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Beyond SO_x reductions from shipping: assessing the impact of NO_x and carbonaceous-particle controls on human health and climate

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Abstract

Historically, cargo ships have been powered by low-grade fossil fuels, which emit particles and particle-precursor vapors that impact human health and climate. We used a global chemical-transport model with online aerosol microphysics (GEOS-Chem-TOMAS) to estimate the aerosol health and climate impacts of four emission-control policies: (1) 85% reduction in sulfur oxide (SO_x) emissions (Sulf); (2) 85% reduction in SO_x and black carbon (BC) emissions (Sulf-BC); (3) 85% reduction in SO_x, BC, and organic aerosol (OA) emissions (Sulf-BC-OA); and (4) 85% reduction in SO_x, BC, OA, and nitrogen oxide (NO_x) emissions (Sulf-BC-OA-NO_x). The SO_x reductions reflect the 0.5% fuel-sulfur cap implemented by the International Maritime Organization (IMO) on 1 January 2020. The other reductions represent realistic estimates of future emission-control policies. We estimate that these policies could reduce fine particulate matter (PM_{2.5})-attributable mortalities by 13 300 (Sulf) to 38 600 (Sulf-BC-OA-NO_x) mortalities per year. These changes represent 0.3% and 0.8%, respectively, of annual PM_{2.5}-attributable mortalities from anthropogenic sources. Comparing simulations, we estimate that adding the NO_x cap has the greatest health benefit. In contrast to the health benefits, all scenarios lead to a simulated climate warming tendency. The combined aerosol direct radiative effect and cloud-albedo indirect effects (AIE) are between 27 mW m⁻² (Sulf) and 41 mW m⁻² (Sulf-BC-OA-NO_x). These changes are about 2.1% (Sulf) to 3.2% (Sulf-BC-OA-NO_x) of the total anthropogenic aerosol radiative forcing. The emission control policies examined here yield larger relative changes in the aerosol radiative forcing (2.1%–3.2%) than in health effects (0.3%–0.8%), because most shipping emissions are distant from populated regions. Valuation of the impacts suggests that these emissions reductions could produce much larger marginal health benefits (\$129–\$374 billion annually) than the marginal climate costs (\$12–\$17 billion annually).

1. Introduction

Between 80% and 90% of global trade, on a per-weight basis, is transported by ships (United Nations Conference on Trade and Development 2018). Ships are powered with low-grade remnants of crude-oil distillation called residual fuels (e.g. heavy fuel oil).

When combusted, these fuels emit air pollutants (e.g. black carbon (BC), organic aerosol (OA), sulfur dioxide (SO₂), nitrogen oxide (NO)) (Eyring *et al* 2010). Once emitted, gas-phase SO₂ and NO emissions may undergo reactions to form particle-phase SO₄²⁻ and NO₃⁻, respectively. These secondary pollutants, along with primary emissions of BC and OA,

are responsible for most of the fine particulate matter ($\text{PM}_{2.5}$) emissions associated with shipping, which is estimated to cause 50 000–400 000 mortalities per year globally (Winebrake *et al* 2009, Partanen *et al* 2013, Sofiev *et al* 2018).

Atmospheric aerosols impact the radiative budget in two major ways: they directly scatter and absorb radiation (direct radiative effect, DRE) and change cloud reflectivity (cloud-albedo aerosol indirect effect, AIE) (Stocker *et al* 2013). Ships contribute to atmospheric SO_4^{2-} , NO_3^- , and OA, which primarily scatter radiation (cooling effect), and BC, which strongly absorbs radiation (warming effect). Ships also impact cloud-condensation nuclei (CCN), affecting cloud albedo.

To combat the adverse health and environmental impacts of shipping, the International Maritime Organization (IMO) introduced a fuel-sulfur limit under an amendment to Annex VI of the International Convention for the Prevention of Pollution from Ships (i.e. MARPOL) (Marine Environment Protection Committee 2008). The policy requires a global 85% reduction in ship fuel sulfur-mass content. The prior limit was 3.5%, and the new limit is 0.5%. This policy went into effect on 1 January 2020. To meet the new standard and avoid fines, ships around the globe can use (1) fuels with less than 0.5% sulfur content, (2) exhaust scrubbers, (3) alternative fuels (e.g. liquefied natural gas), and/or (4) onshore power supplies when docked (Solakivi *et al* 2019, Zhu *et al* 2020).

The health and radiative effects of the 2020 fuel-sulfur cap have been researched extensively (Lauer *et al* 2009, Winebrake *et al* 2009, Tronstad Lund *et al* 2012, Partanen *et al* 2013, Sofiev *et al* 2018). Results vary in magnitude, but all studies conclude that the 2020 fuel-sulfur cap will reduce $\text{PM}_{2.5}$, leading to substantial health benefits, while the SO_4^{2-} reductions will reduce the planetary albedo, leading to a climate warming tendency. Impacts of emission controls on co-emitted pollutants (BC, OA, and nitrogen oxides (NO_x)) have not been investigated systematically. Partanen *et al* (2013), Winebrake *et al* (2009), and Lauer *et al* (2009) investigated coupled fuel-sulfur and OA reductions, but they did not evaluate the impacts of OA independently. Peters *et al* (2012) found that removing all BC and OA emissions from ships had little effect on the AIE, but they did not explicitly investigate the health or DRE impacts of BC and OA from ships. Previous work demonstrated that reducing shipping fuel-sulfur content increases aerosol nitrate (Lauer *et al* 2009, Sofiev *et al* 2018). However, to the best of our knowledge, the impacts of a global shipping NO_x cap have not been evaluated.

