Global health benefits of mitigating ozone pollution with methane emission controls

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Methane (CH_4) contributes to the growing global background concentration of tropospheric ozone (O_3), an air pollutant associated with premature mortality. Methane and ozone are also important greenhouse gases. Reducing methane emissions therefore decreases surface ozone everywhere while slowing climate warming, but although methane mitigation has been considered to address climate change, it has not for air quality. Here we show that global decreases in surface ozone concentrations, due to methane mitigation, result in substantial and widespread decreases in premature human mortality. Reducing global anthropogenic methane emissions by 20% beginning in 2010 would decrease the average daily maximum 8-h surface ozone by ≈1 part per billion by volume globally. By using epidemiologic ozone-mortality relationships, this ozone reduction is estimated to prevent ≈30,000 premature all-cause mortalities globally in 2030, and ≈370,000 between 2010 and 2030. If only cardiovascular and respiratory mortalities are considered, ≈17,000 global mortalities can be avoided in 2030. The marginal cost-effectiveness of this 20% methane reduction is estimated to be ≈$420,000 per avoided mortality. If avoided mortalities are valued at $1 million each, the benefit is ≈$240 per tonne of CH_4 (≈$12 per tonne of CO_2 equivalent), which exceeds the marginal cost of the methane reduction. These estimated air pollution ancillary benefits of climate-motivated methane emission reductions are comparable with those estimated previously for CO_2. Methane mitigation offers a unique opportunity to improve air quality globally and can be a cost-effective component of international ozone management, bringing multiple benefits for air quality, public health, agriculture, climate, and energy.

human health | mortality | tropospheric ozone | air quality

Tropospheric ozone (O_3) is an oxidant that damages agriculture, ecosystems, and materials. Ozone also adversely affects human health and has been associated in epidemiologic studies with daily premature mortality (1–10). Surface O_3 concentrations have historically increased in both polluted and remote regions and now frequently exceed regulatory standards (11–14). Global background surface O_3 concentrations have roughly doubled since preindustrial times (15), primarily because of increases in anthropogenic emissions of nitrogen oxides (NO_x) and methane (CH_4) (16), and are projected to continue to increase (17, 18).

Tropospheric O_3 is formed from photochemical reactions involving NO_x and volatile organic compounds (VOCs). Although nonmethane VOCs are the dominant anthropogenic VOCs contributing to O_3 formation in polluted regions, CH_4 is the primary anthropogenic VOC in the global troposphere (19). Because CH_4 reacts slowly (lifetime of 8–9 yr), it affects global background concentrations of O_3. Because this background underlies the O_3 produced on urban and regional scales, CH_4 mitigation reduces O_3 concentrations by roughly the same amount in polluted regions as in rural regions (19, 20).

Methane and O_3 are also greenhouse gases, which rank behind only carbon dioxide (CO_2) in anthropogenic radiative forcing of climate (21). Consequently, abatement of CH_4 emissions both reduces surface O_3 concentrations everywhere and slows greenhouse warming (19, 20). Methane abatement has been considered a low-cost means of addressing climate change (22, 23), particularly to influence the short-term rate of climate change. However, CH_4 abatement has not been considered for air quality management, mainly because O_3 pollution has traditionally been considered a local and regional problem, and the local benefits of local CH_4 reductions are small.

Here we examine the global reduction in O_3 and consequent decrease in premature human mortalities resulting from CH_4 emission controls. We first estimate the global decrease in surface O_3 concentration due to CH_4 mitigation, using the MOZART-2 global three-dimensional tropospheric chemistry-transport model (24, 25). This spatial distribution of O_3 is then overlaid on projections of population, and avoided premature mortalities are estimated by using daily O_3-mortality relationships from epidemiologic studies (6–9). Results are presented as the number of avoided premature mortalities due to the CH_4 reduction, the marginal cost-effectiveness per avoided mortality (using the marginal cost of CH_4 mitigation), and the monetized benefit per tonne of CH_4 reduced [using a value of a statistical life (VSL)].

Response of Global Surface Ozone to Methane Mitigation

Methods. We consider a CH_4 emission reduction of 65 $Mt{\cdot}yr^{-1}$ (1 Mt = 10^9 kg) (≈20% of current global anthropogenic emissions), which is assumed to be immediate in 2010 and sustained relative to the Intergovernmental Panel on Climate Change Special Report on Emissions Scenarios (SRES) A2 scenario (26) until 2030. A compilation of global CH_4 abatement options in five industrial sectors (27) suggests that 65 $Mt{\cdot}yr^{-1}$ can be reduced by 2010 at a net cost savings, using identified abatement options.

