

## Corrections

### REVIEW

Correction for “A post-Kyoto partner: Considering the stratospheric ozone regime as a tool to manage nitrous oxide,” by David Kanter, Denise L. Mauzerall, A. R. Ravishankara, John S. Daniel, Robert W. Portmann, Peter M. Grabel, William R. Moomaw, and James N. Galloway, which appeared in issue 12, March 19, 2013, of *Proc Natl Acad Sci USA* (110:4451–4457; first published February 25, 2013; 10.1073/pnas.1222231110).

The authors note that on page 4454, left column, 2nd full paragraph, lines 7–9, “For example, oxidation catalysts are able to reduce N<sub>2</sub>O emissions ~70% compared with models without the technology (22)” should instead appear as “For example, advanced three-way catalysts are able to reduce N<sub>2</sub>O emissions ~65% compared with models without the technology (22).”

The authors also note that ref. 22 should appear as:

22. Eggleston HS, Buendia L, Miwa K, Ngara T, Tanabe K, eds (2006) *IPCC Guidelines for National Greenhouse Gas Inventories, Volume 2: Energy* (Institute for Global Environmental Strategies, Hayama, Japan), p 3.22.

[www.pnas.org/cgi/doi/10.1073/pnas.1317243110](http://www.pnas.org/cgi/doi/10.1073/pnas.1317243110)

### BIOPHYSICS AND COMPUTATIONAL BIOLOGY

Correction for “Charge interactions can dominate the dimensions of intrinsically disordered proteins,” by Sonja Müller-Späh, Andrea Soranno, Verena Hirschfeld, Hagen Hofmann, Stefan Rügger, Luc Reymond, Daniel Nettels, and Benjamin Schuler, which appeared in issue 33, August 17, 2010, of *Proc Natl Acad Sci USA* (107:14609–14614; first published July 16, 2010; 10.1073/pnas.1001743107).

The authors note that Eq. 5 appeared incorrectly. The corrected equation appears below.

$$R_g = N^{0.5} \frac{\alpha b}{\sqrt{6}} \left( 1 + \rho \frac{Ka}{1 + Ka} \right)$$

[www.pnas.org/cgi/doi/10.1073/pnas.1317338110](http://www.pnas.org/cgi/doi/10.1073/pnas.1317338110)

### MICROBIOLOGY

Correction for “Repurposing the antimycotic drug flucytosine for suppression of *Pseudomonas aeruginosa* pathogenicity,” by Francesco Imperi, Francesco Massai, Marcella Facchini, Emanuela Frangipani, Daniela Visaggio, Livia Leoni, Alessandra Bragonzi, and Paolo Visca, which appeared in issue 18, April 30, 2013, of *Proc Natl Acad Sci USA* (110:7458–7463; first published April 8, 2013; 10.1073/pnas.1222706110).

The authors note that the following statement should be added to the end of page 7461, right column, line 2:

“While exogenously provided 5-fluorouracil is toxic to *P. aeruginosa* (39), it has been found to inhibit several *P. aeruginosa* virulence-related traits at subinhibitory concentrations (40), though pyoverdine-dependent virulence gene expression was not previously shown as 5-fluorouracil target. Given that 5-fluorouracil affected *P. aeruginosa* growth, while flucytosine did not (see ref. 39 and this work), further studies are required to decipher the different specificities, impacts, and modes of action of flucytosine and 5-fluorouracil treatments on this bacterial pathogen.”

Additionally, the authors note that they omitted references to articles by West et al. and Ueda et al. The complete references appear below.

39. West TP, Chu CP (1986) Utilization of pyrimidines and pyrimidine analogues by fluorescent pseudomonads. *Microbios* 47(192-193):149–157.  
40. Ueda A, Attila C, Whiteley M, Wood TK (2009) Uracil influences quorum sensing and biofilm formation in *Pseudomonas aeruginosa* and fluorouracil is an antagonist. *Microb Biotechnol* 2(1):62–74.

[www.pnas.org/cgi/doi/10.1073/pnas.1316459110](http://www.pnas.org/cgi/doi/10.1073/pnas.1316459110)

# A post-Kyoto partner: Considering the stratospheric ozone regime as a tool to manage nitrous oxide

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Contributed by A. R. Ravishankara, December 31, 2012 (sent for review September 7, 2012)

