Climate and Air Quality Impact of Using Ammonia as an Alternative Shipping Fuel

Anthony Y. H. Wong[1], Noelle E. Selin[2,3], Sebastian D. Eastham[4,5], Christine Mounaïm-Rouselle[6], Yiqi Zhang[7], Florian Allroggen[5]

[1] Centre for Global Change Science, Massachusetts Institute of Technology, Cambridge, MA, USA
[2] Institute for Data, Systems and Society, Massachusetts Institute of Technology, Cambridge, MA, USA
[3] Department of Earth, Atmospheric, and Planetary Sciences, Massachusetts Institute of Technology, Cambridge, MA, USA
[4] Joint Program on the Science and Policy of Global Change, Massachusetts Institute of Technology, Cambridge, MA, USA
[5] Laboratory for Aviation and the Environment, Department of Aeronautics and Astronautics, Massachusetts Institute of Technology, Cambridge, MA, USA
[6] Laboratoire PRISME, Université d'Orléans, Orléans, France
[7] Division of Environment and Sustainability, Hong Kong University of Science and Technology, Clear Water Bay, Hong Kong, China

Abstract

As carbon-free fuel, ammonia has been proposed as an alternative fuel to facilitate maritime decarbonization. Deployment of ammonia-powered ships is proposed as soon as 2024. However, NO_x, NH_3 and N_2O from ammonia combustion could impact air quality and climate. In this study, we assess whether and under what conditions switching to ammonia fuel might affect climate and air quality. We use a bottom–up approach combining ammonia engine experiment results and ship track data to estimate global tailpipe NO_x, NH_3 and N_2O emissions from ammonia-powered ships with two possible engine technologies (NH_3–H_2 (high NO_x, low NH_3 emissions) vs pure NH_3 (low NO_x, very high NH_3 emissions) combustion) under three emission regulation scenarios (with corresponding assumptions in emission control technologies), and simulate their air quality impacts using GEOS–Chem High Performance global chemical transport model. We find that the tailpipe N_2O emissions from ammonia-powered ships have climate impacts equivalent to 5.8% of current shipping CO_2 emissions. Globally, switching to NH_3–H_2 engines avoids 16,900 mortalities from $PM_{2.5}$ and 16,200 mortalities from O_3 annually, while the unburnt NH_3 emissions (82.0 Tg NH_3 yr^{-1}) from pure NH_3 engines could lead to 668,100 additional mortalities from $PM_{2.5}$ annually under current legislation. Requiring NH_3 scrubbing within current Emission Control Areas leads to smaller improvements in $PM_{2.5}$-related mortalities (22,100 avoided mortalities for NH_3–H_2 and 623,900 additional mortalities for pure NH_3 annually), while extending both Tier III NO_x standard and NH_3 scrubbing requirements globally leads to larger improvement in $PM_{2.5}$-related mortalities associated with a switch to ammonia-powered ships (66,500 avoided mortalities for NH_3–H_2 and 1,200 additional mortalities for pure NH_3 annually). Our findings suggest that while switching to ammonia fuel would reduce tailpipe greenhouse gas emissions from shipping, stringent ammonia emission control is required to mitigate the potential adverse effects on air quality.

DO NOT CITE, COPY OR DISTRIBUTE

Introduction

Maritime shipping burns fossil fuels in large diesel engines for energy (propulsion, heat, and electricity), which leads to emissions of CO_2 and air pollutants. The main air pollutants emitted by the maritime transport sector include SO_x ($\equiv SO_2 + SO_4^{2-}$), NO_x ($\equiv NO + NO_2$), non-methane volatile organic compound (NMVOC), CO and carbonaceous aerosols. These are either components or precursors of particulate matter (PM) and ozone (O_3). Exposure to PM, particularly the fine PM (aerodynamic diameter < 2.5 μm, named $PM_{2.5}$) that can reach deep inside the respiratory tract, is estimated to have caused 3.7 – 4.8 million deaths in 2015 by increasing the risk of cardiopulmonary and cerebrovascular diseases (Cohen *et al* 2017). O_3 exposure exerts oxidative stress on the respiratory tract (Nuvolone *et al* 2018), which also leads to increased risk of cardiopulmonary diseases, and therefore another 1.04 – 1.24 millions of respiratory deaths in 2010 globally (Malley *et al* 2017). Shipping emissions are estimated to account for 2.7% of global energy-related CO_2 emissions and caused an estimated 84800 – 103000 annual premature deaths from $PM_{2.5}$ exposure globally in 2015 (Zhang *et al* 2021b), and account for up to 14 and 25% of $PM_{2.5}$ concentration over East Asia and Mediterranean area, respectively (Contini and Merico 2021).

The International Maritime Organization (IMO) has outlined a goal of reducing greenhouse gas (GHG) emissions from international shipping by at least 40% by 2030 compared to the 2008 level (International Maritime Organization 2018). The uses of alternative fuels (e.g. NH_3, H_2, methanol) and other energy solutions (e.g. electrification) are essential for reaching such a decarbonization goal (Balcombe *et al* 2019). NH_3 is one of the main candidates for alternative maritime fuels, and could represent up to 43% of the energy mix of shipping in 2050 (IRENA 2021). Since NH_3 is mainly manufactured with H_2 and N_2 through the Haber-Bosch Process, the carbon footprint of NH_3 production can be reduced by carbon capture (blue NH_3), or using renewable energy for N_2 and H_2 production and the synthesis process (green NH_3) (Valera-Medina *et al* 2021).

Wolfram *et al* (2022) and Bertagni *et al* (2023) summarized scientific concerns about the potential environmental impacts of using NH_3 as a marine fuel. NH_3 combustion may generate additional NO_x and N_2O compared to other fuels (Hinokuma and Sato 2021). NH_3 emission is one of the major source of global $PM_{2.5}$ pollution (e.g. Gu *et al* 2021) by neutralizing H_2SO_4 and HNO_3 in the atmosphere (Jacob 1999). Heo *et al* (2016) find that NH_3 emission leads to much higher $PM_{2.5}$ mortality costs per ton ($23000 – 66000) than SO_2 ($14000 – 24000) and NO_x ($3800 – 14000) in the United States. These show the potential danger of uncontrolled NH_3 emission via worsening $PM_{2.5}$ air quality. Emitted NO_x and NH_3 would then deposit to Earth's surface, causing damages to ecosystems (e.g. soil acidification and eutrophication) and may lead to additional emission of N_2O, which is a potent greenhouse gas and contributes to stratospheric ozone depletion.