The MOZART-2 simulations use uniform global mixing ratios of CH_4, and spatially and temporally distributed emissions of other O_3 precursors, as other studies have done (19, 28). We conduct four simulations with MOZART-2, as shown in Table 1. Simulations I and III use CH_4 mixing ratios and emissions of other O_3 precursors as specified for the Intergovernmental Panel on Climate Change AR-4 2000 and 2030 A2 atmospheric chemistry experiments (29). In the CH_4 reduction cases (simulations II and IV), the decreased CH_4 mixing ratios are the steady-state mixing ratios resulting from a 65 $Mt{\cdot}yr^{-1}$ emission

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Abbreviations: CR, cardiovascular and respiratory; PM, particulate matter; ppbv, part(s) per billion by volume; VOC, volatile organic compound; VSL, value of a statistical life.

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Table 1. Four MOZART-2 simulations conducted in this study

Simulation	Fixed CH_4 mixing ratio, ppbv	Global anthropogenic NO_x emissions, $Mt \cdot yr^{-1}$ as NO_2
I: 2000 base case	1,760	124.8
II: 2000 CH_4 reduction	1,460*	124.8
III: 2030 A2	2,163	212.7
IV: 2030 A2, CH_4 reduction	1,865*	212.7

*Fixed global CH_4 mixing ratios at steady state, corresponding to an emission reduction of 65 $Mt \cdot yr^{-1}$ of CH_4.

reduction versus the corresponding base cases (simulations I and III), assuming a CH_4 feedback factor of 1.4 (28). We do not consider any effects of changes in future climate on O_3 distributions in projecting to 2030 (30, 31), nor do we consider the decrease in global mean temperature due to CH_4 reductions, which could amplify the O_3 decrease that we estimate. MOZART-2 has a horizontal resolution of ≈1.9° by 1.9° and 28 vertical levels. In all cases, we use meteorological fields from the National Centers for Environmental Prediction reanalysis (32), beginning in July 1998, with an 18-month initialization, before focusing on results for the meteorological year 2000.

Results. Between 2000 and 2030 (simulations I and III), we project the population-weighted global average 8-h daily maximum surface O_3 mixing ratio to increase by 12.3 parts per billion by volume (ppbv) (25%) (Table 2), primarily because of projected increases in anthropogenic emissions of NO_x (70%) and CH_4 (48%). The 65 $Mt \cdot yr^{-1}$ CH_4 emission reduction decreases the steady-state population-weighted mean 8-h O_3 by 1.16 ppbv (1.9%, Table 2). This sensitivity is in agreement with other models (18, 19, 28, 33), and these results together suggest that global surface O_3 responds fairly linearly to changes in CH_4 (33). Decreases in O_3 due to CH_4 reductions are widespread globally (Fig. 1), with the largest O_3 decreases occurring over the Middle East, North Africa, and Europe, because of greater down-welling from the free troposphere and greater availability of NO_x. This spatial pattern is similar to previous results (19, 20), suggesting that the pattern is independent of the extent of methane abatement. Methane controls initiated in 2010 will yield ≈81% of this steady-state O_3 change by 2030, assuming exponential decay with a CH_4 perturbation lifetime of ≈12 yr (28).

Table 2. Global average O_3 mixing ratios (ppbv) in the 2000 and 2030 A2 base model runs (simulations I and III), and the steady-state change in O_3 due to a 65 $Mt \cdot yr^{-1}$ reduction in CH_4 emissions, relative to the 2030 base (simulation IV minus simulation III)

Parameter	2000	2030 A2	ΔO_3 2030
24-h average	29.1	33.6	−0.82
8-h daily maximum	31.8	37.1	−0.87
8-h maximum population-weighted	49.4	61.7	−1.16

The steady-state change in O_3 when 65 $Mt \cdot yr^{-1}$ are reduced relative to the 2000 base case (simulation II vs. simulation I) is virtually identical to the change in Table 2 (−1.11 ppbv for population-weighted 8-h O_3), indicating that the projected changes in nonmethane O_3 precursors between 2000 and 2030 have little effect on the O_3 sensitivity to CH_4. This insensitivity presumably reflects the fact that there is little change in hydroxyl radical (OH) concentrations, because of similar emission ratios of NO_x to (CO + VOCs) in 2000 and 2030 (16). Therefore, although the A2 scenario includes larger growth in emissions of O_3 precursors than other SRES scenarios, and larger than the "Current Legislation" scenario of Dentener *et al.* (18), this high growth does not strongly affect the O_3-CH_4 sensitivity.

Indirect Effects of Methane Reductions on Particulate Matter (PM). Methane reductions also indirectly affect PM concentrations through complex oxidant chemistry. MOZART-2 (25) results suggest that CH_4 reductions cause a global net decrease in inorganic PM, because of decreases in hydrogen peroxide that in turn reduce sulfate production. Inorganic PM concentrations also increase at some locations, where the increased gas-phase oxidation (due to increased OH concentrations) dominates the change in sulfate production. Although the global average decrease is only ≈0.5% of the inorganic PM (sulfate, nitrate, and associated ammonium), the decrease is concentrated in populated regions. Confidence in the change in PM is lower than for O_3 because of competing influences on inorganic PM, and because we have neglected changes in organic PM.

