup>. In addition, previous  
 1075 literatures regarding estimates of ‘Surface sink’<sup>58,73</sup>, ‘Lightning’<sup>53,54</sup>, ‘Atmospheric  
 1076 production’<sup>56,57,104</sup>, ‘Aquaculture’<sup>31,62</sup>, and model-based ‘Tropospheric sink’<sup>81</sup> and observed  
 1077 ‘Stratospheric sink’<sup>1</sup> are included in the current synthesis. <sup>a</sup>MacLeod et al.<sup>31</sup> and Hu et al.<sup>62</sup>  
 1078 provide global aquaculture N<sub>2</sub>O emissions in 2013 and in 2009, respectively; and the nutrient  
 1079 budget model<sup>30,60,61</sup> provides N flows in global freshwater and marine aquaculture over the  
 1080 period 1980–2016. <sup>b</sup>Model-based estimates of N<sub>2</sub>O emissions from ‘Inland and coastal waters’  
 1081 include rivers and reservoirs<sup>35,36</sup>, lakes<sup>51</sup>, estuaries<sup>35</sup>, coastal zones (i.e., seagrasses, mangroves,  
 1082 saltmarsh and intertidal saltmarsh)<sup>68</sup>, and coastal upwelling<sup>50</sup>.

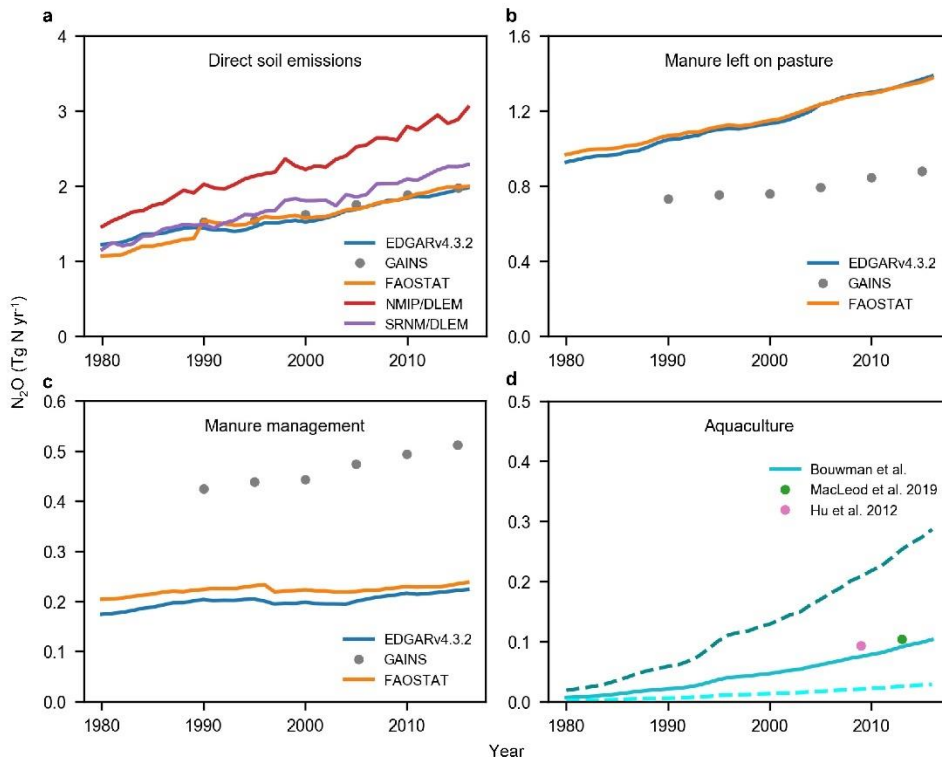
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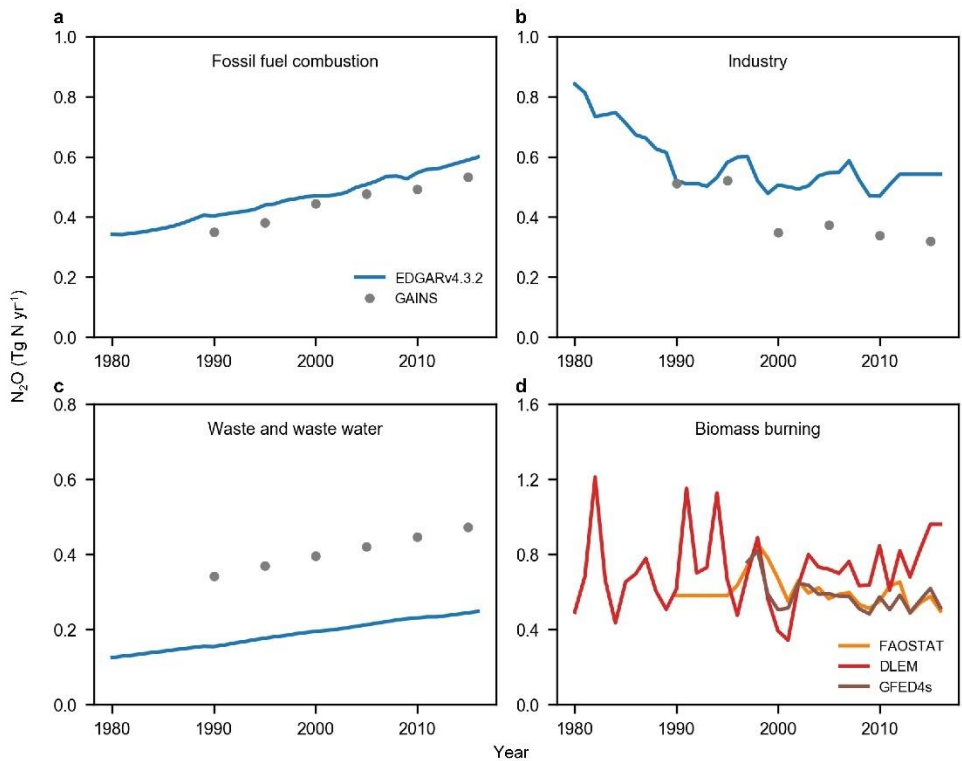


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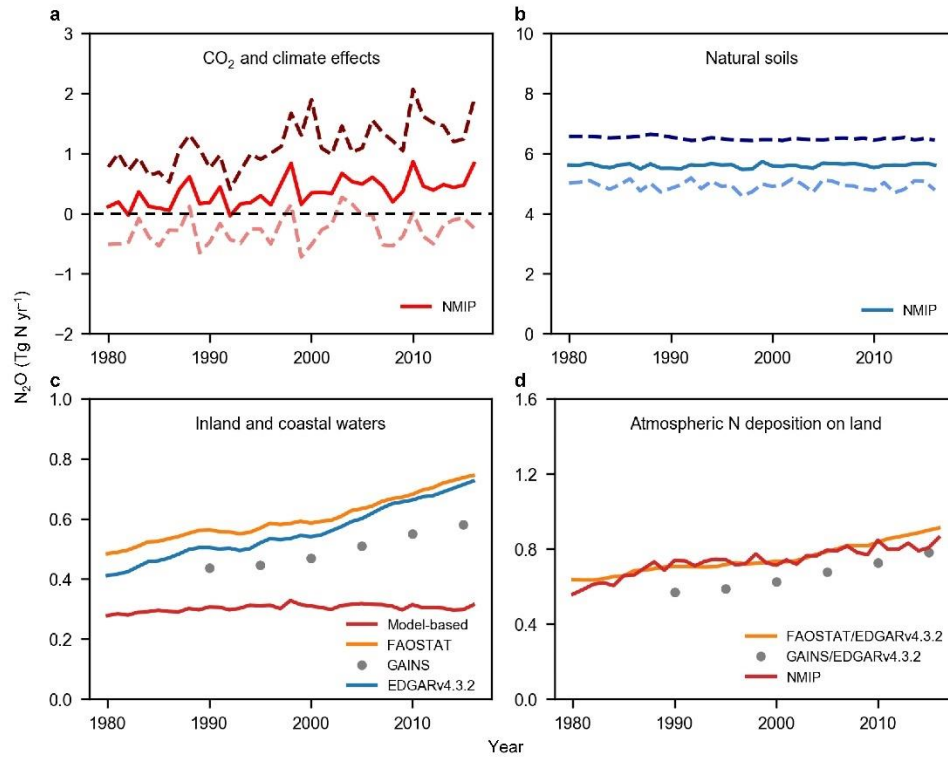
**Extended Data Fig. 3 Comparison of annual total N<sub>2</sub>O emissions at global and regional scales estimated by BU and TD approaches.** The blue lines represent the mean N<sub>2</sub>O emission from BU methods and the shaded areas show minimum and maximum estimates; The gold lines represent the mean N<sub>2</sub>O emission from TD methods and the shaded areas show minimum and maximum estimates.