**Nitrous oxide (N<sub>2</sub>O) is the largest known remaining anthropogenic threat to the stratospheric ozone layer. However, it is currently only regulated under the 1997 Kyoto Protocol because of its simultaneous ability to warm the climate. The threat N<sub>2</sub>O poses to the stratospheric ozone layer, coupled with the uncertain future of the international climate regime, motivates our exploration of issues that could be relevant to the Parties to the ozone regime (the 1985 Vienna Convention and its 1987 Montreal Protocol) should they decide to take measures to manage N<sub>2</sub>O in the future. There are clear legal avenues to regulate N<sub>2</sub>O under the ozone regime as well as several ways to share authority with the existing and future international climate treaties. N<sub>2</sub>O mitigation strategies exist to address the most significant anthropogenic sources, including agriculture, where behavioral practices and new technologies could contribute significantly to reducing emissions. Existing policies managing N<sub>2</sub>O and other forms of reactive nitrogen could be harnessed and built on by the ozone regime to implement N<sub>2</sub>O controls. There are several challenges and potential cobenefits to N<sub>2</sub>O control which we discuss here: food security, equity, and implications of the nitrogen cascade. The possible inclusion of N<sub>2</sub>O in the ozone regime need not be viewed as a sign of failure of the United Nations Framework Convention on Climate Change to adequately deal with climate change. Rather, it could represent an additional valuable tool in sustainable development diplomacy.**

climate gases | nitrogen cycle | stratospheric ozone depletion | global environmental governance

**N**itrous oxide (N<sub>2</sub>O) contributes to two distinct global environmental problems: climate change and stratospheric ozone layer depletion. It is currently the third most significant greenhouse gas (GHG) in terms of climate forcing after carbon dioxide (CO<sub>2</sub>) and methane (CH<sub>4</sub>) (1), and its current emissions will contribute more to stratospheric ozone depletion than the current emissions of any other substance (2). N<sub>2</sub>O is not alone in having these dual impacts—for example, chlorofluorocarbons (CFCs) also exacerbate both environmental problems, and CFC controls have reduced both ozone layer depletion and anthropogenic climate change. However, CFCs and N<sub>2</sub>O are controlled under different international treaties: CFCs under the universally accepted 1987 Montreal Protocol on Substances that Deplete the Ozone Layer and N<sub>2</sub>O under the 1997 Kyoto Protocol to the United Nations Framework Convention on Climate Change (UNFCCC). The latter has not been universally ratified, and its first phase commitments expired at the end of 2012. Although a second commitment period will run through 2020, its emissions reduction targets remain unclear, and the Protocol's membership has shrunk. Because N<sub>2</sub>O emissions are the largest remaining anthropogenic threat to the stratospheric ozone layer, this paper examines the issues that we regard as most relevant if the Parties to the ozone regime decide to consider future N<sub>2</sub>O controls. Our aim should not, however, be interpreted as implicitly endorsing such an outcome.

The ozone regime (the 1985 Vienna Convention and its 1987 Montreal Protocol) is widely regarded as the most effective international environmental institution ever established. The Montreal Protocol has reduced the production and consumption of the ozone-depleting substances (ODSs) it controls by 98% since its inception, while simultaneously delaying the growth of overall anthropogenic radiative forcing by an amount equivalent to 7–12 y of increased CO<sub>2</sub> emissions in the early 21st century (3). Its institutional architecture has elements that various Parties regard as essential to their participation. Every country in the world has ratified the Protocol, and all Parties have legally binding commitments (with developing countries given longer to comply with their commitments, which are often identical to developed country commitments). There is a strong financial mechanism, the Multilateral Fund, funded by developed countries that finances projects in developing countries to cover their incremental costs of complying with their Montreal Protocol commitments. There is also an enforcement mechanism restricting trade in ODS. Well-respected assessment panels, made up of experts from industry, government, international organizations, private consultancies, and academia, provide valuable information and advice to the Parties on the science and environmental effects of ozone depletion as well as the technical and economic feasibility of chemical and process alternatives. The Parties may wish to use this existing institutional architecture if they decide to adopt future N<sub>2</sub>O controls. For example, the Parties could request a scoping report from the Technology and Economics Assessment Panel on the technical and economic feasibility of specific N<sub>2</sub>O control strategies before deciding on a course of action (and possibly establish an N<sub>2</sub>O Technical Options Committee if they do decide to adopt controls) (4). Likewise, the Multilateral Fund could apply general lessons learned from successful projects in the agricultural sector that helped farmers adopt methods to reduce or replace use of the pesticide methyl bromide (5).

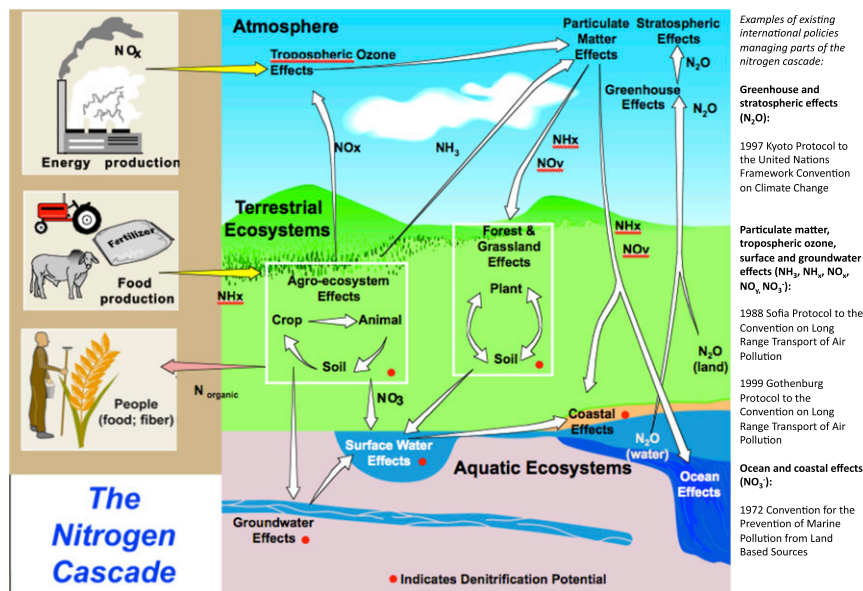
N<sub>2</sub>O is a part of the tightly coupled nitrogen (N) cycle. Increases in anthropogenic emissions have come mainly from agriculture because of the biogeochemical processes of nitrification and denitrification, with additional contributions from stationary and mobile combustion, biomass burning, nitric and adipic acid production, and wastewater treatment (6). The N cycle is best characterized as a chemical cascade, with one N atom able to transform readily among different forms (7) (Fig. 1). As a result, N [specifically, reactive nitrogen (Nr)—all N compounds except N<sub>2</sub>] can contribute to a myriad of environmental problems. In principle, therefore, it can be controlled at a number of points along