Global Mortality Benefits of Reduced Ozone

Methods. Ozone has been associated in epidemiologic studies with adverse health effects including hospital admissions and

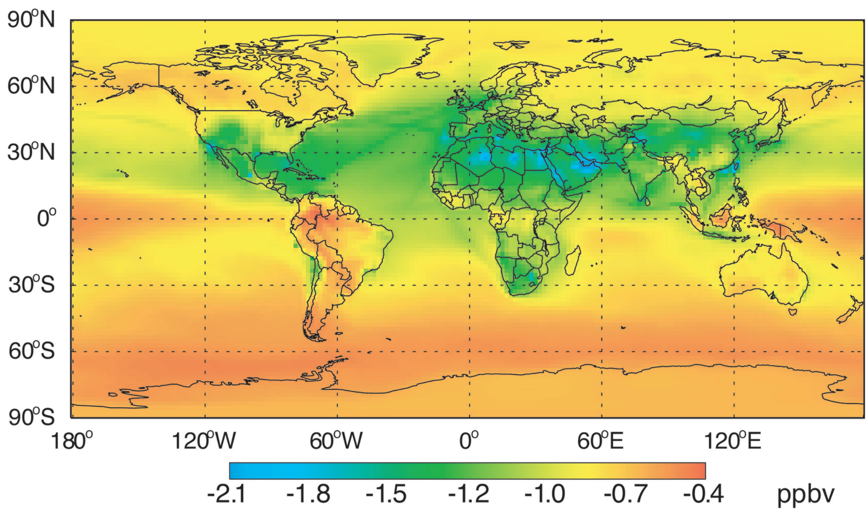

Fig. 1. Change in annual average daily maximum 8-h surface O_3 mixing ratios, at steady state, due to a 65 $Mt \cdot yr^{-1}$ reduction in CH_4 emissions relative to the 2030 A2 base case (simulation IV minus III).

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chronic respiratory conditions, and recent research provides strong evidence for an association with daily premature mortality (1–10). We use the daily O_3-mortality relationship (β) estimated by Bell *et al.* (6), using a distributed lag method for 95 cities in the United States, and apply this relationship globally. Because long-term effects of O_3 on mortality have not been demonstrated (34), we do not consider possible chronic effects of O_3 or years of life lost due to premature mortality. Bell *et al.* (6) directly use a large data set, and therefore their results are not subject to publication bias, which can bias meta-analyses high. The β estimated by Bell *et al.* (6) with a single-day lag is much smaller than the β estimated in three recent meta-analyses (7–9). However, the β of Bell *et al.* (6) with the distributed lag method, used in this study, is much more comparable with the meta-analyses (7–9), which are 22–36% higher. We consider the sensitivity of our results to the uncertainties reported by Bell *et al.* (6) and the meta-analyses (7–9). Although Bell *et al.* (6) focus on the United States, similar results have been reported in North America and Europe (5, 7–9). Few studies of O_3 mortality have been conducted elsewhere, although some such studies suggest associations between O_3 and mortality in other regions (35–37).

Although Bell *et al.* (6) find similar relationships between ozone and mortality over all seasons in the United States, many studies find reduced O_3 impacts in winter, when O_3 concentrations are often lower (5, 8, 9). However, applying seasonal differences in tropical regions is not straightforward. Available studies also show adverse effects of O_3 below current standards, without identifying a clear threshold below which O_3 does not affect mortality (5, 6). Rather than imposing seasonally varying relationships, we assume a low-concentration threshold of 25 ppbv, approximately the preindustrial mixing ratio (13, 15), below which we neglect any effect of O_3 on mortality. We apply this threshold on each day, through all seasons, and consider the sensitivity of our results to the threshold used.

We apply β to the total nonaccident baseline mortality rates, using data for 14 world regions (38). Baseline mortality rates are applied uniformly within each region, and are assumed to be constant into the future. The spatial distribution of population is modeled consistently with the SRES A2 scenario, growing to 9.17 billion in 2030 (26).

Avoided premature mortalities are estimated daily in each model grid square, based on the maximum daily 8-h O_3 mixing ratio in the A2 base and CH_4 control cases. The A2 base and CH_4 control cases are constructed for the period 2000–2030 by interpolating between simulations I, III, and IV. For the A2 base case, 8-h O_3 mixing ratios on each day and in each grid square are interpolated between 2000 and 2030 (simulations I and III) by using a constant percent growth rate. For the CH_4 control case, O_3 decreases begin in 2010 and exponentially approach the steady-state change (simulation IV minus III) with the 12-yr CH_4 perturbation lifetime (see the supporting information, which is published on the PNAS web site).

Results. Table 3 and Fig. 2 show that reducing CH_4 emissions by 65 $Mt{\cdot}yr^{-1}$ in 2010 would prevent ≈30,000 all-cause premature mortalities in the year 2030 (≈0.04% of the total projected mortalities), with ≈370,000 avoided premature mortalities accumulated between 2010 and 2030. These avoided mortalities are distributed globally, with the majority in highly populated regions (Table 3 and Fig. 3). Mortality benefits per million people in 2030 are highest in Africa, which has high baseline mortality rates, followed by Europe and the eastern Mediterranean.