Here, we explore the possible ranges of air quality and climate impacts of transitioning from using fossil fuels to ammonia as the major shipping fuel under different technologies and policies, aiming to highlight the opportunities and challenges of ammonia combustion as a strategy to decarbonize maritime transport.

DO NOT CITE, COPY OR DISTRIBUTE

Method

We use a bottom–up approach to estimate the global NO_x, NH_3 and N_2O emissions from converting the entire fleet into NH_3–powered ships as a function of engine technologies, emission control strategies and policy under 6 scenarios, using results from ammonia engine experiments and ship Automatic Identification System (AIS) data. We then simulate the associated changes in O_3 and $PM_{2.5}$ air quality using a global 3-D chemical transport model (GEOS-Chem High Performance). Finally, we estimate the impacts of simulated changes in O_3 and $PM_{2.5}$ on public health (expressed in annual premature mortalities) using concentration functions derived from epidemiological studies.

Scenarios

Scenario Name	Emission control inside current ECA	Emission control outside current ECA	Equivalent policy scenario
Baseline	Zhang et al. (2021) inventory for 2015 shipping with 0.5% sulphur cap		
Post-2020 NO_x baseline	Baseline with Tier III NO_x (post-2020) standard imposed globally		
$[NH_3–H_2]_{2020}$	SCR	SCR	2020 NO_x limit
$[NH_3–H_2]_{NH3\_ECA\_LIM}$	SCR+NH_3 scrubbing	SCR	Additional NH_3 limit in ECA
$[NH_3–H_2]_{GLOB\_LIM}$	SCR+NH_3 scrubbing	SCR+NH_3 scrubbing	Global NO_x and NH_3 limits
$[Pure\ NH_3]_{2020}$	SCR	None	2020 NO_x limit
$[Pure\ NH_3]_{NH3\_ECA\_LIM}$	SCR+NH_3 scrubbing	None	Additional NH_3 limit in ECA
$[Pure\ NH_3]_{GLOB\_LIM}$	SCR+NH_3 scrubbing	SCR+NH_3 scrubbing	Global NO_x and NH_3 limits

Table 1. Description of the engine technology and policy scenarios considered in this study. SCR refers to Selective Catalytic Reduction (assumed to be 90% effective), which converts NO_x and NH_3 into N_2 in 1:1 ratio under ideal conditions. NH_3 scrubbing is assumed to remove 95% of NH_3 slip after SCR.

In all scenarios, we apply an AIS-based shipping emission model (Zhang *et al* 2019) to estimate the global spatially-resolved pollutant and GHG emissions for every ship track in 2015 following the technology and policy assumptions of each scenario. The emission model calculates ship emissions as a function of engine power demand, ship specifications, emission factors (EF) and activity time. Missing entries in ship specifications are filled based on the lengths and capacities of the associated ships.

Table 1 shows the scenario design of our study. We choose the emission scenario with 0.5% cap on fuel sulphur content from Zhang *et al* (2021b) as our baseline. The "post–2020 NO_x baseline" scenario imposes the most stringent IMO NO_x emissions (Tier III) limit on top of baseline scenario, which represents the emissions from fossil fuel powered ships if all of them were retrofitted to follow IMO emission standards for newly–built ships. 6 counterfactual scenarios are designed to examine the possible range of air quality outcomes from total conversion to ammonia-powered ships given the possible engine technologies (and therefore emission management strategies) and emission regulations (current legislation versus additional NH_3 emission regulations).

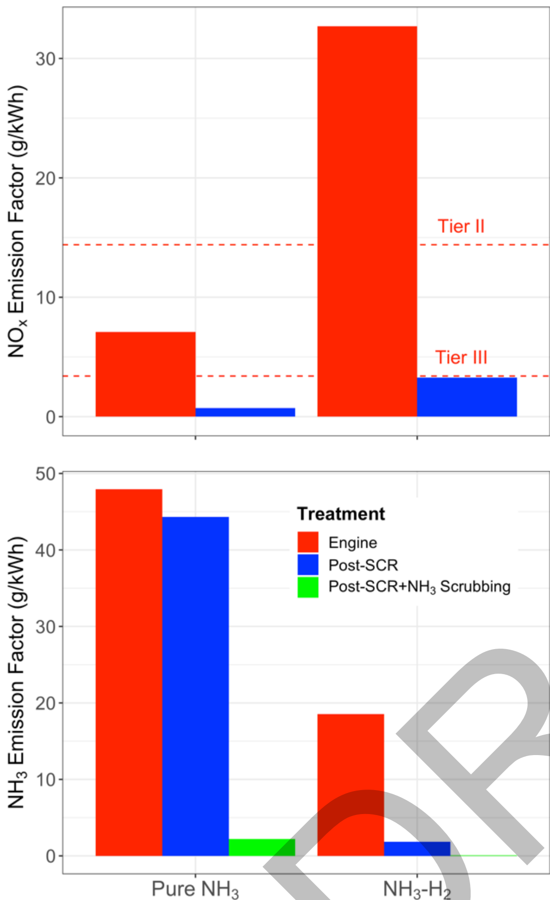

Figure 1. Load-corrected NH_3 and NO_x emission factors (EF) of pure NH_3 and NH_3–H_2 engines, as a function of emission control strategy. Red bar ("Engine") refers to EF from completely untreated engine exhaust. Blue (Post-SCR) and green bars (Post-SCR + NH_3 Scrubbing) refer to EF after implementations of emission control measures. SCR and NH_3 scrubbing are done sequentially. Red dotted lines indicate IMO NO_x regulations for slow engine speed (<130 rpm), which is typical for large engine.

We consider the emissions from ammonia-powered ships with two types of engine technologies. The first type of engine technology considered is pure NH_3 combustion (Mounaïm-Rousselle *et al* 2022). The second type ("NH_3-H_2") is proposed by Imhoff *et al* (2021) based on the experimental data from Lhuillier *et al* (2020). Part of the NH_3 is transferred to a catalytic NH_3 cracker to generate H_2 to improve combustion. This balances NH_3 and NO_x concentration in engine exhaust, allowing both NO_x and NH_3 emissions to be controlled by Selective Catalytic

Reduction (SCR). The derivations of EF and load dependences for the two types of engines, and a discussion about the uncertainty in engine technologies are given as Supplemental Information.

