

Testing high resolution nitrous oxide emission estimates against observations using an atmospheric transport model

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Abstract

Global budgets of atmospheric nitrous oxide (N₂O) – an important greenhouse gas – show considerable uncertainty in the estimates of sources and sinks. Here we present forward runs of an atmospheric transport model to test hypotheses on recently derived source and sink estimates. Most atmospheric N₂O monitoring stations are not well situated for verifying the simulated seasonality in atmospheric N₂O. In addition, the precision of N₂O measurements is not adequate for resolving seasonal trends. The global N₂O inventory used does not account for soil-N₂O consumption in temperate nitrogen-limited ecosystems and observed episodic emissions in temperate ecosystems during thaw periods, autumn and from snow-covered soils. These potential errors and possible underestimation of N₂O emissions from combustion and industrial sources may exaggerate the simulated seasonal trends.

Nitrous oxide occurs in the atmosphere in minute quantities (310 ppb) and is increasing by ~0.8 ppb yr⁻¹ ¹, although the rate is not constant. In 1992 the increase was about half that of the 1980s ². The uncertainty factor for most N₂O source estimates is at least 2 (ref. 1). The causes of this increase and its variability are not well understood. The major global sources of N₂O – soils and oceans – may show seasonal variation in emissions ^{3,4} that may be reflected in the atmospheric N₂O concentration ⁵. Currently, there are 10 monitoring stations worldwide where atmospheric N₂O concentrations are measured ^{6,7}. These stations were established in remote locations where the air is thought to be well mixed and representative of large-scale air masses. There is only one continental station (Niwot Ridge, Colorado).

Monthly inventories of N₂O emission from soils under natural vegetation and from the oceans were used where available, while annual estimates for other sources were calculated on a monthly basis using different criteria (Table 1). The monthly N₂O emission estimates (Figure 1) were used to prescribe a 2.5° resolution global Lagrangian atmospheric transport model ⁵. The simulated N₂O concentrations were ~1 ppb higher in the Northern Hemisphere (NH) than in the Southern Hemisphere (SH). This result is consistent with measured concentrations ⁶, suggesting that the overall latitudinal source distribution is correct. The modelled latitudinal gradient varied with time; smaller differences and the highest N₂O concentrations in the tropics were seen from November to April, and the maximum difference and highest values over Northern mid-latitudes from May to October. Simulated continental concentrations in the order of 310-315 ppb over strong-source regions (Figure 2) agree with high N₂O concentrations observed at the Colorado station in July-September 1991 ⁷.

Table 1. Global monthly 1° x 1° resolution inventories of N₂O sources used to prescribe the atmospheric transport model *

Source	Annual N ₂ O emission (Tg N ₂ O-N yr ⁻¹)†	Time scale	Reference	Major criterion used to produce monthly N ₂ O fluxes
Soils under natural vegetation	4.3 ‡	month	18,19	— §
Arable lands and synthetic N fertilizer use	1.8	month/year	18,19	Growing seasons ¶
Grasslands and animal excreta	2.4	month/year	18	Soils under natural vegetation #
Savanna burning and forest clearing	0.1	month	22,23	— §
Agricultural waste burning	0.1	year	23,24	Growing seasons **
Post-clearing enhanced soil flux	0.4	year	18	Soils under natural vegetation
Fossil-fuel combustion and traffic	0.3	year	18	Constant flux
Biofuel combustion	0.1	year	18	Constant flux
Industry	0.5 ††	year	18	Constant flux
Oceans	3.6 ††	month	4	— §
Total sources	13.6			
Stratospheric loss	10.5			
Atmospheric increase	3.1			

* "Best" estimate of eight different cases ¹⁸.

† Tg = teragram; 1 Tg = 10¹² g

‡ Model result ^{3,19} for different broad ecosystems, including the N₂O emission of 2.3 Tg N₂O-N yr⁻¹ for closed tropical forests ¹⁸ consistent with the estimated 2.4 Tg N₂O yr⁻¹ for "lowland tropical forests" ²⁵; 1 Tg N₂O-N yr⁻¹ from open tropical forests ¹⁸, which is within the range (0.4-1.3 Tg N yr⁻¹) for "dry tropical forests" ²⁶, and 0.5 Tg N₂O-N yr⁻¹ for temperate forests ¹⁸ consistent with the range of other estimates ^{12,15}. No literature estimates are available for comparing the remaining 0.5 Tg N₂O-N yr⁻¹ from "other lands" ¹⁸.