Table 4 shows a large sensitivity to β over the range of uncertainties in Bell *et al.* (6) and three meta-analyses (7–9). The avoided mortalities also vary with the sensitivity of O_3 to CH_4 but are rather insensitive to the low-concentration threshold over the range considered. This insensitivity occurs because regions with low O_3 typically also have low population and small changes in O_3 due to CH_4; O_3 is below 25 ppbv on ≈12% of populated grid square-days in 2030, but the number of avoided mortalities decreases by only 2% relative to the no-threshold case.

The mortality benefits of O_3 decreases are most uncertain in developing nations, where fewer epidemiologic studies exist and the general causes of death differ substantially from those in industrialized nations. As a more conservative estimate, we consider the avoided cardiovascular and respiratory (CR) mortalities, because these may be more closely linked to O_3. We apply the β for CR mortalities from Bell *et al.* (6), which is higher than for total mortalities but not significantly different, to baseline CR mortality rates. In Table 3, ≈17,000 premature CR mortalities can be avoided globally in 2030 by the CH_4 emission reduction, with the greatest per capita benefits in Europe, where relatively more people die of CR causes. Although our estimates of avoided CR mortalities may be more robust in developing nations than total mortalities, they likely miss important decreases in other causes of mortality. Henceforth, we use an uncertainty range from the estimated avoided CR mortalities (≈17,000 in 2030) to the highest number in Table 4 (≈56,000).

Effects of Changes in PM on Mortality. By using the changes in inorganic PM in the previous section and a chronic PM-mortality relationship (34), the avoided 2030 mortalities are estimated to be less than, but comparable with, the O_3 benefit (see the

Table 3. Avoided premature mortalities in 2030 by world region and avoided mortalities per million people in 2030, resulting from decreases in surface O_3 due to a global CH_4 emission reduction of 65 $Mt{\cdot}yr^{-1}$

	Avoided total mortalities in 2030		Avoided CR mortalities in 2030	
Region	Number	Per 10^6 people	Number	Per 10^6 people
Africa	6,920	5.59	2,070	1.68
North America	1,110	2.81	700	1.77
Latin America	1,790	1.88	960	1.01
Southeast Asia	7,790	3.33	4,550	1.95
Western Europe	1,900	3.86	1,260	2.56
Eastern Europe and former Soviet Union	1,790	3.50	1,560	3.06
Eastern Mediterranean	3,150	3.69	1,660	1.94
Western Pacific	500	2.86	310	1.77
East Asia	5,250	2.36	3,610	1.63
Global	30,200	3.29	16,700	1.82

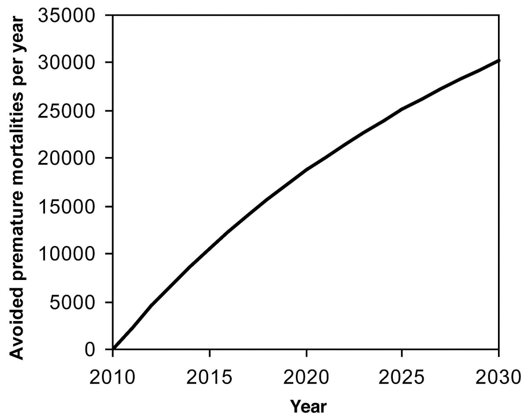

Fig. 2. Avoided global premature mortalities from a 65 $Mt{\cdot}yr^{-1}$ CH_4 emission reduction, beginning in 2010.