Given the uncertainty in ammonia engine designs, the engine technology scenarios do not intent to realistically replicate how ammonia combustion would be implemented on ships. Rather, the two engine technologies considered in our study reflects two extremes of, and therefore provide bounding scenarios for NO_x and NH_3 emission management approaches: 1) with pure NH_3 engine having low NO_x (currently regulated) and very high NH_3 (currently unregulated) emissions, versus 2) NH_3–H_2 engine that strictly maintains the NO_x/NH_3 ratio to allow SCR to simultaneously control both pollutants.

We consider three policy scenarios. The first ("2020") follows the IMO regulations as of 2020. The untreated NO_x EF are 32.7 g/kWh for NH_3–H_2 and 7.08 g/kWh for pure NH_3 engines following the load corrections prescribed by IMO (International Maritime Organization 2008) (fig. 1). Current IMO guidelines (International Maritime Organization 2017) cap NO_x EF for new vessels at 7.7 – 14.4 g/kWh (Tier II limit) when operating outside the Emission Control Area (ECA, mostly includes North America and United States Caribbean Sea as of 2020, and additionally Baltic Sea and North Sea in 2021) and 2 – 3.4 g/kWh (Tier III limit) within ECA, depending on the engines' rated speed. Compliance with such a guideline would require SCR that can remove 90% of NO_x to operate globally for NH_3–H_2 and within ECA only for pure NH_3 engines. The second ("NH_3_ECA_LIMIT") assumes that additional NH_3 scrubbing requirements (assumed to be 95% effective from available technology) (Melse and Ogink 2005, Van der Heyden *et al* 2015, Boero *et al* 2023) are implemented within ECA for both types of engines, while the third ("GLOB_LIM") extends Tier III NO_x compliance and NH_3 scrubbing requirements to the whole globe.

Atmospheric Chemistry Modeling

We use version 13.4.1 of the GEOS-Chem High Performance model (GCHP, https://doi.org/10.5281/zenodo.4429193) (Martin *et al* 2022, Eastham *et al* 2018) to simulate the response of O_3 and $PM_{2.5}$ to pollutant emission changes in each scenario through resolving the chemistry, transport, emission and deposition of relevant chemical species. The model is driven by the Modern-Era Retrospective analysis for Research and Application (MERRA-2) assimilated meteorological fields (Gelaro *et al* 2017). The model is run at a horizontal resolution of ~200km in cubed-sphere configuration (C48) from 1st Oct 2018 to 31st Dec 2019, with the first 3 months of output discarded as spin-up. O_3 is simulated from a coupled O_3-NO_x-VOCs-CO-halogen-aerosols chemical mechanism (Sherwen *et al* 2016). Anthropogenic emissions are from Community Emission Data System (Hoesly *et al* 2018) except the shipping sector. Biogenic VOCs, soil NO_x and sea salt aerosol emissions follow Weng *et al* (2020) and dust emissions follow Meng *et al* (2021). Re-emissions of deposited NO_x and NH_3 are not considered. Formation of secondary inorganic aerosols are simulated by the ISORROPIA II, which considers thermodynamic equilibrium of the NH_4^+–Na^+–SO_4^{2-}–NO_3^-–Cl^-–H_2O (Fountoukis and Nenes 2007). $PM_{2.5}$ concentrations are derived by summing the mass of its constituents at standard conditions to align with the sampling standard used by the United States Environmental Protection Agency (Latimer and Martin 2019). Ship plume chemistry is parameterized by the

PARANOX scheme (Vinken *et al* 2011). Model evaluation is provided as Supplemental Information.

Health Outcome

We estimate the impacts of air quality changes on public health using the global gridded population data at 30 arc-second resolution from the Gridded Population of the World version 4.11 (Center for International Earth Science Information Network - CIESIN - Columbia University 2018). Country-level age distribution and baseline mortality rates are provided by the World Health Organization (WHO) (WHO 2018). We estimate the risk of relative mortality from chronic O_3 and $PM_{2.5}$ exposure under the baseline (RR_{base}) and each alternative scenario i (RR_i) for every age group. The change in the annual mortality for scenario i ($\Delta Mort_i$) due to some disease for that age group is then calculated for each grid cell as:

$$\Delta \mathrm{Mort}_i = \mathrm{Mort}_{base} \frac{\mathrm{RR}_i - \mathrm{RR}_{base}}{\mathrm{RR}_{base}} \quad (1)$$

where $Mort_{base}$ is the number of mortalities due to that disease in 2016. The relative risk is calculated by comparing the simulated exposure-relevant concentration under scenario i to that under the baseline scenario using an appropriate concentration response function (CRF). We use a log-linear CRF for O_3 from Turner *et al* (2016), which estimate a 12% increase (95% confidence interval (CI): 8.0 – 16%) in respiratory mortality per 10 ppb increase in annual mean maximum daily 8-hour average (MDA8) O_3 concentration. For $PM_{2.5}$ we estimate RR for non-communicable diseases and lower respiratory infections using the age-specific non-linear CRFs from the Global Exposure Mortality Model (Burnett *et al* 2018).

We estimate the median and 95% confidence interval of changes in mortalities due to O_3 and $PM_{2.5}$ for each scenario by performing 1,000 random draws of the CRF parameters in a paired Monte-Carlo simulation.

DO NOT CITE, COPY OR DISTRIBUTE

Result

Modelled Shipping Emissions

Scenario	NO_x (Tg/yr)	NH_3 (Tg/yr)	$CO_{2,e}$ (Tg/yr)
Baseline	17.2	0.004	867
Post-2020 NO_x baseline	3.59		
$[NH_3–H_2]_{2020}$	4.43	2.51	
$[NH_3–H_2]_{NH3\_ECA\_LIM}$	4.43	2.21	
$[NH_3–H_2]_{GLOB\_LIM}$	4.43	0.125	50.2
$[Pure\ NH_3]_{2020}$	6.84	82.0	
$[Pure\ NH_3]_{NH3\_ECA\_LIM}$	6.84	71.7	
$[Pure\ NH_3]_{GLOB\_LIM}$	0.762	3.92	

Table 2 Modelled global total nitrogen-based air pollutants (in Tg/yr) and GHG emissions (in Tg $CO_{2,e}$/yr) from different scenarios. $CO_{2,e}$ (equivalent amount of CO_2 in terms of 100-year Global Warming Potential) is calculated as CO_2 emissions + (N_2O emissions × 273).