A cocktail of control strategies and technologies have been shown to reduce BC, OA, and NO_x emissions for ships. These include, but are not limited to, water-in-fuel emulsions, exhaust gas scrubbers, diesel particulate filters (DPF), fuel switching,

slow steaming, exhaust gas recirculation, and various engine modifications (e.g. slide valves and engine load). Below, we provide examples of the most promising measures that would provide the largest reductions in emissions; a comprehensive list is in previous publications (e.g. Azzara *et al* 2014, Comer *et al* 2017, Corbett *et al* 2010). DPFs can efficiently trap and reduce BC emissions by 95%–99% and also provide reductions in OA and $\text{PM}_{2.5}$ but to a slightly lower degree (50%–90%) (Lack *et al* 2012, Johansen 2015). Selective catalytic reduction (SCR) systems inject urea or ammonia into the exhaust to reduce NO_x to molecular nitrogen over a catalyst (Kim *et al* 2020), reducing NO_x emissions by 80%–95% (Azzara *et al* 2014). Finally, switching from residual fuels (e.g. heavy fuel oil) to distillate fuels (similar to regular on-road diesel) (Johnson *et al* 2016) or natural gas (Burel *et al* 2013) have the potential to reduce significantly (>90%) all air pollutant emissions. Distillates enable the use of advanced after-treatment systems developed for on-road diesel engines (e.g. DPF, oxidation catalysts, and SCR) and have produced significant reductions in CO, BC, OA, and NO_x from mobile sources over the past few decades (McDonald *et al* 2013, 2015, Jiang *et al* 2018). Switching to hydrogen, electricity, and wind-assisted propulsion could nearly eliminate all pollutant emissions in the future (Comer 2019).

Here, we used GEOS-Chem-TOMAS, a global chemical-transport model with online aerosol microphysics, to investigate the potential health and radiative effects of four shipping emissions scenarios: (1) 85% reduction in SO_x (Sulf); (2) 85% reduction in SO_x and BC (Sulf-BC); (3) 85% reduction in SO_x , BC, and OA (Sulf-BC-OA); and (4) 85% reduction in SO_x , BC, OA, and NO_x (Sulf-BC-OA- NO_x) relative to business as usual (BAU). Sulf went into effect on 1 January 2020, while the other scenarios represent potential future policies and follow the order that emissions were regulated for terrestrial, heavy-duty diesel engines (May *et al* 2014). For instance, a modern-day on-road diesel would be equipped with an oxidation catalyst (for CO and unburned hydrocarbon removal), regular or catalyzed DPF (for BC and OA removal), and an SCR (for NO_x reduction) in that order. The health and climate impacts of policies beyond Sulf have not been investigated systematically. Our study highlights the need to carefully consider the health-climate tradeoffs of emission-control policies and is relevant for present and future transportation emission-control decisions.

2. Methods

2.1. GEOS-Chem model configuration

We used (GEOS-Chem v12.6.0 2019) with GEOS-FP offline meteorological fields (GEOS-FP; <https://gmao.gsfc.nasa.gov/>) for the year 2013. Simulations were run at $2^\circ \times 2.5^\circ$ resolution, with

47 vertical layers using GEOS-Chem's tropospheric chemistry ('tropchem') and Two Moment Aerosol Sectional (TOMAS) online aerosol microphysics configurations. TOMAS has 15 aerosol size sections (3 nm to 10 μm) (Adams and Seinfeld 2002, Trivittayanurak *et al* 2008, Kodros and Pierce 2017) and includes nucleation, condensation, and coagulation; size-resolved emissions; dry and wet deposition; and aqueous sulfur chemistry. Size-resolved aerosol species were SO_4^{2-} , OA, BC, sea salt, and mineral dust. NH_4^+ and NO_3^- were not size resolved in the model; so, for the radiative effect, we assumed the fraction of ammonium and nitrate in each TOMAS size section followed the aerosol water size distribution, increasing particle size but not number.

We used the Community Emissions Data System (CEDS) emissions inventory for all anthropogenic emissions, including shipping (Hoesly *et al* 2018). We assumed constant shipping emissions for 2013–2020. Shipping emissions have remained roughly constant throughout this timeframe (Olmer *et al* 2017, International Energy Agency 2020), because increases in shipping emissions have been offset by gains in efficiency. Thus, our estimates are representative of present-day health and climate impacts of emissions reductions. Additionally, the estimated health and climate impacts will not differ greatly year to year. The subtraction between the BAU and emission-cap scenarios is not very sensitive to inter-annual variability. In addition to the base SO_2 shipping emissions, we added 3.1% of SO_2 emissions as SO_4^{2-} , assuming a bimodal number distribution with two modes ($\mu = 10$ nm, $\sigma = 1.6$; $\mu = 70$ nm, $\sigma = 2$) (Stevens and Pierce 2014). The SO_4^{2-} emissions represent the subgrid treatment of sulfate chemistry (i.e. 'primary sulfate') (Stevens and Pierce 2014). For secondary OAs, we used Pai *et al* (2020). For natural emissions, we used Global Fire Emissions Data, version 4 (GFED4) for biomass burning (Randerson *et al* 2017); the Jaeglé scheme for sea-salt (Jaeglé *et al* 2011); the Dust Entrainment and Deposition (DEAD) scheme for mineral dust (Zender *et al* 2003); and the Model of Emissions of Gases and Aerosols from Nature version 2.1 (MEGAN2.1) + Guenther 2012 scheme for biogenic emissions (Guenther *et al* 2012).

We simulated six scenarios (table 1). For 'BAU,' all shipping emissions were set to CEDS-2013 levels. For 'No-Anthro,' all anthropogenic emissions were set to zero. 'Sulf' employed an 85% reduction to the CEDS-2013 SO_x emissions levels (which corresponds to the 0.5% fuel-sulfur cap implemented in 2020). This reduction was included in all emission-cap scenarios and was applied first because this control policy is underway. Then, we employed sequential 85% reductions in the CEDS-2013 BC, OA, and NO_x levels. The order of emission caps generally followed the order emissions were regulated for terrestrial heavy-duty diesel engines (May *et al* 2014). We also implemented emissions reductions by pollutant

to isolate species' impacts and because past shipping policies have regulated species rather than technology (e.g. fuel-sulfur cap). The health and climate impacts are sensitive to the order the controls are implemented, due to non-linearities in aerosol chemistry and our impact calculations. We discuss the implications in the 'results & discussion' section.

The timeframe for achieving these emission reductions will depend on how these regulations are imposed (e.g. grandfathering, rate of fleet turnover, retrofitting, best-available technology). These emissions reductions were achieved for terrestrial mobile sources through multiple tiers of regulatory efforts over several decades, noting that the regulatory timelines varied by species (Dallmann and Harley 2010, McDonald *et al* 2013, 2015, Jiang *et al* 2018). As ships might have longer fleet turnover times, complete emissions reductions in the marine sector might take several decades. A review of the literature suggests that emissions controls for ships may reduce BC by up to 99%, OA by up to 70%, and NO_x by up to 95% (Corbett *et al* 2010, Comer *et al* 2017). We chose to reduce each species by 85% for simplicity and congruence with the fuel-sulfur cap noting that the actual emissions reductions may vary both in magnitude and species. However, we do not expect the impacts to change much because the relationship between emissions and atmospheric aerosols is roughly linear in the regime we investigated. For example, if these species were eliminated (i.e. reduced by 100%), the human health and climate impacts would only be 15% higher, which would be much smaller than the inherent uncertainties in those estimates.