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**Fig. 1.** Illustration of the nitrogen cascade showing the sequential effects that a single atom of N can have in various reservoirs after it has been converted from nonreactive  $N_2$  to a reactive form (yellow arrows) and examples of existing international management policies. Adapted with permission from the GEO Yearbook 2003, United Nations Environment Programme (UNEP), 2004 (8) which was based on Galloway et al., 2003 (7).

the cascade. For example, nitrogen oxides ( $NO_x = NO + NO_2$ ), nitrate ( $NO_3^-$ ), and  $N_2O$  are all forms of Nr, which can be controlled as air, water, and ozone and climate pollutants, respectively. Because of the cascade effect, reductions in one form of Nr likely reduce total Nr levels (9). Consequently, controlling  $N_2O$  provides environmental cobenefits (e.g., improved air and water quality) in addition to the direct benefits of reducing ozone depletion and climate change.

The following sections examine the scientific, legal, technical and policy-related issues surrounding potential  $N_2O$  controls under the ozone regime.

### $N_2O$ —Atmospheric Abundance and Ozone-Depleting Properties

**$N_2O$  Emission Sources.** With natural emissions assumed to have remained unchanged, it is believed that anthropogenic activity alone is responsible for the ~20% increase in atmospheric  $N_2O$  concentrations [from 270 to 325 parts per billion by volume (ppbv)] since 1860 (8). Natural emissions are estimated at 10.2 Tg N per year (10) compared with current anthropogenic emissions estimates of 5.5–8.2 Tg N per year (6, 10–12). Consequently, the current stratospheric photochemical sink of  $N_2O$  of ~13.3 Tg N per year (given an atmospheric abundance of 325 ppbv and an atmospheric lifetime of 119 y) is not large enough to offset total annual emissions of 15.7–18.4 Tg N (12); the atmospheric abundance of  $N_2O$  continues to increase as a result. Although the magnitude of total  $N_2O$  anthropogenic emissions is reasonably well-known, attributing anthropogenic  $N_2O$  emissions to various activities or sectors has significant uncertainties.  $N_2O$  from natural and agricultural sources is a product of denitrification (the transformation of  $NO_3^-$  into  $N_2$ ) and nitrification [the transformation of ammonium ( $NH_4^+$ ) into  $NO_3^-$ ] (13).  $N_2O$  is also emitted as an industrial byproduct from other anthropogenic sources (*Feasibility of  $N_2O$  Reductions: Sector-By-Sector Emissions and Mitigation Opportunities*). Source attribution is difficult, because the larger sources are diffuse, variable in time, dispersed across the globe, and occur above a large natural background. In addition, new anthropogenic sources of  $N_2O$ , such as aquaculture, may be growing rapidly (14). Uncertainty in the magnitude of individual sources makes it challenging to predict

emission reductions from specific mitigation actions, particularly in agriculture.

**Contribution of  $N_2O$  to Stratospheric Ozone Layer Depletion.** Both natural and anthropogenic  $N_2O$  emissions are transported to the stratosphere, where ~10% is oxidized by excited atomic oxygen [ $O(^1D)$ ] to form nitric oxide (NO), which together with nitrogen dioxide ( $NO_2$ ), forms a catalytic cycle for ozone destruction (15). There is a balance between ozone production and loss, with  $NO_x$  catalysis a major ozone loss process in the mid- to upper stratosphere. Indeed,  $NO_x$  was found to catalyze stratospheric ozone destruction several years before similar concerns emerged about CFCs (16).

