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Tian, H, Xu, R, Canadell, JG et al. (54 more authors) (2020) A comprehensive quantification of global nitrous oxide sources and sinks. Nature, 586 (7828). pp. 248-256. ISSN 0028-0836

https://doi.org/10.1038/s41586-020-2780-0

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#### A comprehensive quantification of global nitrous oxide sources and sinks 1

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91 Nitrous oxide  $(N_2O)$ , like carbon dioxide, is a long-lived greenhouse gas that accumulates in 92 the atmosphere. The increase in atmospheric  $N_2O$  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  $N_2O$  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  $N_2O$  emissions in emerging economies-particularly Brazil, China and India. Analysis of process-based model 111 112 estimates reveals an emerging  $N_2O$ -climate feedback resulting from interactions between 113 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.

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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, fiber and energy, and a rising source from waste generation and industrial processes<sup>4,11,12</sup>. Since 126 127 1990, anthropogenic  $N_2O$  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, Africa)<sup>2,9,13</sup>. Moreover, we need a complete account of all human activities that accelerate the 134 135 global N cycle and that interact with the biochemical processes controlling the fluxes of N<sub>2</sub>O in both terrestrial and aquatic ecosystems<sup>2,8</sup>. Here we present a comprehensive, consistent analysis 136

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 (BU) and top-down (TD) methods and their crossconstraints. Our assessment enhances understanding of the global N cycle and will inform policy
development for N<sub>2</sub>O mitigation, ideally helping to curb warming to levels consistent with the
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 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 171 172 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 well with the observed 2007–2016 increase in atmospheric abundance of 3.8–4.8 Tg N yr<sup>-1</sup> (see 175 176 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 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 178 179 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

agriculture and other sectors contributed ~52% and ~18%, respectively. Of the remaining

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

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## 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. Specifically, the BU land estimate during 1998–2016 was on average 1.8 Tg N yr<sup>-1</sup> higher than 195 the TD estimate, but showed a slightly slower increasing rate of  $0.8\pm0.2$  Tg N yr<sup>-1</sup> per decade 196 (95% confidence interval; P < 0.05) compared to 1.1±0.6 Tg N yr<sup>-1</sup> per decade (P < 0.05) from 197 198 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 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> 201 202 per decade (P < 0.05), while the TD approach gave a higher but stable value of 5.1 (3.4–7.1) Tg N yr<sup>-1</sup> during 1998–2016 (Table 1). 203 Based on BU approaches, anthropogenic N<sub>2</sub>O emissions increased from 5.6 (3.6-8.7) Tg N yr<sup>-</sup> 204

<sup>1</sup> in the 1980s to 7.3 (4.2–11.4) Tg N yr<sup>-1</sup> in the recent decade at a rate of  $0.6\pm0.2$  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 fertilizer applications is the major source for agricultural emission increases, followed by a small 208 209 but significant increase in emissions from livestock manure and aquaculture. The model-based estimates of direct soil emissions<sup>15-17</sup> exhibit a faster increase than the three inventories used in 210 211 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 rate than suggested by the IPCC Tier 1  $EF^{14}$  (which assumes a linear response), especially after 216 2009 (ref. <sup>18</sup>). The remaining causes of the increase are attributed to other direct anthropogenic 217 218 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).

Our analysis of process-based model estimates indicates that soil N<sub>2</sub>O emissions accelerated substantially due to climate change since the early 1980s, which has offset the reduction due to elevated CO<sub>2</sub> concentration (Extended Data Fig. 6a). Elevated CO<sub>2</sub> enhances plant growth and thus increases N uptake, which in turn decreases soil N<sub>2</sub>O emissions<sup>16,19</sup>. Land conversion from tropical mature forests with higher N<sub>2</sub>O emissions to pastures and other unfertilized agricultural 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 converted croplands or pastures<sup>21</sup> (see Methods; Extended Data Fig. 7). 231 From the ensemble of process-based land model emissions<sup>15,16</sup>, we estimate a global 232 233 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, Perturbed fluxes from climate, atmospheric CO<sub>2</sub>, and land cover change). Previous field 236 237 experiments reported a better fit to local observations of soil N2O emissions when assuming a non-linear response to fertilizer N inputs under varied climate and soil conditions<sup>17,22</sup>. The non-238 239 linear response is likely also associated with long-term N accumulation in agricultural soils from N fertilizer use and in aquatic systems from N loads (the legacy effect)<sup>18,23</sup>, which provides more 240 substrate for microbial processes<sup>18,24</sup>. The increasing N<sub>2</sub>O emissions estimated by process-based 241 242 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).

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

BU approaches give estimates of N<sub>2</sub>O emissions in the five source categories, while TD
approaches only provide total emissions (Fig. 2). BU and TD approaches indicate that Africa was
the largest N<sub>2</sub>O source in the last decade, followed by South America (Fig. 2). BU and TD
approaches agree well in the magnitudes and trends of N<sub>2</sub>O emissions from South Asia and
Oceania (Extended Data Fig. 3j, 1). 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).

Based on the Global N<sub>2</sub>O Model Intercomparison Project (NMIP) estimates<sup>16</sup>, natural soil 261 262 emissions (to different extents) dominated in tropical and sub-tropical regions. Soil N<sub>2</sub>O emissions in the tropics  $(0.1\pm0.04 \text{ g N m}^{-2} \text{ yr}^{-1})$  are about 50% higher than the global average, 263 since many lowland, highly-weathered tropical soils have excess N relative to phosphorus<sup>20</sup>. 264 265 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 induced by livestock manure that is deposited in pastures and rangelands<sup>28,29</sup>. East Asia 270 271 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 play a moderate role in the total 272 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 273

anthropogenic sources together contribute  $N_2O$  emissions of approximately 0.2–0.4 Tg N yr<sup>-1</sup> in 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.

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

286 Trends in anthropogenic emissions varied 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 past 37 years (1980–2016). The 287 288 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

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 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 N2O 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 numbers<sup>15,28</sup>. Rising demand for meat and dairy products has significantly increased global N<sub>2</sub>O 310 311 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 of livestock could further enhance global N<sub>2</sub>O emissions<sup>37,38</sup>. Likewise, increasing demand for 313 314 fish has triggered a five-fold increase in global aquaculture production since the late 1980s<sup>39</sup>, with demand projected to increase further<sup>40</sup>, although this remains a small fraction (<1%) of total 315 316 N<sub>2</sub>O emissions.

The acceleration of global N<sub>2</sub>O emissions resulting from anthropogenic sources is apparent in both BU and TD results and currently tracks the highest Representative Concentration Pathway (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,  $N_2O$ 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

been shown to increase crop yields, reduce N<sub>2</sub>O emissions, increase water quality, and increase farm income<sup>47</sup>. In addition, N<sub>2</sub>O emissions can be efficiently abated in the chemical industry<sup>11,43,48,49</sup>, as has been achieved successfully in nitric acid plants in the European Union where industrial N<sub>2</sub>O emissions dropped from 11% to 3% of total emissions between 2007 and 2012 (ref. <sup>46</sup>). Additional available strategies 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>.