2.2. Health effect calculation

We used the Global Burden of Disease (GBD) framework to estimate the averted $\text{PM}_{2.5}$ -attributable mortalities under each emission cap relative to the BAU. To estimate $\text{PM}_{2.5}$, we summed the aerosol species using the equation found in Note S1. To relate $\text{PM}_{2.5}$ exposures to the risk of mortality, we used the Global Exposure Mortality Models (GEMM) from Burnett *et al* (2018). This concentration–response function is developed from a meta-analysis of numerous epidemiological studies and assumes equal toxicity for all $\text{PM}_{2.5}$ components. We evaluated the four primary causes of adult mortality attributable to $\text{PM}_{2.5}$: lung cancer, ischemic heart disease, stroke, and chronic obstructive pulmonary disease. We provide mean estimates and an uncertainty range ($\pm 2 \times$ standard error) using the Burnett *et al* (2018) coefficients. We used the 'all-age' risk function and subtracted the mortalities under the emissions control from the BAU to calculate the averted $\text{PM}_{2.5}$ -attributable mortalities. We used non-age-specific demographic data, including population density data for 2015 (Socioeconomic Data and Application Center; <https://sedac.ciesin.columbia.edu/>) and country-level baseline mortality rates from

Table 1. Global emission rates of sulfur dioxide (SO₂) and sulfate (SO₄^{2−}) in Tg of SO₂ year^{−1}; black carbon (BC) and organic carbon (OA) in Tg carbon (C) year^{−1}, and nitrogen oxides (NO_x) in Tg of nitrogen dioxide (NO₂) year^{−1} for our simulations: business as usual (BAU), four shipping emissions-cap, and no anthropogenic emissions scenarios. Short names for each scenario are provided in parentheses. Percent reductions are provided in parentheses. Gray shading indicates where an emissions cap was applied.

	SO ₂ + SO ₄ ^{2−}	BC	OA	NO
Business as usual (BAU)	8.98 Tg year ^{−1} (0%)	0.17 Tg year ^{−1} (0%)	0.13 Tg year ^{−1} (0%)	14.65 Tg year ^{−1} (0%)
Fuel-sulfur cap (Sulf)	1.35 Tg year ^{−1} (−85%)	0.17 Tg year ^{−1} (0%)	0.13 Tg year ^{−1} (0%)	14.65 Tg year ^{−1} (0%)
Fuel-sulfur cap + BC cap (Sulf-BC)	1.35 Tg year ^{−1} (−85%)	0.02 Tg year ^{−1} (−85%)	0.13 Tg year ^{−1} (0%)	14.65 Tg year ^{−1} (0%)
Fuel-sulfur cap + BC cap + OA cap (Sulf-BC-OA)	1.35 Tg year ^{−1} (−85%)	0.02 Tg year ^{−1} (−85%)	0.02 Tg year ^{−1} (−85%)	14.65 Tg year ^{−1} (−85%)
Fuel-sulfur cap + BC cap + OA cap + NO _x cap (Sulf-BC-OA-NO _x)	1.35 Tg year ^{−1} (−85%)	0.02 Tg year ^{−1} (−85%)	0.02 Tg year ^{−1} (−85%)	2.20 Tg year ^{−1} (−85%)
No anthropogenic emissions (No-Anthro)	0 Tg year ^{−1} (−100%)	0 Tg year ^{−1} (−100%)	0 Tg year ^{−1} (−100%)	0 Tg year ^{−1} (−100%)

2013 (Institute for Health Metrics and Evaluation; www.healthdata.org/gbd). All inputs were gridded to 0.25° × 0.25° by linear interpolation.

2.3. Radiative effect calculation

The DRE and AIE calculations followed Kodros *et al* (2016), Kodros and Pierce (2017), and Ramnarine *et al* (2019). We used the Rapid Radiative Transfer Model for Global and Regional Model Applications offline to calculate the top-of-the-atmosphere all-sky DRE and AIE (Iacono *et al* 2008). Monthly meteorological variables were from GEOS-FP and aerosol parameters from GEOS-Chem-TOMAS. The DRE was calculated with two black-carbon mixing-state assumptions. The first was an ‘external’ assumption where, for each TOMAS size bin, BC was represented as a separate particle from all other aerosol components. The second was a ‘core-shell’ mixing-state assumption, where, for each TOMAS size bin, BC was represented as a pure core within a homogeneous shell of all other aerosol components. The actual DRE value likely lies somewhere between the external and core-shell estimates. These assumptions affected the calculated aerosol optical depth (AOD), single scattering albedo (SSA), and the asymmetry parameter (Bohren and Huffman 1998). For AIE, we used the cloud-droplet-radius perturbation method of Kodros *et al* (2016), Rap *et al* (2013), and Scott *et al* (2014).

3. Results & discussion

3.1. Changes in PM_{2.5} under emission controls

Simulation data are publicly available (Bilsback *et al* 2020). Overall, the shipping emissions-cap scenarios reduced the global-average surface PM_{2.5} by −0.7% (Sulf) to −1.4% (Sulf-BC-OA-NO_x)