Ravishankara et al. (2) calculated an ozone depletion potential (ODP) for  $N_2O$ . ODP is a metric used to evaluate the efficacy of a species' stratospheric ozone destruction relative to CFC-11. Ravishankara et al. (2) reported an ODP for  $N_2O$  of ~0.02 (i.e., 1 kg  $N_2O$  emissions destroy ~2% of the stratospheric ozone that 1 kg CFC-11 emissions destroy). This value changes slightly with atmospheric conditions (*SI Text*) but is comparable with the ODPs of several substances already controlled under the Montreal Protocol [e.g., hydrochlorofluorocarbon-123 (0.02)].  $N_2O$ 's small ODP can be deceptive, because the mass of  $N_2O$  emitted from anthropogenic activities is much larger than past or projected future CFC emissions (e.g., in 2008, ODP-weighted emissions of  $N_2O$  were approximately double the ODP-weighted emissions of CFC-11). Ravishankara et al. (2) note that, based on ODP-weighted emissions, anthropogenic  $N_2O$  was the fourth most important ODS at the height of CFC emissions, is the most important ODS emitted today, and is projected to remain the most important throughout the 21st century. Furthermore, Ravishankara et al. (2) suggest that, by 2050,  $N_2O$  ODP-weighted emissions could be as large as one-third of the peak of the CFC ODP-weighted emissions, which occurred in the late 1980s. As a result,  $N_2O$  emissions are currently the largest remaining anthropogenic threat to the stratospheric ozone layer.

Considering together future projections of  $CO_2$ ,  $CH_4$ , and  $N_2O$  increases and ODS decreases, it is estimated that the ozone layer should eventually return to its pre-1980 levels, despite continued emissions of  $N_2O$  and other ODSs (17). However, the Parties to the Montreal Protocol continue to consider and pursue ways to

accelerate ozone recovery, such as the destruction of ODS banks and the accelerated hydrochlorofluorocarbon phase out, partly because these actions would deliver both ozone and climate benefits (18). Controlling N<sub>2</sub>O would also bring these cobenefits, because in addition to its ozone-destroying properties, N<sub>2</sub>O is the third most significant GHG behind CO<sub>2</sub> and CH<sub>4</sub>. Because of the N cascade phenomenon, N<sub>2</sub>O controls would likely also deliver air and water quality benefits caused by reduced NO<sub>3</sub><sup>-</sup> leakage and ammonia (NH<sub>3</sub>) volatilization. Moreover, the pre-1980 ozone levels used as a benchmark by the Parties to the Montreal Protocol do not represent a world unaffected by the impacts of ozone depletion, because anthropogenic ozone depletion was occurring well before 1980 (19). Even if anthropogenic N<sub>2</sub>O emissions were fully eliminated tomorrow, N<sub>2</sub>O-induced ozone loss would continue for centuries because of its long atmospheric lifetime, with the associated adverse impacts on human health and natural ecosystems. Consequently, there are still significant environmental benefits to be gained from a reduction in N<sub>2</sub>O emissions, including for the ozone layer.

### Legal Authority of the Ozone Regime

N<sub>2</sub>O is currently included under the UNFCCC and its Kyoto Protocol (Article 4, UNFCCC; Articles 2, 5, and 7 and Annex A-B, Kyoto Protocol) as a GHG. However, as outlined in the previous section, current N<sub>2</sub>O emissions pose the greatest remaining anthropogenic threat to the stratospheric ozone layer. Moreover, with the first phase of binding commitments under the Kyoto Protocol expiring at the end of 2012, uncertainty surrounding what a second Kyoto Protocol commitment period from 2013 to 2020 will entail, and no possibility of a new agreement coming into force before 2015, there is increasing uncertainty as to how the GHGs currently in the Kyoto basket will be regulated in the future. Consequently, it is reasonable to explore the possibility of measures to manage N<sub>2</sub>O under the ozone regime and how they might interact with existing and future international climate regimes.

**Legal Authority to Regulate N<sub>2</sub>O.** Neither the 1985 Vienna Convention (Ozone Convention) nor the Montreal Protocol defines ODSs (Article 1, Ozone Convention and Montreal Protocol). However, the Parties list N<sub>2</sub>O as one of the chemicals that modifies “the chemical and physical properties of the ozone layer” in the Ozone Convention [Article 3 and Annex I at (4)(b), Ozone Convention]. Coupled with the threat N<sub>2</sub>O poses to the stratospheric ozone layer, this listing suggests that it could legally be classified as an ODS for purposes of the Ozone Convention and Montreal Protocol.

The Ozone Convention commits the Parties to regulate all human activities that “have or are likely to have adverse effects resulting from modification or likely modification of the ozone layer” without distinction between products and byproducts [Article 2(2)(b), Ozone Convention]. Therefore, although the Montreal Protocol has previously only controlled ODS production and consumption (whereas N<sub>2</sub>O is mostly an unwanted byproduct, its intentional production and consumption being very limited), the Parties could regulate N<sub>2</sub>O if they choose to do so.