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.

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369 **References** 

370	1	Prather, M. J. et al. Measuring and modeling the lifetime of nitrous oxide including its
371		variability. Journal of Geophysical Research: Atmospheres 120, 5693-5705 (2015).
372	2	Ciais, P. et al. in Climate Change 2013: The Physical Science Basis. Contribution of
373		Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on
374		Climate Change 465-570 (Cambridge University Press, 2014).
375	3	Gidden, M. J. et al. Global emissions pathways under different socioeconomic scenarios
376		for use in CMIP6: a dataset of harmonized emissions trajectories through the end of the
377		century. Geoscientific Model Development 12, 1443-1475 (2019).
378	4	Davidson, E. A. Representative concentration pathways and mitigation scenarios for
379		nitrous oxide. Environmental Research Letters 7, 024005 (2012).
380	5	Hall, B., Dutton, G. & Elkins, J. The NOAA nitrous oxide standard scale for atmospheric
381		observations. Journal of Geophysical Research: Atmospheres 112, D09305 (2007).
382	6	Prinn, R. G. et al. History of chemically and radiatively important atmospheric gases
383		from the Advanced Global Atmospheric Gases Experiment (AGAGE). Earth System
384		<i>Science Data</i> <b>10</b> , 985-1018 (2018).
385	7	Butterbach-Bahl, K., Baggs, E. M., Dannenmann, M., Kiese, R. & Zechmeister-
386		Boltenstern, S. Nitrous oxide emissions from soils: how well do we understand the
387		processes and their controls? Phil. Trans. R. Soc. B 368, 20130122 (2013).
388	8	Tian, H. et al. The terrestrial biosphere as a net source of greenhouse gases to the
389		atmosphere. Nature 531, 225-228 (2016).
390	9	UNEP. Drawing down N <sub>2</sub> O to protect climate and the ozone layer. Report No.
391		9280733583, (United Nations Environment Programme (UNEP), 2013).
392	10	Park, S. et al. Trends and seasonal cycles in the isotopic composition of nitrous oxide
393		since 1940. Nature Geoscience 5, 261-265 (2012).
394	11	Davidson, E. A. & Kanter, D. Inventories and scenarios of nitrous oxide emissions.
395		Environmental Research Letters 9, 105012 (2014).
396	12	Reay, D. S. et al. Global agriculture and nitrous oxide emissions. Nature Climate Change
397		<b>2</b> , 410-416 (2012).
398	13	Syakila, A. & Kroeze, C. The global nitrous oxide budget revisited. <i>Greenhouse Gas</i>
399		Measurement and Management 1, 17-26 (2011).
400	14	IPCC. 2006 IPCC Guidelines for National Greenhouse Gas Inventories., (Japan on behalf
401		of the IPCC, Hayama, Japan, 2006).
402	15	Dangal, S. R. et al. Global nitrous oxide emissions from pasturelands and rangelands:
403		Magnitude, spatio-temporal patterns and attribution. <i>Global Biogeochemical Cycles</i> 33,
404		200-222 (2019).
405	16	Tian, H. Q. et al. Global soil nitrous oxide emissions since the preindustrial era estimated
406		by an ensemble of terrestrial biosphere models: Magnitude, attribution, and uncertainty.
407		<i>Global Change Biology</i> <b>25</b> , 640-659 (2019).

408 17 Wang, Q. et al. Data-driven estimates of global nitrous oxide emissions from croplands. 409 National Science Review 7, 441-452 (2020). Thompson, R. L. et al. Acceleration of global N2O emissions seen from two decades of 410 18 411 atmospheric inversion. Natural Climate Change 9, 993-998 (2019). 412 Zaehle, S., Ciais, P., Friend, A. D. & Prieur, V. Carbon benefits of anthropogenic reactive 19 413 nitrogen offset by nitrous oxide emissions. Nature Geoscience 4, 601-605 (2011). 414 Davidson, E. A. et al. Recuperation of nitrogen cycling in Amazonian forests following 20 415 agricultural abandonment. Nature 447, 995-998 (2007). 416 21 Verchot, L. V. et al. Land use change and biogeochemical controls of nitrogen oxide 417 emissions from soils in eastern Amazonia. Global Biogeochemical Cycles 13, 31-46 418 (1999). 419 22 Shcherbak, I., Millar, N. & Robertson, G. P. Global metaanalysis of the nonlinear 420 response of soil nitrous oxide (N2O) emissions to fertilizer nitrogen. Proceedings of the 421 National Academy of Sciences 111, 9199-9204 (2014). 422 23 Van Meter, K. J., Basu, N. B., Veenstra, J. J. & Burras, C. L. The nitrogen legacy: 423 emerging evidence of nitrogen accumulation in anthropogenic landscapes. Environmental 424 Research Letters 11, 035014 (2016). 425 24 Firestone, M. K. & Davidson, E. A. Microbiological basis of NO and N<sub>2</sub>O production and 426 consumption in soil. Exchange of trace gases between terrestrial ecosystems the 427 atmosphere 47, 7-21 (1989). 428 25 Griffis, T. J. et al. Nitrous oxide emissions are enhanced in a warmer and wetter world. 429 Proceedings of the National Academy of Sciences 114, 12081-12085 (2017). 430 Pärn, J. et al. Nitrogen-rich organic soils under warm well-drained conditions are global 26 431 nitrous oxide emission hotspots. Nature Communications 9, 1135 (2018). 432 Smith, K. The potential for feedback effects induced by global warming on emissions of 27 433 nitrous oxide by soils. *Global Change Biology* **3**, 327-338 (1997). 434 FAOSTAT. The Food and Agriculture Organization of the United Nations Statistics: 28 435 Emissions-Agriculture, Emissions Land Use Trade (Crops and livestock products), 436 Population, Agri-Environmental Indicators (Livestock Manure) (2019). 437 29 Xu, R. et al. Increased nitrogen enrichment and shifted patterns in the world's grassland: 1860–2016. Earth System Science Data 11, 175-187 (2019). 438 439 Beusen, A. H., Bouwman, A. F., Van Beek, L. P., Mogollón, J. M. & Middelburg, J. J. 30 440 Global riverine N and P transport to ocean increased during the 20th century despite 441 increased retention along the aquatic continuum. *Biogeosciences* 13, 2441-2451 (2016). 442 MacLeod, M., Hasan, M. R., Robb, D. H. F. & Mamun-Ur-Rashid, M. Quantifying and 31 443 mitigating greenhouse gas emissions from global aquaculture. FAO, Rome (2019). 444 Buitenhuis, E. T., Suntharalingam, P. & Le Quéré, C. Constraints on global oceanic 32 445 emissions of N<sub>2</sub>O from observations and models. *Biogeosciences* 15, 2161-2175 (2018). 446 33 Manizza, M., Keeling, R. F. & Nevison, C. D. On the processes controlling the seasonal 447 cycles of the air-sea fluxes of O<sub>2</sub> and N<sub>2</sub>O: A modelling study. Tellus B: Chemical and 448 Physical Meteorology 64, 18429 (2012). 449 34 Martinez-Rey, J., Bopp, L., Gehlen, M., Tagliabue, A. & Gruber, N. Projections of oceanic N<sub>2</sub>O emissions in the 21st century using the IPSL Earth system model. 450 451 Biogeosciences 12, 4133-4148 (2015). 452 35 Maavara, T. et al. Nitrous oxide emissions from inland waters: Are IPCC estimates too 453 high? Global Change Biology 25, 473-488 (2019).

