



Supplement of

Global nitrous oxide budget (1980–2020)

Hanqin Tian et al.

Correspondence to: Hanqin Tian (hanqin.tian@bc.edu)

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Extended methodology

S1.1 NMIP-2: global Nitrogen/N₂O Model Inter-comparison Project phase 2

The NMIP2 is a follow-up model intercomparison project of NMIP (Tian *et al.*, 2018), which provides estimates of N₂O emissions from natural and agricultural soils and covers the time period 1850-2020. Eight process-based Terrestrial Biosphere Models (TBMs) participate in NMIP-2. In general, N₂O emissions from soil are regulated at two levels, which are the rates of nitrification and denitrification in the soil and soil physical factors regulating the ratio of N₂O to other nitrous gases (Davidson *et al.*, 2000). For N input to land ecosystems, all eight models considered N fertilizer use, atmospheric N deposition and biological fixation, but five models considered manure as N input. For vegetation processes, all models included dynamic algorithms in simulating N allocation to different living tissues and vegetation N turnover, and simulated plant N uptake using the “Demand and Supply-driven” approach. For soil N processes, all eight models simulated N leaching according to water runoff rate; however, models are different in representing nitrification and denitrification processes and the impacts of soil chemical and physical factors. The differences in simulating nitrification and denitrification processes are one of the major uncertainties in estimating N₂O emissions. Model characteristics in simulating major N cycling processes associated with N₂O emissions in each participating model are briefly described in Table S1.

Table S1. Model characteristics in simulating major N cycling processes

	CLASS IC	DLEM	ELM	ISAM	LPX-Bern	O-CN	ORCHIDEE	VISIT
Open C cycle ^a	Yes	Yes	Yes	Yes	Yes	Yes	Yes	Yes
C-N coupling	Yes	Yes	Yes	Yes	Yes	Yes	Yes	Yes
N pools ^b	(3, 1, 3)	(6,6,8)	(6,4,5)	(6,4,4)	(4,3,8)	(9,6,9)	(9,6,9)	(4,1,4)
Demand and supply-driven plant N uptake	Yes	Yes	Yes	Yes	Yes	Yes	Yes	Yes
N allocation ^c	Dynamic	Dynamic	Dynamic	Dynamic	Dynamic	Dynamic	Dynamic	Dynamic
Nitrification	$f(T, SWC, C_{NH4})$	$f(T, SWC, C_{NH4})$	$f(T, SWC, pH, rh, C_{NH4})$	$f(T, SWC, C_{NH4})$	$f(T, SWC, C_{NH4})$	$f(T, SWC, pH, C_{NH4})$	$f(T, SWC, pH, C_{NH4})$	$f(T, SWC, pH, C_{NH4})$
Denitrification	$f(T, SWC, C_{NO3})$	$f(T, SWC, clay, rh, C_{NO3})$	$f(T, SWC, pH, rh, C_{NO3})$	$f(T, SWC, C_{NO3})$	$f(T, SWC, R_{mb}, C_{NO3})$	$f(T, SWC, pH, R_{mb}, C_{NO3})$	$f(T, SWC, pH, denitrifier, C_{NO3})$	$f(SWC, rh, C_{NO3})$
Mineralization, immobilization	$f(C:N)$	$f(C:N)$	$f(C:N)$	$f(C:N)$	$f(C:N)$	$f(C:N)$	$f(C:N)$	$f(C:N)$
N leaching	$f(runoff, C_{NO3}, C_{NH4})$	$f(runoff, C_{NO3}, C_{NH4})$	$f(runoff, C_{NO3})$	$f(runoff, C_{NO3}, C_{NH4})$	$f(runoff, C_{NO3})$	$f(runoff, C_{NO3}, C_{NH4})$	$f(runoff, C_{NO3}, C_{NH4})$	$f(runoff, C_{NO3})$
NH ₃ volatilization	$f(C_{NH3})$	$f(T, SWC, C_{NH3})$	No	$f(C_{NH3})$	$f(T, SWC, C_{NH3})$	$f(C_{NH3})$	$f(SWC, pH, C_{NH4})$	$f(T, SWC, C_{NH3})$

		$pH,$ C_{NH4}			$pH,$ C_{NH4}			$pH,$ C_{NH4}
Plant N turnover ^d	Dynamic	Dynamic	Dynamic	Dynamic	Dynamic	Dynamic	Dynamic	Dynamic
N resorption	Fixed	$f(C:N)$	Fixed	$f(C:N)$	Fixed	Fixed	Fixed	Fixed
N fixation	$f(N_{limit})$	$f(T,$ $SWC,$ $C_{NH4},$ $C_{NO3})$	$f(T,$ $C:N)$	$f(ET)$	Implied by mass balance	$f(N_{limit})$	Fixed	$f(ET)$
N fertilizer use	Yes	Yes	Yes	Yes	Yes	Yes	Yes	Yes
Manure N use	No	Yes	No	Yes	No	Yes	Yes	Yes
N deposition	Yes	Yes	Yes	Yes	Yes	Yes	Yes	Yes

21 ^a “Open” denotes that excess N can be leached from the system.

22 ^b Numbers of N pools (vegetation pools, litter pools, soil pools).

23 ^c Dynamic denotes time-varied N allocation ratio to different N pools.

24 *T*: soil temperature, *SWC*: soil water content, *clay*: soil clay fraction, *ET*: evapotranspiration, *denitrifier*:
 25 soil denitrifier biomass, *rh*: soil heterogeneous respiration, *N_{limit}*: N limitation of vegetation growth,
 26 *C_{NO3}*: soil NO_3^- content, *C_{NH4}*: soil NH_4^+ content.

27
 28 All NMIP2 models are driven by consistent input datasets (i.e., climate, atmospheric CO₂ concentration,
 29 land cover change, irrigation, atmospheric N deposition, mineral N fertilization, and manure N application
 30 and deposition) and implemented consistent simulation experiments (SH0 – SH12; Table A4). Nitrogen
 31 inputs data used in NMIP2 simulations are from History of anthropogenic Nitrogen inputs (HaNi) dataset
 32 (*Tian et al.*, 2022), which takes advantage of different data sources in a spatiotemporally consistent way to
 33 generate a set of high-resolution (5 arcminutes) gridded N input products from 1850 to 2020. HaNi data set
 34 shows that the total anthropogenic N inputs to global terrestrial ecosystems increased from 29.05 Tg N yr⁻¹
 35 in the 1860s to 267.23 Tg N yr⁻¹ in the 2010s, with the dominant N source changing from atmospheric N
 36 deposition (before the 1900s) to manure N (the 1910s-2000s), and to synthetic fertilizer in the 2010s (Fig.
 37 B3). The climate data used to run historical simulations is the half degree CRU-JRA2.2 6-hourly forcing
 38 over 1901- 2020 (<https://catalogue.ceda.ac.uk/uuid/4bdf41fc10af4caa489b14745c665a6>). Annual CO₂
 39 concentration during 1850-2020 were derived from ice core CO₂ data and NOAA annual
 40 observations(<https://www.esrl.noaa.gov>). Historical distribution of cropland, pasture, rangeland and
 41 irrigation during 1850-2020 were from Land-Use Harmonization 2 (LUH2) dataset (*Hurt et al.*, 2020). The
 42 original dataset of LUH2 is at a resolution of 0.25° x 0.25° longitude/latitude. We aggregated all geo-
 43 referenced input data into a consistent spatial resolution of 0.5° x 0.5° longitude/latitude to run NMIP2
 44 models.

45
 46 NMIP2 models perform a subset of 13 simulations (SH0-SH12) to quantify N₂O emissions from both
 47 agricultural and natural soils during the study period, and to disentangle the effects of multiple
 48 environmental factors on soil N₂O emissions. The SH1 results were taken as the “best estimates” of soil
 49 N₂O emissions because they include the effects of all driving factors that models can take into account. In
 50 the SH0 simulation, driving forces were kept constant at the level in 1850 over the entire simulation period
 51 (1850-2020). According to previous N₂O budget studies, atmospheric N₂O growth rate and Monte-Carlo
 52 method, we suggest the following criteria for the N₂O budget inclusion (Table A6), and the criteria for
 53 carbon components are consistent with TRENDY. By comparing results from factorial simulation
 54 experiments (SH0 - SH12), we attribute changes in soil N₂O emissions to seven natural and anthropogenic
 55 factors, namely, climate (CLIM, including precipitation, humidity, temperature and photosynthetic active
 56 radiation changes), atmospheric CO₂ concentration (CO₂), land cover change (LCC), irrigation (IRRI),

57 atmospheric N deposition (NDEP), mineral N fertilizer use (NFER), and manure N use in cropland
 58 (MANN). In order to understand soil N₂O emissions dynamics caused by crop cultivation, we further
 59 separate the global and regional N₂O emissions into those derived from cropland soils and those from soils
 60 of other land ecosystems. In this study, we attribute the impact of a single factor on cropland N₂O emissions.
 61 Five models (DLEM, ISAM, O-CN, ORCHIDEE, and VISIT) considered the effects of manure N
 62 application in cropland, therefore, we use these five models' results to calculate the manure N effect (SH1-
 63 SH2). Meanwhile, we used results from all the eight models (i.e., CLASSIC, DLEM, ELM, ISAM, LPX-
 64 Bern, O-CN, ORCHIDEE, and VISIT) to calculate the effects of synthetic N fertilizer use (SH1-SH3) and
 65 atmospheric N deposition (SH1-SH4). The effect of N deposition in natural ecosystems (SH1-SH4) and the
 66 effects of CO₂ (SH1-SH7) and climate (SH1-SH8) on global terrestrial ecosystems are calculated from the
 67 eight NMIP2 models mentioned above.
 68

69 **Table S2. Criteria for the N₂O budget inclusion**

Carbon criteria	N ₂ O criteria
(1) Steady state after spin-up, diagnosed from SH0 run: steady-state defined as an offset < 0.10 PgC yr ⁻¹ , drift < 0.05 PgC yr ⁻¹ per century (i.e. first is the average over 1850-2020, second is the slope x 100).	(1) Steady state after spin-up, diagnosed from SH0 run: drift < 0.2 Tg N ₂ O-N yr ⁻¹ per century (i.e. the slope x 100).
(2) Net annual land flux is a carbon sink over the 1990s and 2000s as constrained by global atmospheric and oceanic observations (Keeling & Manning, 2014), diagnosed from SH3 run.	(2) Inside the present-day (2007-2016) land emission range: 7-13 Tg N ₂ O-N yr ⁻¹ , diagnosed from SH1 run. The upper limit was calculated using the maximum total N ₂ O emissions minus the minimum estimates of other sources, and the lower limit was calculated using the minimum total N ₂ O emissions minus the maximum estimates of other sources. The range of total emissions was estimated by a one-box model using atmospheric N ₂ O growth rate, and the range of the sum of other sources was calculated by a Monte-Carlo method using estimates from Tian et al. (2020).
	(3) Inside the pre-industrial land emission range: 3 to 9 Tg N ₂ O-N yr ⁻¹ , diagnosed from SH1 run. This range is derived from the pre-industrial atmospheric burden/N ₂ O lifetime minus ocean and river/ coastal/estuary emissions (Michael J. Prather et al., 2015).

70
71

72 **S1.2 Brief description of algorithms associated with N₂O flux in each NMIP2 model:**

73 **S1.2.1: CLASSIC**

74 The representation of nitrogen cycling in CLASSIC is described in *Asaadi and Arora (2021)* and *Kou*
 75 *Giesbrecht and Arora (2022)*. N₂O production due to both nitrification and denitrification are represented.
 76 N₂O loss during nitrification (I_{N_2O} ; g N m⁻² d⁻¹) is represented with the following equation:

$$77 \quad I_{N_2O} = \eta_{N_2O} f_I(T_{0.5}) f_I(\psi) N_{NH_4} \quad (1)$$

78 η_{N_2O} is a coefficient (d⁻¹), $f_I(T_{0.5})$ is a dimensionless scalar that depends on soil temperature averaged over
 79 the top 0.5m soil depth ($T_{0.5}$), $f_I(\psi)$ is a dimensionless scalar that depends on soil matric potential (ψ), and
 80 N_{NH_4} is the soil ammonium pool (g N m⁻²).

