

Acceleration of global N₂O emissions seen from two decades of atmospheric inversion

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Abstract

Nitrous oxide (N₂O) is the third most important long-lived greenhouse gas and an important stratospheric ozone depleting substance. Agricultural practices and the use of N-fertilizers have greatly enhanced emissions of N₂O. Here we present estimates of N₂O emissions determined from three global atmospheric inversion frameworks during 1998-2016. We find that globally N₂O emissions increased substantially from 2009 and at a faster rate than estimated by the Intergovernmental Panel on Climate Change (IPCC) emission factor (EF) approach. The regions of East Asia and South America made the largest contributions to the global increase. From the inversion-based emissions, we estimate a global EF of $2.3 \pm 0.6\%$, which is significantly larger than the IPCC Tier-1 default for combined direct and indirect emissions of 1.375%. The larger EF and accelerating emission increase found from the inversions suggest that N₂O emission may have a non-linear response at global and regional scales with high levels of N-input.

Main text

Atmospheric N₂O has risen steadily since the mid-20th century^{1,2}, from approximately 290 ppb in 1940 to 330 ppb in 2017^{3,4} - a trend strongly linked to the increase in reactive nitrogen (Nr) in the environment^{5,6}. Nr creation has increased enormously since the mid-20th century largely owing to the Haber-Bosch process (used primarily to produce N-fertilizer), but also to the cultivation of N-fixing crops and the combustion of fossil and bio-fuels⁷. Although increased Nr availability has enabled large increases in food production, it is also associated with a number of environmental problems. Among these is the rise in N₂O emissions: Nr is the substrate of the microbial processes of nitrification and denitrification, both of which produce N₂O as a by-product⁸.

N₂O emissions increased from 10-12 TgN y⁻¹ prior to the industrial era^{5,9} to an average of ~17 TgN/y in the last decade. Agriculture is responsible for the largest part of this change, with emissions increasing from 0.3-1.0 TgN y⁻¹ in 1850 to 3.9-5.3 TgN y⁻¹ in 2010^{5,9,10}. In

48 order to meet ambitious climate targets, non-CO₂ greenhouse gas emissions will also require
49 reductions¹¹. For N₂O, this means reducing agricultural emissions while meeting the growing
50 demand for food and other agricultural products. This will require changes in human diet and
51 agricultural practices, and ultimately, improved nitrogen use efficiency (NUE), that is,
52 increasing Nr in harvest relative to N-input^{12,13}.

53 N-input, in particular N-fertilizer use, is one of the best single predictors of N₂O emissions
54 from agriculture with an estimated emission factor (EF) of ~1% based on emissions measured
55 from soils¹⁴. Emission inventories, used for example in reporting under the United
56 Framework Convention on Climate Change (UNFCCC), are based predominantly on the EF
57 approach. For direct emissions from agricultural land, the default (Tier-1) value used in
58 reporting to the UNFCCC is 1% with an uncertainty range from 0.3% to 3% owing to the
59 variability with agricultural practices, soil properties, and meteorological conditions¹⁴.
60 Similarly, EFs are used to estimate indirect N₂O emissions from ecosystems downstream and
61 downwind of agricultural land, which receive Nr via run-off and atmospheric deposition,
62 amounting to an additional but even more uncertain EF of ~0.375% (Ref 12).

63 Estimates of the global mean EF have also been made by relating observed changes in
64 atmospheric N₂O to N-input, the so-called top-down approach, which includes emissions
65 from agricultural land as well as downstream and downwind ecosystems. Top-down EF
66 estimates vary from ~2 to 5% and strongly depend on the explanatory variable used,
67 specifically whether it includes only newly fixed Nr or all Nr sources^{5,15,16}. While there are
68 differences between the modelled N₂O emissions depending on the explanatory variable, all
69 EF approaches assume a linear response of N₂O to N-input. Conversely, evidence from field
70 experiments suggests the emission response is often nonlinear where N-input is high¹⁷⁻²².
71 However, whether a non-linear response of N₂O emissions is relevant at large scales and
72 globally is unknown.

73 N₂O emissions can be estimated regionally independently of EFs using the atmospheric
74 inversion approach, which utilizes spatiotemporal variations in atmospheric N₂O²³⁻²⁵. Here,
75 we use a global network of N₂O observations to estimate N₂O emissions and their trends
76 during 1998-2016. These are estimated using three independent inversion frameworks and
77 transport models (see Supplementary Tables 1&2), providing a range of estimates
78 representing the systematic uncertainty from errors in modelled transport and stratospheric
79 N₂O loss (see Methods). Using updated datasets of N-input for the whole agricultural system
80 (i.e. including crops and grasslands) and of N-surplus for cropping systems (i.e. the difference
81 between N-input and Nr removed through harvest), we determine the response of the
82 inversion-based emissions to these two explanatory variables and examine the linear
83 assumption.

84 **Emission trends and relation to N-input**

85 From three inversions, we estimate a global mean emission of 17.0 (16.6-17.4) TgN y⁻¹ for
86 1998 to 2016, with 11.3 (10.2-13.2) TgN y⁻¹ from land and 5.7 (3.4-7.2) TgN y⁻¹ from ocean
87 (values in parentheses give the range over three inversions, Supplementary Table 3). The
88 global emissions presented here are consistent with other top-down estimates ranging
89 between 15.7 and 18.3 TgN y⁻¹ for the year 2000^{5,9,23-25}. Similarly, our land emissions
90 estimate is within the range of other top-down estimates of 11.0 to 12.6 TgN y⁻¹, also for the
91 year 2000^{9,23-25}, and the recent estimate from the Nitrogen Model Inter-comparison Project
92 (NMIP)¹⁰ of 10.0 ± 2.0 TgN y⁻¹.