454	36	Yao, Y. et al. Increased global nitrous oxide emissions from streams and rivers in the
455		Anthropocene. Natural Climate Change 10, 138-142 (2020).
456	37	Gerber, P. J. et al. Tackling climate change through livestock: a global assessment of
457		emissions and mitigation opportunities. FAO (2013).
458	38	Herrero, M. et al. Biomass use, production, feed efficiencies, and greenhouse gas
459		emissions from global livestock systems. Proceedings of the National Academy of
460		<i>Sciences</i> <b>110</b> , 20888-20893 (2013).
461	39	Yuan, J. et al. Rapid growth in greenhouse gas emissions from the adoption of industrial-
462		scale aquaculture. Nature Climate Change 9, 318-322 (2019).
463	40	Froehlich, H. E., Runge, C. A., Gentry, R. R., Gaines, S. D. & Halpern, B. S.
464		Comparative terrestrial feed and land use of an aquaculture-dominant world. <i>Proceedings</i>
465		of the National Academy of Sciences 115, 5295-5300 (2018).
466	41	O'Neill, B. C. et al. The Scenario Model Intercomparison Project (ScenarioMIP) for
467		CMIP6. Geoscience Model Development 9, 3461-3482 (2016).
468	42	Gütschow, J. et al. The PRIMAP-hist national historical emissions time series. Earth
469		<i>System Science Data</i> <b>8</b> , 571-603 (2016).
470	43	Winiwarter, W., Höglund-Isaksson, L., Klimont, Z., Schöpp, W. & Amann, M. Technical
471		opportunities to reduce global anthropogenic emissions of nitrous oxide. <i>Environmental</i>
472		<i>Research Letters</i> <b>13</b> , 014011 (2018).
473	44	Zhang, X. <i>et al.</i> Managing nitrogen for sustainable development. <i>Nature</i> <b>528</b> , 51-59
474		(2015).
475	45	Mueller, N. D. <i>et al.</i> Declining spatial efficiency of global cropland nitrogen allocation.
476	16	Global Biogeochemical Cycles <b>31</b> , 245-257 (2017).
477	46	European Environment Agency. Annual European Union greenhouse gas inventory 1990-
478		2017 and inventory report 2019. Submission under the United Nations Framework
479	17	Convention on Climate Change and the Kyoto Protocol, Copenhagen, DK (2019).
480	4/	Cui, Z. <i>et al.</i> Pursuing sustainable productivity with millions of smallholder farmers.
481	40	Nature <b>555</b> , 363-366 (2018).
482	48	Kanter, D. <i>et al.</i> A post-Kyoto partner: considering the stratospheric ozone regime as a
485		1001 to manage mirous oxide. Proceedings of the National Academy of Sciences 110,
404	40	4451-4457 (2015).
405	49	the CDM: A dipic acid A case of carbon lookage. Stockholm Environment Institute
400		(2010)
407		(2010).
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	the 1980s			the 1990s			the 2000s			2007-2016			
Anthropogenic so	mean	min	max	mean	min	max	mean	min	max	mean	min	max	
Direct emissions	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
of N additions in	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
the agricultural	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
sector	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
(Agriculture)	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	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
Other direct	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
sources	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
3001003	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
	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
Indirect emissions from anthropogenic N	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
additions	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
	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
Perturbed fluxes from	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
climate/CO <sub>2</sub> /land cover change	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
Anthropogenic tota		5.6	3.6	8.7	6.2	3.9	9.7	6.7	4.1	10.3	7.3	4.2	11.4
Natural fluxes													
Natural soils baseli	ne	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 wat	ers, estuaries, coastal	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 atmo	spheric production	04	0.2	1.2	04	0.2	1.2	04	0.2	1.2	04	0.2	1.2
Surface sink		-0.01	0.00	-0.3	-0.01	0.0	-0.3	-0.01	0.00	-0.3	-0.01	0.00	-0.3
Natural total		9.9	8.5	12.2	9.8	8.3	12.1	9.8	8.2	12.0	9.7	8.0	12.0
Bottom-up total		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								51	0.1	7.2	51	2.4	7.1
Top-down Ccean								10.8	3.1	12.5	11.8	3.4 10.6	13.8
Top-down total								10.0	9.3	10.0	11.0	10.6	477
source								15.9	15.1	16.9	16.9	15.9	17.7
Top-down Statospheric sink								12.1	11.4	13.1	12.4	11.7	13.3
Observed atmosph							13.3	12.2	14.4	13.5	12.4	14.6	
Change in atmosp							3.7	3.2	4.2	4.3	3.8	4.8	
Atmospheric burden	1462	1442	1482	1493	1472	1514	1531	1510	1552	1555	1533	1577	

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

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  $N_2O$  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  $N_2O$ ,

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.



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  $N_2O$  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  $N_2O$  budget, as reflected in each of the terms. The  $N_2O$ 516 sources and sinks are given in  $Tg N yr^{-1}$ . 517



519 Fig. 2 Regional N<sub>2</sub>O sources in the recent decade (2007–2016) over 11 regions. The Earth's

ice-free land is partitioned into ten regions: North America (NA), South America (SA), Europe (EU),
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:  $30^{\circ}$ – $90^{\circ}N$ ,
- 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  $N_2O$  emission (kg N capita<sup>-1</sup> yr<sup>-1</sup>) during
- 531 2007–2016 is shown in Supplementary Fig. 2.
- 532
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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<sub>2</sub>O 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<sup>-2</sup>) 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



546 Fig. 4 Historical and projected global anthropogenic N<sub>2</sub>O emissions and concentrations.

547 Global anthropogenic N<sub>2</sub>O 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>).

scenarios based on the Shared Socioeconomic Pathways (SSPs) used in CMIP6 (b, d, ref. <sup>41</sup>).
The historical data is represented as the mean of the BU and TD estimates of anthropogenic N<sub>2</sub>O

emissions, while the atmospheric concentration uses the three observation networks available,

551 emissions, while the almospheric 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

value is equal to the 2000–2009 average of the mean of TD and BU estimates. The SSPs are

554 *value is equal to the 2000–2009 average of the mean of TD and DO estimates. The 551's 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.