§ Monthly estimates are included in the inventory.

¶ "Background" emission (0.9 Tg N₂O-N yr⁻¹) from unfertilized arable lands calculated on a monthly basis. Monthly distributions of the fertilizer-induced emission (1.25% of N application ¹⁸) based on the growing season ²⁰ in each agricultural grid cell ²¹, with 60% of N₂O losses within 1 month of N application for growing periods < 180 days. For longer growing seasons allowing cultivation of more than one crop, the fertilizer-induced emission is assumed to occur as a constant flux during the growing period.

"Background" N₂O emission from grasslands (1.4 Tg N₂O-N yr⁻¹) calculated on a monthly basis; N₂O emission from animal excreta is given a monthly distribution identical to the emissions from soils under natural vegetation.

** For growing seasons < 300 days, burning of agricultural waste is assumed to occur at the end of the season (25% during the last month, 50% in the succeeding 30 days and 25% in the 4 weeks thereafter). For growing seasons > 300 days, the burning is assumed to be constant throughout the year.

†† Including nitric and adipic acid production.

‡‡ The original inventory at ~2.8° x 2.8° resolution was converted to 1° x 1° resolution. Due to the scale difference the estimated global oceanic emission is 0.2 Tg N₂O-N yr⁻¹ lower than the original estimate ⁴.

The flask sample concentration measurements show stronger oscillations than the high frequency measurements (Figure 3a-d). Neither concentration measurement shows seasonal trends in the NH and SH. Modelled results for monitoring stations in NH temperate zones showed somewhat higher summer than winter concentrations, according to the seasonality of N₂O emissions (Figure 3a-b). The seasonal variation in the modelled concentrations for the SH was smaller – and more consistent with N₂O concentration measurements – than in the NH (Figure 3c-d), because of lower emission rates and smaller seasonal variation in emissions in the SH relative to the NH (Figure 1).

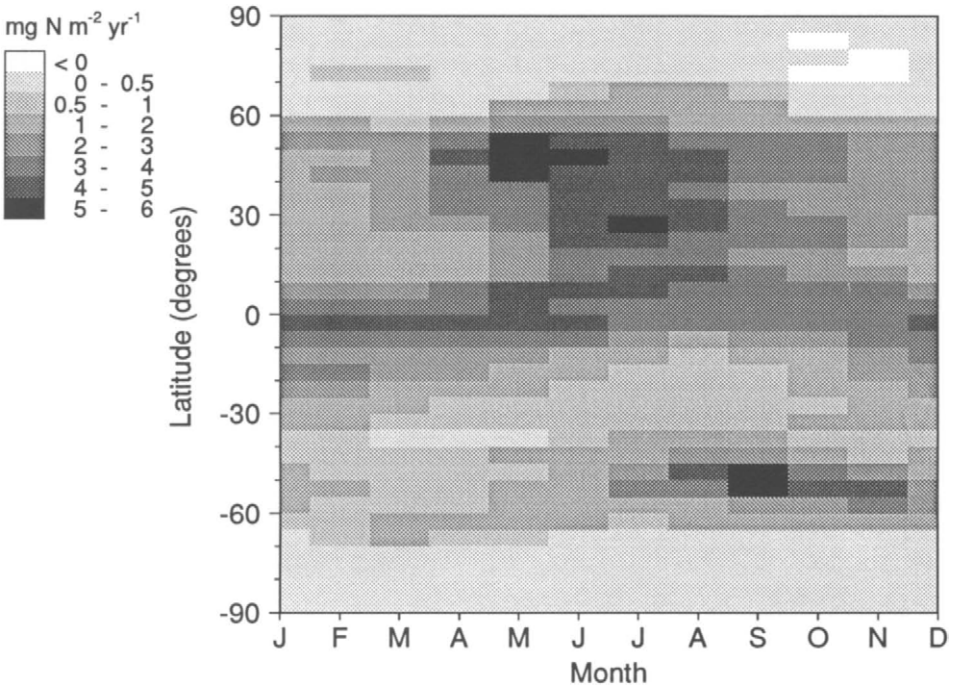


Figure 1. Seasonal-latitude distribution of annual emission rates from all sources listed in Table 1.