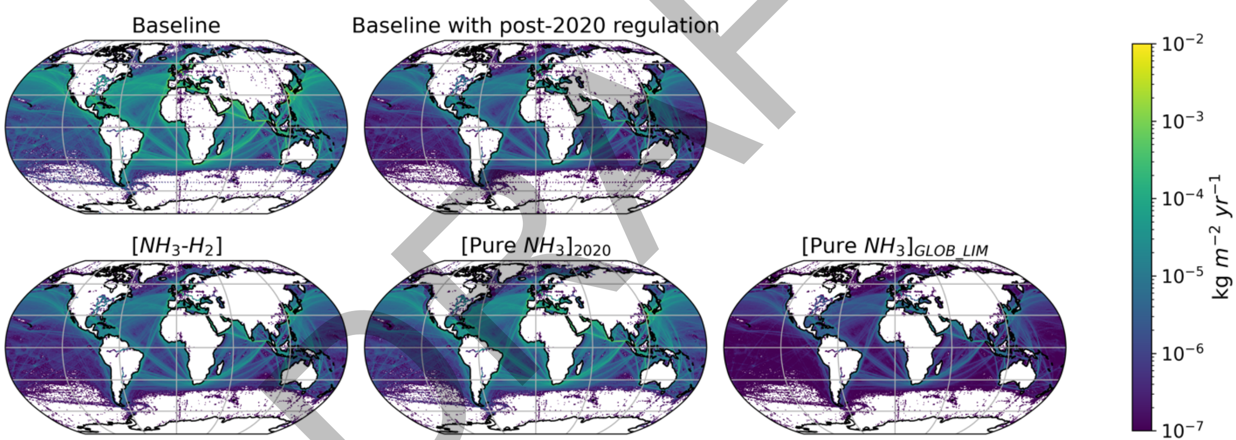

Figure 2. Spatial pattern of annual total NO_x emissions (kg m^{-2} yr^{-1}) under different scenarios.

Table 2 shows the modelled global annual shipping emissions of NO_x, NH_3 and GHG under different scenarios, and Figure 2 shows the spatial distribution of NO_x emissions. Under current regulations (“2020”), ammonia-powered ships have lower NO_x emissions (4.4 Tg NO_x/yr for NH_3–H_2 and 6.9 Tg NO_x/yr for pure NH_3). Such comparison mostly reflects regulatory rather than technological differences, since the older ships in the baseline scenario do not follow the newer and more stringent (Tier II or Tier III) NO_x regulations, while all newly built ammonia-powered ships abide the Tier II regulation outside ECA and Tier III regulations within ECA. To comply with Tier II NO_x regulations, SCR is required for the NH_3–H_2 engine while no NO_x control is needed for the pure NH_3 engine. This leads to higher total post-treatment NO_x emissions from pure NH_3 engines than that from NH_3–H_2 engines, despite pure NH_3 engines has lower pre-treatment NO_x emissions than NH_3–H_2 engines. If the Tier III NO_x regulations is enforced globally ("GLOB_LIM"), the NO_x emission of fossil fuel (3.6 Tg NO_x/yr) and NH_3–H_2 (4.4 Tg NO_x/yr) engines are similar, while pure NH_3 engines (0.8 Tg NO_x/yr) produce the lowest NO_x emissions.

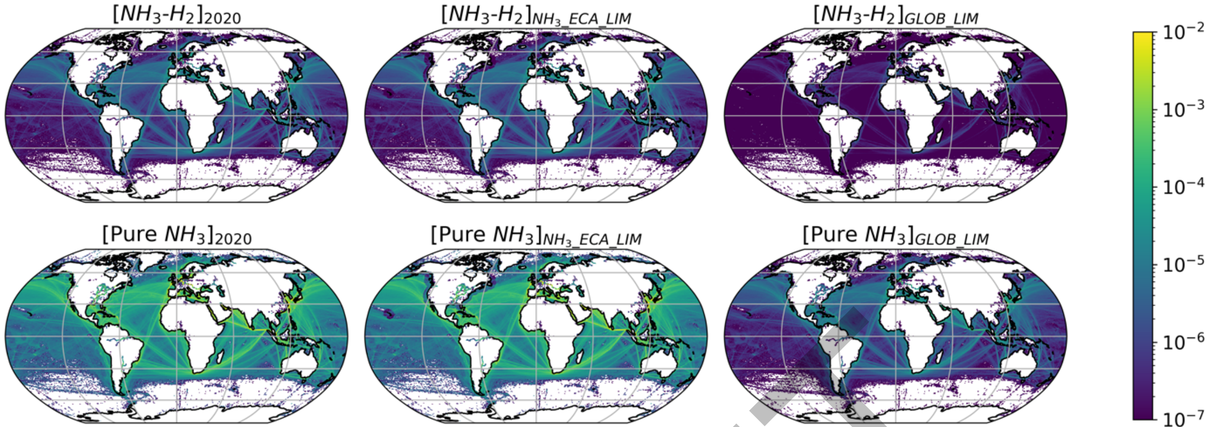

Figure 3. Spatial pattern of annual total NH_3 emissions (kg m^{-2} yr^{-1}) under different scenarios

Figure 3 shows the spatial distribution of modelled NH_3 emissions under different technology and policy scenarios. Under current regulations ("2020"), switching to NH_3-H_2 engines leads to 2.5 Tg/yr NH_3 emissions, while switching to pure NH_3 engines leads to NH_3 emissions (82.0 Tg/yr) that are 32.8 times higher than that from NH_3-H_2 engines. For pure NH_3 engines, SCR can only remove 7% of NH_3 from engine exhaust, leading to high tailpipe NH_3 emissions. In the "NH_3_ECA_LIM" scenario, which requires NH_3 scrubbing over ECA (mostly North American coast and northern Europe), global NH_3 emissions reduce by 12% for both NH_3-H_2 (2.2 Tg/yr) and pure NH_3 (71.7 Tg/yr) engines. In the "GLOB_LIM" scenario, with both SCR and NH_3 scrubbing are required globally, NH_3 emissions fall to 0.1 Tg/yr for NH_3–H_2 engines and 3.9 Tg/yr for pure NH_3 engines.