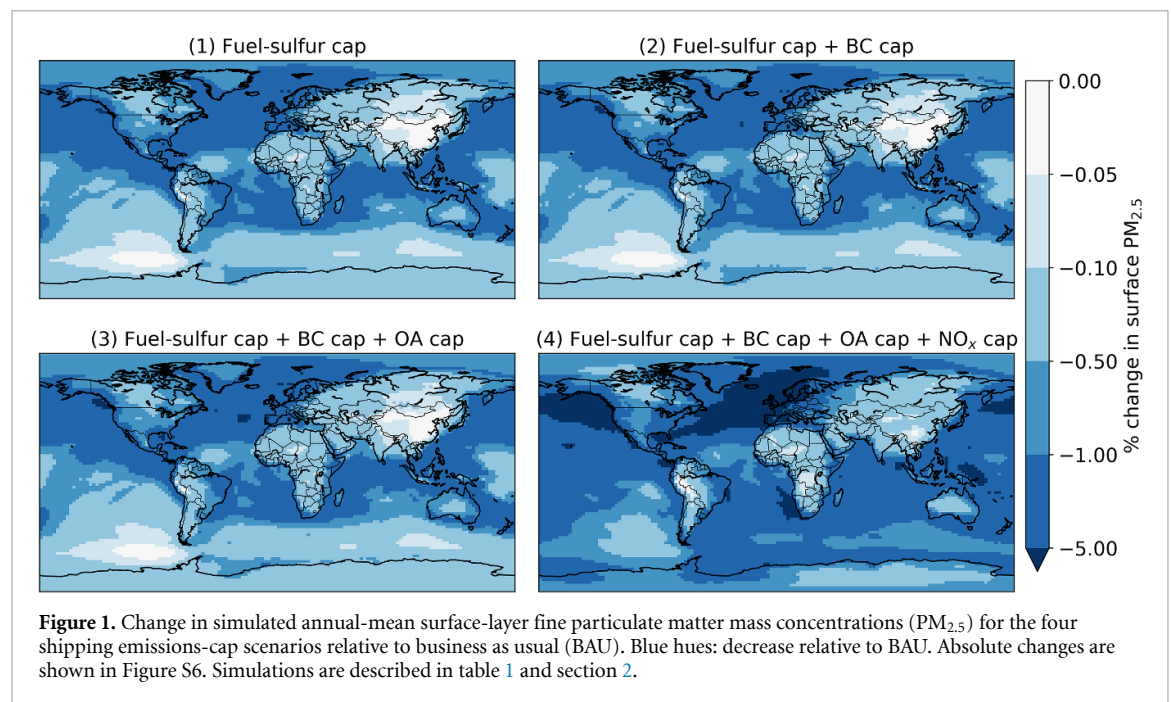
(figure 1, table 2) (figures S1–S5 (available online at <https://stacks.iop.org/ERL/15/124046/mmedia>) show changes by species). Relative to the SO_x and NO_x caps, the BC and OA caps yielded marginal reductions in PM_{2.5} (SO_x: −0.69% and NO_x: −0.64% versus BC and OA: −0.02%), because SO₂ and NO emissions were an order of magnitude larger than BC and OA emissions from ships (table 1). Across all simulations, the largest fractional decreases in surface PM_{2.5} occurred in regions with heavy ship traffic. The largest absolute reductions in PM_{2.5} were also along major shipping routes for Sulf, Sulf-BC, and Sulf-BC-OA (figure S6). However, for the Sulf-BC-OA-NO_x scenario, the largest decrease in absolute PM_{2.5} was over the European continent, far from major shipping routes (figure S6). This change was driven by a reduction of surface NO₃[−] (figure S4). Ammonium nitrate (NH₄NO₃) formation over Europe was likely NO₃[−] limited, thus decreasing NO_x led to a decrease in particle-phase NH₄NO₃ and PM_{2.5}.

3.2. Health effects under emission controls

We estimated that our scenarios could avert between 13 300 (Sulf) and 38 600 (Sulf-BC-OA-NO_x) PM_{2.5}-attributable mortalities per year (figure 2, table 3). Averted mortalities were similar between Sulf, Sulf-BC, and Sulf-BC-OA, but were almost twice as high for Sulf-BC-OA-NO_x. Comparing simulations, we estimated that the NO_x and SO_x caps alone could avert 24 100 and 13 300 mortalities per year, respectively. In contrast, the BC and OA caps could avert 500 and 600 mortalities per year, respectively. Using the value of a statistical life (\$9.7 million per year) from the US Environmental Protection Agency (2016), we estimate that these scenarios would save \$129 billion (Sulf) to \$374 billion (Sulf-BC-OA-NO_x) per year (see Note S2 for details). The US, Europe, Russia, India,

Table 2. Top row, unshaded: Global-mean percent change in mass concentration of fine particulate matter (PM_{2.5}), sulfate (SO₄^{2−}), black carbon (BC), organic carbon (OA), nitrate (NO₃[−]), ammonium (NH₄⁺) and change in number concentration of particles with diameter greater than 10 nm (N10) and greater than 80 nm (N80). Changes are for the four shipping emissions-cap scenarios, relative to the business-as-usual scenario. 25th percentile and 75th percentiles are in parentheses. Bottom row, shaded gray: As above, for the Southern Hemisphere only. Negative numbers: mass reduction relative to BAU. Positive numbers: mass increase relative to BAU. Simulations are described in table 1 and section 2.

	Sulf—BAU	Sulf-BC—BAU	Sulf-BC-OA—BAU	Sulf-BC-OA-NO _x —BAU
Surface PM _{2.5} [%]	−0.7 (−1.2, −0.2)	−0.7 (−1.2, −0.2)	−0.7 (−1.3, −0.2)	−1.4 (−2.5, −0.7)
	−0.7 (−0.7, −0.2)	−0.7 (−0.7, −0.2)	−0.7 (−0.7, −0.2)	−1.5 (−2.0, −0.6)
Column SO ₄ ^{2−} [%]	−1.5 (−1.8, −0.5)	−1.5 (−1.8, −0.5)	−1.5 (−1.8, −0.5)	0.7 (−0.1, 2.9)
	−0.9 (−0.8, −0.4)	−0.9 (−0.8, −0.4)	−0.9 (−0.9, −0.42=)	2.4 (2.3, 3.4)
Column BC [%]	0.0 (0.1, 0.2)	−0.8 (−2.0, −1.1)	−0.8 (−2.0, −1.1)	−0.9 (−2.2, −1.2)
	0.1 (0.1, 0.2)	−0.7 (−1.8, −1.1)	−0.7 (−1.8, −1.1)	−0.8 (−2.2, −1.5)
Column OA [%]	0.1 (0.1, 0.2)	0.1 (0.1, 0.2)	0.0 (0.0, 0.1)	−0.2 (−0.5, −0.2)
	0.1 (0.1, 0.2)	0.1 (0.1, 0.2)	0.0 (0.1, 0.1)	−0.2 (−0.5, −0.4)
Column NO ₃ [−] [%]	0.9 (0.8, 2.0)	0.9 (0.8, 2.0)	0.9 (0.8, 2.0)	−2.6 (−8.3, −1.7)
	1.0 (0.9, 1.2)	1.0 (0.9, 1.2)	1.0 (0.9, 1.2)	−5.9 (−9.6, −7.0)
Column NH ₄ ⁺ [%]	−1.2 (−1.5, −0.4)	−1.2 (−1.5, −0.4)	−1.2 (−1.5, −0.4)	−1.5 (−1.8, −0.6)
	−1.5 (−1.2, −0.4)	−1.5 (−1.2, −0.4)	−1.5 (−1.2, −0.4)	−1.5 (−1.4, −0.7)
900 hPa N10 [%]	−1.1 (−1.1, 0.1)	−1.1 (−1.6, 0.0)	−1.2 (−2.1, −0.1)	−2.4 (−4.7, −0.8)
	−1.0 (−0.9, 0.0)	−1.0 (−1.2, −0.1)	−1.1 (−1.5, −0.1)	−3.0 (−4.3, −0.6)
900 hPa N80 [%]	−0.3 (−0.6, 0.0)	−0.4 (−0.9, −0.1)	−0.6 (−1.3, −0.3)	−1.0 (−2.2, −0.2)
	−0.2 (−0.5, 0.1)	−0.4 (−0.7, −0.1)	−0.5 (−1.1, −0.2)	−0.9 (−1.5, 0.7)