81 N₂O loss during denitrification (E_{N_2O} ; g N m⁻² d⁻¹) is represented with the following equation:

$$E_{N_2O} = \mu_{N_2O} f_E(T_{0.5}) f_E(\theta) N_{NO_3} \quad (2)$$

μ_{N_2O} is a coefficient (d^{-1}), $f_E(T_{0.5})$ is a dimensionless scalar that depends on soil temperature averaged over the top 0.5m soil depth ($T_{0.5}$), $f_E(\theta)$ is a dimensionless scalar that depends on soil moisture (θ), and N_{NO_3} is the soil nitrate pool ($g\ N\ m^{-2}$).

86
87

S1.2.2: DLEM

88 The nitrogen cycle scheme in DLEM2.0 (Xu et al., 2017; Yang et al., 2015; Tian et al. 2020) are similar as
89 DLEM1.0 (Lu and Tian, 2013; Tian et al., 2012b; Tian et al., 2010; Tian et al., 2011; Xu et al., 2011),
90 However, the N_2O emission schemes in DLEM2.0 (Xu et al., 2017) have been modified based on Chatskikh
91 et al. (2005) and Heinen (2006).

92

$$R_{nit} = k_{nit_max} f(T1) f(WFPS) C_{NH_4} \quad (3)$$

94

$$R_{den} = k_{den_max} f(T2) f(WFPS) C_{NO_3} \quad (4)$$

96

97 where R_{nit} is the daily nitrification rate ($g\ N/m^2/d$); R_{den} is the daily denitrification rate ($g\ N/m^2/d$);
98 $f(T1)$ and $f(T2)$ are the impact function of daily soil temperature on nitrification and denitrification,
99 respectively; $f(WFPS)$ is the impact function of water-filled pore space (WFPS) on nitrification,
100 denitrification and N_2O diffusion; k_{nit_max} is the maximum fraction of NH_4^+ -N that is converted to NO_3^- -
101 N or gases (0-1); k_{den_max} is the maximum fraction of NO_3^- -N that is converted to gases (0-1); C_{NH_4} and
102 C_{NO_3} are the soil NH_4^+ -N and NO_3^- -N content ($g\ N/m^2$). N_2O from denitrification and nitrification processes
103 are calculated as,

104

$$R_{N_2O} = (R_{nit} + R_{den}) f(T3) (1 - f(WFPS)) \quad (5)$$

106

107 where R_{N_2O} is the daily N_2O emission rate ($g\ N/m^2/d$); $f(T3)$ is the impact function of daily soil
108 temperature on N_2O diffusion rate from soil pores. The calculation methods for these functions and
109 parameters were described in detail in Xu et al. (2017) and Yang et al. (2015).

110

S1.2.3: ELM

112 The nitrogen dynamics in ELM is simulated based on the theory of equilibrium chemistry approximation
113 (Zhu et al., 2016). Plants, soil microbes, and abiotic factors such as mineral surfaces coexist in the same
114 soil environment and vie for a diverse array of nutrients, including NH_4^+ , NO_3^- . Due to the limited
115 availability of these nutrients, intense competitive interactions are expected. The competition of those
116 limited resources is represented by consumer–substrate networks, therefore, the uptake of nutrient substrate
117 by each consumer is dependent on the relative competitiveness of one consumer over the others. Nutrient
118 consumers' competitiveness is parametrized with kinetic parameters (Zhu et al., 2016). As a result, neither
119 plan nor soil microbes get the first priority to access nutrient substrates. Instead, when the potential nutrient
120 demands (from all nutrient consumers) exceed the supply at a given time step, the allocation of limited
121 nutrients among the consumers affects their performance (e.g., plant growth, soil organic matter
122 accumulation, nitrification, denitrification rates). ELM adopts a multiple-consumer-multiple-substrate
123 competition network (Zhu et al., 2016; Zhu et al., 2019) to simulate (1) nitrogen uptake facilitated by
124 nitrogen carrier enzymes, (2) binding of a nutrient substrate to a particular enzyme precludes it from
125 attaching to other enzymes, and (3) rates and affinities of consumers for different substrates. After the
126 nutrient competition has been resolved, scaling terms ($f(ECA_{nit})$ and $f(ECA_{den})$) will be applied to the
127 potential nitrification and denitrification processes:

128

$$R_{nit} = k_{nit_max} f(\theta) f(T) (1 - f(O)) f(ECA_{nit}) C_{NH_4} \quad (6)$$

130

$$R_{den} = \min(f(deomp), f(C_{NO3}))f(ECA_{den}) \quad (7)$$

where $k_{nit,max}$ is the maximum nitrification rate, $f(\theta)$, $f(T)$, $f(O)$ are soil moisture, temperature, and oxygen scalars, respectively. $f(deomp)$ and $f(C_{NO3})$ are carbon limited and NO₃- limited denitrification rates (Del Grosso et al., 2000).

137 S1.2.4: ISAM

138 ISAM model contains detailed calculations of the terrestrial ecosystem's organic and mineral N cycle (Yang
139 et al., 2009). The major N processes in ISAM include biological fixation, leaching, mineralization and
140 immobilization, plant uptake, nitrification, and denitrification. The soil biogeochemistry module of ISAM
141 shares the same ten soil layers (to 3.5 m depth) as the soil biogeophysics and calculates the vertical transport
142 of SOC and N (Shu et al., 2020; Yang et al., 2009). N₂O emission in ISAM N₂O is produced as a byproduct
143 of nitrification and denitrification (Xu et al., 2021). N₂O module explicitly accounts for the vertical transport
144 of C, N, and O₂ within every soil layer for both saturated and unsaturated soil conditions by accounting for
145 the process of oxygen diffusing into the soil from the atmosphere and the soil oxygen supply. The model
146 also explicitly accounts for the effects of anoxic and oxic environments on nitrification (N_{ni}, Eq. 6) and
147 denitrification (N_{de}, Eq. 7). Both environments are calculated based on the fraction of anoxic soil depending
148 on soil O₂ concentration, which is non-linearly correlated with the chemical pathways forming N₂O.

$$149 N_{ni} = NH_4^+ \times (1 - e^{-F_{te,m} \times F_{sm,m} \times r_{ni}}) \times F_{pH,m,ni} \times R_d \quad (8)$$

$$150 N_{de} = NO_3^- \times r_{de} \times Rh \times F_{pH,m,de} \times R_d \quad (9)$$

151 where NH_4^+ and NO_3^- are ammonium and nitrate pool sizes; $F_{te,m}$ is temperature modifier; $F_{sm,m}$ is soil
152 moisture modifier; r_{ni} and r_{de} are base nitrification and denitrification rates; $F_{pH,m,ni}$ and $F_{pH,m,de}$ are pH
153 modifiers for nitrification and denitrification; R_d is relative soil anoxic fraction; $Rh (= 1 - R_d)$ is heterotrophic
154 respiration.

155 Under anoxic soil conditions, N₂O is produced through denitrification, while under oxic soil conditions,
156 more N₂O is produced from nitrification. The model accounts for soil NH₄⁺ volatilization at the soil surface
157 when NH₄⁺ in NH₄⁺-containing fertilizers (e.g., urea) is converted to ammonia gas, depending upon pH
158 (Huang and Gerber, 2015). The soil NH₄⁺ volatilization in the model is also affected by the anoxic condition,
159 which increases under a higher temperature and relatively lesser soil anoxic condition. The model accounts
160 for the impacts of pH on nitrification, denitrification, and volatilization rates (Li et al., 2000; Xu-Ri and
161 Prentice, 2008). We prescribe the soil pH from the Global Soil Dataset for Earth System Modeling dataset
162 (GSDE) (Shangguan et al., 2014).

163 S1.2.5: LPX-Bern

164 The implementation of nitrogen dynamics in LPX-Bern is based on the work of Xu-Ri and Prentice (2008).
165 Nitrogen uptake by plants is governed by their demand and the availability of nitrogen in two soil pools
166 representing ammonium and nitrate. Nitrogen from deposition and fertilization are added to these inorganic
167 soil pools. Losses include ammonium volatilization, nitrate leaching as well as N₂O and NO production
168 during nitrification and N₂O, NO and N₂ production during denitrification. Aerobic nitrification of
169 ammonium is dependent on soil temperature (T_{soil}) and indirectly on soil water content due to the
170 partitioning of wet and dry soil:

$$171 R_{nit} = \max_{nit} f_1(T_{soil}) C_{NH4,dry} \quad (10)$$

172 where $\max_{nit} = 0.92 \text{ day}^{-1}$ is the daily maximum nitrification rate at 20°C.

180 Anaerobic denitrification of nitrate in wet soil depends on labile carbon availability and soil temperature:

$$181 \\ 182 R_{den} = R_{mb}/(R_{mb} + K_{mb})f_2(T_{soil})C_{NO3,wet}/(C_{NO3,wet} + K_n) \quad (11)$$

183
184 The parameters K_{mb} and K_n are taken from *Xu-Ri and Prentice* (2008) and R_{mb} is the microbiological soil
185 respiration. The amount of nitrogen lost as N_2O due to nitrification and denitrification is modelled as a
186 function of soil temperature, water content and the respective process rate.

187 188 **S1.2.6: O-CN**

189 The treatment of inorganic soil nitrogen dynamics in O-CN follows largely *Xu-Ri and Prentice* (2008). O-
190 CN (*Zaehle and Friend*, 2010) considers N losses to NH_3 volatilisation, NO_x , N_2O and N_2 production and
191 emission, as well as NH_4 and NO_3 leaching. Inorganic nitrogen dynamics in the soil are tightly coupled to
192 plant uptake and net mineralization. The anaerobic volume fraction of the soil is estimated by an empirical
193 function of the fractional soil moisture content (*Zaehle et al.*, 2011). The fraction of ammonium in the
194 aerobic part of the soil is subject to nitrification, according to:

$$195 \\ 196 R_{nit} = v_{max_{nit}}f(T1)f(pH1)C_{NH4} \quad (12)$$

197
198 where $f(pH1)$ is the soil pH response functions for nitrification (*Li et al.*, 1992; *Xu-Ri and Prentice*, 2008),
199 and $v_{max_{nit}}$ is the maximum daily nitrification rate under 20°C and favourable pH conditions (*Xu-Ri and*
200 *Prentice*, 2008).

201
202 Gross denitrification of the fraction of nitrate under anoxic conditions is modelled as:

$$203 \\ 204 R_{den} = R_{mb}/(R_{mb} + K_{mb})f(T2)f(pH2)C_{NO3}/(C_{NO3} + K_n) \quad (13)$$

205
206 where $f(pH2)$ is the soil pH response functions for denitrification (*Li et al.*, 1992; *Xu-Ri and Prentice*, 2008),
207 R_{mb} is the soil microbial respiration rate, and K_{mb} and K_n parameters taken from *Li et al.* (1992).

208
209 The N_2O production from nitrification and denitrification is then calculated as:

$$210 \\ 211 R_{N2O} = a_{nit}f(T1)R_{nit} + b_{den}f(T2)f(pH3)R_{den} \quad (14)$$

212
213 where a_{nit} and b_{den} are fraction loss constants, $f(pH3)$ is a pH-modifier changing the degree of denitrification
214 producing N_2O versus NO_x or N_2 (*Zaehle et al.*, 2011). Emissions of volatile compounds are simulated
215 using the empirical emission of *Xu-Ri and Prentice* (2008).

216 217 **S1.2.7: ORCHIDEE**

218 Modeling of the mineral N dynamics by the ORCHIDEE model originates from the formulations used in
219 the O-CN (*Zaehle and Friend*, 2010). It is composed of five pools for ammonium/ammoniac, nitrate, NO_x ,
220 nitrous oxide, and di-nitrogen forms. N_2O production in both nitrification and denitrification processes are
221 represented.