Table 2 also shows the long-lived GHG emissions from each scenario, given as the equivalent amount of CO_2 ($CO_{2,e}$) in terms of 100-year Global Warming Potential (GWP_{100}) using a conversion factor of 273 from N_2O emission to $CO_{2,e}$ (Smith *et al* 2021). $CO_{2,e}$ from the baseline scenario does not include GHG other than CO_2 (mainly CH_4 and N_2O), which contribute to less than 3% of global shipping $CO_{2,e}$ during 2013 – 2015 (Olmer *et al* 2017). We find that the tailpipe $CO_{2,e}$ from the ammonia-powered fleet is 5.8% of that from the current fossil-fuel-powered fleet. Our analysis (see Supplemental Information) also shows that the "secondary N_2O emissions" from reactive nitrogen deposition (Wolfram *et al* 2022) is not a problem for NH_3-H_2 engine as the total reactive nitrogen emissions are lower than current fleets. For pure NH_3 engine, the net climate effects from nitrogen deposition are likely to be smaller than reduction in tailpipe GHG emissions (817.2 Tg $CO_{2,e}$/yr) from switching to ammonia-powered ships, showing the potential of blue and green ammonia as a climate-friendly shipping fuel, though considerable uncertainties exist on how CO_2 uptake and N_2O emissions respond to nitrogen deposition. This analysis, however, does not fully consider the life cycle GHG emissions (e.g. energy, methane slip) of NH_3 production.

Impacts on Air Quality

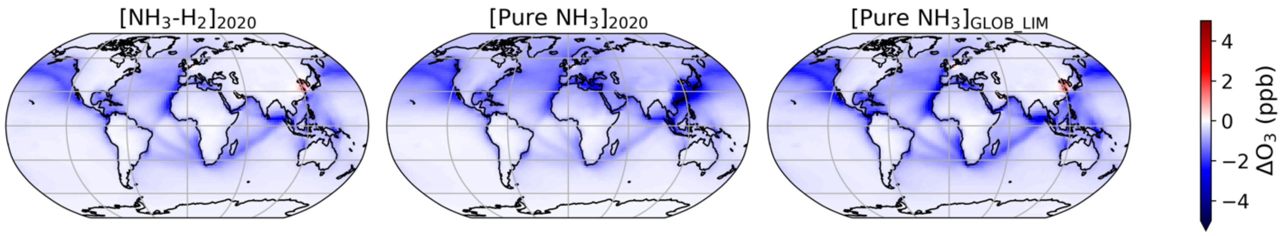

Figure 4. Changes in annual mean MDA8 O_3 concentration (ΔO_3, ppb) for different ammonia-powered ship scenarios

Figure 4 shows the modelled global changes in annual mean MDA8 O_3 due to converting current fleet to ammonia-powered ships with different technology and policy options. Generally, the lower NO_x emissions from ammonia-powered ships reduce annual mean MDA8 O_3. Under all scenarios, global population-weighted average MDA8 O_3 decreases (-0.27 ppb for [NH_3-H_2]$_{2020}$, -1.13 ppb for [Pure NH_3]$_{2020}$, -0.37 ppbv for [Pure NH_3]$_{GLOB\_LIM}$). The greatest reductions in population-weighted O_3 are simulated over coastal and island nations (e.g. 1.5 to 1.9 ppb for Sri Lanka and Djibouti, 1.4 to 2.2 ppb for Panama, 1.4 to 1.7 ppb for Jamaica). However, over highly NO_x–saturated coasts near northern China, northern Europe, and Persian Gulf, local increases in surface O_3 are simulated, especially under the scenarios with greater NO_x reductions ([NH_3-H_2]$_{2020}$ and [Pure NH_3]$_{GLOB\_LIM}$). Over North Sea, the NO_x–saturation leads to further increases in MDA8 O_3 as NO_x emissions become lower, increasing the population-weighted O_3 from 1 ppb under [Pure NH_3]$_{2020}$ to up to 1.5 ppb under [Pure NH_3]$_{GLOB\_LIM}$ over the Netherlands. Over East Asia, population-weighted MDA8 O_3 decreases by 2.4 ppb under the scenario with least NO_x reduction ([Pure NH_3]$_{2020}$), but increases by 0.2 ppb under [Pure NH_3]$_{GLOB\_LIM}$ and [NH_3-H_2]$_{2020}$ as NO_x emissions become lower. This shows the importance of local chemical environment in controlling the response of O_3 pollution to marine NO_x control.

In addition, we find substantial sensitivity of O_3 response to assumptions in ship plume chemistry (mainly NO_x lifetime, see Supplemental Material), which could be a major source of uncertainties. This shows the importance of understanding the plume chemistry of NH_3 ship in capturing the O_3 response.

DO NOT CITE, COPY OR DISTRIBUTE

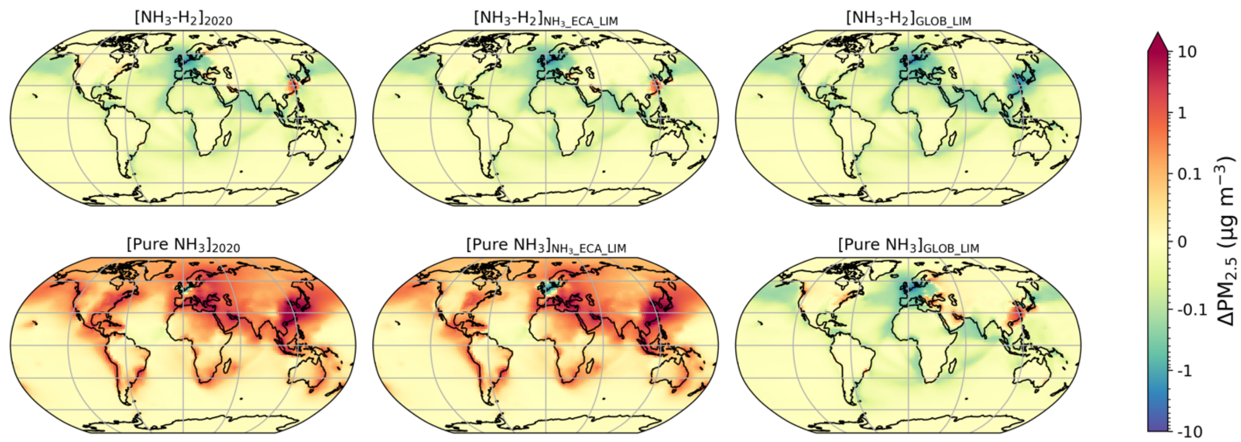

Figure 5 Spatial patterns of changes in annual mean $PM_{2.5}$ concentration ($\Delta PM_{2.5}$, μg m^{-3}) for all ammonia-powered ships scenarios.