557

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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 (NMIP<sup>16</sup>, Supplementary Tables 16–17) and provided here reflect a situation without 587 588 consideration of land use change (e.g., deforestation) and without consideration of indirect anthropogenic effects via global change (i.e., climate, elevated CO<sub>2</sub>, and atmospheric N 589 590 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, estuaries, coastal zones' includes coastal upwelling<sup>50</sup> and inland and coastal waters that were 592 593 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, we assume that these values are constant during 1980–2016. Yao et al.<sup>36</sup> provided annual 595 596 riverine N<sub>2</sub>O emissions using DLEM during the same period. Here, we averaged estimates from Yao et al. with that from Maavara et al.<sup>35</sup>. In addition, we estimated N<sub>2</sub>O emissions from global 597 598 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 estuaries and lakes is still based on the long-term averaged values provided by Maavara et al.<sup>35</sup> 600 and Lauerwald et al.<sup>51</sup>, respectively. We considered the riverine emissions in the year 1900 as 601 602 equivalent to the natural emission for the DLEM estimate assuming that the N load from land was negligible in that period<sup>52</sup>. We quantified the contribution of natural sources to total 603 604 emission from reservoirs, lakes, and estuaries at 44% (36%-52%), with consideration of all N inputs (i.e., inorganic, organic, dissolved, particulate forms). We combined the estimate from 605

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	NH <sub>2</sub> and NO <sub>2</sub> . This category is in any case very small and the anthropogenic enhancement effect
611	is uncertain. Lightning produces NO <sub>x</sub> , the median estimate of which is 5 Tg N yr <sup>-1</sup> (ref. $^{53}$ ). We
612	assumed an EF of 1% (ref. $^{54}$ ) and a global estimate of 0.05 (0.02–0.09) Tg N yr <sup>-1</sup> from lightning.
613	Atmospheric production of $N_2O$ results from the reaction of $NH_2$ with $NO_2$ (refs. <sup>55,56</sup> ), N with
614	NO <sub>2</sub> , and oxidation of N <sub>2</sub> by $O(^{1}D)^{57}$ , all of which constitute an estimated source of 0.3 (0.2–1.1)
615	Tg N yr <sup>-1</sup> . 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 $N_2O$ 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 N2O emissions, the same as the NMIP. Thus, we
622	add the DLEM-based estimate of pasture $N_2O$ 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 N2O 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 N2O 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 $\pm$ 0.01 Tg N yr <sup>-1</sup> per decade (P < 0.05; Extended Data Fig. 4b-c).
643	Meanwhile, global aquaculture N2O 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±0.01 Tg N yr <sup>-1</sup> 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 $-0.03\pm0.04$ Tg N yr <sup>-1</sup> per decade (P = 0.3) since
655	the1980s (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, N <sub>2</sub> 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
663 664	(c) Indirect emissions from anthropogenic N additions. Data were obtained from various sources and considered N deposition on land and ocean ('N deposition on land' and 'N
663 664 665	<ul> <li>(c) Indirect emissions from anthropogenic N additions. Data were obtained from various sources and considered N deposition on land and ocean ('N deposition on land' and 'N deposition on ocean'), as well as the N leaching and runoff from upstream ('Inland and coastal</li> </ul>
663 664 665 666	<ul> <li>(c) Indirect emissions from anthropogenic N additions. Data were obtained from various</li> <li>sources and considered N deposition on land and ocean ('N deposition on land' and 'N</li> <li>deposition on ocean'), as well as the N leaching and runoff from upstream ('Inland and coastal waters'). The emission from 'N deposition on ocean' was provided by Suntharalingam et al.<sup>67</sup>,</li> </ul>
663 664 665 666 667	<ul> <li>(c) Indirect emissions from anthropogenic N additions. Data were obtained from various</li> <li>sources and considered N deposition on land and ocean ('N deposition on land' and 'N</li> <li>deposition on ocean'), as well as the N leaching and runoff from upstream ('Inland and coastal waters'). The emission from 'N deposition on ocean' was provided by Suntharalingam et al.<sup>67</sup>, while emission from 'N deposition on land' was the ensemble mean of an average of three</li> </ul>
<ul> <li>663</li> <li>664</li> <li>665</li> <li>666</li> <li>667</li> <li>668</li> </ul>	<ul> <li>(c) Indirect emissions from anthropogenic N additions. Data were obtained from various</li> <li>sources and considered N deposition on land and ocean ('N deposition on land' and 'N</li> <li>deposition on ocean'), as well as the N leaching and runoff from upstream ('Inland and coastal waters'). The emission from 'N deposition on ocean' was provided by Suntharalingam et al.<sup>67</sup>, while emission from 'N deposition on land' was the ensemble mean of an average of three</li> <li>inventories: FAOSTAT/EDGAR v4.3.2, GAINS/EDGAR v4.3.2, and NMIP. FAOSTAT and</li> </ul>
<ul> <li>663</li> <li>664</li> <li>665</li> <li>666</li> <li>667</li> <li>668</li> <li>669</li> </ul>	(c) Indirect emissions from anthropogenic N additions. Data were obtained from various sources and considered N deposition on land and ocean ('N deposition on land' and 'N deposition on ocean'), as well as the N leaching and runoff from upstream ('Inland and coastal waters'). The emission from 'N deposition on ocean' was provided by Suntharalingam et al. <sup>67</sup> , while emission from 'N 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
<ul> <li>663</li> <li>664</li> <li>665</li> <li>666</li> <li>668</li> <li>669</li> <li>670</li> </ul>	(c) Indirect emissions from anthropogenic N additions. Data were obtained from various sources and considered N deposition on land and ocean ('N deposition on land' and 'N deposition on ocean'), as well as the N leaching and runoff from upstream ('Inland and coastal waters'). The emission from 'N deposition on ocean' was provided by Suntharalingam et al. <sup>67</sup> , while emission from 'N 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 N leaching or N deposition, while EDGAR v4.3.2 did not. Here, we treated 'Indirect
<ul> <li>663</li> <li>664</li> <li>665</li> <li>666</li> <li>669</li> <li>670</li> <li>671</li> </ul>	(c) Indirect emissions from anthropogenic N additions. Data were obtained from various sources and considered N deposition on land and ocean ('N deposition on land' and 'N deposition on ocean'), as well as the N leaching and runoff from upstream ('Inland and coastal waters'). The emission from 'N deposition on ocean' was provided by Suntharalingam et al. <sup>67</sup> , while emission from 'N 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 N leaching or N deposition, while 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
<ul> <li>663</li> <li>664</li> <li>665</li> <li>666</li> <li>667</li> <li>668</li> <li>669</li> <li>670</li> <li>671</li> <li>672</li> </ul>	(c) Indirect emissions from anthropogenic N additions. Data were obtained from varioussources and considered N deposition on land and ocean ('N deposition on land' and 'Ndeposition on ocean'), as well as the N leaching and runoff from upstream ('Inland and coastalwaters'). The emission from 'N deposition on ocean' was provided by Suntharalingam et al. <sup>67</sup> , Iwhile emission from 'N deposition on land' was the ensemble mean of an average of threeinventories: FAOSTAT/EDGAR v4.3.2, GAINS/EDGAR v4.3.2, and NMIP. FAOSTAT andGAINS documented the sector 'Indirect agricultural N2O emissions' by separating estimatesfrom N leaching or N deposition, while EDGAR v4.3.2 did not. Here, we treated 'Indirectagricultural N2O emissions' from EDGAR v4.3.2 as 'Inland and coastal waters' emissions fordata synthesis. Only EDGAR v4.3.2 provided an estimate of indirect emission from non-