222
223 The potential daily rate of nitrification, R_{nit} , occurs only on the aerobic fraction of the soil and is a function
224 of temperature, pH, and ammonium concentration (C_{NH4}):

$$225 \\ 226 R_{nit} = (1 - f(WFPS))f(T1)f(pH1)k_{nit}C_{NH4} \quad (15)$$

227
228 where k_{nit} is the reference potential NO_3^- production per mass unit of ammonium.

229

230 S1.2.8: VISIT

231 The nitrogen cycle scheme of VISIT is composed of three organic soil nitrogen pools (microbe, litter, and
232 humus), two inorganic soil nitrogen pools (ammonium and nitrate), and vegetation pools. Fertilizer is
233 considered as an input to the ammonium and nitrate pools at a fixed ratio, and manure as an input into the
234 litter organic nitrogen pool. N₂O emissions through nitrification and denitrification are estimated using the
235 scheme developed by *Parton et al.* (1996). Nitrification-associated N₂O emission (R_{nit,N_2O}) is evaluated as
236 follows,

$$237 R_{nit,N_2O} = f(WFPS)f(pH1)f(T1)(K_{max} + F_{max}f(NH_4)) \quad (16)$$

239 where K_{max} is the soil-specific turnover coefficient; F_{max} is the parameter of maximum nitrification gas flux;
240 and $f(NH_4)$ is the effect of soil ammonium on nitrification. Denitrification-associated N₂O emission
241 (R_{den,N_2O}) is evaluated by the following equation:

$$242 R_{den,N_2O} = R_{den}(1 + R_{N_2/N_2O}) \quad (17)$$

$$243 R_{den} = \min(f(NO_3), f(CO_2)) \times f(WFPS) \quad (18)$$

247 where R_{N_2/N_2O} is the fractionation coefficient, which is also a function of WFPS, soil nitrate, and
248 heterotrophic respiration, $f(NO_3)$ is the maximum denitrification rate in high soil respiration rate condition,
249 $f(CO_2)$ is the maximum denitrification rate in high NO₃⁻ levels, and $f(WFPS)$ is the effect of WFPS on
250 denitrification rate.

251 N₂O production by nitrification ($R_{N_2O,nit}$, g N-N₂O/m²/d) is expressed as a function of the potential daily rate
252 of nitrification (R_{nit} , g N-NO₃⁻/m²/d), temperature and the water content as shown in *Zhang et al.* (2002).

$$253 R_{N_2O,nit} = f(WFPS)f(T1)R_{nit}p_{N_2O,nit} \quad (19)$$

254 where $p_{N_2O,nit}$ (g N-N₂O (g N-NO₃⁻)⁻¹) is the reference N₂O production per mass unit of NO₃⁻ produced by
255 nitrification. The denitrification occurs on the anaerobic fraction of the soil which is computed as a function
256 of the water-filled porosity ($f(WFPS)$) and is controlled by temperature, pH, soil NO concentration and
257 denitrifier microbial activity (a_{microb} , g m⁻²) (*Li et al.*, 2000).

$$258 R_{N_2O,den} = f(WFPS)f(T2)f(pH)f(NO)p_{N_2O,den}a_{microb} \quad (20)$$

259 where $f(NO)$ is a Michaelis-Menten shape function and $p_{N_2O,den}$ is the reference N₂O production per mass
260 unit of denitrifier microbes.

261 S2 The FAOSTAT inventory

262 The FAOSTAT emissions data (*FAO*, 2022) are computed at Tier 1 following *IPCC* (2006), Vol. 4. The
263 overall equation is as follows:

264 Direct emissions are estimated at the country level, using the formula:

$$265 Emission = A * EF \quad (21)$$

266 where emission represents kg N yr⁻¹; A represents the amount of N in the following items (annual synthetic
267 N applications/manure applied to soils/manure left on pasture/manure treated in manure management
268 systems/crop residue/biomass burned amount) in kg N yr⁻¹; EF = Tier 1, default IPCC emission factors,
expressed in kg N/kg N.

279
280 Indirect emissions are estimated at the country level, using the formula:

281
282
$$Emission = A_{v\&l} * EF \quad (22)$$

283
284 where emission represents kg N yr⁻¹; $A_{v\&l}$ represents the fraction of manure/synthetic N fertilizers that
285 volatilize as NH₃ and NO_x and are lost through runoff and leaching in kg N yr⁻¹; EF = Tier 1, default IPCC
286 emission factors, expressed in kg N/kg N.

287
288 Synthetic N fertilizers: N₂O from synthetic fertilizers is produced by microbial processes of nitrification
289 and denitrification taking place on the addition site (direct emissions), and after volatilization/redeposition
290 and leaching processes (indirect emissions).

291
292 Manure management: The term manure includes both urine and dung (i.e., both liquid and solid material)
293 produced by livestock. N₂O is produced directly by nitrification and denitrification processes in the manure,
294 and indirectly by nitrogen (N) volatilization and redeposition processes.

295 Manure applied to soils: N₂O is produced by microbial processes of nitrification and denitrification taking
296 place on the application site (direct emissions), and after volatilization/redeposition and leaching processes
297 (indirect emissions).

298
299 Manure left on pastures: N₂O is produced by microbial processes of nitrification and denitrification taking
300 place on the deposition site (direct emissions), and after volatilization/redeposition and leaching processes
301 (indirect emissions).

302
303 Crop Residue: N₂O emissions from crop residues consist of direct and indirect emissions from nitrogen (N)
304 in crop residues left on agricultural fields by farmers and from forages during pasture renewal (following
305 the definitions in the IPCC guidelines (IPCC, 2006)). Specifically, N₂O is produced by microbial processes
306 of nitrification and denitrification taking place on the deposition site (direct emissions), and leaching
307 processes (indirect emissions).

308
309 Cultivation of organic soils: The FAOSTAT domain “Cultivation of organic soils” contains estimates of
310 direct N₂O emissions associated with the drainage of organic soils – histosols – under cropland and grazed
311 grassland.

312
313 Burning-savanna: N₂O emissions from the burning of vegetation biomass in the land cover types: Savanna,
314 Woody Savanna, Open Shrublands, Closed Shrublands, and Grasslands. Burning-crop residues: N₂O
315 produced by the combustion of a percentage of crop residues burnt on-site. Burning-biomass: N₂O
316 emissions from the burning of vegetation biomass in the land cover types: Humid tropical forests, other
317 forests, and organic soils.

318 319 **S3 The EDGAR v7.0 inventory**

320 The new online version, EDGAR v7.0 (https://edgar.jrc.ec.europa.eu/dataset_ghg70) incorporates a full
321 differentiation of emission processes with technology-specific emission factors and additional end-of-pipe
322 abatement measures and as such updates and refines the emission estimates. The emissions are modelled
323 based on the latest scientific knowledge and available global statistics primarily from International Energy
324 Agency (IEA, 2021) for energy related sectors, FAO statistics (FAO, 2022) for agriculture, which were
325 complemented for the rest of sectors with United States Geological Survey (USGS), International Fertiliser
326 Association (IFA), Gas Flaring Reduction Partnership (GGFR)/U.S. National Oceanic and Atmospheric
327 Administration (NOAA) and World Steel Association (worldsteel) recent statistics; the methods are those
328 recommended by IPCC (2006). Official data submitted by the Annex I countries to the United Nations

329 Framework Convention on Climate Change (UNFCCC) and to the Kyoto Protocol are used to some extent,
330 particularly regarding control measures implemented since 1990 that are not described by international
331 statistics. A fast-Track approach was used to extend the N₂O emission time series for the latest years up to
332 2021 (Crippa *et al.*, 2021; Crippa *et al.*, 2022).

333
334 The N₂O emission factors for direct soil emissions of N₂O from the use of synthetic fertilizers, from manure
335 used as fertilizers, and from crop residues are taken from *IPCC* (2006), which updated the default IPCC
336 emission factor in the IPCC Good Practice Guidance (2000) with a 20% lower value. N₂O emissions from
337 the use of animal waste as fertilizer are estimated considering both the loss of N that occurs from manure
338 management systems before manure is applied to soils and the additional N introduced by bedding material
339 (Janssens-Maenhout *et al.*, 2019). N₂O emissions from fertilizer use and CO₂ from urea fertilization are
340 estimated based on IFA and FAO recent statistics.

341
342 N₂O emissions from manure management are based on the distribution of manure management systems
343 from Annex I countries reporting to the UNFCCC, Zhou *et al.* (2007) for China and *IPCC* (2006) for the
344 rest of the countries.

345
346 Different N₂O emission factors are applied to tropical and non-tropical regions. N and dry matter content
347 of agricultural residues are estimated from the cultivation area and yield for 24 crop types from *FAO* (2022)
348 and using emission factors of *IPCC* (2006).

349
350 Indirect N₂O emissions from leaching and runoff of nitrate are estimated from N input to agricultural soils.
351 Leaching and runoff are assumed to occur in all agricultural areas except non-irrigated dryland regions,
352 which are identified with maps of FAO Geonetwork (<https://www.fao.org/land-water/databases-and-software/geonetwork/en/>). The fraction of N lost through leaching and runoff is based on the study of Van
353 Drecht *et al.* (2003). The updated emission factor for indirect N₂O emissions from N leaching and run-off
354 from the *IPCC* (2006) guidelines is selected, while noting that it is 70% lower than the mean value of the
355 1996 IPCC Guidelines and the IPCC Good Practice Guidance *IPCC* (1996; 2000).

356
357 Indirect N₂O emissions from atmospheric deposition of N of NO_x and NH₃ emissions from non-agricultural
358 sources, mainly fossil fuel combustion, are estimated using N in NO_x and NH₃ emissions from these sources
359 as activity data, based on EDGAR v7.0 database for these gases. The same emission factor from *IPCC*
360 (2006) is used for indirect N₂O from atmospheric deposition of N from NH₃ and NO_x emissions, as for
361 agricultural emissions (Janssens-Maenhout *et al.*, 2019).

362
363 The uncertainties for EDGAR N₂O emissions estimated by Solazzo *et al.* (2021) are based primarily on the
364 uncertainties in emissions factors and activity data statistics from the *IPCC* (2006). Globally, these
365 emissions are accurate within an interval of ±113 for energy, -12% to +16% for industrial processes and
366 product use, -225 to +302 for agriculture, -159% to 203% for waste and ±112% for others; the most
367 uncertain emissions are those related to N₂O from waste and agriculture.

368 369 370 **S4 The UNFCCC inventory (need description of UNFCCC)**

371 The UNFCCC collects detailed data on GHG emissions from its parties. Following extensive guidance
372 developed by IPCC (Buendia *et al.*, 2019; Eggleston *et al.*, 2006), parties to the convention prepare national
373 GHG inventories, including emissions (and sinks) of N₂O. All anthropogenic activities are covered, in
374 agriculture both direct and indirect N₂O emissions are included. While IPCC basically provides emission
375 factor approaches, parties are encouraged to take account of national specificities, use national factors and
376 data, wherever available, or develop emission models, with adequate scientific proof provided.
377 Combustion-related emissions and emissions from industrial processes may take advantage of emission

378 monitoring or specific plant operation conditions, if provided. Emission processes that are not associated
 379 with anthropogenic activities are also not covered in the inventories.

380
 381 Obligations and quality of data provided differ strongly by country category. High scrutiny is put on GHG
 382 inventories from countries listed in Annex-I of the convention (Annex-I countries include most European
 383 countries, U.S. and Canada, Australia and New Zealand, and Japan). Annex-I countries are obliged to
 384 provide annual national inventories in considerable detail and have to be very transparent also in terms of
 385 methodology used and underlying information. Each year, time-series of emissions and underlying data
 386 since 1990 (in a few cases, alternative base years are used) up to the pre-previous year are freshly provided
 387 in April each year (e.g., in April 2023 data up to the year 2021 had to be provided), leading to a
 388 homogeneous data series. Reports and emission data are provided (to UNFCCC, and to all users from the
 389 UNFCCC web site at <https://unfccc.int/reports>) in standardized format such that they can be transferred to
 390 databases. National results are routinely being checked and evaluated by expert teams in form of specific
 391 internal and external audits to assure data quality and consistency.