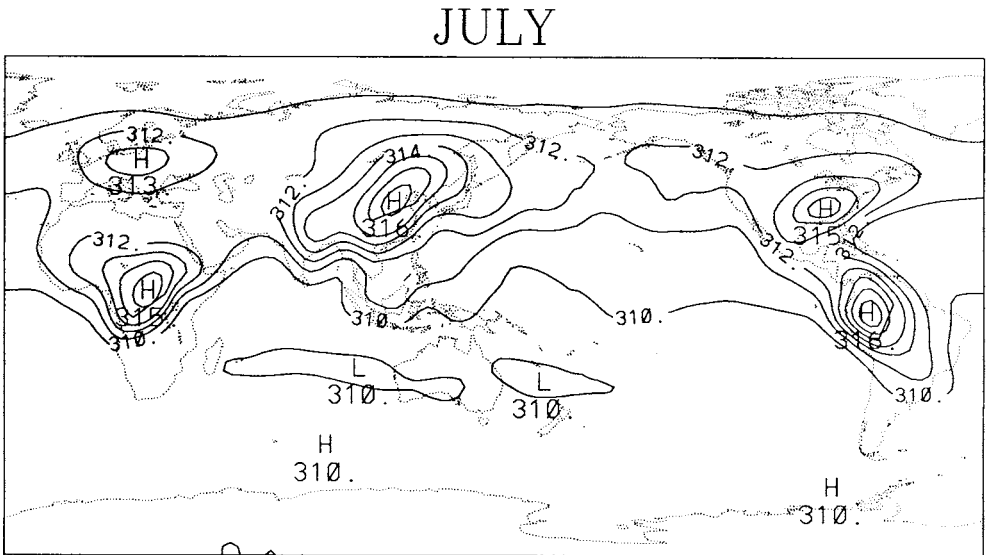


Figure 2. Simulated global distribution of N₂O concentration for July.