674 NH<sub>x</sub>/NO<sub>y</sub> 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 estimated by Yao et al.<sup>36</sup> considered annual N inputs and other environmental factors (i.e., 680 681 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 intertidal N<sub>2</sub>O emissions were undated from Murray et al<sup>68</sup>. Coastal waters with low disturbance 684 generally either have low N<sub>2</sub>O emissions or act as a sink for  $N_2O^{69,70}$ . Here, coastal zone 685 686 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 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 688 689 2007–2016 (Table 1). The N<sub>2</sub>O emissions from inland and coastal waters increased at a rate of 690  $0.03\pm0.00$  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  $0.06\pm0.01$  Tg N yr<sup>-1</sup> per decade (P < 0.05) reported in the three estimates (FAOSTAT/EDGAR 696 697 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 in 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).

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  $CO_2$ , 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 developed by Houghton et al.<sup>72</sup> for accounting for carbon flows due to land use. In this study, an 707 708 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 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 711 712 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 713 714 uses data collected by a review analysis<sup>74</sup>, based upon which the second one further considers observations from Verchot et al.<sup>21</sup> and Keller and Reiners<sup>75</sup>. In the first function, x (unit: year) 715 716 indicates pasture age in years after deforestation and y (unitless; 0-1) indicates the ratio of 717 pasture  $N_2O$  emission over the  $N_2O$  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

initial forest clearing and the gradually declining emission rates of converted crops/pastures<sup>21,76</sup>. 721 722 Using these two curves and the baseline, we kept track of the  $N_2O$  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 al.<sup>72</sup> and McGuire et al.<sup>77</sup> to simulate its influences on carbon and N cycles. Moreover, through 728 using the sites of field observation from Davidson et al.<sup>20</sup> and Keller and Reiners<sup>75</sup>, we estimated 729 N<sub>2</sub>O emission from secondary tropical forests based on the algorithm: y = 0.0084x + 0.2401 ( $R^2$ 730 731 = 0.44). x (unit: year) indicates secondary forest age and y (unitless; 0-1) indicates the ratio of secondary forest N<sub>2</sub>O emission over that of a reference mature forest. The difference between 732 733 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 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 739 measuring the mean surface mixing ratios<sup>78</sup> and that of converting surface mixing ratios to a 740 global mean abundance<sup>80</sup>, we estimate a  $\pm 1.4\%$  uncertainty in the burden. Annual change in 741 742 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 AR5 (ref.<sup>2</sup>). There shows an agreement of the 743

stratospheric loss from atmospheric chemistry transport models (TD modeled chemical sink<sup>18,81</sup>) and from satellite observations with a photolysis model (observed photochemical sink<sup>1</sup>), which differ only by ~1 Tg N yr<sup>-1</sup>. The satellite-based lifetime, 116±9 years, gives an overall uncertainty in the annual loss of ±8%. The tropospheric loss of N<sub>2</sub>O from reaction with O(<sup>1</sup>D) is included in observed atmospheric chemical sink (Table 1) and is small (~1% of the stratospheric 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  $N_2O$ 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 agricultural soils, the largest emission source. Previous global N<sub>2</sub>O assessments<sup>52,82,83</sup> based on 753 754 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 global inventories (EDGAR v4.3.2, FAOSTAT, and GAINS) that are based on the IPCC 2006 757 guidelines<sup>14</sup>. The main reason is that indirect emissions from leaching and groundwater were 758 759 overestimated in previous studies<sup>85</sup>. Correspondingly, projections of atmospheric N<sub>2</sub>O concentrations based on these overestimated emissions<sup>82</sup> led to biased estimates. For example, 760 Mosier and Kroeze<sup>82</sup> expected atmospheric N<sub>2</sub>O concentrations to be 340–350 ppb in the year 761 762 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

soil emissions based on these new guidelines may decrease by 20%–25%. Integrating estimates
relying on the IPCC methodology with estimates by process-based models provides for a more
balanced assessment in this paper. We also added information from assessments<sup>86,87</sup> that derived
agricultural emissions as the difference between atmospheric terms and other emissions like
combustion, industry and nature, and they gave comparable magnitudes (4.3–5.8 Tg N yr<sup>-1</sup>) to
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 variety of TD and BU methods. TD approaches, consisting of four inversion frameworks<sup>88-91</sup>, 774 775 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 strongly determined by the number of atmospheric N<sub>2</sub>O measurements<sup>18</sup>. Inversions are 778 779 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.

BU approaches are subject to uncertainties in various sources from land<sup>16</sup> and oceans<sup>32</sup>. For process-based models (e.g. 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 multiple 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

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 these regions may produce more N<sub>2</sub>O than is simulated by the models<sup>32</sup>. The current generation 803 804 of global ocean biogeochemistry models are not sufficiently accurate to represent the high N2O production processes in low-oxygen zones, and their associated variability (see refs. <sup>34,93,94</sup> for 805 806 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.

Regardless of the tier approach used, GHG 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, 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 N2O 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 $N_2O$ 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 N2O
824	emissions from global peatland degradation are still lacking, although we have accounted for
825	$N_2O$ 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	$cycle^{99}$ contribute to increasing N <sub>2</sub> O emissions, which, however, have not been well established
828	in the current estimates of the global N2O budget.
829	Statistics. Through using the Mann-Kendall test in R-3.4.4, we checked the significance of
830	trends in annual N2O emissions from each sub-sector based on the BU approach.

## 832 **References**

833 50 Nevison, C. D., Lueker, T. J. & Weiss, R. F. Quantifying the nitrous oxide source from
834 coastal upwelling. *Global Biogeochemical Cycles* 18, GB1018 (2004).

Lauerwald, R. *et al.* Natural lakes are a minor global source of N<sub>2</sub>O to the atmosphere. *Global Biogeochemical Cycles* 33, 1564-1581 (2019).

Kroeze, C., Mosier, A. & Bouwman, L. Closing the global N<sub>2</sub>O budget: a retrospective analysis 1500–1994. *Global Biogeochemical Cycles* 13, 1-8 (1999).