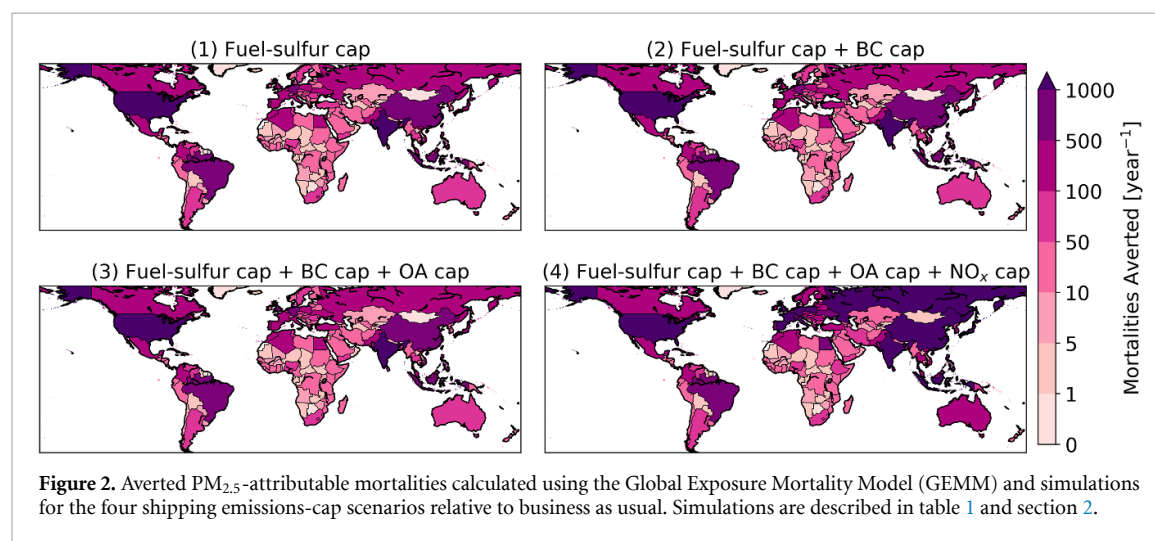
Brazil, and China had the most substantial health benefits, following the changes in PM_{2.5} and population (figures 1 and S6).

Using our BAU simulation, we estimate that PM_{2.5} from natural and anthropogenic sources leads to 6.2 million mortalities per year (uncertainty range: 4.5–7.6 million). The 2017 GBD estimate (4.3 million mortalities per year in 2013) falls within this range (GBD 2017 Risk Factor Collaborators 2018). Using our No-Anthro and BAU simulations, we estimate that anthropogenic PM_{2.5} leads to 4.8 million mortalities per year (uncertainty range: 3.4–5.9 million). Using this estimate, we find that Sulf, Sulf-BC, and Sulf-BC-OA could reduce anthropogenic PM_{2.5}

mortality by 0.28%, while Sulf-BC-OA-NO_x could reduce anthropogenic PM_{2.5} mortality by 0.80%. Independently, the SO_x cap could reduce anthropogenic PM_{2.5} mortality by 0.28%, the addition of BC and OA caps could reduce mortality by 0.01%, and the NO_x cap reduced mortality by 0.50%. Air pollution from shipping primarily impacts coastal regions and port cities (Sofiev *et al* 2018), whereas most ship emissions are over remote oceans. Similar results have been found for aviation (Grobler *et al* 2019), highlighting that the mortality burden per mass emissions is much larger for sources that are co-located with human population centers (Lelieveld *et al* 2015).

Table 3. Global averted $\text{PM}_{2.5}$ -attributable mortalities, aerosol direct radiative effect (DRE, external and core shell mixing), cloud-albedo indirect effect (AIE) and DRE + AIE, for the four shipping emissions-cap scenarios simulations. Changes are relative to business as usual (BAU). Uncertainty ranges are in parentheses. For the averted $\text{PM}_{2.5}$ mortalities: global mean estimate and $\pm 2 \times$ standard error. For the radiative effects, top row, unshaded: global mean, 25th percentile, and 75th percentiles in parentheses. Bottom row, shaded: As above, for the Southern Hemisphere only.

	Sulf	Sulf-BC	Sulf-BC-OA	Sulf-BC-OA- NO_x
Averted $\text{PM}_{2.5}$ mortalities (year^{-1})	13 300 (9800, 16 200)	13 800 (10 200, 16 800)	14 500 (10 700, 17 500)	38 600 (28 500, 46 600)
DRE (external) (mW m^{-2})	16.9 (2.6, 16.9)	16.1 (2.3, 15.9)	16.1 (2.3, 15.8)	19.4 (1.8, 18.4)
DRE (core-shell) (mW m^{-2})	6.4 (0.9, 7.0)	6.1 (0.7, 6.7)	6.1 (0.7, 6.6)	9.7 (0.9, 12.2)
AIE (mW m^{-2})	10.1 (0.0, 12.3)	15.1 (0.3, 17.3)	22.1 (0.7, 24.2)	21.1 (−0.4, 31.9)
DRE (external) + AIE (mW m^{-2})	8.7 (0.0, 11.6)	12.3 (0.1, 15.7)	17.1 (0.2, 21.0)	11.6 (−2.3, 17.9)
DRE (external) + AIE (mW m^{-2})	27.0 (5.0, 28.2)	31.2 (5.3, 32.1)	38.2 (6.5, 38.8)	40.5 (1.4, 48.5)
DRE (external) + AIE (mW m^{-2})	15.1 (0.5, 18.5)	18.5 (0.6, 22.1)	23.1 (1.1, 27.3)	21.2 (−0.9, 31.5)