392
 393 National information is highly relevant also for non-Annex I countries to the UNFCCC and is being
 394 collected and distributed by UNFCCC as well. Requirements are much less stringent, however, as parties
 395 are expected to provide data only according to their own capabilities and the support they get from other
 396 countries. The so-called Biannual Update Reports are to be prepared every other year only. While in
 397 principle following the same IPCC guidance, commitments to format, timing, and quality assessment are
 398 by far less stringent, and the own ambition level of the respective party (country) may determine much of
 399 the outcome. In any case, self-reporting of a country always also means the party is willing to take the
 400 responsibility of the emissions reported.

401
 402 The “EDGAR/UNFCCC” dataset used in this paper utilizes the database for Annex-I countries for
 403 emissions from fossil-fuel consumption, industrial processes, waste and wastewater, and merges with the
 404 respective set derived from EDGARv7.0 for all remaining countries.

405 406 **S5.1 The SRNM model: Flux upscaling model**

407 The SRNM model (Wang et al., 2020) was applied to simulate direct cropland-N₂O emissions. In SRNM,
 408 N₂O emissions were simulated from N application rates using a quadratic relationship, with spatially
 409 variable model parameters that depend on climate, soil properties, and management practices. The original
 410 version of SRNM was calibrated using field observations only from China (Zhou et al., 2015). In this study,
 411 we used the global N₂O observation dataset to train it to create maps of gridded annual emission factors of
 412 N₂O and the associated emissions at 5-minute resolution from 1901 to 2014 (Cui et al., 2021). The gridded
 413 EF and associated direct cropland-N₂O emissions are simulated based on the following equation:

414
 415

$$416 \quad E_{ijt} = \alpha_{ij} N_{ijt}^2 + \beta_{ij} N_{ijt} + \varepsilon_{ijt}, \quad \forall i \quad (23)$$

417 where

$$418 \quad \alpha_{ij} \sim N\left(\sum_k (x_k \lambda_{ijk}), \sigma_{ijk}^2\right), \quad \beta_{ij} \sim N\left(\sum_k (x_k \phi_{ijk}), \sigma_{ijk}^{\prime 2}\right) \quad (24)$$

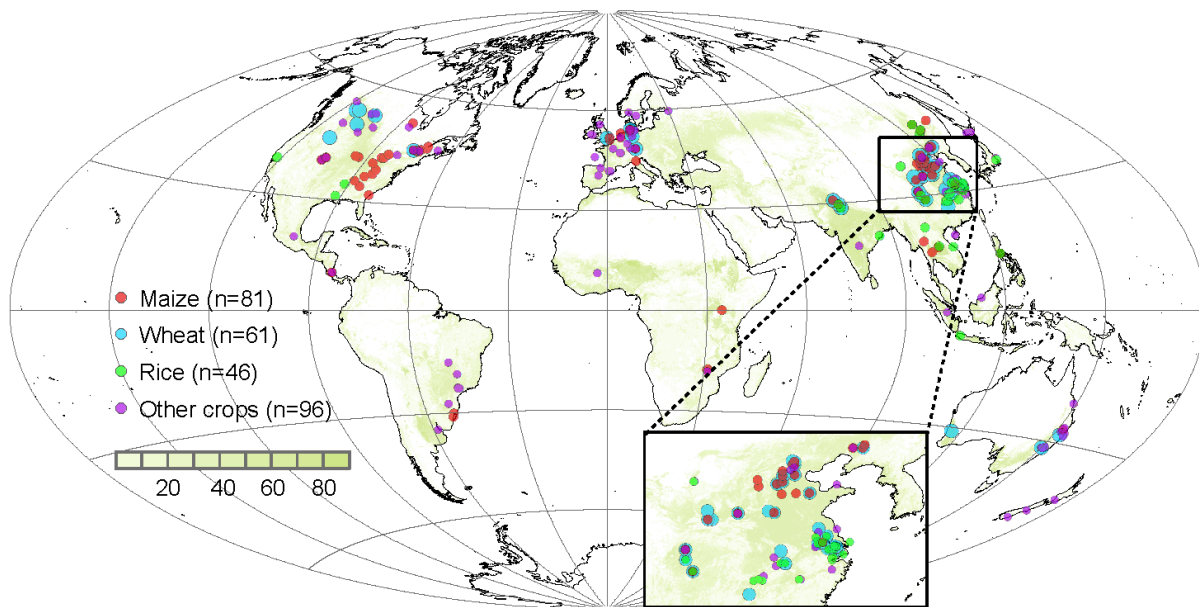
$$419 \quad \lambda_{ijk} \sim N(\mu_{ijk}, \omega_{ijk}^2), \quad \phi_{ijk} \sim N(\mu'_{ijk}, \omega_{ijk}^{\prime 2}), \quad \varepsilon_{ijt} \sim N(0, \tau^2) \quad (25)$$

420 and i denotes the sub-function of N₂O emission ($i=1, 2, \dots, I$) that applies for a sub-domain division W_i of
 421 six climate or soil factors, j represents the type of crop ($j=1-2$, 1 for upland crops and 2 for paddy rice), k is
 422 the index of climate or soil factors ($k=1-6$, i.e., soil pH, clay content, SOC, BD, the sum of cumulative
 423 precipitation and irrigation, mean daily air temperature). W_i denotes a set of the range of multiple x_k . E_{ijt}

424 denotes direct N₂O emission flux (kg N ha⁻¹ yr⁻¹) estimated for crop type j in year t in the i th sub-domain,
425 N_{ijt} is N application rate (kg N ha⁻¹ yr⁻¹), and a_{ij} and b_{ij} are defined as summation of the product of x_k and l_{ijk}
426 over k . The random terms l and f are assumed to be independent and normally distributed, representing the
427 sensitivity of a and b to x_k . e is the model error. m and $m\phi$ are the mean effect of x_k for a and b , respectively.
428 s , $s\phi$, w , $w\phi$, and t are standard deviations. Optimal sub-domain division, associated parameters mean
429 values and standard deviations were determined by using the Bayesian Recursive Regression Tree version
430 2 (BRRT v2), constrained by the extended global cropland-N₂O observation dataset. The detailed
431 methodological approach of the BRRT v2 is described by Zhou et al (2015).
432

433 **S5.2 Global cropland N₂O observation dataset**

434 We aggregated cropland N₂O flux observation data from 180 globally distributed observation sites from
435 online databases, on-going observation networks, and peer-reviewed publications (Figure S1). Chamber-
436 based observations were only included in this dataset. These data repositories are as follows: the
437 NitroEurope, CarbonEurope, GHG-Europe (EU-FP7), GRACenet, TRAGnet, NANORP, and 14 meta-
438 analysis datasets (Decock, 2014; Harris et al., 2014; Helgason et al., 2005; Hénault et al., 2005; Hickman
439 et al., 2014; Kim et al., 2013a; Kim et al., 2013b; Lehuger et al., 2011; Leppelt et al., 2014; Rochette and
440 Janzen, 2005; Sacks et al., 2010; Shcherbak et al., 2014; Stehfest and Bouwman, 2006; Walter et al., 2015).
441 Four types of data were excluded from our analysis: (i) observations without a zero-N control for
442 background N₂O emission, (ii) observations from sites that used controlled-release fertilizers or nitrification
443 inhibitors, (iii) observations not covering the entire crop growing season, (iv) observations made in
444 laboratory or greenhouse. We then calculated cropland-N₂O emissions as the difference between observed
445 N₂O emission (E) and background N₂O emission (E_0). Values of EF were estimated for each nonzero N
446 application rate (N_a) as direct cropland-N₂O emission divided by N_a : $EF = (E - E_0)/N_a$. This yielded a
447 global dataset of direct cropland-N₂O emissions, N-rate-dependent N₂O EFs and fertilization records from
448 each site (i.e., 1,052 estimates for upland crops from 152 sites and 154 estimates for paddy rice from 28
449 sites), along with site-level information on climate, soils, crop type, and relevant experimental parameters.
450 Total numbers of sites and total measurements in the dataset were more than doubled those for previous
451 datasets of N₂O EF. The extended global N₂O observation network covered most of fertilized croplands,
452 representing a wide range of environmental conditions globally. For each site in our dataset, the variables
453 included four broad categories: N₂O emissions data, climate data (cumulative precipitation and mean daily
454 air temperature), soil attributes (soil pH, clay content, SOC, BD), and management-related or experimental
455 parameters (N application rate, crop type). More details on global cropland N₂O observation dataset can be
456 found in Cui et al. (2021).
457



458
 459 **Figure S1 Global observation dataset of N₂O EF for direct soil emissions.** Green area indicates the
 460 harvested areas of all crops derived from the Earthstat. Sites are indicated in different colors for maize,
 461 wheat, rice, and other crops.
 462

463 **S5.3 Gridded input datasets:**

464 The updated SRNM model was driven by many input datasets, including climate, soil properties,
 465 agricultural management practices (e.g., fertilization, tillage, irrigation), as well as the historical distribution
 466 of cropland. Cumulative precipitation and mean daily air temperature over the growing season were
 467 acquired from the CRU TS V4.06 climate dataset (0.5-degree resolution) (Harris *et al.*, 2014), where
 468 growing season in each grid cell was identified following Sacks *et al.* (2010) The patterns of SOC, clay
 469 content, BD, and soil pH were acquired from the HWSD v1.2 ((Berdanier and Conant, 2012), 1-km
 470 resolution). Both climate and soil properties were re-gridded at 5-arc-minute spatial resolution using a first-
 471 order conservative interpolation widely used in the CMIP5 model intercomparison (Yang *et al.*, 2017). The
 472 annual cropland area at 5-arc-minute spatial resolution from 1961 to 2020 was obtained from the History
 473 Database of the Global Environment (HYDE 3.2.1) (Goldewijk *et al.*, 2017).
 474

475 For fertilization, crop-specific N fertilizer inputs (including synthetic N fertilizers, crop residues and
 476 manure), fertilizer types, and placement during 1961-2020 were obtained from Adalibieke *et al.*, (2023).
 477 The frequency (i.e., one or multiple times) of N fertilization were the same as Cui *et al.* (2021) and we
 478 assumed that the frequency remained constant during the study period. For tillage, the fraction of tillage by
 479 crop during 1961-2020 was obtained from Adalibieke *et al.*, (2023), which was constructed with the country
 480 and province (or state) level no-tillage area data during 1961-2020 and downscaled to grid with the method
 481 of Porwollik *et al.* (2019). For irrigation, the History Database of the Global Environment (HYDE version
 482 3.2) (Goldewijk *et al.*, 2017) and the MIRCA2000 dataset (Portmann *et al.*, 2010) were used to compile the
 483 global crop-specific irrigation proportion data from year 1961 to 2020. Categories of cropland in HYDE
 484 provided new distinctions with irrigated and rain-fed crops (upland crops, other than rice), irrigated and
 485 rain-fed rice during 1960-2017. The national-level dataset of “Agricultural area actually irrigated” was
 486 obtained from (FAO, 2022), which was used to scale the baseline year 2015 maps of irrigated area from
 487 HYDE over the period 2016-2020. The area of irrigated upland crops from HYDE was first disaggregated

488 into 21 crops based on the irrigated proportion from MIRCA2000 for per grid cell. We assumed an even
489 share of irrigated area by each upland crop during the period 1961-2020, like MIRCA2000. Finally, the
490 crop-specific irrigated area was masked by reporting harvested area, then the irrigated proportion of each
491 crop can be calculated as the crop-specific irrigated area divided by the physical area of each crop. For rice,
492 we further divided irrigated rice into continuously and intermittently flooded systems as provided by *Cui*
493 *et al.* (2021), and we assumed that the irrigation proportion was kept the same during the study period.
494