1092  
 1093 **Extended Data Fig. 4 Global agricultural N<sub>2</sub>O emissions.** **a**, Direct emission from agricultural  
 1094 soils associated with mineral fertilizer, manure and crop residue inputs, and cultivation of  
 1095 organic soils based on EDGAR v4.3.2, GAINS, FAOSTAT, NMIP/DLEM, and SRNM/DLEM  
 1096 estimates. NMIP/DLEM or SRNM/DLEM means the combination of N<sub>2</sub>O emission by NMIP or  
 1097 SRNM from croplands with N<sub>2</sub>O emission from intensively managed grassland (pasture) by  
 1098 DLEM. **b**, Direct emission from the global total area under permanent meadows and pasture, due  
 1099 to manure N deposition (left on pasture) based on EDGAR v4.3.2, FAOSTAT, and GAINS  
 1100 estimates. **c**, Emission from manure management based on FAOSTAT, GAINS, and EDGAR  
 1101 v4.3.2. **d**, Aquaculture N<sub>2</sub>O emission based on a nutrient budget model<sup>30</sup>, MacLeod et al.<sup>31</sup>, and  
 1102 Hu et al.<sup>62</sup>; the solid line represents the ‘best estimate’ that is the product of EF (1.8%) and N  
 1103 waste from aquaculture provided by the nutrient budget model; the dashed lines represent the  
 1104 minimum and maximum values.  
 1105

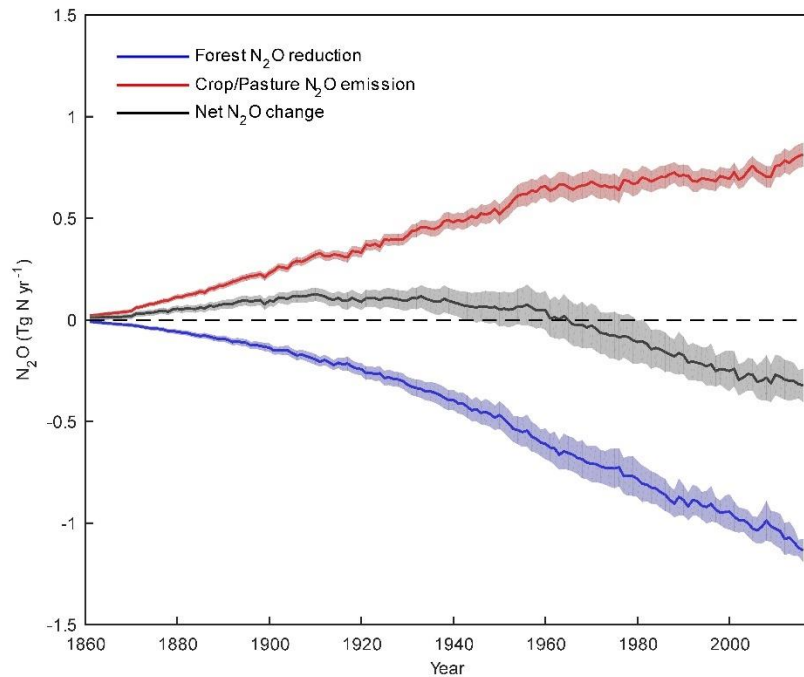


1106  
 1107 **Extended Data Fig. 5 Global N<sub>2</sub>O emission from other direct anthropogenic sources. a,**  
 1108 **Emission from fossil fuel combustion based on EDGAR v4.3.2 and GAINS estimates. b,**  
 1109 **Emission from industry based on EDGAR v4.3.2 and GAINS estimates. c, Emission from waste**  