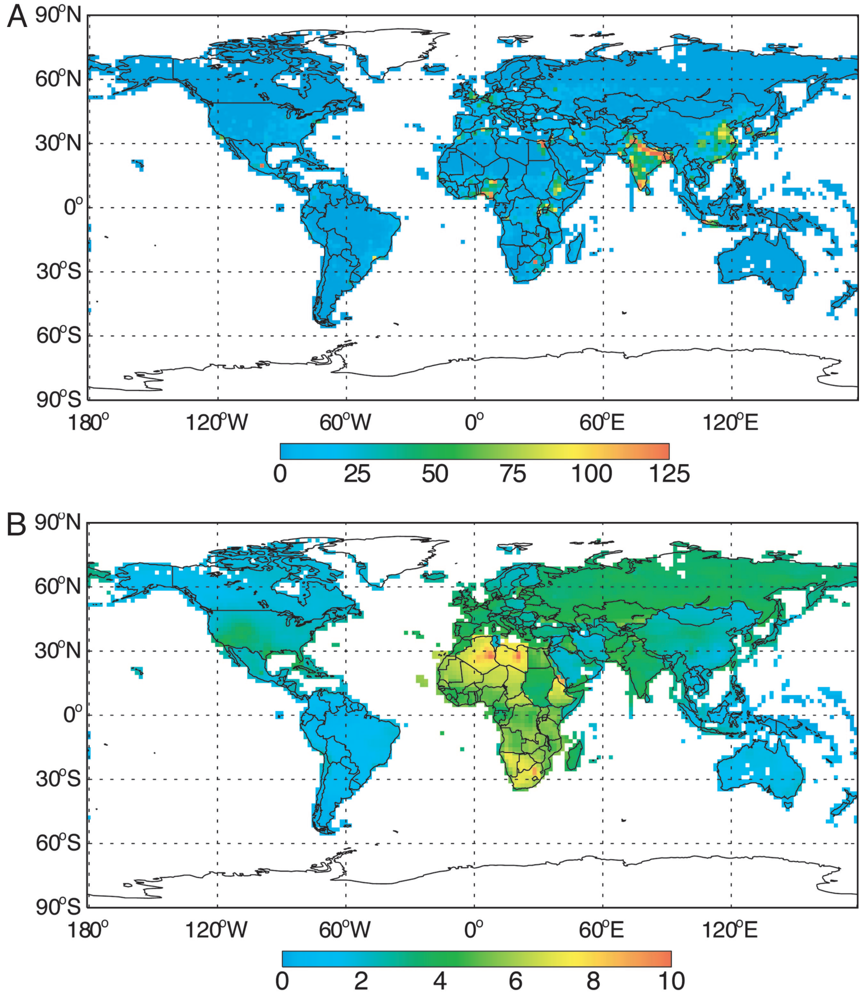

Fig. 3. Estimated avoided premature mortalities in 2030. (*A*) Total. (*B*) Per million people.

supporting information). The effects of CH_4 reductions on both inorganic and organic PM should be further investigated, including changes in PM precursor emissions from energy sources possibly displaced because of increased CH_4 availability.

Policy Analysis of Ozone Control by Means of Methane Mitigation

A compilation of global CH_4 abatement measures from five industrial sectors (27) shows that ≈41 Mt·yr^{-1} can be reduced at a negative marginal cost (net cost-savings, through natural gas recovery), which can be justified regardless of health benefits.

Table 4. Sensitivity of global 2030 avoided premature mortalities to uncertain parameters

Parameter	Base value	Range	Avoided mortalities over range
β*	0.043	0.027–0.079	19,300–55,800
O_3 sensitivity to CH_4†	—	−35% to +35%	19,600–40,700
Low-concentration threshold, ppbv	25	40–0	26,900–30,900

Using base values gives 30,200 avoided premature mortalities.

*Percent excess mortality per ppbv change in 8-h O_3 mixing ratio. The range spans the 95% confidence intervals of four studies (6–9).

†Range applied as scaling factors to the change in daily 8-h O_3 in each grid square.

The 65 Mt·yr^{-1} reduction has a marginal cost of ≈$100 per tonne of CH_4 (2000 U.S. dollars), whereas the net cost of this reduction is negative. We combine this marginal cost with the all-cause avoided premature mortalities, which we convert to a constant annualized benefit between 2010 and 2030 at a 5% yr^{-1} discount rate (see the supporting information), yielding $420,000 per avoided mortality ($230,000–$760,000) as the marginal cost-effectiveness of reducing 65 Mt·yr^{-1}. The 65 Mt·yr^{-1} reduction would be justified, in cost-benefit terms, for any globally averaged VSL >$420,000.

If we use $1 million as a reasonable globally averaged VSL (39), the monetized benefit of reducing CH_4 emissions is $240 per tonne of CH_4 ($140–$450), or $12 per tonne of CO_2 equivalent ($7–$22), which exceeds the marginal cost of the 65 Mt·yr^{-1} reduction (≈$100 per tonne of CH_4). This estimate neglects increases in the VSL as incomes grow, and only considers 20 yr of benefits, whereas the O_3 reductions will continue growing beyond 2030 as population also grows. The monetized benefit scales proportionally with the assumed globally averaged VSL.

Because CH_4 reductions have recently traded in international markets at $10–$20 per tonne of CO_2 equivalent, these results suggest that current climate-motivated CH_4 emission reductions can roughly be justified by their benefits to air quality and health, irrespective of other benefits of CH_4 and O_3 reductions. Furthermore, although the ancillary benefits of CO_2 mitigation for air quality and health have received attention (40), the ancillary benefits of CH_4 mitigation have not. Our estimate for CH_4 of \$12 per tonne of CO_2 equivalent is comparable with the range estimated previously for CO_2 of \$0.5–\$140 per tonne of CO_2 (41). Unlike the ancillary benefits of CO_2 mitigation, however, the ancillary benefits of CH_4 mitigation do not depend on the location or means of CH_4 abatement, because the health benefits of CH_4 mitigation result from reactions involving the CH_4 itself, and CH_4 emissions affect O_3 globally regardless of emission location.

The compilation of CH_4 abatement measures used in this study (27) considers five industrial sectors (coal, oil, and natural gas operations, landfills, and wastewater treatment) for which methane abatement opportunities are well understood. Because this compilation neglects abatement opportunities in the large agricultural sector, it may underestimate the availability of low-cost CH_4 options, which would suggest that CH_4 mitigation is more cost-effective than estimated here. On the other hand, a separate compilation by the U.S. Environmental Protection Agency (42–44) suggests that less CH_4 can be reduced at low cost (see the supporting information and ref. 20).