Moreover, the Montreal Protocol’s historical focus on production and consumption could provide advantages to reducing agricultural N<sub>2</sub>O emissions in particular: such an approach would allow a focus on fertilizer consumption rather than N<sub>2</sub>O emissions—the method currently used under the climate regime. Emphasizing consumption would likely be a more manageable (and measurable) approach to tackling the problem than attempting to control emissions. N<sub>2</sub>O emissions from agriculture are spatially and temporally heterogeneous and difficult to measure, whereas data on fertilizer production and consumption are readily available (20, 21). N<sub>2</sub>O emissions could then be derived from fertilizer consumption data by emissions factors (22) and biogeochemical computer models

(23). Although the default emission factors for direct and indirect emissions from fertilizer and manure published by the Intergovernmental Panel on Climate Change are generally consistent with the global budget of N<sub>2</sub>O, a key research objective is to find a metric that adequately estimates N<sub>2</sub>O emissions from fertilizer consumption on more local scales.

**How N<sub>2</sub>O Could Be Regulated Under the Ozone Regime.** There are two ways the Parties could currently regulate N<sub>2</sub>O under the Ozone Convention. First, the Parties to the Montreal Protocol could choose to amend the treaty to add N<sub>2</sub>O as a controlled substance and establish control measures to reduce N<sub>2</sub>O emissions [Article 2 (10), Montreal Protocol]. Second, the Parties to the Ozone Convention could choose to adopt and ratify a new protocol under Article 8 of the Ozone Convention tailored to specifically regulate N<sub>2</sub>O (Articles 2 and 8, Ozone Convention) (a detailed analysis of these two options is in the *SI Text*).

**Interaction with the Existing Climate Regime.** Establishing control measures for N<sub>2</sub>O under the ozone regime is consistent with the UNFCCC’s principles and ultimate objective (Articles 2–3, UNFCCC). The UNFCCC explicitly directs its Parties to “seek and utilize, where appropriate, the services and cooperation of, and information provided by, competent international organizations and intergovernmental ... bodies” such as the Montreal Protocol [Article 7 (2)(1), UNFCCC]. The Parties to its Kyoto Protocol have already implemented this mandate by delegating responsibility for reducing GHG emissions from two entire sectors of the economy, aviation and bunker fuels, to the International Civil Aviation Organization and the International Maritime Organization, respectively [Article 2 (2), Kyoto Protocol]. Legal provisions could be added to both treaties to reduce potential conflicts of authority, which was suggested in discussions of the proposed hydrofluorocarbon amendments to the Montreal Protocol (24) (*SI Text*).

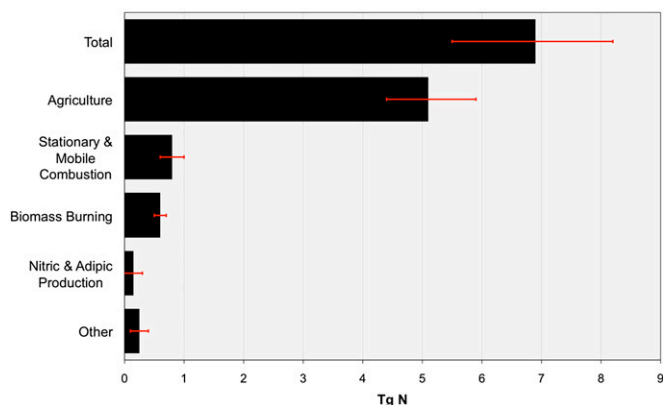
**Interaction with the Post-2012 Climate Regime.** The details of the Kyoto Protocol’s second commitment period are still being negotiated, with 2015 now the target date for securing a new agreement to succeed the Kyoto Protocol. This negotiation schedule means that, from a legal perspective, the post-2012 climate regime could be tailored to accommodate N<sub>2</sub>O control measures under the ozone regime with very little difficulty. Thus, not only is N<sub>2</sub>O regulation under the ozone regime legally possible, but working toward an agreement now would likely be more straightforward than after any potential new climate agreement is in place.

### Feasibility of N<sub>2</sub>O Reductions: Sector-By-Sector Emissions and Mitigation Opportunities

The majority of anthropogenic N<sub>2</sub>O is emitted from agriculture, stationary and mobile combustion, biomass burning, and nitric and adipic acid production (Fig. 2). We, therefore, focus on these sectors in this section (although other sectors, such as wastewater, are developing methods to reduce N<sub>2</sub>O emissions as well) (25, 26). Each sector has introduced (or is in the process of developing) technologies and/or practices to reduce emissions. We outline some mitigation opportunities below, starting with the smaller sources and concluding with agriculture, where future mitigation efforts may yield the largest reductions and yet, could also prove to be the most difficult to implement.

**Nitric/Adipic Acid Production.** Together, nitric and adipic acid production are responsible for less than 4% of global N<sub>2</sub>O emissions (11, 27). Nitric acid is mainly used as a feedstock for synthetic fertilizer, with additional uses in the production of explosives and adipic acid. N<sub>2</sub>O forms as a result of ammonia oxidation, the first stage in nitric acid production (22). Adipic acid is primarily used

### Anthropogenic N<sub>2</sub>O Emissions by Sector



**Fig. 2.** Sector-by-sector contribution to anthropogenic N<sub>2</sub>O emissions in 2005. Smaller sources, such as wastewater treatment and aquaculture, are included in the "Other" bar. Error bars represent the range of leading estimates taken from US Environmental Protection Agency (6), Davidson (10), Syakila and Kroeze (11), and Crutzen et al. (12) with the total error bar representing the sum of the individual error bars.

as a feedstock in synthetic fiber production, with N<sub>2</sub>O formation the result of using nitric acid to oxidize cyclohexanone and/or cyclohexanol (28). Abatement technologies exist for both nitric and adipic acid production, most notably catalytic reduction techniques, with reduction efficiencies of up to 98% (22), and they have been (or will soon be) installed in most developed countries (*SI Text*).