Figure 5 shows the modelled changes in annual mean surface $PM_{2.5}$. Under $[NH_3\text{-}H_2]_{2020}$, population-weighted $PM_{2.5}$ increases by 0.21 μg m^{-3} (0.4%) over East Asia (definition of regions follows Giorgi *et al* (2001)). Smaller increases are simulated over western North America (0.08 μg m^{-3}), though the percentage increase (1.7%) is higher since the baseline population-weighted $PM_{2.5}$ (4.82 μg m^{-3}) is low. $PM_{2.5}$ levels are mostly reduced over other regions in the world, especially over northern Europe and Mediterranean Basin, where population-weighted $PM_{2.5}$ decreases by 0.70 (4%) and 0.16 (0.6%) μg m^{-3}, respectively. Under $[NH_3\text{-}H_2]_{NH3\_ECA\_LIM}$, population-weighted $PM_{2.5}$ is reduced by 0.82 μg m^{-3} (4.8%) and 0.055 μg m^{-3} (0.7%) over northern Europe and the United States, respectively, as NH_3 emission control is enforced over those regions. Under $[NH_3\text{-}H_2]_{GLOB\_LIM}$, both Tier III NO_x and NH_3 emission limit are extended globally, resulting in reduced $PM_{2.5}$ levels over the whole globe. Particularly, the negative impacts from NH_3 emission over Mediterranean Basin and East Asia are successfully mitigated, resulting in 0.33 (1.4%) and 0.62 μg m^{-3} (1.2%) of reduction in population-weighted $PM_{2.5}$, respectively.

Pure NH_3 engines have high NH_3 emission, leading to higher $PM_{2.5}$ levels than NH_3-H_2 engines under the same policy scenarios. Under $[\text{Pure } NH_3]_{2020}$, $PM_{2.5}$ increases globally expect over the North Sea. Reduction in NO_x emissions lead to lower population-weighted $PM_{2.5}$ over Netherlands (1.86 μg m^{-3}, 9.0%), Denmark (0.50 μg m^{-3}, 3.2%), and Belgium (0.35 μg m^{-3}, 2.0%). The largest increases in population-weighted $PM_{2.5}$ are simulated over East Asia (11.4 μg m^{-3}, 21.2%), North Africa (3.40 μg m^{-3}, 5.5%), Mediterranean Basin (3.36 μg m^{-3}, 14.6%), Southeast Asia (2.7 μg m^{-3}, 14.2%), western North America (1.20 μg m^{-3}, 24.8%) and eastern North America (1.88 μg m^{-3}, 21.7%). Under $[\text{Pure } NH_3]_{NH3\_ECA\_LIM}$, the increase of $PM_{2.5}$ over northern Europe (0.058 μg m^{-3}, 0.34% vs 0.74 μg m^{-3}, 4.3% under $[\text{Pure } NH_3]_{2020}$), eastern North America (0.35 μg m^{-3}, 7.2%) and western North America (0.55 μg m^{-3}, 6.3%) are partially mitigated by the NH_3 emission control. When NH_3 emission control is required globally ($[\text{Pure } NH_3]_{GLOB\_LIM}$), the spatial pattern of $PM_{2.5}$ changes largely resembles that from $[NH_3\text{-}H_2]_{2020}$ due to comparable combined NO_x+NH_3 emissions (4.7 Tg/yr for $[\text{Pure } NH_3]_{GLOB\_LIM}$ vs 6.9 Tg/yr for

[NH_3-H_2]$_{2020}$). Despite having lower combined NO_x+NH_3 emissions, [Pure NH_3]$_{GLOB\_LIM}$ has higher $PM_{2.5}$ levels than [NH_3-H_2]$_{2020}$ due to higher NH_3 emissions (3.9 Tg/yr for [Pure NH_3]$_{GLOB\_LIM}$ vs 2.5 Tg/yr for [NH_3-H_2]$_{2020}$) globally except over northern Europe.

In addition, we find that NH_3 could potentially form $PM_{2.5}$ with anions and acids in sea spray, which implies extra sensitivity of $PM_{2.5}$ to NH_3 emissions that could not be controlled by reducing NO_x and SO_x emissions alone (see Supplemental Information).

Health Impacts

Scenario	$\Delta M_{PM2.5}$	ΔM_{O3}
[NH_3-H_2]$_{2020}$	-16,900 (-24,000; -10,000)	-16,200 (-23,300; -9,000)
[NH_3-H_2]$_{NH3\_ECA\_LIM}$	-22,100 (-29,800; -8,700)	-15,900 (-23,000; -8,700)
[NH_3-H_2]$_{GLOB\_LIM}$	-66,500 (-78,800; -54,400)	-12,600 (-19,900; -5,200)
[Pure NH_3]$_{2020}$	+668,100 (+542,600; +797,300)	-73,100 (-94,600; -51,100)
[Pure NH_3]$_{NH3\_ECA\_LIM}$	+623,900 (+504,000; +747,300)	-69,700 (-90,300; -48,700)
[Pure NH_3]$_{GLOB\_LIM}$	+1,200 (-10,200; +12,700)	-22,400 (-31,600; -13,000)
Post-2020 NO_x Baseline	-46,200 (-54,800; -37,700)	-13,000 (-21,100; -4,800)

Table 3. Estimated changes in annual global mortality attributable to $PM_{2.5}$ ($\Delta M_{PM2.5}$) and O_3 (ΔM_{O3}) from each scenario. Parentheses indicates 95% confidence interval (CI) of the estimates from 1000 Monte-Carlo simulations.

Table 3 shows the changes in annual global mortality attributable to O_3 (ΔM_{O3}) and $PM_{2.5}$ ($\Delta M_{PM2.5}$) for each scenario. We estimate that current shipping emissions leads to 87,400 and 16,900 mortalities from $PM_{2.5}$ and O_3, respectively. The lower NO_x emissions from ammonia-powered ships provide significant O_3 air quality benefit, reducing annual O_3-related mortality by 12,600 to 73,100. Despite the lack of primary PM (BC, OC) and secondary PM precursors (SO_2, NMVOC) emissions other than NO_x and NH_3, ammonia-powered ships lead to worse $\Delta M_{PM2.5}$ (-22,100 to +668,100) than fossil fuel powered ships with similar NO_x regulation ("Post-2020 NO_x Baseline", -46,200) except the scenario with lowest NH_3 emissions ([NH_3-H_2]$_{GLOB\_LIM}$), -66,500). This highlights the importance of NH_3 as a $PM_{2.5}$ precursor in coastal environment, and therefore minimizing tailpipe NH_3 emission to mitigate the negative air quality impacts from ammonia-powered ships.