Methane mitigation also benefits climate, because it reduces the radiative forcing of both CH_4 and O_3. The 65 $Mt \cdot yr^{-1}$ CH_4 reduction would decrease global radiative forcing by 0.14 $W \cdot m^{-2}$, from CH_4 and O_3 together (at steady state). In contrast, reductions in NO_x emissions decrease O_3 forcing but increase CH_4 forcing (45), with a net effect that could be positive or negative depending on location (46).

Methane is also an important source of global energy, and capturing half of the 65 $Mt \cdot yr^{-1}$ for energy use would provide $\approx$2% of current global natural gas production. The reductions in O_3 concentrations would also result in benefits to human health (morbidity) and agriculture (47), which we previously estimated to be smaller than the monetized benefits of avoided mortalities estimated here (20). Methane mitigation may further benefit air quality and climate by removing other pollutants (e.g., VOCs) through the same actions that reduce CH_4 emissions, and by increasing the availability of natural gas, which may reduce emissions of CO_2 and air pollutants from the combustion of other fossil fuels. In addition, because the reductions in O_3 are widespread globally, CH_4 mitigation may increase the net primary productivity of plants, causing increased uptake of CO_2 (48). Finally, methane mitigation may affect stratospheric O_3, but the direction of that influence is not certain (49).

The effects of CH_4 mitigation on surface O_3 concentrations are widespread globally, and are delayed. These characteristics differ from other means of controlling O_3, as well as most actions to manage air quality, which abate local and regional pollution over hours to weeks. Because of its global impacts, with small local benefits, CH_4 mitigation for air quality purposes (as for climate) will best be implemented at national and international levels. Furthermore, the potential for reducing O_3 through CH_4 mitigation is limited to a few parts per billion by volume. Methane mitigation is therefore most appropriate for international and long-term (decadal) O_3 management, where CH_4 mitigation for background O_3 is complementary to local and regional O_3 management through reductions in emissions of NO_x and nonmethane VOCs (20).

Important uncertainties in this study lie in the relationship between O_3 and mortality, and between CH_4 emissions and global surface O_3 concentrations. Because CH_4 affects O_3 globally, this research highlights the need to improve understanding of O_3 mortality in developing nations, and of the relationship between O_3 and mortality at low concentration, including consideration of possible thresholds. Future research should also investigate the effects of CH_4 mitigation on PM concentrations, and its implications for air quality, public health, and climate. Finally, future research should further examine opportunities to abate CH_4 emissions, emphasizing the large agricultural sector.

Conclusions

As background O_3 concentrations increase, meeting national O_3 standards increasingly becomes an international problem (50–52). Methane mitigation reduces surface O_3 everywhere, offering a unique opportunity to improve air quality globally. We estimate that reducing $\approx$20% of current global anthropogenic CH_4 emissions, which can be achieved at a net cost-savings by using identified technologies, will reduce O_3 mixing ratios globally by $\approx$1 ppbv and prevent $\approx$30,000 premature mortalities globally in 2030 and $\approx$370,000 mortalities between 2010 and 2030. If these mortalities are valued at \$1 million each, the monetized benefit is $\approx$\$240 per tonne of CH_4, or $\approx$\$12 per tonne of CO_2 equivalent. These benefits exceed the marginal costs of the 20% anthropogenic CH_4 reduction ($\approx$\$100 per tonne of CH_4) and demonstrate that CH_4 mitigation has ancillary benefits to air quality and human health that are comparable with those previously estimated for CO_2. Methane mitigation benefits air quality, public health, agriculture, climate, and energy, and should increasingly be considered a cost-effective component of international long-term O_3 management.