**Biomass Burning.** Biomass burning accounts for 8–9% of global N<sub>2</sub>O emissions (10, 11), with emissions from both the oxidation of atmospheric N<sub>2</sub> and organic N in the biomass. Major sources include burning involved in forest to cropland conversion, burning of agricultural waste, and wildfires (29). Mitigation opportunities vary greatly depending on the timing and location of the burning, resource management objectives, vegetation type, and available resources. They include reducing the acreage burned (for example, through conservation tillage or burning in alternate years), wildfire management through prevention education and fuel management, and improving burning practices to increase combustion efficiency and reduce fuel consumption and loading (30).

**Stationary and Mobile Combustion.** Stationary and mobile combustion account for 8–14% of global N<sub>2</sub>O emissions (6, 11). N<sub>2</sub>O from mobile sources is largely formed by catalytic converters used to control NO<sub>x</sub>, carbon monoxide (CO), and hydrocarbons in tailpipe emissions (31) (*SI Text*). Technical improvements to reduce N<sub>2</sub>O emissions are being gradually introduced into new car models. For example, oxidation catalysts are able to reduce N<sub>2</sub>O emissions ~70% compared with models without the technology (22). Stationary combustion sources (mainly coal power plants) emit N<sub>2</sub>O caused by the oxidation of both atmospheric N<sub>2</sub> and organic N in the fuel. Emissions depend on the amount of organic N in the fuel and temperature and oxygen levels during combustion (32). Technologies and practices to reduce emissions include a variety of selective catalytic reduction techniques (33), with N<sub>2</sub>O reduction efficiencies of up to 80% (34).

**Agricultural Emissions.** N<sub>2</sub>O from agriculture accounts for 62–84% of anthropogenic emissions (10–12), which are driven mostly by the large application of synthetic N fertilizer and manure to farmland. The central cause of N<sub>2</sub>O emissions from agriculture is a lack of synchronization between soil availability and crop demand for N—with, on average, ~50% of N applied to agricul-

tural soils lost to the environment (35). The ultimate control on emissions would be a decrease in the amount of Nr created to grow food, because eventually, the majority of Nr added to the soils is lost to the environment. This lost Nr has the potential to undergo nitrification and denitrification, the two main microbial processes that produce N<sub>2</sub>O. There are both behavioral and technological strategies that can partially remedy this dislocation and could reduce N<sub>2</sub>O emissions substantially.

**Behavioral practices.** Behavioral practices focus on using fertilizer more efficiently by (i) applying the right type in the right amount in the right place at the right time (36), (ii) lessening the impacts of fertilizer after it escapes the farm (in its various chemical forms), and (iii) educating the consumer about N-smart consumption practices. Implementing these practices involves considerable educational efforts, particularly in developing countries, and in some cases, additional technology to precisely pinpoint N requirements within a field is needed. Many of these practices incur little or zero additional cost to the farmer, because they lead to lower fertilizer use and/or increased yields, potentially increasing profit margins as a result (6, 37)—although they have to contend with the predominant view among many farmers that the risks of yield decline from underapplication outweigh the costs of fertilizer overapplication (38). Another dimension of behavior to consider is the consumption of agricultural products. There are significant changes to consumer behavior, including a decrease in meat consumption and food wastage, that could mitigate Nr pollution considerably (39). Tools such as the Nitrogen Footprint Calculator have been shown to be effective in educating consumers about how their resource choices contribute to Nr losses to the environment (40). Recommended behavioral practices include (more details in *SI Text*):

- Use of cover crops and residue recycling to reduce and reuse surplus Nr.

- Improvement of the timing and placement of fertilizer application to better synchronize crop N demand with soil supply.

- Watershed management to stimulate complete denitrification to N<sub>2</sub>.

- Livestock management by altering animal diets and implementing responsible waste management practices.

**Technological options.** Technological advancements in improving fertilizer use efficiency are advantageous, because farmers are not required to change the way that they apply fertilizer N—the remedy is embedded in the product (although they are often most effective combined with behavioral practices). The Intergovernmental Panel on Climate Change Third Assessment Report estimated that these technologies could potentially cut global N<sub>2</sub>O emissions from N fertilizer use by 30% (41). A more recent metaanalysis of field trials estimates that two of the most widely used technologies to improve fertilizer use efficiency, nitrification inhibitors and controlled-release fertilizers, reduce N<sub>2</sub>O emissions by ~35% on average (42) (*SI Text*). Their use is growing, particularly in intensive agricultural regions such as the Midwest United States, where the gains from N use efficiency can outweigh the purchase costs (43). Wood chip-filled trenches, known as denitrifying bioreactors, also show promise by increasing complete denitrification to N<sub>2</sub> (44, 45).