Under currently legislation ("2020"), switching to NH_3–H_2 engine reduces annual global mortalities from $PM_{2.5}$ (16,900) and O_3 (16,200) in comparable magnitudes. While providing substantial benefits from reducing O_3–related mortality (-73,100), switching to pure NH_3 engines increases in $PM_{2.5}$–related mortality (+668,100). Since current ECA are mostly over North America and northern Europe, additional NH_3 emissions control over current ECA ("NH3_ECA_LIM") only provides marginal benefits in terms of $PM_{2.5}$–related mortalities (5,200 (31%) for NH_3–H_2 engines and 44,200 (7%) for pure NH_3 engines) since most of the increases in $PM_{2.5}$ occur overs East Asia, North Africa, Southeast Asia and Mediterranean region. In contrast, when both Tier III NO_x and NH_3 emission controls are extended globally ("GLOB_LIM"), the negative impacts of pure NH_3 drivetrains on $PM_{2.5}$ (1,200 additional mortalities) can be mitigated to a level that could be offset by the benefits on O_3 (22,400 avoided mortalities). For NH_3–H_2 engines, the low NH_3 emissions, and therefore global reduction in $PM_{2.5}$ level, lead to substantial reduction in $PM_{2.5}$-related mortalities (-66,500) equivalent to 79% of that from current shipping emissions.

Discussion

Using blue and green NH_3 to facilitate decarbonization of maritime transport has been gaining traction among the industry, while concerns have been raised about the consequences (e.g. secondary N_2O emissions, air pollution, eutrophication, soil acidification) of such large additional reactive nitrogen production and emission into the Earth System (Baessler *et al* 2019, Wolfram *et al* 2022). Despite the uncertainties in the drivetrain design, fuel mix, emission factors and plume chemistry of ammonia-powered ships as they are not yet deployed in real world, an early evaluation using currently available information can provide information to help stakeholders identify the potential climate and air quality issues and formulate mitigation measures.

We combine results from engine experiments and ship activity data to estimate the possible GHG and air pollutant emissions and impacts from ammonia-powered ships. We find that the GWP attributable to tailpipe N_2O emissions from ammonia-powered fleet is a small fraction (5.8%) of that of the current fleet. Our findings confirm the potential of blue and green NH_3 as a climate-friendly shipping fuel. However, the impacts of large reactive nitrogen deposition over land ecosystems on GHG balance remain highly uncertain.

We find that the public health impacts of switching from fossil fuel to ammonia depends largely on the technology and policy choices. If tuned to balance NO_x and NH_3 concentration from engine exhaust to allow simultaneous reduction of NO_x and NH_3 emissions using well-optimized exhaust post-treatment systems with highly efficient combustion modes, deployment of ammonia combustion technology can lead to net health benefits by reducing both O_3 and $PM_{2.5}$ levels. If the engines are tuned to have lower NO_x emissions than NH_3–H_2 combustion, which is more compatible with current NO_x–focused regulatory framework, the unburnt NH_3 emission, if unmitigated, can lead to large increases in $PM_{2.5}$, and consequently 668,100 additional global $PM_{2.5}$–related mortalities annually. Imposing NH_3 emission regulation over current ECA only mitigates 7% of the increases in annual $PM_{2.5}$-related mortalities from pure NH_3 engines, since the largest negative impacts are expected over East Asia, which is not

DO NOT CITE, COPY OR DISTRIBUTE

currently part of any ECAs. Extending stringent control of NO_x and NH_3 emissions to the globe provides substantial air quality benefits.

Our study assumes total conversion to ammonia-powered ships, while in reality ammonia-powered ships will operate alongside SO_x-emitting fossil fuel powered ships, which would increase the sensitivity of $PM_{2.5}$ to NH_3 emissions. This shows the urgency of updating shipping emission regulations in anticipation of the real–world deployment of ammonia-powered-ships. Particularly, given the availability of effective (> 95%) NH_3 removal strategies, priority should be given towards developing and enforcing working NH_3 emission regulations. More stringent control of SO_x and NO_x emissions, which is foreseeable in the future, could be another viable strategy to reduce the $PM_{2.5}$ formation from unburnt NH_3 emissions (Bauer *et al* 2016).

The practicality and efficacy of SCR for ammonia engines remain highly uncertain. The lack of sulfur and particulate poisoning of catalyst, and not requiring a separate NH_3 source to operate could potentially lead to cheaper SCR operation since catalyst and urea recharge are estimated to account for at least 61% of the total cost of SCR ownership and operation (Zhang *et al* 2021a). However, NH_3 combustion generates more H_2O than diesel combustions (see Supplemental Information), which limits the efficacy of SCR (Kuta *et al* 2023, Xiang *et al* 2024). Excessive tailpipe N_2O emissions can result from mistuned SCR and ammonia oxidation systems (Yates *et al* 2005), which could potentially offset the climate benefits. Optimizing the SCR systems for ammonia engines is crucial to limiting their potential air quality and climate impacts.

Our study shows the feasibility of NH_3 to be a climate-friendly shipping fuel despite the concern of tailpipe N_2O emission, and highlights the adverse effects of unburnt NH_3 emissions on $PM_{2.5}$ air quality, which can be mitigated by emission control measures feasible under current technology. Apart from tailpipe emissions, NH_3 leakages also occur over the whole value chain (e.g. production, distribution, bunkering, fueling) (Bertagni *et al* 2023), which can deteriorate the $PM_{2.5}$ air quality over localities near the NH_3 supply chain if unabated (Rathod *et al* 2023). Development and enforcement of new NH_3 emission regulations is critical for ammonia-powered ships to provide positive impact on air quality and prevent negative impacts from excessive nitrogen deposition, alongside reducing GHG emissions.

Data Availability

EF, shipping emission maps, GCHP modelled annual mean MDA8 O_3 and $PM_{2.5}$, and the scripts for data analysis are available on Zenodo (https://zenodo.org/records/11237986)

Acknowledgements

This research is supported by a grant from the MIT Climate and Sustainability Consortium. The MERRA-2 data used in this research have been provided by the Global Modeling and

Assimilation Office (GMAO) at NASA Goddard Space Flight Center. The AIS data used in this study are managed by the Institute for the Environment and Environmental Central Facility of the Hong Jong University of Science and Technology. We thank Professor Andrew Babbin for his input on the fraction of reactive nitrogen deposition being converted into N_2O over the ocean.