839	53	Schumann, U. & Huntrieser, H. The global lightning-induced nitrogen oxides source.
840		Atmospheric Chemistry and Physics 7, 3823-3907 (2007).
841	54	De Klein, C. et al. N <sub>2</sub> O emissions from managed soils, and CO <sub>2</sub> emissions from lime and
842		urea application. IPCC Guidelines for National Greenhouse Gas Inventories, Prepared
843		by the National Greenhouse Gas Inventories Programme 4, 1-54 (2006).
844	55	Dentener, F. J. & Crutzen, P. J. A three-dimensional model of the global ammonia cycle.
845		Journal of Atmospheric Chemistry 19, 331-369 (1994).
846	56	Röckmann, T., Kaiser, J., Crowley, J. N., Brenninkmeijer, C. A. & Crutzen, P. J. The
847		origin of the anomalous or "mass-independent" oxygen isotope fractionation in
848		tropospheric N2O. Geophysical Research Letters 28, 503-506 (2001).
849	57	Kaiser, J. & Röckmann, T. Absence of isotope exchange in the reaction of N <sub>2</sub> O+O( <sup>1</sup> D)
850		and the global $\Delta^{17}$ O budget of nitrous oxide. <i>Geophysical Research Letters</i> <b>32</b> (2005).
851	58	Schlesinger, W. H. An estimate of the global sink for nitrous oxide in soils. Global
852		<i>Change Biology</i> <b>19</b> , 2929-2931 (2013).
853	59	Syakila, A., Kroeze, C. & Slomp, C. P. Neglecting sinks for N <sub>2</sub> O at the earth's surface:
854		does it matter? Journal of Integrative Environmental Sciences 7, 79-87 (2010).
855	60	Bouwman, A. F. et al. Hindcasts and future projections of global inland and coastal
856		nitrogen and phosphorus loads due to finfish aquaculture. Reviews in Fisheries Science
857		<b>21</b> , 112-156 (2013).
858	61	Bouwman, A. F. et al. Global hindcasts and future projections of coastal nitrogen and
859		phosphorus loads due to shellfish and seaweed aquaculture. Reviews in Fisheries Science
860		<b>19</b> , 331-357 (2011).
861	62	Hu, Z., Lee, J. W., Chandran, K., Kim, S. & Khanal, S. K. Nitrous oxide (N <sub>2</sub> O) emission
862		from aquaculture: a review. Environmental science technology 46, 6470-6480 (2012).
863	63	Williams, J. & Crutzen, P. J. Nitrous oxide from aquaculture. Nature Geoscience 3, 143
864		(2010).
865	64	Andela, N. et al. A human-driven decline in global burned area. Science 356, 1356-1362
866		(2017).
867	65	Yang, J. et al. Spatial and temporal patterns of global burned area in response to
868		anthropogenic and environmental factors: Reconstructing global fire history for the 20th
869		and early 21st centuries. Journal of Geophysical Research: Biogeosciences 119, 249-263
870		(2014).
871	66	Dennis, R. A review of fire projects in Indonesia (1982-1998). Cifor (1999).
872	67	Suntharalingam, P. et al. Quantifying the impact of anthropogenic nitrogen deposition on
873		oceanic nitrous oxide. Geophysical Research Letters 39, L07605 (2012).
874	68	Murray, R. H., Erler, D. V. & Eyre, B. D. Nitrous oxide fluxes in estuarine environments:
875		response to global change. Global Change Biology 21, 3219-3245 (2015).
876	69	Erler, D. V. et al. Applying cavity ring - down spectroscopy for the measurement of
877		dissolved nitrous oxide concentrations and bulk nitrogen isotopic composition in aquatic
878		systems: Correcting for interferences and field application. <i>Limnology and</i>
879		<i>Oceanography: Methods</i> <b>13</b> , 391-401 (2015).
880	70	Murray, R., Erler, D., Rosentreter, J. & Eyre, B. Seasonal and spatial N <sub>2</sub> O concentrations
881		and emissions in three tropical estuaries. Marine Chemistry 221, 103779 (2020).
882	71	Vernberg, F. J. & Vernberg, W. B. The coastal zone: past, present, and future. Univ of
883		South Carolina Press (2001).

884	72	Houghton, R. et al. Changes in the Carbon Content of Terrestrial Biota and Soils between
885		1860 and 1980: A Net Release of CO <sub>2</sub> to the Atmosphere. <i>Ecological monographs</i> 53,
886		235-262 (1983).
887	73	Davidson, E. A. The contribution of manure and fertilizer nitrogen to atmospheric nitrous
888		oxide since 1860. Nature Geoscience 2, 659-662 (2009).
889	74	van Lent, J., Hergoualc'h, K. & Verchot, L. V. Reviews and syntheses: Soil N <sub>2</sub> O and NO
890		emissions from land use and land-use change in the tropics and subtropics: a meta-
891		analysis. <i>Biogeosciences</i> <b>12</b> , 7299-7313 (2015).
892	75	Keller, M. & Reiners, W. A. Soil-atmosphere exchange of nitrous oxide, nitric oxide, and
893		methane under secondary succession of pasture to forest in the Atlantic lowlands of Costa
894		Rica. Global Biogeochemical Cycles 8, 399-409 (1994).
895	76	Melillo, J. M. et al. Nitrous oxide emissions from forests and pastures of various ages in
896		the Brazilian Amazon. Journal of Geophysical Research: Atmospheres 106, 34179-34188
897		(2001).
898	77	McGuire, A. et al. Carbon balance of the terrestrial biosphere in the twentieth century:
899		Analyses of CO <sub>2</sub> , climate and land use effects with four process-based ecosystem models.
900		Global Biogeochemical Cycles 15, 183-206 (2001).
901	78	Dlugokencky, E., Steele, L., Lang, P. & Masarie, K. The growth rate and distribution of
902		atmospheric methane. Journal of Geophysical Research: Atmospheres 99, 17021-17043
903		(1994).
904	79	Prather, M. et al. Annex II: Climate system scenario tables. Cambridge, United Kingdom
905		and New York, NY, USA (2013).
906	80	Prather, M. J., Holmes, C. D. & Hsu, J. Reactive greenhouse gas scenarios: Systematic
907		exploration of uncertainties and the role of atmospheric chemistry. Geophysical Research
908		Letters <b>39</b> , L09803 (2012).
909	81	Prather, M. J. & Hsu, J. Coupling of Nitrous Oxide and Methane by Global Atmospheric
910		Chemistry. Science 330, 952-954 (2010).
911	82	Mosier, A. & Kroeze, C. Potential impact on the global atmospheric N <sub>2</sub> O budget of the
912		increased nitrogen input required to meet future global food demands. Chemosphere-
913		<i>Global Change Science</i> <b>2</b> , 465-473 (2000).
914	83	Mosier, A. et al. Closing the global N <sub>2</sub> O budget: nitrous oxide emissions through the
915		agricultural nitrogen cycle. Nutrient cycling in Agroecosystems 52, 225-248 (1998).
916	84	IPCC. Revised 1996 IPCC guidelines for national greenhouse gas inventories. Hayama,
917		<i>Japan</i> (1997).
918	85	Nevison, C. in IPCC, Background Papers: IPCC Expert Meetings on Good Practice
919		Guidance and Uncertainty Management in National Greenhouse Gas Inventories. IPCC
920		National Greenhouse Gas Inventories Programme, Technical Support Unit. 381-397
921		(2000).
922	86	Crutzen, P. J., Mosier, A. R., Smith, K. A. & Winiwarter, W. N <sub>2</sub> O release from agro-
923		biofuel production negates global warming reduction by replacing fossil fuels.
924		Atmospheric Chemistry and Physics 8, 389-395 (2008).
925	87	Smith, K. A., Mosier, A. R., Crutzen, P. J. & Winiwarter, W. The role of N <sub>2</sub> O derived
926		from crop-based biofuels, and from agriculture in general, in Earth's climate.
927		Philosophical Transactions of the Royal Society of London B: Biological Sciences 367,
928		1169-1174 (2012).