495 **S6 Global N flow in aquaculture**

496 We applied the IMAGE-GNM aquaculture nutrient budget model for shellfish and finfish (*Bouwman et al.*,
497 2013; *Bouwman et al.*, 2011) to calculate the nutrient flows in aquaculture production systems. These flows
498 comprise feed inputs, retention in the fish, and nutrient excretion. Individual species within crustaceans,
499 seaweed, fish and molluscs are aggregated to the International Standard Statistical Classification of Aquatic
500 Animals and Plants (ISSCAAP) groups (*FAO*, 2022), for which production characteristics are specified.
501 Feed and nutrient conversion rates are used for each ISSCAAP group to calculate the feed and nutrient
502 intake based on production data from *FAO* (*FAO*, 2020). Feed types include home-made aquafeeds and
503 commercial compound feeds with different feed conversion ratios that also vary in time due to efficiency
504 improvement; in addition, the model accounts for algae in ponds, that are often fertilized with commercial
505 fertilizers or animal manure, consumed by omnivore fish species like carp. A special case is the filter-
506 feeding bivalves that filter seston from the water column, and excrete pseudofeces, feces and dissolved
507 nutrients. Based on production data and tissue/shell nutrient contents, the model computes the nutrient
508 retention in the fish. Using apparent digestibility coefficients, the model calculates outflows in the form of
509 feces (i.e., particulate nutrients) and dissolved nutrients. Finally, nutrient deposition in pond systems and
510 recycling are calculated. For computing the N₂O emissions, we consider the amount of N released to the
511 environment, i.e., the difference between N intake and N in the harvested fish, which includes all the
512 nutrient excretion. Since in pond cultures part of that N is managed, we made the amount of N recycling
513 explicit, as well as ammonia emissions from ponds. This is to avoid double counting when computing N₂O
514 emissions from crop production.
515

516 **S7 Continental Shelves N₂O fluxes**

517 N₂O emissions from the global ocean do not include the contribution from continental shelves and are added
518 here using the extended mask of *Laruelle et al.* (2017) to delineate the coastal ocean. This mask excludes
519 estuaries and inland water bodies, while its outer limit is set 300 km away from the shoreline. Within this
520 coastal ocean domain, gridded N₂O emissions were calculated using one data-driven estimate and three
521 high-resolution model estimates from two distinct models, all interpolated on the same 0.25° x 0.25° grid.
522 Models and data are each covering different time-periods and only one climatology is provided, keeping
523 the original timespan of each product: 1988-2017 for the observation-based product that relied on a random-
524 forest (RF) algorithm to interpolate N₂O data (*Yang et al.*, 2020) from the MEMENTO database (MEM-
525 RF) (*Kock and Bange*, 2015), 1998-2018 for the estimate relying on the high-resolution configuration
526 (*Berthet et al.*, 2019) of the global ocean-biogeochemical component of CNRM-ESM2-1 (CNRM-0.25°),
527 1998-2013 and 2006-2013 for the estimates relying on the ECCO-Darwin model running at 1/3° (ECCO-
528 Darwin1) and 1/6° (ECCO-Darwin2), respectively. The resulting climatology can be considered as broadly
529 representative of the last 2-3 decades. Each product is further described as follows:
530

531 **S7.1 MEM-RF**

532 The N₂O air-sea flux reconstruction by *Yang et al.* (2020) is based on a synthesis of over 158,000
533 observations of N₂O mixing ratio, partial pressure, and concentration in the surface ocean from the
534 MEMENTO database (<https://memento.geomar.de>) (*Kock and Bange*, 2015) and additional cruises
535 (Dataset S1) (*Yang et al.*, 2020). N₂O measurements are converted to surface N₂O mixing ratio anomalies
536 using observations from the NOAA atmospheric flask dataset (*Hall et al.*, 2007), and extrapolated to a 0.25-

537 degree resolution global monthly climatology using an ensemble of 100 random forest realizations. The
538 random forest algorithm predicts N₂O mixing ratio anomalies based on their relationship to oceanographic
539 predictors that include hydrographic variables, nutrients, oxygen, chlorophyll, net primary production, and
540 seafloor depth. Reconstructed mixing ratio climatologies are used to estimate air-sea fluxes by applying a
541 commonly used gas exchange parameterization (Wanninkhof, 2014). Two formulations of piston velocity
542 are adopted: one based on a quadratic dependence on wind speed (Wanninkhof, 2014), and one that
543 explicitly accounts for bubble-mediated fluxes (Liang et al., 2013). Sea ice cover, surface temperature,
544 salinity and atmospheric pressure are taken from ERA5 reanalysis (Hersbach et al., 2017). Calculations are
545 performed with two high-resolution wind products (ERA5 and CCMP) that are available at 0.25, 6-hourly
546 resolution for the period from 1988 to 2017, yielding four permutations of the piston velocity. The resulting
547 ensemble of 400 global N₂O air-sea flux estimates is averaged in time to obtain monthly mean climatologies.
548 A description of the dataset and methods is presented in Yang et al. (2020). The code used to produce these
549 datasets is archived on a public GitHub repository at <https://github.com/yangsi7/mapping-ocean-n2o> (DOI:
550 10.5281/zenodo.3757194).

551

552 **S7.2 CNRM-0.25°**

553 N₂O fluxes have been inferred from the global ocean-biogeochemical component of CNRM-ESM2-1
554 (Séférian et al., 2019) run at 0.25° horizontal resolution with 75 vertical levels in the ocean. This high-
555 resolution configuration is described in Berthet et al. (2019) and is based on the NEMOV3.6 oceanic model
556 (Madec, 2008), the multi-category sea ice model GELATOV6 (Salas y Mélia, 2002) and the PISCESv2-gas
557 model for marine biogeochemistry (Aumont et al., 2015), which includes an updated version of (Martinez-
558 Rey et al., 2015) for the marine N₂O module. The simulation was first spun-up during 300 years under
559 preindustrial conditions and then has been forced by the OMIP2-compliant JRA55-do-1-5 atmospheric
560 reanalysis (Tsuji no et al., 2020; Tsuji no et al., 2018) considering the historical evolution of CO₂ and N₂O
561 in the atmosphere since the year 1850. Boundary conditions for nitrogen deposition and riverine inputs are
562 prescribed from monthly climatologies. The suboxic production of N₂O uses the oxygen-dependent
563 formulation of Jin and Gruber (2003) and is enhanced at low oxygen concentrations. This formulation
564 encompasses N₂O production during remineralization, nitrification and grazing, as well as a sink term
565 corresponding to N₂O consumption under anoxic conditions by denitrification. The oceanic N₂O partial
566 pressure is computed based on the surface N₂O concentration and the N₂O solubility in the ocean. Sea-to-
567 air N₂O fluxes are then computed using the standard gas exchange parameterization of Wanninkhof (1992;
568 2014).

569

570 **S7.3 ECCO-Darwin & ECCO2-Darwin**

571 For this study we generated global air-sea fluxes of nitrous oxide (N₂O) from the global ocean by using two
572 models that include the same biogeochemical component but embedded in two different ocean physical
573 settings, ECCO2-Darwin and ECCO-Darwin.

574

575 The first model, ECCO2-Darwin model, is a global physical-biogeochemical ocean model with nominal
576 horizontal grid of 1/6 of degree therefore eddy-permitting at lower latitudes.

577 The second model is ECCO-Darwin, a global physical-biogeochemical ocean model with nominal
578 horizontal grid resolution of 1/3 of degree (Carroll et al., 2020). The ECCO-Darwin model is forced with
579 an atmospheric forcing corresponding to the 1992-present optimized with adjoint technique in order to
580 realistically represent the observed physical ocean climate variability. An extensive description of this
581 model run of ECCO2-Darwin including the optimized atmospheric forcing spanning from 2004 to 2013
582 can be found in Manizza et al., (2019, 2023) while for ECCO-Darwin a more detailed model description
583 can be found in Carroll et al., (2020). Both models have 50 vertical levels and in the top 100 m the model
584 is vertically resolved with 10-meter grid boxes.

585

586 The Darwin biogeochemical/ecological model used for this study explicitly represents the cycle of carbon,
587 oxygen, phosphorus, silica, and iron in the global ocean. It also has an ecosystem component representing
588 five groups of phytoplankton and two groups of zooplankton (Manizza et al., 2019, Carroll et al., 2020).
589

590 For this version of the model, we implemented a parameterization of the oceanic cycle of N₂O using the
591 scheme of Nevison et al., (2003) based on the oceanic oxygen cycle previously represented in ECCO2-
592 Darwin model (Ganesan et al., 2020). The air-sea gas flux of N₂O was parameterized according to
593 Wanninkhof (1992).
594

595 In the ECCO2-Darwin simulation the 2004-2005 period was discarded, and we used the 2006-2013 period
596 only for our analysis. However, the ECCO-Darwin numerical simulation was run for the 1992-2014 period,
597 but we discarded the inclusion of the output relative to the 1992-1996 period in our analysis due to the
598 model adjustment in this initial part of our numerical simulation. The results of these simulations were also
599 used in the study of Resplandy et al. (2023).
600

601 **S8 Open Ocean N₂O fluxes**

602 N₂O is produced in the open ocean by microbial activity during organic matter cycling in the subsurface
603 ocean, and its production pathways are influenced by the local environmental oxygen level. In the oxic
604 ocean N₂O is produced as a byproduct during the oxidation of ammonia to nitrate, mediated by ammonia
605 oxidizing bacteria and archaea. N₂O is also produced and consumed in sub-oxic and anoxic waters through
606 the action of marine denitrifiers during the multi-step reduction of nitrate to gaseous N. The oceanic N₂O
607 distribution therefore displays significant heterogeneity with background levels of 10-20 nmol/l in the well-
608 oxygenated ocean basins, high concentrations (> 40 nmol/l) in hypoxic waters, and N₂O depletion in the
609 core of ocean oxygen minimum zones (OMZs).
610

611 For this synthesis open ocean N₂O emissions to the atmosphere were compiled from four global ocean
612 biogeochemistry models/Earth System models that simulate the production, consumption and circulation
613 of oceanic N₂O (Table 6). N₂O flux exchange between ocean and atmosphere is derived using gas-exchange
614 parameterizations applied to modeled surface ocean N₂O. Versions of the four submitting models also
615 participated in the previous N₂O budget synthesis (Tian et al., 2020a). Model details and updates to the
616 previous N₂O budget synthesis are summarized below.

617 The models differ in aspects of physical configuration (e.g., spatial resolution), meteorological forcing
618 applied at the ocean surface, and in their parameterizations of ocean biogeochemistry; specific details on
619 individual models are provided in the publications listed in Table 1. Towards this N₂O budget synthesis,
620 modelling groups reported grid-resolved (1°×1° horizontal resolution) monthly estimates of ocean-
621 atmosphere N₂O fluxes for the period 1980-2020 (or for as many years as possible in that period).
622

623 **S8.1 U. Bern: Bern-3D**

624 N₂O fluxes are derived from the Bern-3D Earth System Model of Intermediate Complexity which includes
625 a prognostic marine biogeochemistry model (based on (Parekh et al., 2008) and (Tschumi et al., 2011)).
626 Configuration of the model for simulation of N₂O is outlined in Battaglia and Joos (2018). Model
627 simulations were run at a native resolution of horizontal resolution of 41 by 40 grid cells and 32
628 logarithmically scaled vertical layers. Modifications of the biogeochemistry model relevant for the N₂O
629 cycle include the assignment of organic matter remineralization to aerobic and anaerobic pathways
630 dependent on mean grid-cell dissolved oxygen level. N₂O is produced by nitrification as a product of
631 remineralization rate and a specified N₂O yield which has a functional form dependent on oxygen level (see
632 details in (Battaglia and Joos, 2018)). N₂O consumption by denitrification processes is represented by a
633 first-order kinetics formulation which also includes a dependence on local oxygen level to account for the
634 relative importance of denitrification-related N₂O production and consumption processes in each gridcell.
635 Measurements of dissolved N₂O (surface and interior) from the MEMENTO database (Kock and Bange,

636 2015) together with an ensemble of model runs are used to constrain the model parameters governing N₂O
637 production and consumption mechanisms. From a pre-industrial equilibrium state the model is forced by
638 historical CO₂ emissions, non-CO₂ radiative forcing, and land-use changes. N₂O in the atmosphere is
639 prescribed based on historical data.