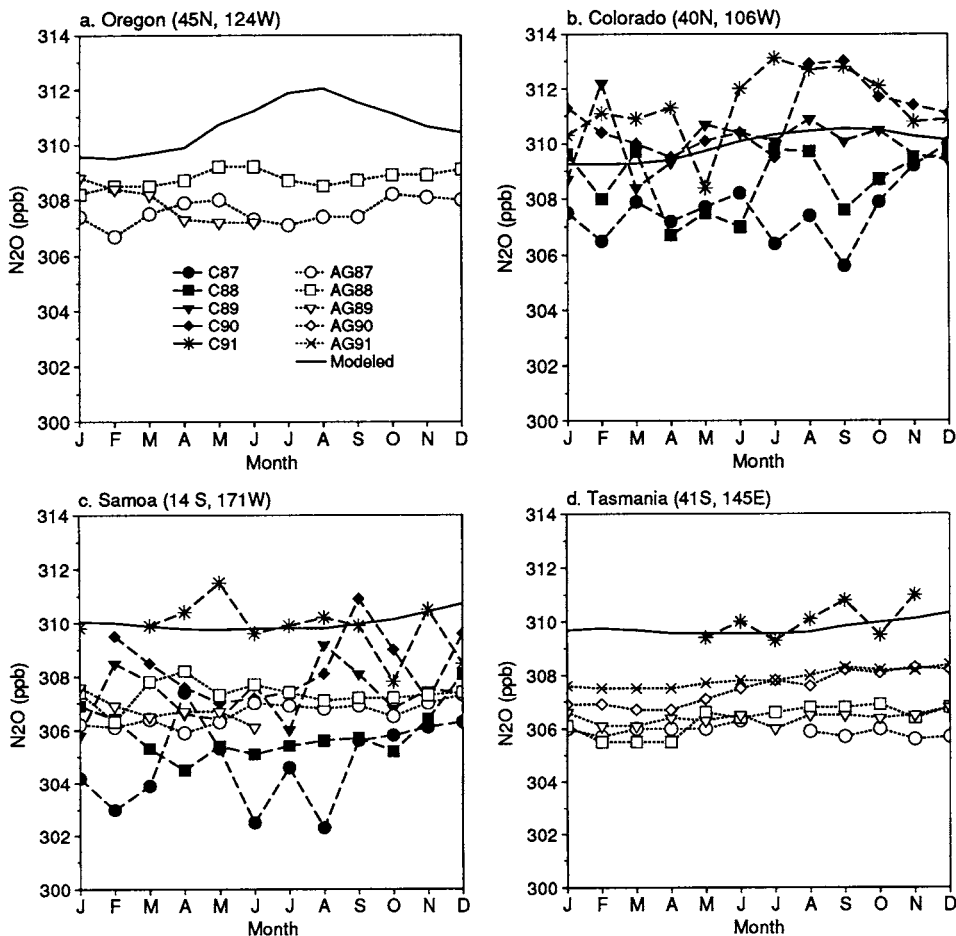


Figure 3. Monthly mean N_2O concentration for different years from Atmospheric Lifetime Experiment and its successor the Global Atmospheric Gases Experiment (AG) high frequency (4 - 12 measurements per day) real-time in situ gas chromatographic measurements (not corrected for pollution events)⁶ and Climate Monitoring and Diagnostics Laboratory (C) measurements in weekly flask samples⁷ as well as simulated monthly concentrations at the locations: Cape Meares, Oregon (a); Niwot Ridge, Colorado (b); American Samoa (c); Cape Grim, Tasmania (d).

Because the model correctly reproduces the latitudinal gradient and seasonality of CFC-11 and other tracers⁸ and is in general agreement with other N_2O modelling studies⁴, we assume that significant model inconsistency with atmospheric observations is not caused by biases in the model. The simulated seasonal variability of concentrations may be in error due to misrepresentations in the emission inventories.

Net annual consumption of N_2O in soils is reported for grasslands⁹ and temperate forest soils¹⁰, while other studies report episodic soil N_2O uptake^{11,12}. The uptake is attributed to consumption of N_2O by denitrification under conditions of low nitrification, leading to low nitrate concentrations⁹. In

nitrogen-limited cold temperate and boreal forests preferential immobilisation of N occurs in the forest floor and its associated decomposers¹³. Immobilisation and slow mineralisation of N may create conditions conducive to N₂O consumption. Uptake of N₂O may also occur in nitrogen-poor wetlands³. In our model, N₂O consumption in the spring to autumn period would lead to a lower seasonal amplitude in the NH.

The model used to calculate monthly N₂O emissions from soils (Table 1) was based on the relationships between N₂O fluxes and soil temperature and moisture, leading to the highest fluxes in temperate forests in spring and summer. However, elevated N₂O fluxes were observed in temperate forests in autumn^{14,15}, in arable lands during thaw periods¹⁶ and from snow covered soils¹⁷. These episodic emissions from vast temperate forest areas may account for an important global contribution, causing a decrease in variability of the simulated NH N₂O emission and concentrations.

Industrial N₂O emissions may not show a significant seasonality, but combustion-related N₂O emission may be higher in winter than in summer. Higher estimates for these sources prevailing in the NH¹⁸ could lead to a decrease in the simulated seasonal NH oscillation. The major part of the N₂O from other source candidates, such as industrial and chemical processes, medical and industrial use of N₂O, production and use of explosives, corona power losses from electric transmission lines, and coastal and freshwater systems, stems from the 30°N-90°N zone¹⁸. Emissions of N₂O from most of these sources probably lack seasonal patterns. Their inclusion would further reduce the simulated seasonal amplitude of the NH N₂O concentration.

The precision of the N₂O concentration measurements is 0.15% or 0.5 ppb, but the standard deviation in the daily and monthly averages is much higher^{6,7}. Different concentration measurements vary considerably in the amplitude and temporal pattern of the oscillations (Figure 3c). As CO₂ is considered not to interfere with N₂O measurements⁶ (J. Elkins, personal communication), the inconsistencies between different observations could be the result of other interferences (e.g. H₂O) in the N₂O concentration measurements. The predicted seasonal variations are in the order of 1 ppb for the NH and <1 ppb for the SH. Resolving an annually averaged latitudinal gradient may be testing the limits of current measurement technology. In addition, there is a lack of N₂O monitoring stations in continental interiors. Testing hypotheses on sources and sinks of atmospheric N₂O with the available observational data is therefore difficult.

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References

- 1 Khalil, M.A.K. & Rasmussen, R.A. *J. geophys. Res.* **97**, 14651-14660 (1992).
- 2 Swanson, T.H., Elkins, J.W., Butler, J.H., Montzka, S.A., Myers, R.C., Thompson, T.M., Baring, T.J., Cummings, S.O., Dutton, G.S., Hayden, A.H., Lobert, J.M., Holcomb, G.A., Sturges, W.T. & Gilpin, T.M. in *Climate Monitoring and Diagnostics Laboratory No. 21, Summary Report 1992* 59-75 (U.S. Department of Commerce, National Oceanic and Atmospheric Administration, Environmental Research Laboratories, Boulder, Colorado, 1993).
- 3 Bouwman, A.F., Fung, I., Matthews, E. & John, J. *Global biogeochem Cycles* **7**, 557-597 (1993).