93 Top-down methods, including atmospheric inversions, estimate the source as the sum of the
94 observed change in atmospheric N₂O abundance and the amount lost in the stratosphere. As
95 the stratospheric loss is not constrained directly by observations this term has considerable

96 uncertainty, which is propagated into the source estimate. We calculate that stratospheric loss
97 contributes 1.1 TgN y⁻¹ to the discrepancy in the source estimate based on the range of
98 modelled atmospheric lifetimes, 118 to 129 years, and a median abundance of 1522 TgN
99 (Supplementary Table 3) (the lifetimes and abundance are comparable to previous findings²⁶).
100 The discrepancy, however, is larger than the range in source estimates, indicating
101 compensating effects in the inversions.

102 From 2000 the atmospheric growth rate increased steadily from a mean of 0.68 ppb y⁻¹ for
103 2000-2005 to 0.98 ppb y⁻¹ for 2010-2015, with significant bi- to tri-annual periodicity (Figure
104 1). Prior to 2000, calibration accuracy and measurement precision were significantly poorer,
105 hence the growth rate for 1998 to 2000 is more uncertain. Our discussion, therefore, focuses
106 on trends from 2000 onwards. Previous studies found a correlation between inter-annual
107 variability in the growth rate and El Niño-Southern Oscillation (ENSO) and attributed it to
108 changes in soil and ocean emissions^{27,28}. El Niño is associated with lower growth rates, likely
109 owing to reduced rainfall in tropical and subtropical regions²⁹ and suppressed upwelling in
110 the eastern tropical Pacific³⁰. One study also hypothesized an influence from stratosphere to
111 troposphere transport on inter-annual variability³¹. The increasing trend, however, is likely
112 due to increasing emissions; based on the inversions, emissions increased from 16.3 (15.5-
113 17.1) TgN y⁻¹ for 2000-2005 to 17.9 (17.3-18.5) TgN y⁻¹ for 2010-2015. This increase is
114 significantly larger than prior estimates, which showed an increase of 0.5 (0.4-0.6) TgN y⁻¹.
115 A change of this magnitude cannot be explained by any known mechanism through the sink,
116 as it would require an increase in atmospheric lifetime of ~20 years, and such a change is
117 unrealistic over this time scale. The atmospheric models used in this study show no trend in
118 lifetime for this period. The growth in emissions is 90% due to emissions over land (Figure
119 2) including the land-ocean aquatic continuum and inland water bodies (the spatial resolution
120 of the inversions does not allow these components to be resolved separately).

121 An increase in emissions is consistent with global trends in total N-input and crop N-surplus,
122 which grew by 59 and 18 TgN, respectively, during 2000-2013 (the last year for which data
123 are available) (Figure 3). We include synthetic fertilizer applied to crop and grasslands and
124 total animal excretion, biologically fixed nitrogen in crops and grassland, and NO_x
125 deposition from non-agricultural sources (Methods). A similar trend in N-input and N-
126 surplus is seen for China, with increases of 15 and 8 TgN, respectively, as well as for South
127 Asia (i.e., India, Nepal, Bangladesh and Pakistan) and to a lesser extent Brazil. We limit our
128 focus to the global scale and the five countries/regions in Figure 2 because the inversions in
129 other regions are not well constrained due to sparse observations and thus rely on the prior
130 estimates.

131 The regional trends in N-input and N-surplus are consistent with the N₂O emissions derived
132 from the inversions. Emissions were found to increase in China by 0.40 (0.34-0.47) TgN y⁻¹
133 between 2000-2005 and 2010-2015 - significantly larger than prior estimates of 0.23 (0.18-
134 0.32) TgN y⁻¹. Although there is an offset between INV1/INV2 and INV3 for Global land
135 and China, the trends are very similar. The offset is largely due to residual dependence of the
136 posterior on the prior estimates: INV3 used a larger land (and lower ocean) prior compared
137 to INV1/INV2. The uncertainty in all regions was reduced by the inversions (Supplementary
138 Figure 5). The change in South Asia was significantly smaller than in China, 0.14 (0.11-0.16)
139 TgN y⁻¹ but larger than indicated by prior estimates of only 0.03-0.05 TgN y⁻¹. In USA and
140 Europe, emissions were fairly stable over the past nearly two decades. In Brazil, there was
141 an increase between the two periods of 0.26 (0.23-0.29) TgN y⁻¹, but it was small compared
142 to the year-to-year variability in emissions of 0.22 TgN y⁻¹. The five regions of focus account
143 for ~50% of the global increase between the two time periods, while Africa accounts for
144 ~20%, Central and South America (excluding Brazil) account for ~10%, Southeast Asia and

145 Oceania account for 8%, and 10% was due to changes in ocean emissions (Supplementary
146 Figure 6).

147 **Estimation of emission factors**

148 Using the inversion emission trends and N-input data, we estimated EFs globally and
149 regionally. To calculate EFs, we subtracted estimates of the non-soil emissions (i.e., from
150 industry, energy and waste sectors from EDGAR-v4.3.2 (Supplementary Figure 7) and
151 biomass burning from GFED-v4.1s) from the total emissions to give the contribution from
152 soil, which we assume is proportional to N-input. Second, we subtracted the mean of the soil
153 emissions from each inversion over 1998-2016 to remove any offset between inversions.
154 Figure 4 shows scatter plots of N₂O emission anomalies from all inversions versus N-input.
155 The linear regression coefficients provide an estimate of the EF for additional emissions
156 resulting from additional N availability. The EFs were statistically significant ($P < 0.05$)
157 globally, for China, Brazil and South Asia, but not for USA and Europe where changes in N-
158 input and N₂O emission were small compared to the scatter in the data (Supplementary Table
159 4). The emissions are generally higher than proportionate (and more scattered) at the upper
160 range of N-input globally and for China and Brazil, but using non-linear regressions led to
161 only marginal improvements with no difference between quadratic versus exponential
162 functions. Regressions were also calculated relative to N-surplus but no improvement in the
163 correlation or reduction in the residual standard error was found (Supplementary Table 5 and
164 Figure 8).