640
641

S8.2 CNRM: CNRM-ESM2-1

642 N₂O fluxes are provided by the CNRM-ESM2-1 Earth System model. The ocean model component is based
643 on NEMO Version 3.6 (*Madec et al.*, 2017) and coupled to the GELATO sea ice model (*Salas y Mélia*,
644 2002) Version 6 and the marine biogeochemical model PISCESv2-gas (*Aumont et al.*, 2015). The spatial
645 model resolution follows the eORCA1L75 grid, with a nominal horizontal resolution of 1° and with higher
646 resolution in the tropics (increasing to $\sim(1/3)^\circ$). The model has 75 vertical levels with higher resolution
647 towards the ocean surface. The simulations were forced at the surface by the atmospheric state of JRA55-
648 do v1.5.0 (*Tsujino et al.*, 2018). Atmospheric N₂O concentration is given as annual means as specified by
649 CMIP6 protocols and is linearly interpolated in time. Parameterization of the marine N₂O cycle follows that
650 of *Martinez-Rey et al.* (2015) with some modifications. N₂O production is driven by an oxygen-dependent
651 yield of N₂O, which encompasses production from denitrification and nitrification processes. This
652 formulation also assumes a constant background yield representing N₂O production by nitrification and a
653 consumption of N₂O in suboxic conditions. Originally implemented by *Martinez-Rey et al.* (2015), the
654 marine N₂O parameterization has benefited from a recoding and an improved calibration presented in
655 *Berthet et al.* (2023). Further details of the model biogeochemistry and configuration are provided by
656 *Séférian et al.* (2019) and *Berthet et al.* (2019).

657
658

S8.3 UVic2.9

659 N₂O model fluxes are derived from the UVic2.9, Earth System Model of Intermediate Complexity with
660 prescribed monthly climatological winds (*Kalnay et al.*, 1996) and ice sheets (*Peltier*, 2004), configuration
661 outlined in *Landolfi et al.* (2017). Oceanic subsurface N₂O production is parameterized following (*Zamora*
662 *and Oschlies*, 2014), as a function of O₂ consumption with a linear O₂ dependency, inherently including
663 both nitrification and denitrification. In O₂-deficient waters ($<4 \text{ mmol m}^{-3}$), denitrification becomes a sink
664 of N₂O consumed at a constant rate. The gradient driving the air-sea N₂O gas exchange, is computed online
665 based on departure of the surface ocean concentration from the saturation value using the solubility
666 coefficients of *Weiss and Price* (1980) and time-varying prescribed atmospheric N₂O concentrations
667 (historical and RCP8.5). The model was spun-up for 6000 years with preindustrial boundary conditions
668 (solar and volcanic and aerosol forcing, fixed atmospheric CO₂ of 280 ppm and N₂O of 276 ppb, and
669 preindustrial atmospheric N deposition).

670

S8.4 UEA: NEMO-PlankTOM10.2

672 N₂O model fluxes are derived from the NEMO-PlankTOM10.2 ocean model. The physical circulation
673 component is NEMO v3.1 (*Madec*, 2008), with horizontal resolution of 2° longitude, and a variable
674 latitudinal resolution (average $\sim 1^\circ$) with higher resolution in the tropics and polar regions. The model has
675 30 vertical layers, with variable resolution ranging from 10m in the upper 100m to 500m at depths of 5000
676 m. The biogeochemical component relies on the marine ecosystem model PlankTOM10, which includes
677 representation of 10 plankton functional types (*Le Quéré et al.*, 2016). It has been extended by *Buitenhuis*
678 *et al.* (2018) to include nitrogen cycle processes, and prognostic and diagnostic models of N₂O production.
679 N₂O is produced from nitrification and denitrification pathways, with oxygen dependent yields employed
680 to account for varying production and consumption processes in hypoxic waters. Nitrogen cycle parameters
681 are optimized using ocean databases of dissolved N₂O (MEMENTO, *Kock and Bange* (2015)) nitrification
682 rates (*Yool et al.*, 2007), and surface ammonium concentrations (*Johnson et al.*, 2015; *Paulot et al.*, 2015).
683 Further details on model configuration are provided in (*Buitenhuis et al.*, 2018).

684

685 **S9 Net N₂O emission from land cover change**

686 This section mainly involves the calculation of post-deforestation N₂O emissions, deforestation induced
687 N₂O reduction and N₂O emissions from forest regrowth (afforestation or reforestation). The methods
688 include both bookkeeping and process-based modeling.

689

690 **S9.1 Deforestation area, crop/pasture expansion and secondary forests**

691 The LUH2 v2h (land use harmonization, <http://luh.umd.edu>) land use data was used to derive the
692 deforestation area and its partition between crops and pastures during 1860–2020. LUH2 categorizes forest
693 lands into forested primary land and potentially forested secondary land, while croplands are divided into
694 C3 annual crops, C3 perennial crops, C4 annual crops, C4 perennial crops, and C3 N-fixing crops.

695

696 In the empirical computation of deforestation induced N₂O emissions, all sub-classes within each land use
697 type were treated the same. Thus, only the annual transition area from forests to croplands or managed
698 pasture was needed. In the process-based estimates, the DLEM model was improved to further account for
699 the classifications of primary forests, secondary forests (further partitioned into established and newly
700 converted by an age threshold of 15 years), croplands/pastures /rangelands (further partitioned into
701 established and newly converted by an age threshold of seven nine years). Each land use type can be divided
702 into several different plant functional types (PFTs). Specifically, within a grid cell, cropland can only be
703 dominated by only one crop type. The pastures and rangelands can be either C3 type or C4 type. To generate
704 the historical spatial distribution of PFTs, a potential vegetation map and the accompanied composition
705 ratio map of each natural PFT acquired from the Synergetic Land Cover Product (SYNMAP) were jointly
706 used with LUH2 v2h.

707

708 **S9.2 Methods**

709 The bookkeeping method was mainly applied to the tropical areas, where forests generally have high N₂O
710 emissions. Specifically, the average tropical forest N₂O emission rate of 1.974 kg N₂O-N ha⁻¹ yr⁻¹ was
711 adopted as the baseline. Two logarithmic response curves of soil N₂O emissions (normalized to the baseline)
712 after deforestation were developed: $y = -0.31 \ln(x) + 1.53$ and $y = -0.454 \ln(x) + 2.21$. This form of
713 the response functions can effectively reproduce the short-lived increase in soil N₂O emissions after initial
714 forest clearing and the gradually declining emission rates of converted crops/pastures (*Melillo et al.*, 2001;
715 *Verchot et al.*, 1999). Using these two curves and the baseline, we kept track of the N₂O reduction of tropical
716 forests and the post-deforestation crop/pasture N₂O emissions at an annual timescale.

717

718 There are not many studies on N₂O emissions from secondary tropical forests that regrowth after crop or
719 pasture abandonment. *Sullivan et al.* (2019) lumped together all forms of N "gas loss" including NO and
720 N₂O for secondary forests across the tropics and the results showed gas loss gradually increases with age
721 since the regrowth of secondary forest. *Keller and Reiners* (1994) also reported a gradual recovery of soil
722 nitrate and soil emissions of N₂O and nitric oxide (NO) during 20 years of secondary forest succession,
723 which however did not return to the level of the primary forests. In this study, using field observations from
724 *Davidson et al.* (2007) and *Keller and Reiners* (1994), we regressed normalized N₂O emissions (relative to
725 a reference mature forest) of secondary tropical forests with their ages as $y = 0.0084x + 0.2401$ ($R^2 = 0.44$;
726 unit of x is year). The derived y values were multiplied by tropical forest N₂O emissions estimated by
727 NMIP2 models that do not distinguish secondary forests from primary forests.

728

729 The DLEM model was run with varying climate and CO₂ with other factors held constant to estimate forest
730 baseline emissions and unfertilized crop/pasture emissions from 1860-2020. The climate data were acquired
731 from CRUJRA, which is a fusion of the CRU and JRA reanalysis products at a spatial resolution of 0.5° ×
732 0.5° and a daily time-step. The atmospheric CO₂ data were obtained from NOAA GLOBLVIEW-CO2
733 dataset (<https://www.esrl.noaa.gov>), which are derived from atmospheric and ice core measurements. In

734 the tropical area, both estimates from the DLEM model and the bookkeeping method were adopted, whereas
735 in extra-tropical area, we only adopted the DLEM outputs.

736

737 **S10 Inland water, estuaries, and coastal vegetation**

738

739 **S10.1 Dynamic Land Ecosystem Model-Terrestrial/Aquatic Continuum (DLEM-TAC)**

740 To quantify N₂O emissions from global inland waters (rivers, lakes, and reservoirs), we use a process-based
741 coupled terrestrial-aquatic model, which builds up on the Dynamic Land Ecosystem Model (DLEM).
742 DLEM-TAC is a fully distributed, process-based land surface model which couples the major land
743 processes (terrestrial hydrology, plant phenology and physiology, soil biogeochemistry) and aquatic
744 dynamics (lateral transport and in-stream biogeochemistry) (*Pan et al.*, 2021; *Tian et al.*, 2015; *Tian et al.*,
745 2020b; *Yao et al.*, 2020). The land component of DLEM-TAC explicitly simulates the carbon, nitrogen,
746 and water fluxes between plants, soil, and atmosphere, and the surface and drainage runoff and nitrogen
747 load from the land module are used as input for the aquatic module. The simulated nitrogen load
748 includes dissolved inorganic nitrogen (DIN), dissolved organic nitrogen (DON), particulate organic
749 nitrogen (PON), and runoffs, sewers as the initial condition of the aquatic module.

750

751 DLEM-TAC aquatic module calculated lateral water transport and the associated aquatic biogeochemical
752 processes by adopting a scale-adaptive scheme. The water transport scheme, which coupled hillslope flow,
753 subnetwork flow, and main channel flow, simulated the water transport processes within grid cells. In the
754 aquatic module, lakes and reservoirs were linked with small streams and large rivers, forming a river-lake-
755 reservoir corridor (*Wollheim et al.*, 2008)). Particularly, lakes that are linked to small streams are typically
756 small in size and are defined as small lakes, while those linked to large rivers are usually had large size and
757 are defined as large lakes; similarly, reservoirs that are linked to main channels are considered as large
758 reservoirs, while those that are linked to small streams are considered as small reservoirs. The incoming
759 flow of a linked river-lake-reservoir corridor drains to lakes first, and the outflow rate of lakes and reservoirs
760 is determined based on the predefined residence time obtained from the global lake dataset (*Lehner et al.*,
761 2011; *Messenger et al.*, 2016; *Yao et al.*, 2022). The aquatic N module was developed based on the scale
762 adaptive water transport scheme, including lateral transport, decomposition of organic matter, particle
763 organic matter deposition, nitrification, and denitrification. The detailed description could be found in the
764 previous studies (*Pan et al.*, 2021; *Tian et al.*, 2020b; *Yao et al.*, 2020).

765

766 Following our previous work referring to the development of water transport and biogeochemistry, we
767 developed an inland water N₂O module within the aquatic biogeochemical component of the DLEM
768 framework (*Yao et al.*, 2020). The net fluxes of dissolved N₂O (including physical and biogeochemical
769 processes) in the main channel (high-order streams) and subnetwork (small rivers) are estimated as:

770

$$(ΔM_{N_2O}) / Δt = Fa + Y_{water} + D - R - E \quad (26)$$

771 where M_{N₂O} is the total mass of dissolved N₂O in the main channel or subnetworks (g N), Δt is the time
772 step, Fa is advective N₂O fluxes (g N d⁻¹), Y_{water} is the N₂O production within the water column (g N d⁻¹),
773 D is the dissolved N₂O from rainfall to rivers (i.e. deposition) (g N d⁻¹) with an initial concentration equal
774 to the atmospheric equilibrium N₂O concentration, R is the flux from N₂O reduction (g N d⁻¹) to nitrogen
775 gas, and E is the riverine N₂O efflux (g N d⁻¹) through the air-water interface.