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1. U.S. Environmental Protection Agency (1996) *Air Quality Criteria for Ozone and Related Photochemical Oxidants* (Office of Research and Development, U.S. Environmental Protection Agency, Washington, DC), EPA Publication EPA/600/P-93/004cF.
2. Levy, J. I., Carrothers, T. J., Tuomisto, J. T., Hammitt, J. K. & Evans, J. S. (2001) *Environ. Health Perspect.* **109,** 1215–1226.
3. Thurston, G. D. & Ito, K. (2001) *J. Exposure Anal. Environ. Epidemiol.* **11,** 286–294.
4. Anderson, H. R., Atkinson, R. W., Peacock, J. L., Marston, L. & Konstantinou, K. (2004) *Meta-Analysis of Time-Series Studies and Panel Studies of Particulate Matter (PM) and Ozone (O₃)* (WHO, Geneva).
5. Gryparis, A., Forsberg, B., Katsouyanni, K., Analitis, A., Touloumi, G., Schwartz, J., Samoli, E., Medina, S., Anderson, H. R., Niciu, E. M., *et al.* (2004) *Am. J. Respir. Crit. Care Med.* **170,** 1080–1087.
6. Bell, M. L., McDermott, A., Zeger, S. L., Samet, J. M. & Dominici, F. (2004) *J. Am. Med. Assoc.* **292,** 2372–2378.
7. Bell, M. L., Dominici, F. & Samet, J. M. (2005) *Epidemiology* **16,** 436–445.
8. Ito, K., De Leon, S. F. & Lippman, M. (2005) *Epidemiology* **16,** 446–457.
9. Levy, J. I., Chemerynski, S. M. & Sarnat, J. A. (2005) *Epidemiology* **16,** 458–468.
10. Schwartz, J. (2005) *Am. J. Respir. Crit. Care Med.* **171,** 627–631.
11. Volz, A. & Kley, D. (1988) *Nature* **332,** 240–242.
12. Marenco, A., Gouget, H., Nedelec, P., Pages, J. P. & Karcher, F. (1994) *J. Geophys. Res.* **99,** 16617–16632.
13. Vingarzan, R. (2004) *Atmos. Environ.* **38,** 3431–3442.
14. Lelieveld, J., van Aardenne, J., Fischer, H., de Reus, M., Williams, J. & Winkler, P. (2004) *Science* **304,** 1483–1487.
15. Lelieveld, J. & Dentener, F. J. (2000) *J. Geophys. Res.* **105,** 3531–3551.
16. Wang, Y. H. & Jacob, D. J. (1998) *J. Geophys. Res.* **103,** 31123–31135.
17. Prather, M., Gauss, M., Berntsen, T., Isaksen, I., Sundet, J., Bey, I., Brasseur, G., Dentener, F., Derwent, R., Stevenson, D., *et al.* (2003) *Geophys. Res. Lett.* **30,** 1100.
18. Dentener, F., Stevenson, D., Cofala, J., Mechler, R., Amann, M., Bergamaschi, P., Raes, F. & Derwent, R. (2005) *Atmos. Chem. Phys.* **5,** 1731–1755.
19. Fiore, A. M., Jacob, D. J., Field, B. D., Streets, D. G., Fernandes, S. D. & Jang, C. (2002) *Geophys. Res. Lett.* **29,** 1919.
20. West, J. J. & Fiore, A. M. (2005) *Environ. Sci. Technol.* **39,** 4685–4691.
21. Ramaswamy, V., Boucher, O., Haigh, J., Hauglustaine, D., Haywood, J., Myhre, G., Nakajima, T., Shi, G. Y. & Solomon, S. (2001) in *Climate Change 2001: The Scientific Basis*, eds. Houghton, J. T., Ding, Y., Griggs, D. J., Noguer, M., van der Linden, P. J., Dai, X., Maskell, K. & Johnson, C. A. (Cambridge Univ. Press, Cambridge, U.K.), pp. 349–416.
22. Reilly, J., Prinn, R., Harnisch, J., Fitzmaurice, J., Jacoby, H., Kicklighter, D., Melillo, J., Stone, P., Sokolov, A. & Wang, C. (1999) *Nature* **401,** 549–555.