929	88	Thompson, R. L. et al. Nitrous oxide emissions 1999 to 2009 from a global atmospheric
930		inversion. Atmospheric Chemistry and Physics 14, 1801-1817 (2014).
931	89	Wells, K. C. et al. Simulation of atmospheric N <sub>2</sub> O with GEOS-Chem and its adjoint:
932		evaluation of observational constraints. Geoscience Model Development 8, 3179-3198
933		(2015).
934	90	Wilson, C., Chipperfield, M., Gloor, M. & Chevallier, F. Development of a variational
935		flux inversion system (INVICAT v1. 0) using the TOMCAT chemical transport model.
936		Geoscientific Model Development 7, 2485-2500 (2014).
937	91	Patra, P. K. et al. Improved Chemical Tracer Simulation by MIROC4. 0-based
938		Atmospheric Chemistry-Transport Model (MIROC4-ACTM). Sola 14, 91-96 (2018).
939	92	Tian, H. Q. et al. The Global N <sub>2</sub> O Model Intercomparison Project. Bulletin of the
940		American Meteorological Society 99, 1231-1252 (2018).
941	93	Suntharalingam, P. et al. Anthropogenic nitrogen inputs and impacts on oceanic N <sub>2</sub> O
942		fluxes in the northern Indian Ocean: The need for an integrated observation and
943		modelling approach. Deep Sea Research Part II: Topical Studies in Oceanography 166,
944		104-113 (2019).
945	94	Battaglia, G. & Joos, F. Marine N <sub>2</sub> O Emissions From Nitrification and Denitrification
946		Constrained by Modern Observations and Projected in Multimillennial Global Warming
947		Simulations. Global Biogeochemical Cycles 32, 92-121 (2018).
948	95	Galloway, J. et al. The impact of animal production systems on the nitrogen cycle. Vol.
949		1, Island Press (2010).
950	96	Steinfeld, H., Mooney, H. A., Schneider, F. & Neville, L. E. Livestock in a changing
951		landscape, volume 1: drivers, consequences, and responses. Vol. 1, Island Press (2013).
952	97	IPCC. 2019 Refinement to the 2006 IPCC Guidelines for National Greenhouse Gas
953		Inventories. Hayama, Japan (2019).
954	98	Elberling, B., Christiansen, H. H. & Hansen, B. U. High nitrous oxide production from
955		thawing permafrost. Nature Geoscience 3, 332-335 (2010).
956	99	Wagner-Riddle, C. et al. Globally important nitrous oxide emissions from croplands
957		induced by freeze-thaw cycles. Nature Geoscience 10, 279-283 (2017).
958	100	Suntharalingam, P. et al. Estimates of Oceanic Nitrous-oxide Emissions from Global
959		Biogeochemistry Models. American Geophysical Union, Fall Meeting 2018 (2018).
960	101	Janssens-Maenhout, G. et al. EDGAR v4.3.2 Global Atlas of the three major greenhouse
961		gas emissions for the period 1970–2012. Earth System Science Data 11, 959-1002
962		(2019).
963	102	Tubiello, F. et al. Estimating greenhouse gas emissions in agriculture: a manual to
964		address data requirements for developing countries. FAO, Rome (2015).
965	103	Van Der Werf, G. R. et al. Global fire emissions estimates during 1997-2016. Earth
966		<i>System Science Data</i> <b>9</b> , 697-720 (2017).
967	104	Dentener, F. Global maps of atmospheric nitrogen deposition, 1860, 1993, and 2050.
968		Data set. Available on-line ( <u>http://daac</u> . ornl. gov/) from Oak Ridge National Laboratory
969		Distributed Active Archive Center, Oak Ridge, TN, USA (2006).
970	105	Riahi, K. et al. The Shared Socioeconomic Pathways and their energy, land use, and
971		greenhouse gas emissions implications: An overview. Global Environmental Change 42,
972		153-168 (2017).
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
- paper. Additional description on data availability for atmospheric N<sub>2</sub>O observations from
   NOAA, AGAGE and CSIRO networks is provided in the Supplementary Information. The
- NOAA, AGAGE and CSIRO networks is provided in the Supplementary Information. The data
   presented here are made available in the belief that their dissemination will lead to greater
- 980 presented here are made available in the benef 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
- providers. Researchers interested to use results made available in the repository are encouraged,
- 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
- and Global Change Research at Auburn University (https://auburn.box.com/).
- 992

# 993 Acknowledgements

This paper is the result of a collaborative international effort under the umbrella of the Global
Carbon Project (a project of Future Earth and a research partner of the World Climate Research

995 Carbon Project (a project of Future Earth and a research partner of the world Chinate Research 996 Programme) and International Nitrogen Initiative. This research was made possible partly by