776

777 Input data. The driving data of DLEM-TAC include the climate variables, atmospheric CO₂ concentration,
778 land use change, nitrogen (N) deposition, N fertilizer, and manure application with a spatial resolution of
779 0.5°× 0.5°. The daily climate variables (precipitation, mean temperature, maximum temperature, minimum
780 temperature, and shortwave radiation) were obtained from the CRUNCEP dataset (<https://vesg.ipsl.upmc.fr>)

781 for 1901-2019. Climate data of each year during 1850-1900 was randomly sampled from 1901-1920.
782 Annual atmospheric CO₂ concentration from 1900-2019 was obtained from the NOAA GLOBALVIEW-
783 CO₂ dataset (<https://www.esrl.noaa.gov>). The annual land use change data was derived from a potential
784 natural vegetation map (synergetic land cover product) and a prescribed cropland area dataset from the
785 history database of the global environment v.3.2 (HYDE 3.2, <ftp://ftp.pbl.nl/hyde>). The data of N fertilizer,
786 manure N application, and N deposition data was obtained from (*Tian et al.*, 2022).
787

788 In the aquatic module, the required channel dataset included channel slope, channel width, and channel
789 length generated from the Hydrosched dataset (*Lehner et al.*, 2008) and DDM30 dataset (*Döll and Lehner*,
790 2002). The flow direction and distance data were obtained from the Dominant River Tracing (DRT) dataset.
791 For modeling water dynamics in lakes and reservoirs, we generated 0.5 grid level surface water area,
792 upstream area, volume, depth, and average residence time for lakes based on the Hydrolakes dataset
793 (*Messenger et al.*, 2016), while the GRand database provided the same information for reservoirs (*Lehner*
794 *et al.*, 2011).
795

796 Simulation protocol. DLEM-TAC simulations include three steps: equilibrium run, spin-up run and two
797 transit runs, one with dam operation close, and another one with dam operation open. First, the equilibrium
798 run is required to obtain the initial and steady condition of carbon, nitrogen, and water pool at the pre-
799 industrial level in each grid cell (*Thornton and Rosenbloom*, 2005). In this step, we held all the driving
800 forces such as climate data, atmospheric CO₂ concentration, land use data, and nitrogen additions consistent
801 with the first year's data we used in the simulation. Second, we conducted a 30-year spin-up run by
802 randomly selecting climate data within the 1850s (*Tian et al.*, 2012a). This step can alleviate the disturbance
803 of driving data changes in the transit run. Then we conduct the natural flow simulation with the dam model
804 temporarily closed, and all the driving forces change over time. After the natural flow simulation, we set
805 up a management flow simulation with the dam module open, specifically the dam module needs natural
806 flow in the previous run as model input.
807

808 **S10.2 Mechanistic Stochastic Modeling of N₂O emissions from large rivers, lakes, reservoirs, and** 809 **estuaries:**

810
811 To calculate the cascading loads of TN and TP delivered to each water body along the river-reservoir-
812 estuary continuum, we spatially routed all reservoirs from the GRand database (*Lehner et al.*, 2011), with
813 river networks from Hydroscheds 15s (*Lehner et al.*, 2008) and, at latitudes above 50°N, Hydro1K
814 (<http://edc.usgs.gov/products/elevation/gtopo30/hydro/>), which were in turn connected to estuaries as
815 represented in the “Worldwide Typology of Nearshore Coastal Systems” of *Dürr et al.* (2011). In addition,
816 the global database HydroLAKES (*Messenger et al.*, 2016) was used to topologically connect 1.4 million
817 lakes with a minimum surface area of 0.1 km² within the river network. Note that besides natural lakes,
818 HydroLAKES includes updated information on 6,796 reservoirs from the GRand database, which was used
819 in the study of *Maavara et al.* (2019). In order to estimate the TN and TP loads to each water body, we then
820 relied on a spatially explicit representation of TN and TP mobilization from the watershed into the river
821 network (see (*Maavara et al.*, 2019) for details (*Bouwman et al.*, 2009; *Van Drecht et al.*, 2009)).
822

823 For the estimation of N₂O emission, we applied two distinct model configurations, respectively named DS1
824 and DS2 in *Maavara et al.* (2019). DS1 estimates N₂O emissions from denitrification and nitrification based
825 on an EF of 0.9%, which is in the mean of published values (*Beaulieu et al.*, 2011), and the assumption that
826 N₂O production equals N₂O emissions (*Maavara et al.*, 2019). For DS2, the reduction of N₂O to N₂ during
827 denitrification if N₂O is not evading sufficiently rapidly from the water body is considered. The fluxes in
828 the model represent lumped sediment-water column rates and were resolved at the annual timescale. The
829 use of water residence time as an independent variable in both the mechanistic model and the upscaling
830 process introduces an important kinetic refinement to existing global N₂O emission estimates. Rather than

831 applying an average EF (directly scaling N₂O emissions to N inputs) to all water bodies, the use of water
832 residence time explicitly adjusts for the extent of N₂O production and emission that is kinetically possible
833 within the timeframe available in a given water body. Simulated N₂O emission rates were evaluated against
834 UNFCCC measurement-based upscaling methods applied to reservoirs (*Deemer et al.*, 2016) and rivers
835 (*Hu et al.*, 2016) as well as a UNFCCC observation-driven regional estimate of lake N₂O emissions based
836 on literature data (*Lauerwald et al.*, 2019).

837

838 **S10.3 Meta analysis-based N₂O emissions from large rivers**

839 Data from 70 published studies between 1998 and 2016 that provided N₂O emission from streams and rivers
840 were compiled by *Hu et al.* (2016). The N₂O emission factors (EF = N₂O /DIN) and emission rates (ER =
841 EF * DIN load, kg N₂O-N yr⁻¹) were calculated for each studied river. Exploratory multiple regression
842 analyses were conducted to predict EF and ER using various combinations of factors (N concentrations,
843 loads, yields, DOC: DIN, discharge, and watershed area) and different functions. Among them, DIN yield
844 (kg N yr⁻¹ km⁻²) was identified as the best predictor of EF and ER. Using the optimal model and DIN loads
845 for 6400 global rivers calculated by the NEWS2-DIN-S model (*McCrackin et al.*, 2014), we estimated
846 global riverine N₂O emissions (*Hu et al.*, 2016).

847

848 **S10.4 Stream and river N₂O emissions combining machine-learning and model-based upscaling**

849 *Marzadri et al.* (2021) developed a novel approach that combines a data-driven Random Forest Machine
850 Learning (RM-ML) model with a physically-based upscaling model to predict near global (neglecting
851 Arctic and Antarctic areas) riverine N₂O emissions flux (F*N₂O given by the ratio between the flux of N₂O,
852 FN₂O, and the in-stream flux of dissolved inorganic nitrogen species FDIN) from both surface (i.e. water
853 column) and subsurface (i.e. benthic zone and hyporheic zone) riverine environments. In particular, to
854 capture the local scale processes responsible for N₂O emissions and to provide estimations at different
855 spatial scales (from local reach up to the global scale) the model compute two different denitrification
856 Damköhler numbers (given by the ratio between a characteristics time of transport and a characteristics
857 time of denitrification (*Marzadri et al.*, 2021; *Marzadri et al.*, 2017)) starting from the hydro-morphological
858 and biogeochemical information provided by the RM-ML model. Model results at the local reach scale
859 shows that nearly 50% of the riverine N₂O emissions comes from small streams (i.e. widths lower than 10
860 m, although they represent only the 13% of the total riverine surface area worldwide) while at the large
861 scale predict a near-global annual riverine N₂O emissions around 72.8 GgN₂O – N/yr.

862

863 **S10.5 Meta-analysis based N₂O emissions from estuaries and coastal vegetation**

864 N₂O emissions from estuaries and coastal vegetated ecosystems were obtained from a meta-analysis of
865 observed N₂O fluxes (*Rosentreter et al.*, 2023). In brief, the meta-data analysis relies on a categorization of
866 estuaries into ‘tidal systems and deltas’, ‘lagoons’, and ‘fjords’. Water-air N₂O fluxes from 123 estuary
867 sites globally were then compiled from peer-reviewed publications until the end of 2020. Coastal vegetation
868 comprises ‘mangrove’, ‘salt marsh’, and ‘seagrass’ ecosystems and N₂O sediment-water-air fluxes were
869 compiled from 55 sites globally from peer-reviewed publications until the end of 2020. A non-parametric
870 bootstrapping method (1000 iterations of the median of samples) was used to resample N₂O fluxes per unit
871 area for each estuary and coastal vegetation type in each of the 18 regions using the ‘boot’ function in the
872 package ‘boot’ in R software. Results from the bootstrapping output (minimum, Q1, median, mean, Q3,
873 maximum) were then scaled to the surface area of each estuary and coastal vegetation type in each of the
874 18 regions. If an ecosystem type had less than three sites in a region, we applied the global statistics of this
875 type in this region. Note that the meta-data analysis only provides flux assessments at the scale of the 18
876 regions.

877

878 **S11 Atmospheric inversion models**

879 Emissions were estimated using four independent atmospheric inversion frameworks (see Table 1). The
 880 frameworks all used a Bayesian inversion method. The approach used here finds the maximum posteriori
 881 (MAP), or optimal, estimate of emissions, that is, those, which when coupled to a model of atmospheric
 882 transport, provide the best agreement to observed N₂O mixing ratios while being guided by their prior
 883 probability. In this particular case, where both the likelihood and prior probability are assumed to be
 884 distributed normally, the optimal emissions are equivalent to those that minimize the cost function,
 885

$$886 \quad J(\mathbf{x}) = \frac{1}{2}(\mathbf{x} - \mathbf{x}_b)^T \mathbf{B}^{-1}(\mathbf{x} - \mathbf{x}_b) + \frac{1}{2}(\mathbf{y} - H(\mathbf{x}))^T \mathbf{R}^{-1}(\mathbf{y} - H(\mathbf{x})) \quad (27)$$

887
 888 where \mathbf{x} and \mathbf{x}_b are, respectively, vectors of the multivariate means of the posterior and prior emission
 889 distributions, \mathbf{B} is the prior error covariance matrix for emissions, \mathbf{y} is a vector of observed N₂O mixing
 890 ratios, \mathbf{R} is the observation error covariance matrix, and $H(\mathbf{x})$ is the model of atmospheric transport (for
 891 details on the inversion method see (Tarantola, 2005)). The optimal emissions, \mathbf{x} , were found by solving
 892 the first order derivative of equation (21):
 893

$$894 \quad J'(\mathbf{x}) = \mathbf{B}^{-1}(\mathbf{x} - \mathbf{x}_b) + (H'(\mathbf{x}))^T \mathbf{R}^{-1}(\mathbf{y} - H(\mathbf{x})) = 0 \quad (28)$$

895
 896 where $(H'(\mathbf{x}))^T$ is the sensitivity of the atmospheric observations to emissions, derived from an adjoint
 897 model of transport. In frameworks INVICAT, PyVAR-CAMS and GEOS-Chem, equation (5b) was solved
 898 using a variational approach (Thompson et al., 2014; Wells et al., 2015; Wilson et al., 2014), which uses a
 899 descent algorithm and computations involving the forward and adjoint models. In framework MIROC4-
 900 ACTM (Patra et al., 2018), equation (22) was solved directly by computing a transport operator, \mathbf{H} from
 901 integrations of the forward model, such that $\mathbf{H}\mathbf{x}$ is equivalent to $H(\mathbf{x})$, and taking the transpose of \mathbf{H} (Patra
 902 et al., 2022).
 903