Research into genetically engineering crops to improve their N use efficiency is ongoing (46). Recent papers have shown that, with the same amount of N fertilizer, certain genetically engineered crops could increase yields by over 30% compared with traditional crops (47). However, genetically engineering N use efficiency is a controversial issue, with a number of skeptics (48).

## Policies Currently Influencing N<sub>2</sub>O Emissions

Policies currently exist at the local, national, regional, and international level that directly or indirectly control N<sub>2</sub>O emissions. These initiatives could potentially be harnessed into a comprehensive global effort to reduce N<sub>2</sub>O emissions.

**Controls.** The Kyoto Protocol to the UNFCCC is the only international agreement that currently lists N<sub>2</sub>O as a controlled substance. Although its Clean Development Mechanism has issued credits for projects reducing industrial N<sub>2</sub>O emissions (e.g., nitric/adipic acid production), none has been issued for agriculture-related projects. National climate policies, such as the United Kingdom Climate Change Act and the Australian Carbon Tax, also cover N<sub>2</sub>O, although with no elements focused specifically on N<sub>2</sub>O, because it is part of a basket of GHGs.

To the best of our knowledge, four policies, at various levels of government, explicitly aim to reduce N<sub>2</sub>O emissions from agriculture: the Alberta Quantification Protocol for Agricultural N<sub>2</sub>O Emission Reductions in Canada, Australia's Carbon Farming Initiative, and protocols under the American Carbon Registry and the US Climate Action Reserve (passed in 2010, 2011, and 2012, respectively), all of which issue credits for on-farm reductions of N<sub>2</sub>O emissions.

In terms of nonagricultural N<sub>2</sub>O emissions, the European Union Emissions Trading Scheme will begin issuing credits for N<sub>2</sub>O reductions at nitric acid plants beginning in 2013 (49). These credits will contribute toward reaching their 21% GHG reduction target by 2020 (from a 2005 baseline). The US Environmental Protection Agency and the Department of Transportation recently set a cap on tailpipe emissions of 0.010 g per mile for N<sub>2</sub>O emissions starting in 2017 model passenger vehicles and light trucks (although current emissions are already significantly below this cap) (50).

**Indirect Controls.** Certain policies indirectly reduce N<sub>2</sub>O emissions by controlling other forms of Nr. These policies could be important as an additional infrastructure already in place for reducing N<sub>2</sub>O emissions, although their focus is on other forms of Nr pollution. They include the 1991 European Union Nitrate Directive (now part of the 2000 Water Framework Directive), which sets limits on the use of N fertilizer in nitrate vulnerable zones and in turn, reduces the potential for N<sub>2</sub>O emissions in waterways downstream of leaching (51). Member states, such as The Netherlands, that have implemented this Directive have seen reductions in N<sub>2</sub>O emissions of up to 30% (52). The Chesapeake Bay watershed has a similar focus, with the US Environmental Protection Agency recently setting a target of a 25% reduction below current levels of Nr pollution entering the watershed by 2025 (53).

The Convention on the Long Range Transport of Air Pollution and its 1988 Sofia Protocol and 1999 Gothenburg Protocol set national limits on NO<sub>x</sub> and NH<sub>3</sub> emissions for its Parties (which include most countries in Europe, the United States, and Canada but no countries outside the OECD). These controls, in turn, will likely reduce N<sub>2</sub>O emissions through the N cascade. The Long Range Transport of Air Pollution's Task Force on Reactive Nitrogen focuses on strategies for reducing Nr pollution, and includes an expert panel on Nr and climate change.

## Challenges and Cobenefits to Controlling Agricultural N<sub>2</sub>O Emissions

There are at least three primary challenges and cobenefit opportunities if the ozone regime, or any other international forum, is to control agricultural N<sub>2</sub>O emissions: how to reduce N<sub>2</sub>O without adding to food security concerns, how to ensure reduction targets are equitable across nations, and how to responsibly manage the tightly coupled nature of the N cycle.

**Food Security.** It is impossible to produce food without N inputs of some kind given its fundamental role in protein generation (54).

Thus, even a phase-down, rather than the traditional Montreal Protocol approach of phasing-out specific chemicals (in this case, certain N fertilizers), would be exceedingly difficult. If N<sub>2</sub>O is to be reduced without adding to food security concerns, methods must be developed to measure and implement reductions of N<sub>2</sub>O emissions in ways that preserve agricultural yields while reducing additional forms of Nr pollution to the surrounding environment. An important avenue to explore, therefore, is how to increase the efficiency of fertilizer use. There are a number of metrics for fertilizer use efficiency, each with a slightly different focus (55). Use of these metrics may provide an opportunity to maintain food security while reducing N<sub>2</sub>O emissions and other environmental impacts resulting from excess Nr in the environment.