- 997 Andrew Carnegie Fellowship Award no. G-F-19-56910; NSF grant nos. 1903722,1243232 and
- 998 1922687; NASA grant nos. NNX14AO73G, NNX10AU06G, NNX11AD47G and
- 999 NNX14AF93G; NOAA grant nos. NA16NOS4780207 and NA16NOS4780204; National Key R
- 1000 & D Program of China (grant nos. 2017YFA0604702 and 2018YFA0606001), National Natural
- 1001 Science Foundation of China (Grant no. 41961124006), CAS grants (KFJ-STSZDTP-0;
- 1002 SKLURE2017-1-6), and OUC-AU Joint Center Program. Additional funding support includes:
- 1003 E.T.B, P.R., G.P.P., R.L.T., P.S. acknowledge funding support from VERIFY project (EC H2020
- 1004 grant no. 776810). P.S. also acknowledges funding from the EC H2020 grant No 641816
- 1005 (CRESCENDO); A.I. acknowledges funding support from JSPS KAKENHI grant (no.
- 1006 17H01867). G.B., F.J, and S.L. acknowledge support by Swiss National Science Foundation
- 1007 (#200020\_172476) and by the EC H2020 grant no. 821003 (Project 4C) and no. 820989 (Project
- 1008 COMFORT). A.L. acknowledges support by DFG project SFB754/3. S.Z. acknowledges support
- 1009 by the EC H2020 grant no. 647204. K.C.W. and D.B.M. acknowledge support from NASA (IDS
- 1010 Grant #NNX17AK18G) and NOAA (Grant #NA13OAR4310086); P.A.R. acknowledges NASA
- 1011 Award NNX17AI74G, M.M. acknowledges support from the Scottish Government's Rural and
- 1012 Environment Science and Analytical Services Division (RESAS) Environmental Change
- 1013 Programme (2016-2021); B.E. acknowledges the support from ARC Linkage Grants
- 1014 LP150100519 and LP190100271; M.P. acknowledges US Department of Energy, DE-
- 1015 SC0012536; Lawrence Livermore National Laboratory, B628407 and NASA MAP program,
- 1016 NNX13AL12G; S.B. was supported by the EC H2020 with the CRESCENDO project (grant no.
- 1017 641816) and by the COMFORT project (grant no. 820989). S.B. also acknowledges the support
- 1018 of the team in charge of the CNRM-CM climate model; F.Z. acknowledges the support from the

1019 National Natural Science Foundation of China (41671464). Supercomputing time was provided

- 1020 by the Météo-France/DSI supercomputing center. P.K.P. is partly supported by Environment
- 1021 Research and Technology Development Fund (#2-1802) of the Ministry of the Environment,
- 1022 Japan. R.L. acknowledges support from the French state aid managed by the ANR under the "Investissements d'avenir" programme with the reference ANR-16-CONV-0003. NOAA ground-1023
- 1024
- based observations of atmospheric N<sub>2</sub>O are supported by NOAA's Climate Program Office under 1025 the Atmospheric Chemistry Carbon Cycle and Climate (AC4) theme. The AGAGE stations
- 1026 measuring N<sub>2</sub>O are supported by NASA (USA) grants NNX16AC98G to MIT, and
- 1027 NNX16AC97G and NNX16AC96G to SIO, and by BEIS (UK) for Mace Head, NOAA (USA)
- 1028 for Barbados, and CSIRO and BoM (Australia) for Cape Grim. We also thank Dr. Steve Frolking
- 1029 and two anonymous reviewers for constructive comments and suggestions that have helped
- 1030 improve this paper. The statements made and views expressed are solely the responsibility of the 1031 authors.
- 1032

#### 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. led atmospheric inversions teaming with M.P.C., T.M., D.B.M., P.K.P., K.C.W., and C.W.; H.T. 1037 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., 1038 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 1042 data of N<sub>2</sub>O flux in aquaculture. G.R.W. and J.Y. provided data of N<sub>2</sub>O emissions from biomass 1043 burning, F.Z. provided cropland N<sub>2</sub>O flux data from a statistical model and field observations. G.J.M., F.N.T. and W.W. provided N<sub>2</sub>O inventory data. M.J.P. and D.J.R. provided data of 1044 1045 stratospheric and tropospheric sinks. G.P.P. provided RCP and SSP scenarios data and analysis. 1046 B.H., E.D. and J.E. provided a global N<sub>2</sub>O monitoring dataset of NOAA/ESRL GMD. R.P. and R.W. provided a global N<sub>2</sub>O monitoring dataset of AGAGE stations. P.K. provided a global N<sub>2</sub>O 1047 monitoring dataset of CSIRO. All coauthors reviewed and commented on the manuscript. 1048 1049

- 1050 Competing interests: The authors declare no competing interests.
- 1051
- 1052 **Additional information**
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- 1054 1055
- 1056 Correspondence and requests for materials should be addressed to H.T.

Supplementary information is available for this paper at https://

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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

as 12-month moving averages. Growth rates are not calculated prior to 1995 due to insufficient

1064 data and higher uncertainties on the measurements.



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 used in Table 1 and Figs. 1-3. We utilize both approaches, including 22 BU and five TD 1069 estimates of N<sub>2</sub>O fluxes from land and oceans. For sources estimated by BU, we include six 1070 process-based terrestrial biosphere modeling studies<sup>16</sup>; five process-based ocean biogeochemical 1071 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 1072 statistical model SRNM based on spatial extrapolation of field measurements<sup>17</sup>; and four GHG 1073 inventories: EDGAR v4.3.2<sup>101</sup>, FAOSTAT<sup>102</sup>, GAINS<sup>43</sup>, and GFED4s<sup>103</sup>. In addition, previous 1074 literatures regarding estimates of 'Surface sink'58,73, 'Lightning'53,54, 'Atmospheric 1075 production<sup>56,57,104</sup>, 'Aquaculture<sup>31,62</sup>, and model-based 'Tropospheric sink'<sup>81</sup> and observed 1076 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> provide global aquaculture N<sub>2</sub>O emissions in 2013 and in 2009, respectively; and the nutrient 1078 budget model<sup>30,60,61</sup> provides N flows in global freshwater and marine aquaculture over the 1079 period 1980-2016. Model-based estimates of N2O emissions from 'Inland and coastal waters' 1080 include rivers and reservoirs<sup>35,36</sup>, lakes<sup>51</sup>, estuaries<sup>35</sup>, coastal zones (i.e., seagrasses, mangroves, 1081 saltmarsh and intertidal saltmarsh)<sup>68</sup>, and coastal upwelling<sup>50</sup>. 1082 1083 1084



1085

1086 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

1089 represent the mean N<sub>2</sub>O emission from TD methods and the shaded areas show minimum and

- 1090 maximum estimates.
- 1091





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 estimates. NMIP/DLEM or SRNM/DLEM means the combination of N<sub>2</sub>O emission by NMIP or 1096 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 estimates. c, Emission from manure management based on FAOSTAT, GAINS, and EDGAR 1100 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 Hu et al.<sup>62</sup>; the solid line represents the 'best estimate' that is the product of EF (1.8%) and N 1102 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.





1114 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 N 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

1110 N20 Huxes due to changing CO2 and chinate. **b**, Global natural son N20 emissions which

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

abandonment from 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

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

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  $N_2O^{-1}$ 

1130 emission from inland waters and estuaries, while site-level emission rates of  $N_2O$  were upscaled

- 1131 to estimate global  $N_2O$  fluxes from the global seagrass area<sup>68</sup>.
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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 represents the mean N<sub>2</sub>O emission from post-deforestation pulse effect (i.e., crop/pasture N<sub>2</sub>O 1138 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 N2O 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

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

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

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

line shows the ensemble direct N<sub>2</sub>O emissions from livestock manure based on EDGAR v4.3.2,
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

direct N<sub>2</sub>O 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>167 <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

applied) are shown in gray regardless of the radiative forcing level they reach in 2100.