We found a smaller health benefit from the 2020 fuel-sulfur cap than previous studies (Partanen *et al* (2013): 48 200 mortalities averted, Winebrake *et al* (2009): ~41 200 mortalities averted, and Sofiev *et al* (2018): 137 000 mortalities averted). The model in this study was run at $2.0^\circ \times 2.5^\circ$ while Partanen *et al* (2013) was run at $1.9^\circ \times 1.9^\circ$; Winebrake *et al* (2009) was run at $2.8^\circ \times 2.8^\circ$; and Sofiev *et al* (2018) was run at $0.1^\circ \times 0.1^\circ$. Differences in model resolution may have contributed to differences in NO_3^- estimates due to nonlinear chemistry. However, all studies estimated similar changes in $\text{PM}_{2.5}$ (less than a few percent). The choice of concentration–response function was one of the primary reasons for interstudy differences because the concentration–response function is the largest source of uncertainty in health impact assessments (Ford and Heald 2016, Ford *et al* 2018, Kodros *et al* 2018). The GEMM (used in this work) is a meta-analysis of some of the most recent epidemiology studies that use a range of globally-relevant outdoor air pollution exposures (Burnett *et al* 2018). The GEMM is likely one of the best concentration–response functions for estimating the global health impacts of outdoor air pollution. Differences in

baseline mortality rates, population estimates, emissions, and $\text{PM}_{2.5}$ estimates may also contribute to interstudy differences.

3.3. Changes in aerosol mass and number under emission controls

We found both increases and decreases in the column mass of SO_4^{2-} , BC, OA, and NO_3^- , depending on the scenario, while NH_4^+ decreased across all scenarios (figures S7–S11, table 2). Relative to BAU, global-average column SO_4^{2-} decreased for Sulf, Sulf-BC, and Sulf-BC-OA and increased for Sulf-BC-OA- NO_x (table 2). The NO_x cap decreased atmospheric oxidant concentrations (figures S12 and S13), which increased the chemical lifetime of SO_2 such that more SO_2 was transported to the free troposphere before being oxidized to SO_4^{2-} , where it has a longer lifetime. Surface-level SO_4^{2-} generally decreased for Sulf-BC-OA- NO_x (figure S1), while column-level SO_4^{2-} generally increased (figure S7), suggesting a changing SO_4^{2-} lifetime. The ozone (O_3) reductions in Sulf-BC-OA- NO_x will likely have health benefits (figure S12). We did not evaluate them because the resolution of our simulations does not capture city-scale NO_x -saturated O_3 chemistry.

For Sulf, Sulf-BC, and Sulf-BC-OA, column NO_3^- increased, across most of the globe, relative to the BAU (table 2, figure S10). In these regions, the atmosphere was in the chemical $\text{SO}_4^{2-}\text{NO}_3^-$ substitution regime (at least some of the time), because when SO_4^{2-} decreased and enough HNO_3 was present, NH_4NO_3 formed, increasing column NO_3^- . However, column NO_3^- decreased across much of the globe for Sulf-BC-OA- NO_x , relative to BAU, which indicates that much of the atmosphere was nitric acid (HNO_3) limited rather than NH_4^+ limited (at least some of the time).

The global-average BC and OA column changes were small (table 2). For Sulf-BC, Sulf-BC-OA, and Sulf-BC-OA- NO_x , column BC decreased due to the BC emissions cap, while for Sulf, BC mass increased (table 2, figure S8). Reduced SO_x emissions reduced the H_2SO_4 vapor available to condense onto BC. Thus, the BC-containing particles became less hygroscopic, reducing their wet deposition rates. These changes increased the BC atmospheric lifetime and loading. Likewise, the global-average OA increased for Sulf and Sulf-BC (table 2, figure S9). For Sulf-BC-OA, OA decreased in the Northern Hemisphere but increased in parts of the Southern Hemisphere because the OA from other sources had a longer lifetime relative to the BAU simulation. For Sulf-BC-OA- NO_x , global-average OA decreased because SO_4^{2-} increased in the Southern Hemisphere, decreasing OA lifetime. If the SO_x emissions cap was not included, the BC and OA impacts would likely be larger.

Relative to BAU, the number concentration of particles with diameter larger than 10 nm (N10) and 80 nm (N80) decreased over the shipping lanes for the four emission caps (figures S14 and S15, table 2). However, number concentrations increased downwind of the shipping lanes due to aerosol microphysical feedbacks (Westervelt *et al* 2014). Reduced particle concentrations over the shipping lanes led to a smaller condensation sink and increased the concentrations of vapors available for nucleation and growth, yielding increases in N10 and N80 away from the shipping lanes. Increases were larger for N10 than N80, which is characteristic of this microphysical feedback (Westervelt *et al* 2014).