165 Globally, we find an EF of $2.3 \pm 0.6\%$ for the change in total soil N₂O emission relative to
166 the change in total N-input, including N-fertilizer, manure, biological nitrogen fixation
167 (BNF), and NO_x deposition from non-agricultural sources (Figure 5). Our N-input differs
168 slightly from the IPCC 2006 reporting guidelines, which includes (in addition to synthetic
169 fertilizer and manure) Nr from crop residues and mineralization of soil organic matter where
170 soil Nr stocks are changing due to land use or management¹⁴. On the other hand, our N-input
171 includes total livestock excretion and not only that applied as manure as in the IPCC 2006
172 method. While the IPCC 2006 method does not directly include BNF, it assumes that Nr from
173 BNF is relevant for N₂O production when left on fields in crop residue. We do not have
174 estimates of Nr from mineralization of soil organic matter from land use or management, but
175 this term is likely to be small compared to other N-inputs. Furthermore, our EF estimates
176 assume that trends in natural emissions of N₂O are negligible over the study time period.
177 Since changes in N₂O emissions due to anthropogenic N-input to natural ecosystems is
178 counted as an anthropogenic emission, changes in natural N₂O emissions are primarily
179 related to climatic changes. Natural emissions changed by an estimated $0.7 \pm 0.5 \text{ TgN y}^{-1}$
180 since the pre-industrial era and, therefore, likely have negligible impact on our EFs for 2000-
181 2013¹⁰.

182 The IPCC (Tier-1) method gives one EF for direct and another for indirect emissions,
183 whereas we calculate the total EF relative to N-input. To compare the two methods, we
184 estimate the IPCC total EF by adding the equations for direct and indirect emissions (using
185 default parameters) and dividing by total N-input, giving an EF of 1.375% (see Methods).
186 Our global mean EF is higher than the IPCC value but is sensitive to positive emission
187 anomalies in 2010 and 2013 (Figure 2); excluding these values gives an EF that is not
188 statistically different from the IPCC value. A longer time series of inversion-based emissions
189 would help in determining the EF more accurately. However, our estimate of 2.3% agrees
190 well with that of a previous top-down study⁵, which found an EF of ~2.5% (Figure 5). Ref 5
191 estimated separate EFs for manure and N-fertilizer, of 2% and 2.5%, respectively, and found
192 this gave a better fit to top-down estimated N₂O emissions throughout the 20th century
193 compared to one EF for total N-input. This was because in the first half of the 20th century

194 Nr in manure was not only derived from contemporaneous N-fixation but was also mined
195 from agricultural soils. Over the past two decades, N-mining from soils occurred only in a
196 few countries, and manure Nr is predominantly derived from fertilizer Nr used to grow crops
197 for livestock feed. Consistent with this, we find for the last nearly two decades that the fit to
198 N₂O emissions did not improve if N-fertilizer and manure were considered separately as
199 explanatory variables. A higher EF than the IPCC default, is also plausible considering the
200 evidence of a non-linear response of N₂O emission to high levels of N-input^{10,17-22}, which is
201 discussed below.

202 For China, we find an EF of $2.1 \pm 0.4\%$, and this estimate is insensitive to emission anomalies.
203 A high EF for China is credible given the very high rates of fertilizer application, low crop
204 NUE (defined as the output/input ratio for cropping systems, Supplementary Figure 9), and
205 possibility of a non-linear response of N₂O emission^{10,17-22,32,33}. However, our EF for China
206 is associated with systematic uncertainty owing to uncertain trends in non-soil emissions, in
207 particular from industry, which differ substantially between inventories. If the non-soil
208 emission trend is underestimated the EF would be overestimated and vice-versa. For example,
209 using the GAINS inventory estimate for non-soil emissions (instead of EDGAR-v4.32), the
210 EF for China would be only $1.4 \pm 0.4\%$ and not statistically different from the IPCC default.
211 The most important difference between EDGAR and GAINS is the change in emissions from
212 adipic acid production - in EDGAR these are reduced by ~90% between 2005 and 2010
213 whereas in GAINS they increase by a factor of ~2 (Supplementary Figure 7). The discrepancy
214 arises from assumptions made about adipic acid plants that became operational after 2005,
215 specifically their contribution to the total adipic acid production and what emission
216 abatement technologies they use^{34,35}. If the GAINS emissions were correct then the increase
217 in emissions from adipic acid production would account for nearly 20% of the total increase
218 in China's emissions since 2005. Trend differences between EDGAR and GAINS have
219 negligible impact on the global EF calculation and for the other regions in our study.

220 For Brazil, we calculate an EF of $2.6 \pm 0.7\%$. This value is sensitive to emission anomalies,
221 specifically in 2010 and 2013 (as for the global EF). Removing these anomalies reduces the
222 EF to $2.1 \pm 0.7\%$. Our high EF for Brazil is puzzling due to the relatively high NUE, ~50%,
223 a low portion of synthetic fertilizer in the total N-input, and predominantly low EF values
224 measured at the plot scale (median 0.38%, range 0.13 to 5.14% in cropland)³⁶. Several
225 speculative explanations are possible, including insufficient field sampling of soil EFs among
226 the rapidly changing agricultural management systems³⁷, declining NUE in expanding cereal
227 production³⁸, underestimated BNF in pastures and sugar cane production³⁹, confounding
228 effects of ENSO on the large emissions from Amazon forest soils or from fire⁴⁰, varying
229 deforestation trends, as well as growth and intensification of cropland and livestock
230 management^{41,42}.

231 For South Asia, we find an EF of $0.8 \pm 0.4\%$, which was not sensitive to emission anomalies
232 and is lower than the IPCC default. Although South Asia has a low NUE, it uses a much
233 smaller portion of synthetic fertilizer in total N-input than China, and has lower intensity of
234 synthetic fertilizer application over crop area, 96 kgN ha⁻¹ compared to 281 kgN ha⁻¹ in China
235 for the mean over 2000-2013.