904 Each of the inversion frameworks used a different model of atmospheric transport with different horizontal
 905 and vertical resolutions (see Table 1). The transport models TOMCAT and LMDz5, used in INVICAT and
 906 PyVAR-CAMS respectively, were driven by ECMWF ERA-5 and ERA-Interim wind fields respectively,
 907 MIROC4-ACTM was driven by JRA-55 wind fields, and GEOS-Chem was driven by MERRA-2 wind
 908 fields. While INVICAT, PyVAR-CAMS, and GEOS-Chem optimized the emissions at the spatial
 909 resolution of the transport model, MIROC4-ACTM optimized the error in the emissions aggregated into 84
 910 land and ocean regions. All frameworks optimized the emissions with monthly temporal resolution. The
 911 transport models included an online calculation of the loss of N₂O in the stratosphere due to photolysis and
 912 oxidation by O(¹D) resulting in mean atmospheric lifetimes of between 118 and 129 years, broadly
 913 consistent with recent independent estimates of the lifetime of 116±9 yr (Prather et al., 2015)).
 914

915 All inversions used N₂O measurements of discrete air samples from the National Oceanic and Atmospheric
 916 Administration Carbon Cycle Cooperative Global Air Sampling Network (NOAA). In addition, discrete
 917 measurements from the Commonwealth Scientific and Industrial Research Organisation network (CSIRO)
 918 as well as in-situ measurements from the Advanced Global Atmospheric Gases Experiment network
 919 (AGAGE), the NOAA CATS network, and from individual sites operated by University of Edinburgh (UE),
 920 National Institute for Environmental Studies (NIES), the Finnish Meteorological Institute (FMI) and the
 921 Japan Meteorological Agency (JMA) were included in INVICAT, PyVAR-CAMS and GEOS-Chem.
 922 Measurements from networks other than NOAA were corrected to the NOAA calibration scale, NOAA-
 923 2006A, using the results of the WMO Round Robin inter-comparison experiment
 924 (<https://www.esrl.noaa.gov/gmd/ccgg/wmorr/>), where available. For AGAGE and CSIRO, which did not
 925 participate in the WMO Round Robins, the data at sites where NOAA discrete samples are also collected
 926 were used to calculate a linear regression with NOAA data, which was applied to adjust the data to the

927 NOAA-2006A scale. For the remaining CSIRO sites where there were no NOAA discrete samples, the
928 mean regression coefficient and offset from all other CSIRO sites were used. The inversions used the
929 discrete sample measurements without averaging, and hourly or daily means of the in-situ measurements,
930 depending on the particular inversion framework.

931
932 Each framework applied its own method for calculating the observation space uncertainty, the square of
933 which gives the diagonal elements of the observation error covariance matrix R . The observation space
934 uncertainty accounts for measurement and model representation errors and is equal to the quadratic sum of
935 these terms. Typical values for the observation space uncertainty were between 0.3 and 0.5 ppb for all
936 inversion frameworks.

937
938 Prior mean emissions were based on estimates from terrestrial biosphere and ocean biogeochemistry models
939 as well as from inventories. INVICAT, PyVAR-CAMS and GEOS-Chem used the same prior estimates for
940 emissions from natural and agricultural soils from the model OCN v1.1 (Zaehle *et al.*, 2011) and for biomass
941 burning emissions from GFEDv4.1s. For non-soil anthropogenic emissions (namely those from energy,
942 industry and waste sectors), INVICAT, PyVAR-CAMS, and GEOS-Chem used EDGAR v5. MIROC4-
943 ACTM used the VISIT model (Inatomi *et al.*, 2010; Ito *et al.*, 2018) for emissions from natural soils and
944 EDGAR 4.2 for all anthropogenic emissions, including agricultural waste burning, but did not explicitly
945 include a prior estimate for wildfire emissions.

946
947 For the prior mean estimate of ocean fluxes, INVICAT, PyVAR-CAMS and GEOS-Chem used the
948 prognostic version of the PlankTOM-v10.2 model (Buitenhuis *et al.*, 2018) with a global total source 2.5
949 TgN yr⁻¹. Prior uncertainties were estimated in all the inversion frameworks for each grid cell (INVICAT,
950 PyVAR-CAMS and GEOS-Chem) or for each region (MIROC4-ACTM) and the square of these
951 uncertainties formed the diagonal elements of the prior error covariance matrix B . INVICAT, PyVAR-
952 CAMS and GEOS-Chem estimated the uncertainty as proportional to the prior value in each grid cell, but
953 MIROC4-ACTM set the uncertainty uniformly for land regions at 1 Tg N yr⁻¹ and for ocean regions at 0.5
954 Tg N yr⁻¹. INVICAT also included off-diagonal covariances in B corresponding to a spatial correlation
955 between flux uncertainties of 500 km and assumed a semi-exponential distribution of uncertainties so as to
956 restrict the possibility of negative fluxes.

957

958

959 **S12 Atmospheric N₂O Observation Networks**

960

961 **S12.1 The NOAA Network:**

962 For atmospheric N₂O observations from the NOAA network (Dutton *et al.* 2023), we used global mean
963 mixing ratios from the NOAA Global Monitoring Laboratory (GML) (combined dataset based on
964 measurements from five different measurement programs [HATS old flask instrument, HATS current flask
965 instrument (OTTO), the Carbon Cycle and Greenhouse Gases (CCGG) group Cooperative Global Air
966 Sampling Network (<https://www.esrl.noaa.gov/gmd/ccgg/flask.php>), HATS in situ (RITS program), and
967 HATS in situ (CATS program)]. CCGG provides uncertainties with each measurement (see site files:
968 ftp://aftp.cmdl.noaa.gov/data/greenhouse_gases/n2o/flask/surface/). The CCGG measurements for N₂O
969 analysis from more than 50 sites globally was changed to tunable infrared laser direct absorption
970 spectroscopy (TILDAS) in mid-2019 from gas chromatography. About 40 sites of them (mostly Marine
971 Boundary Layer sites) are used to calculate CCGG monthly mean global N₂O levels. Monthly mean
972 observations from different NOAA measurement programs are statistically combined to create a long-term
973 NOAA/ESRL GML dataset. Uncertainties (1 sigma) associated with monthly estimates of global mean
974 N₂O, are ~1 ppb from 1977–1987, 0.6 ppb from 1988–1994, 0.3–0.4 ppb from 1995–2000, and 0.1 ppb
975 from 2001–2017. NOAA data are generally more consistent after 1995, with standard deviations on the
976 monthly mean mixing ratios at individual sites of ~0.5 ppb from 1995–1998, and 0.1–0.4 ppb after 1998.

977 A detailed description of these measurement programs and the method to combine them are available via
 978 <https://www.esrl.noaa.gov/gmd/hats/combined/N2O.html>.

979
 980 **S12.2 The AGAGE network:**
 981 The Advanced Global Atmospheric Gases Experiment (AGAGE) global network (and its predecessors ALE
 982 and GAGE) (Prinn et al., 2018) has made continuous high-frequency gas chromatographic (GC)
 983 measurements with electronic capture detection (ECD) of N₂O at five globally distributed sites since 1978.
 984 Improved GC/ECD methods have been deployed over time resulting in N₂O measurement precision of
 985 about 0.35% in ALE, 0.13% in GAGE (Prinn et al., 1990) and 0.05% in AGAGE (Prinn et al., 2008; 2018).
 986 We used the global mean of AGAGE N₂O measurements during 1980–2020 which are reported on the
 987 Scripps Institution of Oceanography SIO-16 scale. Further information on AGAGE stations, instruments,
 988 calibration, uncertainties and access to data is available at the AGAGE Data website:
 989 <https://www.osti.gov/dataexplorer/biblio/dataset/1841748>.

990
 991 **S12.3 The CSIRO network:**
 992 The CSIRO flask network (Francey et al., 2003) consists of nine sampling sites distributed globally and has
 993 been in operation since 1992. Flask samples are collected approximately every two weeks and shipped back
 994 to CSIRO GASLAB for analysis. Samples were analyzed by gas chromatography with electron capture
 995 detection (GC-ECD). One Shimadzu gas chromatograph labelled “Shimadzu-1” (S1) has been used over
 996 the entire length of the record and the measurement precision for N₂O from this instrument is about 0.1%.
 997 N₂O data from the CSIRO global flask network are reported on the NOAA-2006A N₂O scale and are
 998 archived at the World Data Centre for Greenhouse Gases (WDCGG: <https://gaw.kishou.go.jp/>). Nine sites
 999 from the CSIRO network were used to calculate the annual global N₂O mole fractions. Smooth curve fits
 1000 to the N₂O data from each of these sites were calculated using the technique outlined in Thoning et al.
 1001 (1989), using a short-term cut-off of 80 days. The smooth curve fit data were then placed on an evenly
 1002 spaced latitude (5 degree) versus time (weekly) grid using the Kriging interpolation technique. Finally, the
 1003 gridded data were used to calculate the global annual values.

1004
 1005 **Table S3 Factors used to convert N₂O in various units (by convention Unit 1=Unit 2 × conversion)**

Unit 1	Unit 2	Conversion
Tg N ₂ O (teragrams of N ₂ O)	Tg N (teragrams of nitrogen)	1.57
Tg N (teragrams of nitrogen)	g N (grams of nitrogen)	10 ⁻¹²
Tg N (teragrams of nitrogen)	ppb (parts per billion)	4.79

1006
 1007 **Table S4 Atmospheric N₂O dry mole fraction measured by different observing networks during 2000-**
 1008 **2022.**

ppb	NOAA	AGAGE	CSIRO	Min	Max
2000	315.58	316.18	315.48	315.48	316.18
2001	316.33	316.95	316.12	316.12	316.95
2002	316.99	317.54	316.67	316.67	317.54
2003	317.64	318.26	317.31	317.31	318.26
2004	318.24	318.99	317.99	317.99	318.99

2005	318.98	319.71	318.83	318.83	319.71
2006	319.93	320.39	319.58	319.58	320.39
2007	320.59	321.16	320.34	320.34	321.16
2008	321.54	322.11	321.45	321.45	322.11
2009	322.24	322.91	322.22	322.22	322.91
2010	323.04	323.77	323.08	323.04	323.77
2011	324.21	324.68	324.09	324.09	324.68
2012	325.01	325.65	324.99	324.99	325.65
2013	325.92	326.61	325.89	325.89	326.61
2014	327.06	327.66	326.93	326.93	327.66
2015	328.13	328.52	327.99	327.99	328.52
2016	328.94	329.36	328.77	328.77	329.36
2017	329.75	330.37	329.68	329.68	330.37
2018	330.87	331.53	330.90	330.87	331.53
2019	331.85	332.35	331.66	331.66	332.35
2020	333.06	333.48	332.70	332.70	333.48
2021	334.33	334.81	334.03	334.03	334.81
2022	335.71	336.09	335.57	335.57	336.09

1009

1010 **Table S5: Uncertainty in future projections of atmospheric N₂O dry mole fraction.**

ppb	SSP1-1.9		SSP1-2.6		SSP2-4.5		SSP3-7.0		SSP4-3.4		SSP4-6.0		SSP5-8.5	
Year	Min	Max	Min	Max	Min	Max	Min	Max	Min	Max	Min	Max	Min	Max
2020	330.4	331.1	330.4	331.1	331.0	331.6	331.4	332.0	331.2	331.5	331.2	331.4	331.2	331.9
2030	335.1	336.9	335.5	337.0	337.6	339.6	339.5	342.2	337.5	338.6	338.8	339.6	339.5	341.1
2040	336.2	341.1	336.8	342.0	343.2	347.3	347.9	353.4	340.5	345.7	346.3	349.2	349.2	350.7
2050	336.2	344.6	337.8	345.7	348.5	354.3	356.1	364.9	343.3	353.3	353.5	359.2	359.4	361.2

1011

1012 **References:**

1013

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