**Equity.** The high degree of heterogeneity in the global distribution of fertilizer use has important implications for how N<sub>2</sub>O controls could be implemented. In economies across Europe, North America, South America, and Asia, fertilizer is widely and abundantly used. By contrast, sub-Saharan Africa greatly underfertilizes much of its land because of a lack of infrastructure (e.g., paved roads), farmer education, and affordable fertilizer (56). The priority in Africa is to increase yields substantially and distribute those yields more effectively. Existing, widely applied, and cost-effective technologies would be sufficient to achieve these needed improvements (57).

Consequently, agricultural N<sub>2</sub>O controls require a nuanced regulatory approach. This nuance could be encapsulated in, for example, a per capita threshold of fertilizer use, below which a country is exempt from controls until that threshold has been crossed. This approach would allow countries in regions such as sub-Saharan Africa to improve agricultural productivity and thus, their food security without constraints on fertilizer application. Although such a rule would allow considerable growth in fertilizer use in these countries (accompanied by increased N<sub>2</sub>O emissions and other Nr pollution from these regions), the level of fertilizer use in regions like sub-Saharan Africa is so small relative to more industrialized regions (58) that an increase in per capita consumption is likely both necessary for food security and environmentally feasible given that greater reductions can currently be achieved in more industrialized regions.

**Nitrogen Cascade.** Controlling N<sub>2</sub>O involves managing an important and unique facet of the N cycle which sets it apart from other biogeochemical cycles and requires a more nuanced regulatory approach. Because of the facile conversion among its different oxidative states, one N atom can cascade through a variety of chemical forms, each with a different effect on the environment. For example, if urea is applied to a field, a portion of it can be volatilized as NH<sub>3</sub>, which can then react to form ammonium sulfate [(NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub>] and NO<sub>3</sub><sup>-</sup> aerosols, constituents of fine particulate matter that reduce visibility and have adverse impacts on human health. When removed from the atmosphere in precipitation, Nr can be oxidized to NO<sub>3</sub><sup>-</sup> (through nitrification with some N<sub>2</sub>O production), where it can eutrophy surface water and contaminate aquifers. NO<sub>3</sub><sup>-</sup> can be converted to N<sub>2</sub>O through denitrification and emitted to the atmosphere, leading to increased radiative forcing and stratospheric ozone depletion. This multitude of potential environmental impacts poses a unique challenge to policymakers when deciding at what point to intervene in the N cascade. Ideally, it is at the point of Nr formation (e.g., the Haber-Bosch process). It can also be where cost minimization and optimal environmental protection coincide (e.g., precluding future healthcare costs caused by air pollution) (59, 60). Formulating N<sub>2</sub>O controls for the agriculture sector around the concept of fertilizer efficiency (i.e., better synchronizing soil N supply with crop N demand) could take this aspect of the N cycle into account, not only complementing the ozone regime's historical focus on controlling production and consumption (*Legal*

Authority of the Ozone Regime, Legal Authority to Regulate  $N_2O$ ) but also providing a potentially effective way to prevent a damaging N cascade. This approach, in turn, would help mitigate climate change and air and water pollution in addition to protecting the stratospheric ozone layer.

## Conclusion

This paper has explored the central issues that would surround a potential decision of the Parties to the ozone regime to control  $N_2O$ . Although the science underpinning  $N_2O$ 's ozone destructiveness is complex, it is well-understood, and  $N_2O$  is now the largest remaining anthropogenic threat to the stratospheric ozone layer. Moreover,  $N_2O$  reductions could deliver significant climate cobenefits because of its potency as a GHG.  $N_2O$  reductions could also reduce the prevalence of other forms of Nr pollution, such as  $NO_3^-$ -induced eutrophication and  $NO_x$  and  $NH_4^+$  linked production of fine particulate and ozone air pollution, due to the N cascade phenomenon. The legal authority of both the Montreal Protocol and its parent convention to control  $N_2O$  is clear, and a partnership with the existing and future international regimes is a real possibility. Mitigation strategies exist across all major sectors, although there are particularly acute challenges in agriculture caused by the diffuse nature of emissions and the need to balance food security concerns. There are a number of existing policies that could be harnessed to implement

$N_2O$  controls if the Parties to the ozone regime were to decide to control it. Finally, given how essential N is in food production and the ease with which it can contribute to a myriad of environmental problems, controlling  $N_2O$  requires a nuanced approach that any international agreement would find challenging.

If the Parties to the ozone regime were to decide to control  $N_2O$ , it need not be viewed as symptomatic of a failure of the UNFCCC to adequately deal with climate change. Climate change is an exceptionally multifaceted, challenging issue, and—as anticipated by the UNFCCC itself—multiple regimes may need to act simultaneously to address it. Moreover, given how the driving forces behind climate change and other environmental problems often overlap, it may be wise to widen the scope of tools and institutions to approach these issues. Consequently, if  $N_2O$  is transferred to the ozone regime, it could mark a potentially welcome expansion of the horizons of sustainable development diplomacy.

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