236 **Evaluation of the emission factor approach**

237 Globally, the inversion-based soil N₂O emissions grew at a faster rate than predicted with
238 the IPCC Tier-1 EF from 2009 (Figure 6). The increase in emissions from 2000-2005 to
239 2010-2013, of 1.55 (1.44-1.71) TgN y⁻¹, is also more than double that predicted by the IPCC
240 EF, of 0.59 TgN y⁻¹. Using the EF calculated here (2.3%) tended to overestimate the response
241 between 2005-2009 and underestimate it after 2009, when the N-surplus was particularly

242 high. Although a non-linear (quadratic or exponential) function did not markedly improve
243 the residual standard error in the regressions of N₂O emission versus N-input (owing to large
244 scatter in the data), there are reasons to think the response may be non-linear, as suggested
245 from field-based studies¹⁷⁻²². Mechanisms proposed for a non-linear response with large N-
246 surplus include: 1) more available Nr substrate for nitrification and denitrification⁴³, 2) high
247 soil concentrations of NO₃⁻ associated with a higher N₂O to N₂ ratio from denitrification⁴⁴,
248 3) Nr availability to microorganisms exceeding carbon availability leading to higher rates of
249 N₂O emission⁴⁵, and 4) Nr stimulating microbial mobilization of N bound in soil organic
250 matter⁴⁶. We compared the inversion-based soil emissions with the non-linear models in Refs
251 17 and 18 (Supplementary Figure 10) and found that both give slightly higher estimates after
252 2009 compared to the IPCC EF, but still underestimate the emissions.

253 In China, the emissions similarly increased at a faster rate than estimated by the IPCC EF
254 after 2009. Although the agreement is better in the scenario where the industrial emissions
255 followed the trend in GAINS, if N-input remained at the same high level after 2013, then the
256 IPCC Tier-1 EF would considerably underestimate the emissions also in this scenario from
257 2013. For Brazil, the IPCC EF again underestimates the growth in emissions after 2009, but
258 for South Asia, it reproduces the trend seen in the inversion-based estimates.

259 USA and Europe differ from the other regions in that they have stable and decreasing N-
260 input, respectively. In USA, the nearly flat inversion-based emissions are consistent with EF
261 estimates. The notable negative emission anomaly for 2000-2005, however, is not captured,
262 as it is not due to a change in N-input but rather likely to EF changes driven by meteorological
263 conditions. Precipitation data⁴⁷ and the Palmer Drought Severity Index⁴⁸ (PDSI) for the USA
264 in regions with non-negligible N₂O emissions show persistent dry conditions during 1999-
265 2003, which may have led to a decrease in the EF during that time (Supplementary Figure
266 11). In the other regions studied, however, there was no clear relationship between N₂O
267 emission anomaly and precipitation, PSDI, or soil temperature. For Europe, the emissions
268 estimated using the EF approach are close to those from the inversions. Although the EF
269 approach shows a small decrease, of 0.01 TgN y⁻¹ between 2000-2005 and 2010-2013, no
270 trend is seen in the inversion-based estimate, but it may be that any trend related to N-input
271 is still too small to be captured by global scale inversions.

272 **Conclusions and implications**

273 N₂O emissions increased globally by 1.6 (1.4-1.7) TgN y⁻¹ between 2000-2005 and 2010-
274 2015, however the rate of increase from 2009 is underestimated using the IPCC Tier-1 default
275 EF. We hypothesize that this is due to an increase in the EF associated with a growing N-
276 surplus. This suggests that the Tier-1 method, which assumes a constant EF, may
277 underestimate emissions when the rate of N-input and the N-surplus are high. This has been
278 demonstrated at field scale, but here we show this likely also applies at regional and global
279 scales. We therefore recommend moving towards IPCC Tier-2 approaches and using region-
280 specific EFs, especially for high N-input and/or N-surplus conditions, but this would require
281 a body of field measurements to determine accurate values for these EFs. Alternatively,
282 process-based modelling (as used in the IPCC Tier-3 method) validated against observations
283 could help estimate emissions where the N-input and/or N-surplus is high. Our results show
284 that reducing N-surplus (and improving NUE) in high N-input regions should have a more
285 than proportionate outcome in reducing N₂O emissions.

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412 **Methods**

413 Emissions were estimated using three independent atmospheric inversion frameworks (see
414 Supplementary Table 1). The frameworks all used the Bayesian inversion method, which
415 finds the optimal emissions, that is, those, which when coupled to a model of atmospheric
416 transport, provide the best agreement to observed N₂O mixing ratios while remaining with
417 the uncertainty limits of the prior estimates. In other words, the emissions that minimize the
418 cost function:

$$419 \quad J(\mathbf{x}) = \frac{1}{2}(\mathbf{x} - \mathbf{x}_b)^T \mathbf{B}^{-1}(\mathbf{x} - \mathbf{x}_b) + \frac{1}{2}(\mathbf{y} - H(\mathbf{x}))^T \mathbf{R}^{-1}(\mathbf{y} - H(\mathbf{x})) \quad (1)$$

420 where \mathbf{x} and \mathbf{x}_b are, respectively, vectors of the optimal and prior emissions, \mathbf{B} is the prior
421 error covariance matrix, \mathbf{y} is a vector of observed N₂O mixing ratios, \mathbf{R} is the observation
422 error covariance matrix, and $H(\mathbf{x})$ is the model of atmospheric transport (for details on the
423 inversion method see Ref. 49). The optimal emissions, \mathbf{x} , were found by solving the first
424 order derivative of equation (1):

$$425 \quad J'(\mathbf{x}) = \mathbf{B}^{-1}(\mathbf{x} - \mathbf{x}_b) + (H'(\mathbf{x}))^T \mathbf{R}^{-1}(\mathbf{y} - H(\mathbf{x})) = 0 \quad (2)$$

426 where $(H'(\mathbf{x}))^T$ is the adjoint model of transport. In frameworks INV1 and INV2, equation
427 (2) was solved using the variational approach^{50,51}, which uses a descent algorithm and
428 computations involving the forward and adjoint models⁵². In framework INV3, equation (2)
429 was solved directly by computing a transport operator, \mathbf{H} from integrations of the forward
430 model, such that $\mathbf{H}\mathbf{x}$ is equivalent to $H(\mathbf{x})$, and taking the transpose of \mathbf{H} ⁵³.