 1110 **and waste water based on EDGAR v4.3.2 and GAINS estimates. d, Emission from biomass**  
 1111 **burning based on FAOSTAT, DLEM, and GFED4s estimates.**  
 1112

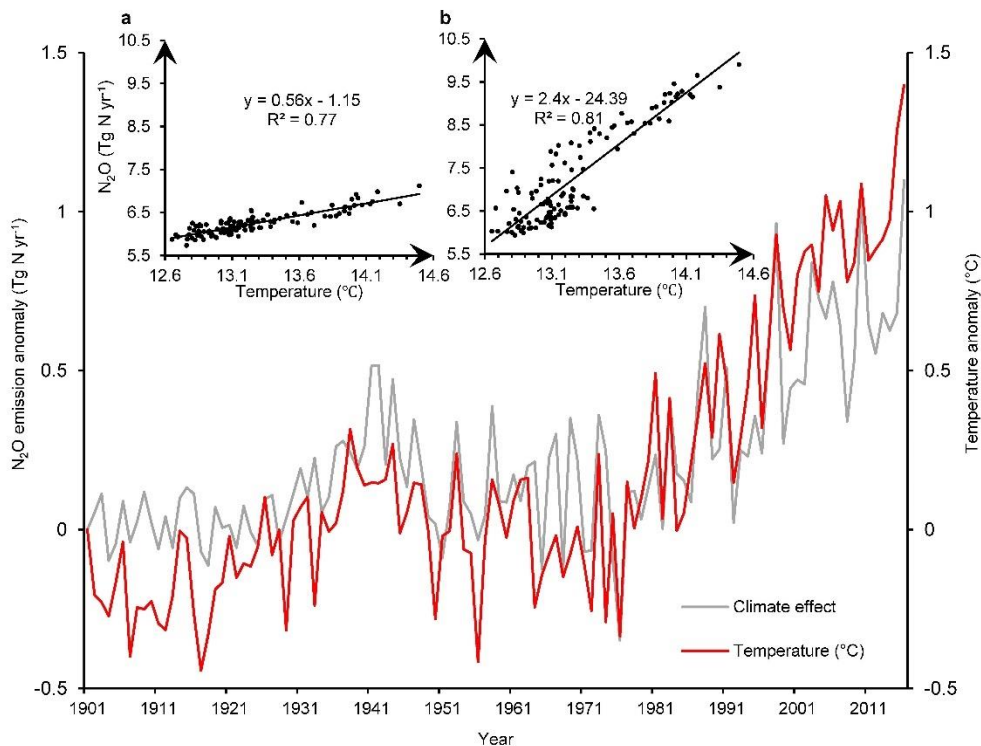


1113  
 1114 **Extended Data Fig. 6 Global N<sub>2</sub>O emissions from natural soils, inland and coastal waters**  
 1115 **and due to change in climate, atmospheric CO<sub>2</sub> and N deposition. a,** Changes in global soil  
 1116 N<sub>2</sub>O fluxes due to changing CO<sub>2</sub> and climate. **b,** Global natural soil N<sub>2</sub>O emissions without  
 1117 consideration of land use change (e.g., deforestation) and without consideration of indirect  
 1118 anthropogenic effects via global change (i.e., climate, elevated CO<sub>2</sub>, and atmospheric N  
 1119 deposition). The estimates are based on NMIP estimates during 1980–2016 including six  
 1120 process-based land biosphere models. Here, we also subtracted the difference between with and  
 1121 without consideration of secondary forests emissions that grow back after pasture or cropland  
 1122 abandonment from natural soil emissions based on NMIP estimates. The solid lines represent the  
 1123 ensemble and dashed lines show the minimum and maximum values. **c,** Global anthropogenic  
 1124 N<sub>2</sub>O emission from inland waters, estuaries, coastal zones based on models (model-based),  
 1125 FAOSTAT, GAINS, and EDGAR v4.3.2 estimates. **d,** Emission due to atmospheric N deposition  
 1126 (NDEP) on land based on NMIP, FAOSTAT/EDGAR v4.3.2, and GAINS/EDGAR v4.3.2.  