- 4 Nevison, C. Cooperative PhD thesis, No. 147 (Stanford University and National Center for Atmospheric Research, 1994).
- 5 Taylor, J.A. *Mathematics and Computers in Simulation* **33**, 597-602 (1992).
- 6 Prinn, R., Cunbold, D., Rasmussen, R., Simmonds, P., Alyea, F., Crawford, A., Fraser, P. & Rosen, R. *J. geophys. Res.* **95**, 18369-18385 (1990).
- 7 Montzka, S.A., Elkins, J.W., Butler, J.H., Thompson, T.M., Sturges, W.T., Swanson, T.H., Myers, R.C., Gilpin, T.M., Baring, T.J., Cummings, S.O., Holcomb, G.A., Lobert, J.M. & Hall, B.D. in *Climate monitoring and Diagnostics Laboratory No. 20, Summary Report 1991* 60-81 (U.S. Department of Commerce, National Oceanic and Atmospheric Administration, Environmental Research Laboratories, Boulder, Colorado, 1992).
- 8 Taylor, J.A. in *Computer modelling in the environmental sciences* (eds D.G. Farmer & Rycroft, M.J.) 233-241 (Clarendon Press, Oxford, 1991).
- 9 Ryden, J.C. *J. Soil Sci.* **34**, 355-365 (1983).
- 10 Castro, M.S., Steudler, P.A., Melillo, J.M., Aber, J.D. & Millham, S. *Biogeochemistry* **18**, 119-135 (1993).
- 11 Keller, M., Goreau, T.J., Wofsy, S.C., Kaplan, W.A. & McElroy, M.B. *Geophys. res. Lett.* **10**, 1,156-1,159 (1983).
- 12 Bowden, R.D., Steudler, P.A., Melillo J.M., & Aber, J.D. *J. geophys. Res.* **95**, 13,997-14,005 (1990).
- 13 Vogt, K.A., Grier C.C. & Vogt, D.J. *Adv. ecol. Res.* **15**, 303-377 (1986).
- 14 Goodroad, L.L. & Keeney, D.R. *J. environ. Qual.* **13**, 448-452 (1984).
- 15 Schmidt, J., Seiler, W., & Conrad, R. *J. atmosph. Chem.* **6**, 95-115 (1988).
- 16 Dörsch, P., Flessa, H., & Beese, F. *Mitt. Deutschen Bodenkundl. Gesellsch.* **72**, 495-498 (1993).
- 17 Sommerfeld, R.A., Mosier, A.R. & Musselman, R.C. *Nature* **361**, 140-142 (1993).
- 18 Bouwman, A.F., van der Hoek, K.W. & Olivier, J.G.J. *J. geophys. Res.* (in the press).
- 19 Kreileman, G.J.J. & Bouwman, A.F. *Water air soil Pollut.*, **76** 231-258 (1994).
- 20 Leemans, R. & Van den Born, G.J. *Water air soil Pollut.* **76**, 133-161 (1994).
- 21 Olson, J.S., Watts, J.A. & Allison, L.J. ORNL 5862. Environmental Sciences Division Publication No.1997 (Oak Ridge National Laboratory, Tennessee, National Technical Information Service, U.S. Dept. Commerce, 1983).
- 22 Hao, W.M., Liu, M.H. & Crutzen, P.J. in *Fire in the Tropical Biota. Ecological Studies 84* (ed. J.G. Goldammer) 440-462 (Springer Verlag, Berlin, 1990).
- 23 Crutzen, P.J. & Andreae, M.O. *Science* **250**, 1669-1678 (1990).
- 24 Andreae, M.O. in *Global biomass burning* (ed. Levine, J.S.) 3-28 (MIT Press, Cambridge, U.S.A., 1991).
- 25 Matson, P.A. and Vitousek, P.M. *Bioscience* **40**, 667-672 (1990).
- 26 Vitousek, P.M., Matson, P., Volkman, C., Mass, J.M., and Garcia, G. *Global Biogeochem. Cycles* **3**, 375-382 (1989).