431 Each of the inversion frameworks used a different model of atmospheric transport with
432 different horizontal and vertical resolutions (see Supplementary Table 1). The transport

433 models TOMCAT and LMDz, used in INV1 and INV2 respectively, were driven by ECMWF
434 ERA-Interim wind fields, and the model, MIROC4-ACTM, used in INV3, was driven by
435 JRA-55 wind fields. While INV1 and INV2 optimized the emissions at the spatial resolution
436 of the transport model, INV3 optimized the error in the emissions aggregated into 84 land
437 and ocean regions⁵³. All frameworks optimized the emissions with monthly temporal
438 resolution. The transport models included an online calculation of the loss of N₂O in the
439 stratosphere due to photolysis and oxidation by O(¹D) resulting in mean atmospheric
440 lifetimes of between 118 and 129 years, broadly consistent with recent independent estimates
441 of the lifetime of 116 ± 9 years²⁶.

442 The inversions used N₂O measurements of discrete air samples from the National Oceanic
443 and Atmospheric Administration Carbon Cycle Cooperative Global Air Sampling Network
444 (NOAA) and the Commonwealth Scientific and Industrial Research Organisation network
445 (CSIRO). In addition, we used measurements from in-situ instruments in the Advanced
446 Global Atmospheric Gases Experiment network (AGAGE), the NOAA CATS network, and
447 from individual sites operated by University of Edinburgh (UE), National Institute for
448 Environmental Studies (NIES) and the Finish Meteorological Institute (FMI) (see
449 Supplementary Figure 1). Measurements from networks other than NOAA were corrected to
450 the NOAA calibration scale, NOAA-2006A⁵⁴, using the results of the WMO Round Robin
451 inter-comparison experiment (<https://www.esrl.noaa.gov/gmd/ccgg/wmorr/>). Frameworks
452 INV1 and INV2 used a total of 83 discrete air sampling sites, 15 in-situ sampling sites and
453 discrete air samples from the NOAA network of ships and moorings, and INV3 used 37
454 discrete air sampling sites. Daily average observations were assimilated in INV1 and INV3,
455 while INV2 assimilated hourly afternoon values for low altitude sites and nighttime values
456 for mountain sites to minimize errors in the modeled mixing ratios from errors in the modeled
457 planetary boundary layer heights and local mountain-valley circulation.

458 Each framework applied its own method for calculating the uncertainty in the observation
459 space, the square of which gives the diagonal elements of the observation error covariance
460 matrix **R**. The observation space uncertainty accounts for measurement and model
461 representation errors and is equal to the quadratic sum of these terms. INV1 assumed a
462 measurement uncertainty of 0.4 ppb and, in addition, estimated the model representation error
463 as the mixing ratio gradient across the grid cell in which the observation is located and the
464 surrounding ones, resulting in a mean total uncertainty of 0.48 ppb. INV2 assumed a
465 measurement uncertainty of 0.3 ppb and estimated the representation error in the same way
466 as INV1, resulting in a mean total uncertainty of 0.50 ppb. INV3 used a measurement
467 uncertainty of 0.32 ppb and estimated the representation error as 1-sigma standard deviation
468 of daily observations at each site.

469 Prior emissions were used in all frameworks and were based on existing estimates from
470 terrestrial biosphere and ocean biogeochemistry models as well as from inventories (see
471 Supplementary Table 2). INV1 and INV2 used the same prior estimates for emissions from
472 natural and agricultural soils from the model OCN-v1.1, for ocean emissions from the model
473 PlankTOM5, and for biomass burning emissions from the Global Fire Emissions Database
474 (GFED-v4.1s). OCN parameterizes N₂O emissions from nitrification and denitrification in
475 soils and accounts for N-input from N-fertilizer, manure, atmospheric deposition, and
476 biological nitrogen fixation. The model is driven by CRU-NCEP meteorological data and
477 uses inter-annually varying N-input⁵⁵. PlankTOM5 uses the observed correlation between
478 apparent oxygen utilisation and excess N₂O in oxic waters to estimate the open ocean source
479 of N₂O production and the increased yield of N₂O in suboxic waters from both nitrification
480 and denitrification as an additional source in oxygen minimum zones⁵⁶. The model,
481 PlankTOM5, is incorporated into the ocean general circulation model, NEMO v3.1, which is

482 forced with NCEP meteorology. For non-soil anthropogenic emissions (namely those from
483 energy, industry and waste sectors), both INV1 and INV2 use the Emission Database for
484 Greenhouse Gas Research (EDGAR) but differing versions (see Supplementary Table 2).
485 INV3 used GEIA (Global Emissions Initiative) for emissions from natural soils and ocean
486 emissions from Manizza et al. 2012⁵⁷. Manizza et al. model ocean emission using the
487 correlation of apparent oxygen utilization and excess N₂O in oxic waters and their model is
488 incorporated into the MIT General Circulation Model. For soil and non-soil anthropogenic
489 emissions, INV3 used a third version of EDGAR (see Supplementary Table 2), which also
490 includes agricultural burning but they did not specifically account for wildfire emissions in
491 the prior estimates.

492 Prior uncertainties were estimated in all the inversion frameworks for each grid cell (INV1
493 and INV2) or for each region (INV3) and square of the uncertainties formed the diagonal
494 elements of the prior error covariance matrix **B**. INV1 and INV2 estimated the uncertainty
495 as proportional to the prior value in each grid cell, and INV2 set lower and upper limits for
496 the uncertainty of 3×10^{-9} and 5×10^{-8} kgN m⁻² h⁻¹, respectively. INV3, on the other hand, set
497 the uncertainty uniformly for the land regions at 1 TgN y⁻¹ and for the ocean regions at 0.5
498 TgN y⁻¹. INV2 was the only framework to account for spatial and temporal correlations in
499 the errors (resulting in off-diagonal elements in the prior error covariance matrix) using an
500 exponential decay model with distance and time with correlation scale lengths of 500 km
501 over land and 1000 km over ocean and 90 days.