 1127 FAOSTAT/EDGAR v4.3.2 or GAINS/EDGAR v4.3.2 means the combination of agricultural  
 1128 source from FAOSTAT or GAINS with non-agricultural source from EDGAR v4.3.2. A process-  
 1129 based model DLEM<sup>36</sup> and a mechanistic stochastic model<sup>35,51</sup> were used to estimate N<sub>2</sub>O  
 1130 emission from inland waters and estuaries, while site-level emission rates of N<sub>2</sub>O were upscaled  
 1131 to estimate global N<sub>2</sub>O fluxes from the global seagrass area<sup>68</sup>.

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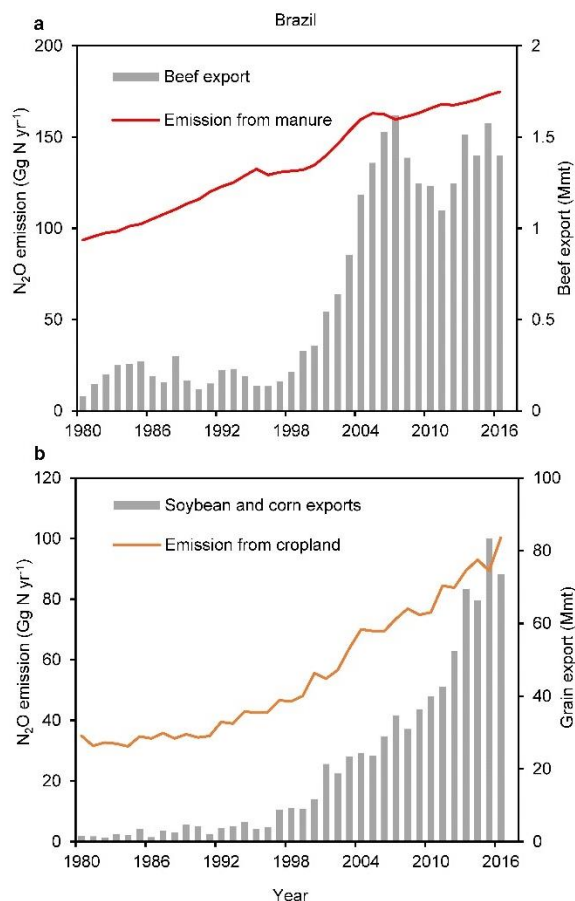
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 1135 **Extended Data Fig. 7 Global N<sub>2</sub>O dynamics due to land cover changes.** The blue line  
 1136 represents the mean forest N<sub>2</sub>O reduction caused by the long-term effect of reduced mature forest  
 1137 area (i.e., deforestation) and shaded areas show minimum and maximum estimates; the red line  
 1138 represents the mean N<sub>2</sub>O emission from post-deforestation pulse effect (i.e., crop/pasture N<sub>2</sub>O  
 1139 emissions from legacy N of previous forest soil, not accounting for new fertilizer N added to  
 1140 these crop/pasture lands) and shaded areas show minimum and maximum estimates; the gray line  
 1141 represents the mean net deforestation emission of N<sub>2</sub>O and shaded areas show minimum and  
 1142 maximum estimates.  
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 1146 **Extended Data Fig. 8 Global simulated N<sub>2</sub>O emission anomaly due to climate effect and**  
 1147 **global annual land surface temperature anomaly during 1901–2016.** Global N<sub>2</sub>O emission  
 1148 anomalies are the ensemble of six process-based land biosphere models in NMIP. The  
 1149 temperature data were obtained from the CRU-NCEP v8 climate dataset  
 1150 (<https://vesg.ipsl.upmc.fr>). The above left figure **a**) shows the correlation between average global  
 1151 annual land surface temperature and simulated N<sub>2</sub>O emissions (i.e., the result of SE6 experiment  
 1152 in NMIP<sup>16</sup>) considering annual changes in climate but keeping all other factors (i.e., N fertilizer,  
 1153 manure, NDEP, elevated CO<sub>2</sub>, and land cover change) at the level of 1860. The above right  
 1154 figure **b**) shows the correlation between average global annual land surface temperature and  
 1155 simulated N<sub>2</sub>O emissions (i.e., the result of SE1 experiment in NMIP<sup>16</sup>) considering annual  
 1156 changes in all factors during 1860–2016.