23. Hayhoe, K., Jain, A., Pitcher, H., MacCracken, C., Gibbs, M., Wuebbles, D., Harvey, R. & Kruger, D. (1999) *Science* **286,** 905–906.
24. Horowitz, L. W., Walters, S., Mauzerall, D. L., Emmons, L. K., Rasch, P. J., Granier, C., Tie, X. X., Lamarque, J. F., Schultz, M. G., Tyndall, G. S., *et al.* (2003) *J. Geophys. Res.* **108,** 4784.
25. Tie, X., Madronich, S., Walters, S., Edwards, D. P., Ginoux, P., Mahowald, N., Zhang, R., Luo, C. & Brasseur, G. (2005) *J. Geophys. Res.* **110,** D03204.
26. Nakicenovic, N., Alcamo, J., Davis, G., de Vries, B., Fenhann, J., Gaffin, S., Gregory, K., Grubler, A., Jung, T. Y., Kram, T., *et al.* (2000) *Special Report on Emissions Scenarios* (Cambridge Univ. Press, Cambridge, U.K.).
27. International Energy Agency Greenhouse Gas R&D Programme (2003) *Building the Cost Curves for the Industrial Sources of Non-*CO_2 *Greenhouse Gases* (International Energy Agency, Cheltenham, U.K.), PH4/25.
28. Prather, M., Ehhalt, D., Dentener, F., Derwent, R., Dlugokencky, E., Holland, E., Isaksen, I., Katima, J., Kirchhoff, V., Matson, P., *et al.* (2001) in *Climate Change 2001: The Scientific Basis*, eds. Houghton, J. T., Ding, Y., Griggs, D. J., Noguer, M., van der Linden, P. J., Dai, X., Maskell, K. & Johnson, C. A. (Cambridge Univ. Press, Cambridge, U.K.), pp. 239–287.
29. Stevenson, D., Dentener, F. J., Schultz, M. G., Ellingsen, K., van Noije, T. P. C., Wild, O., Zeng, G., Amann, M., Atherton, C. S., Bell, N., *et al.*, *J. Geophys. Res.*, in press.
30. Hogrefe, C., Lynn, B., Civerolo, K., Ku, J. Y., Rosenthal, J., Rosenzweig, C., Goldberg, R., Gaffin, S., Knowlton, K. & Kinney, P. L. (2004) *J. Geophys. Res.* **109,** D22301.
31. Knowlton, K., Rosenthal, J. E., Hogrefe, C., Lynn, B., Gaffin, S., Goldberg, R., Rosenzweig, C., Civerolo, K., Ku, J. Y. & Kinney, P. L. (2004) *Environ. Health Perspect.* **112,** 1557–1563.
32. Kalnay, E., Kanamitsu, M., Kistler, R., Collins, W., Deaven, D., Gandin, L., Iredell, M., Saha, S., White, G., Woollen, J., *et al.* (1996) *Bull. Am. Meteorol. Soc.* **77,** 437–471.
33. Shindell, D. T., Faluvegi, G., Bell, N. & Schmidt, G. A. (2005) *Geophys. Res. Lett.* **32,** L040803.
34. Pope, C. A., Burnett, R. T., Thun, M. J., Calle, E. E., Krewski, D., Ito, K. & Thurston, G. D. (2002) *J. Am. Med. Assoc.* **287,** 1132–1141.
35. Borja-Aburto, V. H., Castillejos, M., Gold, D. R., Bierzwinski, S. & Loomis, D. (1998) *Environ. Health Perspect.* **106,** 849–855.
36. O'Neill, M. S., Loomis, D. & Borja-Aburto, V. H. (2004) *Environ. Res.* **94,** 234–242.
37. Kim, S. Y., Lee, J. T., Hong, Y. C., Ahn, K. J. & Kim, H. (2004) *Environ. Res.* **94,** 113–119.
38. World Health Organization (2004) *The World Health Report 2004: Changing History* (WHO, Geneva).
39. Markandaya, A., Halsnaes, K., Lanza, A., Matsuoka, Y., Maya, S., Pan, J., Shogren, J., Seroa de Motta, R. & Zhang, T. (2001) in *Climate Change 2001: Mitigation*, eds. Metz, B., Davidson, O., Swart, R. & Pan, J. (Cambridge Univ. Press, Cambridge, U.K.), pp. 451–498.
40. Cifuentes, L., Borja-Aburto, V. H., Gouveia, N., Thurston, G. & Davis, D. L. (2001) *Science* **293,** 1257–1259.
41. Hourcade, J.-C., Shukla, P., Cifuentes, L., Davis, D., Edmonds, J., Fisher, B., Fortin, E., Golub, A., Hohmeyer, O., Krupnick, A., *et al.* (2001) in *Climate Change 2001: Mitigation*, eds. Metz, B., Davidson, O., Swart, R. & Pan, J. (Cambridge Univ. Press, Cambridge, U.K.), pp. 499–559.
42. U.S. Environmental Protection Agency (1999) *U.S. Methane Emissions 1990–2010: Inventories, Projections, and Opportunities for Reductions* (U.S. Environmental Protection Agency, Washington, DC), EPA Publication 430-R-99-013.
43. U.S. Environmental Protection Agency (2003) *International Analysis of Methane and Nitrous Oxide Abatement Opportunities: Report to the Energy Modeling Forum, Working Group 21* (U.S. Environmental Protection Agency, Washington, DC), www.epa.gov/methane/pdfs/methodologych4.pdf.
44. Delhotal, K. C., de la Chesnaye, F. C., Gardiner, A., Bates, J. & Sankovski, A., *Energy J.*, in press.
45. Fuglestvedt, J. S., Berntsen, T. K., Isaksen, I. S. A., Mao, H. T., Liang, X. Z. & Wang, W. C. (1999) *Atmos. Environ.* **33,** 961–977.
46. Naik, V., Mauzerall, D., Horowitz, L., Schwarzkopf, M. D., Ramaswamy, V. & Oppenheimer, M. (2005) *J. Geophys. Res.* **110,** D24306.
47. Mauzerall, D. L. & Wang, X. P. (2001) *Annu. Rev. Energy Environ.* **26,** 237–268.
48. Felzer, B., Kicklighter, D., Melillo, J., Wang, C., Zhuang, Q. & Prinn, R. (2004) *Tellus* **56,** 230–248.
49. World Meteorological Organization (2003) *Scientific Assessment of Ozone Depletion: 2002* (Global Ozone Research and Monitoring Project, World Meteorological Organization, Geneva), Report No. 47.
50. Akimoto, H. (2003) *Science* **302,** 1716–1719.
51. Holloway, T., Fiore, A. & Hastings, M. G. (2003) *Environ. Sci. Technol.* **37,** 4535–4542.
52. Keating, T. J., West, J. J. & Farrell, A. E. (2004) in *Intercontinental Transport of Air Pollution*, ed. Stohl, A. (Springer, Berlin), pp. 295–320.