502 The optimized emissions were interpolated to $1^\circ \times 1^\circ$ (see Supplementary Figure 2) and the
503 regional emissions were calculated by integrating the gridded emissions within each region
504 or country. For each region, estimates of the non-soil anthropogenic emissions (i.e., from
505 industry, energy and waste sectors) from EDGAR-v4.3.2 and the biomass burning emissions
506 from GFED-v4.1s were subtracted from the total emissions from the inversions to give only
507 the contribution from soil, which is assumed to be proportional to N-input. This assumes that
508 the error in the estimate for non-soil anthropogenic emissions is substantially smaller than
509 that in the soil emissions (Supplementary Figure 7).

510 The inversions were validated by integrating the forward models with the posterior emissions
511 and comparing the simulated mixing ratios with independent observations, i.e., observations
512 that were not assimilated in the inversions. We compared with CONTRAIL (Comprehensive
513 Observation Network for TRace gases by AirLiner, [http://www.jal-
514 foundation.or.jp/shintaikikansokue/contrail_index.htm](http://www.jal-foundation.or.jp/shintaikikansokue/contrail_index.htm)), which has N₂O observations at
515 regular intervals across the Pacific since 2005 (Supplementary Figure 3). All three inversions
516 showed a similar level of performance with differences typically of <0.5 ppb. We also
517 compared with aircraft profile measurements over USA from NOAA from sites with data for
518 the early 2000s (Supplementary Figure 4). We found that INV1 tended to underestimate N₂O
519 in the lower troposphere over the contiguous USA for the early 2000s, hence we did not
520 include the emissions data for USA prior to 2005 in our analyses.

521 We calculated N inputs to the whole agricultural system including crops and grasslands. Total
522 inputs correspond to synthetic fertilizer application, animal excretion (even if finally not
523 reaching crops or grasslands), biological nitrogen fixation, and NO_x deposition on
524 agricultural land. Total outputs correspond to crop and animal production. Total surplus is
525 calculated as the difference between inputs and outputs. In this budget, we neglected the
526 small part of crop production that is locally consumed by livestock. Synthetic fertilizer
527 application is based on the FAOSTAT dataset (<http://www.fao.org/home/en/>) with several
528 inputs from the International Fertilizer Association (<https://www.fertilizer.org/>). Total
529 animal excretion is calculated using the FAOSTAT livestock inventory and dynamic
530 excretion factors, biological N fixation is calculated from crop productivities⁵⁸ and

531 atmospheric deposition was from Ref 59. Grassland nitrogen fixation was based on the
 532 grassland production estimated following Ref 60 and validated through comparison with the
 533 IMAGE model⁶¹. We consider 20% of grass species to be N fixing legumes and that their N
 534 fixation is equal to 1.4 times the N from aerial production to also account for below ground
 535 biomass production, which would otherwise not be included⁵⁸. N output in harvested crops
 536 is based on crop productivity and N content of 177 crops, utilizing data from the FAOSTAT
 537 database. See also the detailed methodology in Refs 32 and 60. We consider the N-surplus
 538 and NUE of cropping systems, as they are widely used as an indicator of the agronomic and
 539 environmental performance of agricultural systems.

540 Emission factors were determined by a linear regression of N₂O soil emission versus total
 541 N-input. The total N-input consisted of sources of N from synthetic fertilizer (N_{SF}), organic
 542 fertilizer and manure (N_{ON}), biological nitrogen fixation (N_{BNF}) and NO_x deposition from
 543 non-agricultural sources. This emission factor represents the total of direct and indirect
 544 emissions. The emission factors calculated in this study were compared to the IPCC Tier-1
 545 default values, where the total IPCC EF was calculated by taking the weighted average of the
 546 direct (EF_{dir}) and indirect factors for deposition (EF_{dep}) and leaching (EF_{leach}) according to:

$$547 \quad EF_{tot} = EF_{dir} + EF_{dep} \left(f_{SF} \frac{N_{SF}}{N_{tot}} + f_{ON} \frac{N_{ON}}{N_{tot}} \right) + EF_{leach} f_{leach} \quad (3)$$

548 where f_{SF} and f_{ON} are the fractions of synthetic and organic fertilizer volatilized, respectively,
 549 and f_{leach} is the fraction of N lost by leaching and runoff¹². The modelled N₂O emission
 550 (F_{N_2O}) using the IPCC emission factors was calculated as:

$$551 \quad F_{N_2O} = EF_{dir} (N_{SF} + N_{ON} + N_{BNF}) + EF_{dep} (N_{SF} f_{SF} + N_{ON} f_{ON}) + \\ EF_{leach} (N_{SF} + N_{ON} + N_{BNF}) f_{leach} \quad (4)$$

552 using the N-input dataset described above.

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588 nitrogen, phosphorus, and carbon fluxes: The generalized representation of agro-food
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616 **Author contributions**

617 RLT designed the study, contributed inversion results and prepared the manuscript; LL
618 prepared the N-data and contributed to the manuscript; PKP, CW and MPC contributed
619 inversion results and to the manuscript; KCW, AG, ENK, WW and EAD helped with the
620 analysis and contributed to the manuscript; HT and JCG contributed to the manuscript.

621 **Competing interests statement**

622 The authors declare that they have no competing interests.

623 **Data availability**

624 Atmospheric observations used in the inversions are available from the databases indicated
625 in the Acknowledgements. The CONTRAIL data used in the validation of the inversion
626 results are available on request to H. Matsueda (MRI-JMA). The inversion output data are
627 available from <http://doi.org/10.5281/zenodo.3384591> and the N-data are available from
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629 following authors on reasonable request: C. Wilson (c.wilson@leeds.ac.uk) for INV1; R.
630 Thompson (rlt@nilu.no) for INV2; and P. Patra (prabir@jamstec.go.jp) for INV3.