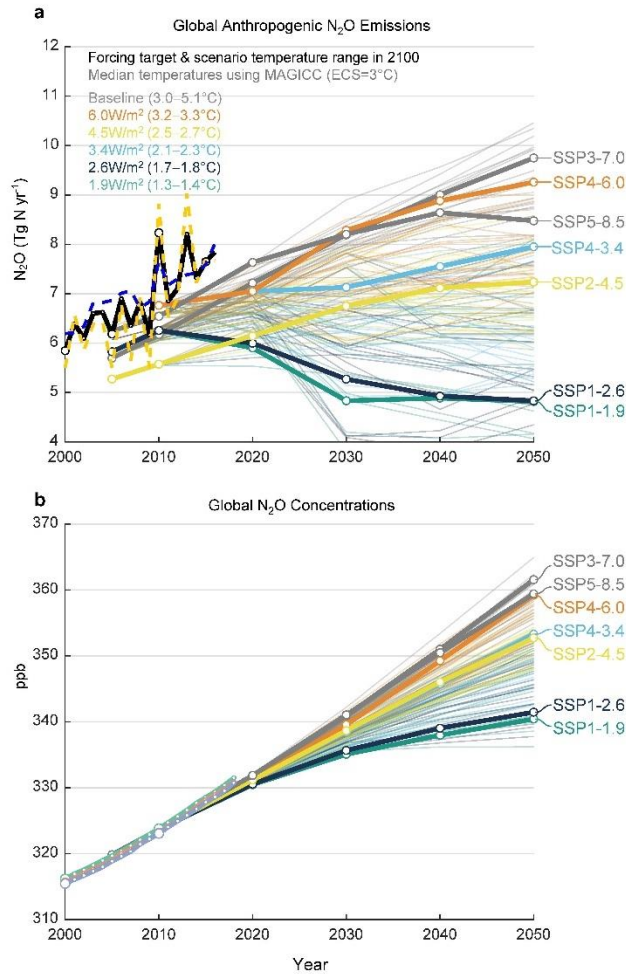
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 1160 **Extended Data Fig. 9 Direct soil emissions and agricultural product trades in Brazil.** **a**, Red  
 1161 line shows the ensemble direct  $N_2O$  emissions from livestock manure based on EDGAR v4.3.2,  
 1162 GAINS, and FAOSTAT, the sum of ‘manure left on pasture’ and ‘manure management’; The  
 1163 gray columns show the amount of beef export by Brazil. **b**, Orange line shows the ensemble  
 1164 direct  $N_2O$  emissions from croplands due to N fertilization based on NMIP and SRNM; The gray  
 1165 columns show the amount of soybean and corn exports by Brazil. The data of beef and cereal  
 1166 product trades were adapted from the ABIEC (beef) and FAOSTAT (soybean and corn). Mmt yr<sup>-1</sup>  
 1167 <sup>1</sup> represents millions of metric tons per year.  
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 1171 **Extended Data Fig. 10 An extension of Fig. 4 to provide a comparison of anthropogenic**  
 1172 **N<sub>2</sub>O emissions (a) and atmospheric N<sub>2</sub>O concentrations (b) in the unharmonized SSPs<sup>105</sup>.**  
 1173 The emission and concentration data are as in Fig. 4. The unharmonized emissions from the  
 1174 Integrated Assessment Models (IAMs)<sup>105</sup> show a large variation due to different input data and  
 1175 model assumptions. Comparison with Fig. 4b, d illustrates the modifications to the IAM scenario  
 1176 data for use in CMIP6. All baseline scenarios (SSP3-7.0 and SSP5-8.5; without climate policy  
 1177 applied) are shown in gray regardless of the radiative forcing level they reach in 2100.