3.4. Radiative effects under emission controls

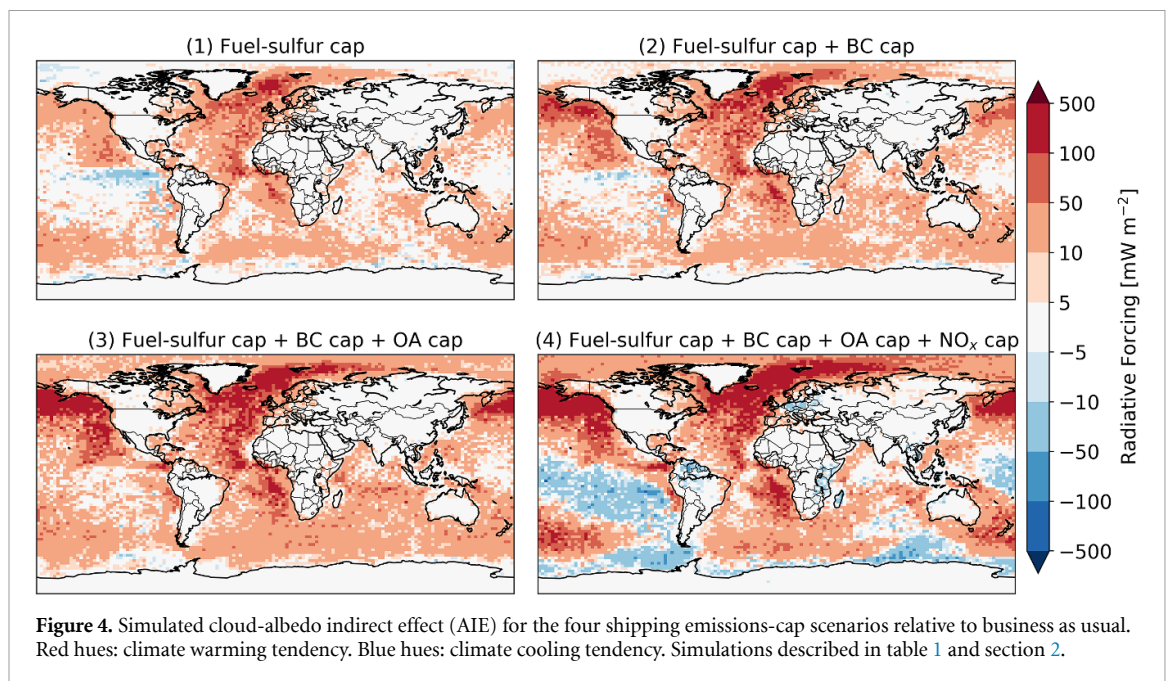
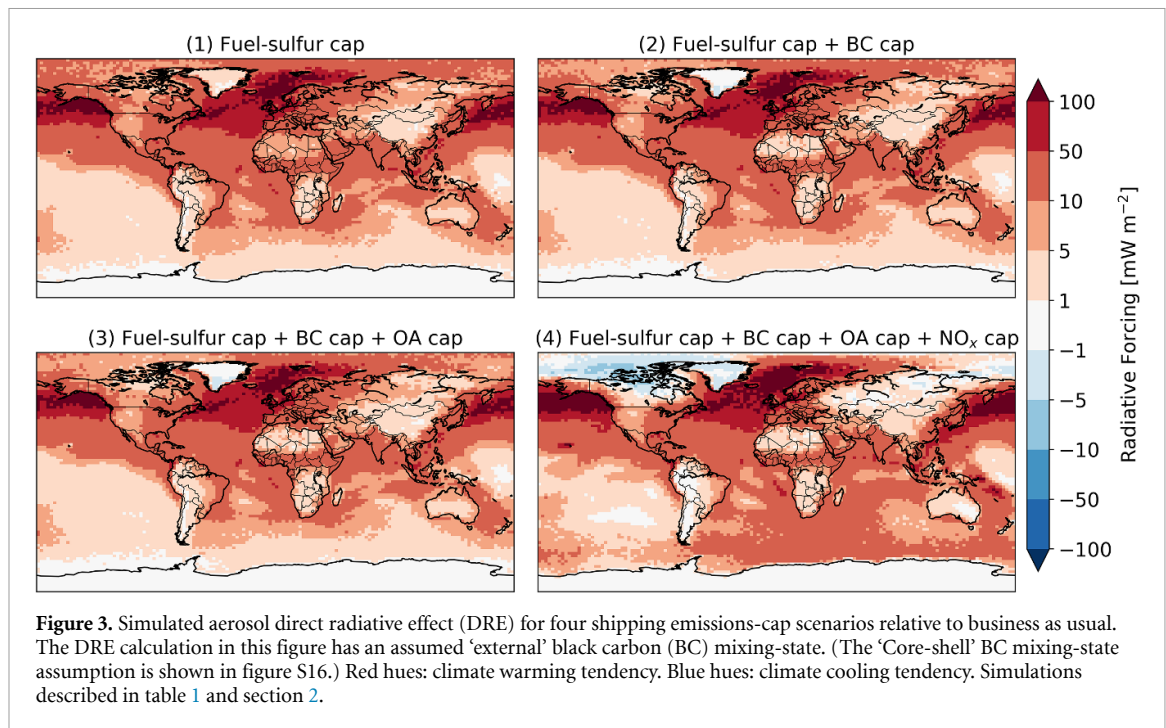
We estimated a global-average climate warming tendency (positive DRE) for all emission caps, relative to BAU. For the external mixing-state assumption, the DRE ranged from 17 mW m^{-2} (Sulf-BC-OA) to 19 mW m^{-2} (Sulf-BC-OA- NO_x) (table 3). The DRE did not differ substantially between the two mixing-state assumptions (table 3, figure S16). The BC and OA caps had a smaller impact on the DRE than the SO_x and NO_x caps because of lower emission rates. The largest DRE was along

the North-America/Europe shipping routes and the Asia/North-America shipping routes (250 mW m^{-2} for Sulf-BC-OA- NO_x) (figure 3). There were DRE changes across the globe, including over the Arctic, which is particularly susceptible to changes in the aerosol radiative forcing (Azzara *et al* 2015, Croft *et al* 2016).

For AIE, a global-average climate warming tendency (positive AIE) was estimated for all emissions-caps, relative to BAU. AIE ranged from 10 mW m^{-2} (Sulf) to 21 mW m^{-2} (Sulf-BC-OA- NO_x) (table 3). The AIE followed similar patterns to the N80 changes (figures 4 and S15). The largest AIE (up to 460 mW m^{-2} for Sulf-BC-OA- NO_x) was along the eastern US-to-Europe shipping routes, US-to-Asia shipping routes, and off the West African coast. The BC and OA caps had a larger impact on AIE than DRE; unlike SO_2 and NO , they are emitted as primary particles, directly influencing particle number and CCN along shipping routes.

Combining the DRE and AIE, the global anthropogenic aerosol forcing increased by 27 mW m^{-2} (Sulf) to 41 mW m^{-2} (Sulf-BC-OA- NO_x) (figure S16). Using our No-Anthro and BAU simulations, we estimate that the global-averaged combined DRE and AIE from anthropogenic aerosols is -1300 mW m^{-2} . This value is within the range given by the International Panel on Climate Change (mean: -900 mW m^{-2} ; range: -1900 to -100 mW m^{-2}) (Stocker *et al* 2013). Thus, our scenarios decrease the cooling tendency of anthropogenic aerosols by 2.1% (Sulf), 2.4% (Sulf-BC), 3.0% (Sulf-BC-OA), and 3.2% (Sulf-BC-OA- NO_x). Comparing simulations, the forcing of individual caps ranged from 2.3 and 27 mW m^{-2} . To estimate what effect these forcings may have on surface temperature, we used an estimate of the Earth's equilibrium climate sensitivity is $\sim 0.8 \text{ K (W m}^{-2})^{-1}$ (Stocker *et al* 2013). With this value, we estimate that our scenarios could increase global temperatures between $\sim 0.02 \text{ K}$ (Sulf) and $\sim 0.03 \text{ K}$ (Sulf-BC-OA- NO_x). Using the Dynamic Integrated Climate-Economy model (DICE 2013-R), we estimate that our emission caps would cost the global economy between \$12 billion (Sulf) to \$17 billion (Sulf-BC-OA- NO_x) per year in 2020 dollars (see Note S2 for details).