631

Figure 1. Observed and modelled global mean growth rates of N_2O . Observed growth rates are shown based on the NOAA discrete sampling network and, for comparison, the AGAGE network. Modelled growth rates were calculated by sampling 4D mixing ratio fields at the times and locations of the NOAA observations. All growth rates were calculated with annual time steps and are shown as 1-year running averages.

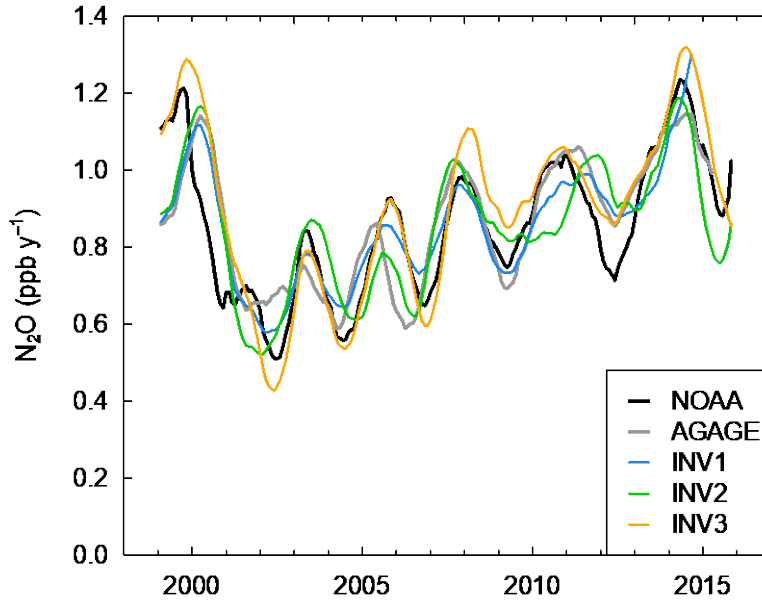


Figure 2. Annual N_2O emissions from the atmospheric inversions for 1998 to 2016 (units TgN y^{-1}). Dashed lines show the prior and solid lines the posterior emissions. INV1 data prior to 2005 for USA are shown as a dotted line as these data are more uncertain (see Methods).

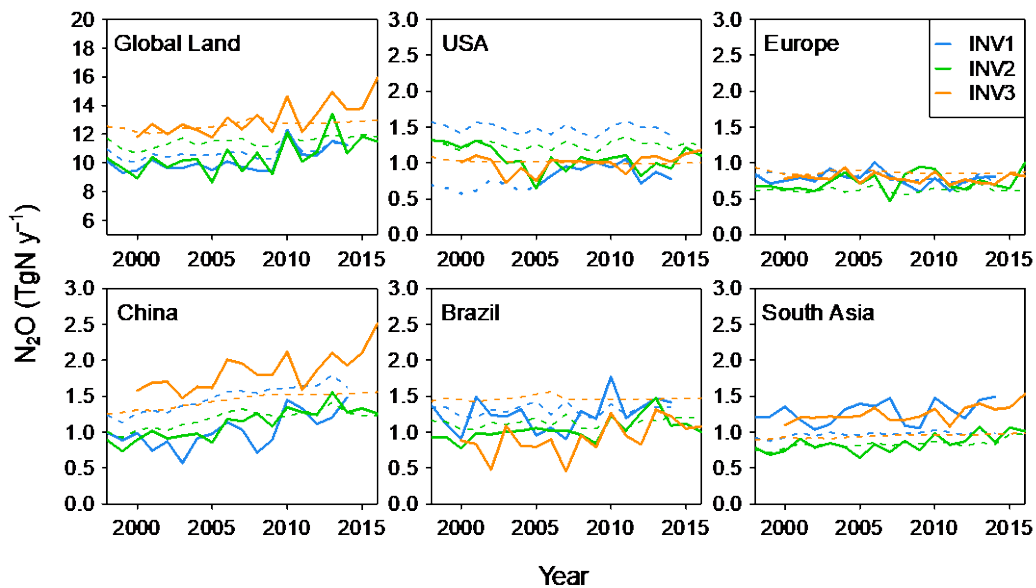


Figure 3. N-inputs to world crops and grasslands (units TgN y^{-1}) and N-surplus in the cropping systems. (N-fert is synthetic fertilizer, N-fixed is biologically fixed N, NO_x -dep is NO_x deposition, N-surplus is surplus only for cropping systems).