Previous combined DRE and AIE estimates for the 0.5% sulfur-fuel cap (Sulf) have varied widely. Our estimates agree with Sofiev *et al* (2018), who estimated a combined DRE and AIE of 71 mW m^{-2} , but are much lower than Partanen *et al* (2013) (330 mW m^{-2}) and Laurer *et al* (2009) ($\sim 270 \text{ mW m}^{-2}$). The latter two estimates are on the order of one third of the total anthropogenic radiative forcing from aerosols (Stocker *et al* 2013). The high forcing estimate in Laurer *et al* (2009) was driven by a large AIE, while Partanen *et al* (2013) did not estimate the DRE and AIE separately. Both Partanen *et al* (2013) and Laurer *et al* (2009) included



the aerosol-cloud lifetime effect, while Sofiev *et al* (2018) and our study did not. We did not include the simulated lifetime effect due to its large uncertainties (Stocker *et al* 2013). Had all studies included the lifetime effect (or omitted it), there might be better interstudy agreement. Differences in model resolution or the subgrid treatment of sulfate chemistry (i.e. ‘primary sulfate’) could contribute to radiative forcing estimate differences (Stevens and Pierce 2014). Partanen *et al* (2013) emitted 2.5% of SO₂ emissions from ships as SO₄²⁻ as single Aitken mode ($\mu = 44$ nm), unlike our assumptions (section 2.1). These differences can lead to large changes

in CCN and radiative forcing estimates (Stevens and Pierce 2014).

3.5. Future work

In the future, shipping rates and routes may change, particularly in the Arctic; as Arctic sea ice melts, shipping will likely increase. Corbett *et al* (2010) predicted 5% of global shipping traffic will use Arctic routes by 2050. Future work could consider these impacts. In marine stratocumulus clouds, the CCN from ship tracks may turn open-cell stratocumulus to closed-cell stratocumulus, increasing the liquid water path (LWP) and cloud-albedo (Christensen and

Stephens 2011, Goren and Rosenfeld 2015). Future work could examine the impacts of LWP and cloud-lifetime changes, ideally with cloud-resolving models; although, Malavelle *et al* (2017) suggested that these cloud lifetime effects are minor in some regions.

We added the SO_x, BC, OA, and NO_x caps following the order that emission controls have generally been implemented for terrestrial, heavy-duty diesel engines (May *et al* 2014). Future work could involve investigating a combined SO_x and NO_x cap (Sulf-NO_x) since NO_x are currently regulated in some areas (e.g. emission control areas) and may be regulated globally in the future (Brynnolf *et al* 2014). We expect the averted PM_{2.5}-attributable mortality and DRE for a Sulf-NO_x scenario to be similar to Sulf-BC-OA-NO_x (because the mass of BC and OA emissions are small relative to SO_x and NO_x). However, the AIE could differ more between Sulf-NO_x and Sulf-BC-OA-NO_x (because primary BC and OA directly influence CCN).

4. Policy implications

We estimated the averted PM_{2.5}-attributable mortalities and aerosol radiative effects (DRE and AIE) of four shipping emissions-control scenarios. Sulf is being realized this year, while Sulf-BC, Sulf-BC-OA, and Sulf-BC-OA-NO_x represent potential future emission control policies that generally follow the order that emissions have been regulated for terrestrial, heavy-duty diesels (May *et al* 2014). These policies could avert 13 300 (Sulf) to 38 600 (Sulf-BC-OA-NO_x) mortalities annually. The BC and OA caps' health benefits were smaller than the SO_x or NO_x caps because BC and OA emissions from ships are smaller by mass. Future emissions control policies targeting NO_x emissions will likely provide the largest health benefit, because Sulf-BC-OA-NO_x averted nearly twice the number of mortalities of the other scenarios.

In contrast to the health benefits, we estimated a climate warming tendency for all scenarios, with a combined DRE and AIE of 27–41 mW m⁻². The SO_x and NO_x caps had a greater impact on the aerosol radiative forcing than the BC and OA caps. However, per mass of emissions, the BC and OA caps had a large effect on AIE, because BC and OA are emitted as primary particles, which more directly impact CCN relative to particle precursors like SO_x and NO_x. The NO_x cap also decreased atmospheric oxidant concentrations, which increased the chemical lifetime of SO₄²⁻.

Overall, our aerosol-focused emissions caps would lead to larger relative climate effects than health effects. Our scenarios could reduce anthropogenic PM_{2.5} mortalities by 0.3%–0.8% but would decrease the cooling effect of anthropogenic aerosols by 2.1%–3.2% (warming the planet). However, when comparing the health and climate costs, we estimate that the health benefits (\$129–\$374 billion

annually) outweigh the climate costs (\$12–\$17 billion annually). Additionally, the ratio of health benefits to climate costs may be further improved with targeted policies. For example, current emissions levels could be allowed in remote areas where emissions lead to climate cooling effects with minimal health impacts. Alternatively, emission-control regulations could target coastal shipping vessels or vulnerable regions. Cruise ships emit more BC (which strongly absorbs radiation) than container ships on a per-mass basis (Comer *et al* 2017) and spend substantial time in port cities and are therefore more likely to impact human health. Regions, like the Arctic, are especially vulnerable to positive climate forcing, so reducing BC emissions in the Arctic may provide substantial climate benefits (Azzara *et al* 2015). Finally, focusing on reducing greenhouse gases (e.g. through fuel switching (Comer 2019, Zhou *et al* 2020)) in conjunction with aerosol emissions caps, may provide simultaneous health and climate benefits.

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Data availability

The data used in this work is (available at <http://dx.doi.org/10.25675/10217/208319>). The data were created using the GEOS-Chem model v12.6.0. The code can be (accessed at: <https://doi.org/10.5281/zenodo.3507501>).

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