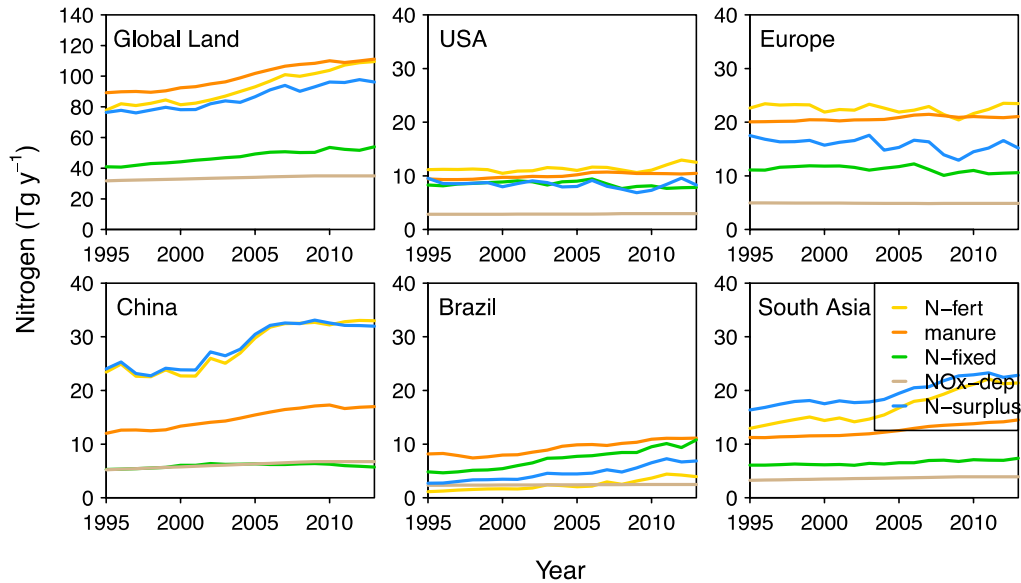


Figure 4. Scatter plots of the N_2O emission anomalies versus N-input (units TgN y^{-1}). The emissions were corrected for the non-soil component and the anomalies were calculated relative to the mean for 1998 to 2013. The symbols are colour-coded by year (circles = INV1, squares = INV2, diamonds = INV3). The solid line shows the regression and the dotted lines the confidence range. In the case that the regression is not significant ($P > 0.05$) a dashed line is used for the regression. (INV1 was excluded for USA owing to the poorer model-observation comparison for 1998-2005).

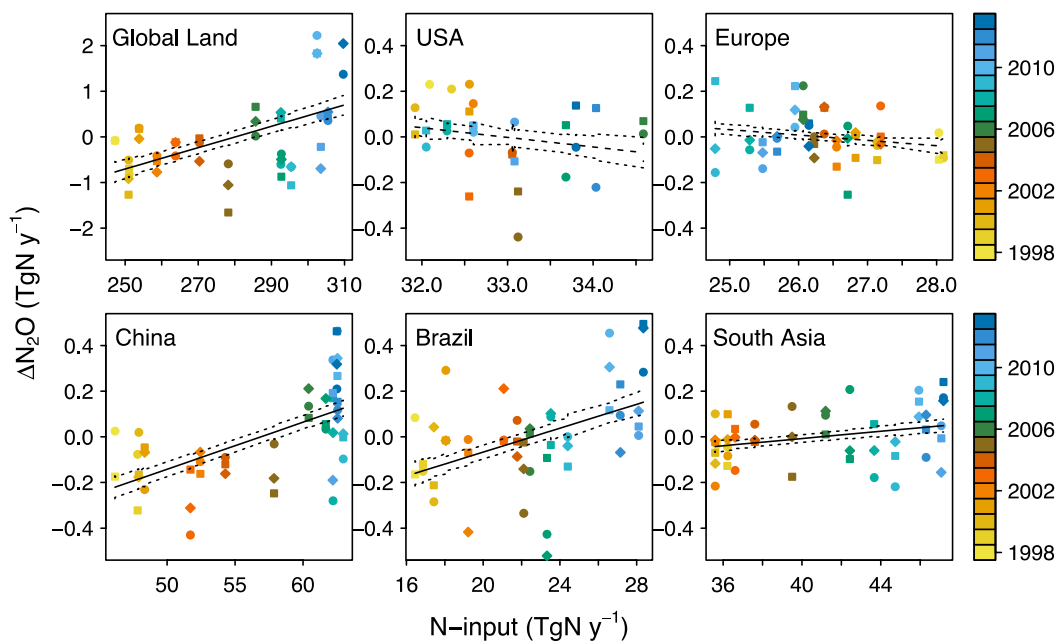


Figure 5. Comparison of emission factors (EF) from this study and from recent literature. The white to red circles are the EFs calculated over all inversions in this study and the colour indicates the correlation coefficient (see legend). The grey points are the EFs calculated from the individual inversions where the correlation was significant (circles = INV1, squares = INV2, diamonds = INV3). A second EF is shown (red diamond) for China using the GAINS estimate for the non-soil anthropogenic emissions. For the values reported by this study, the error bars show the standard error and for the other studies, they show the reported uncertainty.

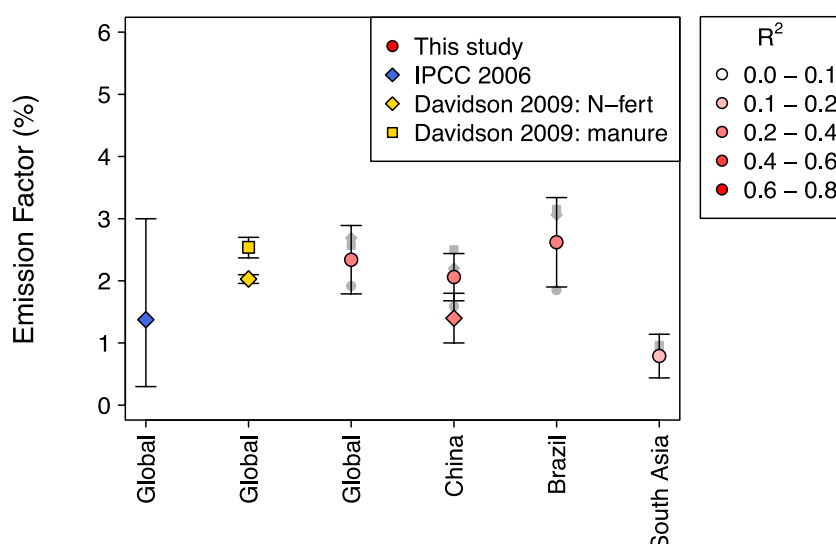


Figure 6. Comparison of N_2O emissions from the inversions (corrected for the non-soil component) with those calculated using the EF approach (units $TgN\ y^{-1}$). The inversion results are shown as the mean (black line) and range (grey shading). A scalar value was added to the emissions time series' so that they matched the inversion mean in the year 2000. The EF results are shown using the IPCC value (blue) and the linear fit from this study (green). For USA and Europe the regional EFs from this study were not significant so the global EF from this study was used instead. For China, the emissions corrected using GAINS for the non-soil component (instead of EDGAR-v4.32) are also shown (black dotted line).