References

Baessler C, Bonn A, Klotz S, Schröter M and Seppelt R 2019 *Atlas of Ecosystem Services: Drivers, Risks, and Societal Responses* (Cham: Springer International Publishing : Imprint: Springer)

Balcombe P, Brierley J, Lewis C, Skatvedt L, Speirs J, Hawkes A and Staffell I 2019 How to decarbonise international shipping: Options for fuels, technologies and policies *Energy Conversion and Management* **182**

Bauer S E, Tsigaridis K and Miller R 2016 Significant atmospheric aerosol pollution caused by world food cultivation *Geophysical Research Letters* **43** 5394–400

Bertagni M B, Socolow R H, Martirez J M P, Carter E A, Greig C, Ju Y, Lieuwen T, Mueller M E, Sundaresan S, Wang R, Zondlo M A and Porporato A 2023 Minimizing the impacts of the ammonia economy on the nitrogen cycle and climate *Proc. Natl. Acad. Sci. U.S.A.* **120** e2311728120

Boero A, Mercier A, Mounaïm-Rousselle C, Valera-Medina A and Ramirez A D 2023 Environmental assessment of road transport fueled by ammonia from a life cycle perspective *Journal of Cleaner Production* **390**

Burnett R, Chen H, Szyszkowicz M, Fann N, Hubbell B, Pope C A, Apte J S, Brauer M, Cohen A, Weichenthal S, Coggins J, Di Q, Brunekreef B, Frostad J, Lim S S, Kan H, Walker K D, Thurston G D, Hayes R B, Lim C C, Turner M C, Jerrett M, Krewski D, Gapstur S M, Diver W R, Ostro B, Goldberg D, Crouse D L, Martin R V, Peters P, Pinault L, Tjepkema M, Van Donkelaar A, Villeneuve P J, Miller A B, Yin P, Zhou M, Wang L, Janssen N A H, Marra M, Atkinson R W, Tsang H, Thach T Q, Cannon J B, Allen R T, Hart J E, Laden F, Cesaroni G, Forastiere F, Weinmayr G, Jaensch A, Nagel G, Concin H and Spadaro J V 2018 Global estimates of mortality associated with longterm exposure to outdoor fine particulate matter *Proceedings of the National Academy of Sciences of the United States of America* **115** 9592–7

Center for International Earth Science Information Network - CIESIN - Columbia University 2018 Gridded {Population} of the {World}, {Version} 4 ({GPWv}4): {National} {Identifier} {Grid}, {Revision} 11 Online: https://doi.org/10.7927/H4TD9VDP

Cohen A J, Brauer M, Burnett R, Anderson H R, Frostad J, Estep K, Balakrishnan K, Brunekreef B, Dandona L, Dandona R, Feigin V, Freedman G, Hubbell B, Jobling A, Kan H, Knibbs L, Liu Y, Martin R, Morawska L, Pope C A, Shin H, Straif K, Shaddick G, Thomas M, van Dingenen R, van Donkelaar A, Vos T, Murray C J L and Forouzanfar M H 2017

Estimates and 25-year trends of the global burden of disease attributable to ambient air pollution: an analysis of data from the Global Burden of Diseases Study 2015 *The Lancet* **389** 1907–18

Contini D and Merico E 2021 Recent Advances in Studying Air Quality and Health Effects of Shipping Emissions *Atmosphere* **12** 92

Eastham S D, Long M S, Keller C A, Lundgren E, Yantosca R M, Zhuang J, Li C, Lee C J, Yannetti M, Auer B M, Clune T L, Kouatchou J, Putman W M, Thompson M A, Trayanov A L, Molod A M, Martin R V and Jacob D J 2018 GEOS-Chem high performance (GCHP v11-02c): A next-generation implementation of the GEOS-Chem chemical transport model for massively parallel applications *Geoscientific Model Development* **11**

Fountoukis C and Nenes A 2007 ISORROPIAII: A computationally efficient thermodynamic equilibrium model for K+-Ca2+-Mg2+-NH4+-Na+-SO42--NO3--Cl--H2O aerosols *Atmospheric Chemistry and Physics* **7** 4639–59

Gelaro R, McCarty W, Suárez M J, Todling R, Molod A, Takacs L, Randles C A, Darmenov A, Bosilovich M G, Reichle R, Wargan K, Coy L, Cullather R, Draper C, Akella S, Buchard V, Conaty A, da Silva A M, Gu W, Kim G K, Koster R, Lucchesi R, Merkova D, Nielsen J E, Partyka G, Pawson S, Putman W, Rienecker M, Schubert S D, Sienkiewicz M and Zhao B 2017 The modern-era retrospective analysis for research and applications, version 2 (MERRA-2) *Journal of Climate* **30** 5419–54

Giorgi F, Hewitson B, Christensen J, Hulme M, Von Storch H, Whetton P, Jones R, Mearns L and Fu C 2001 Regional Climate Information Evaluation and Projections *Climate Change 2001: The Scientific Basis. Contribution of Working Group I to the Third Assessment Report of the Intergovernmental Panel on Climate Change*

Gu B, Zhang L, Dingenen R V, Vieno M, Grinsven H J V, Zhang X, Zhang S, Chen Y, Wang S, Ren C, Rao S, Holland M, Winiwarter W, Chen D, Xu J and Sutton M A 2021 Abating ammonia is more cost-effective than nitrogen oxides for mitigating PM2.5 air pollution *Science* **374**

Heo J, Adams P J and Gao H O 2016 Public Health Costs of Primary PM2.5 and Inorganic PM2.5 Precursor Emissions in the United States *Environmental Science and Technology* **50**

Hinokuma S and Sato K 2021 Ammonia combustion catalysts *Chemistry Letters* **50**

Hoesly R M, Smith S J, Feng L, Klimont Z, Janssens-Maenhout G, Pitkanen T, Seibert J J, Vu L, Andres R J, Bolt R M, Bond T C, Dawidowski L, Kholod N, Kurokawa J I, Li M, Liu L, Lu Z, Moura M C P, O'Rourke P R and Zhang Q 2018 Historical (1750-2014) anthropogenic emissions of reactive gases and aerosols from the Community Emissions Data System (CEDS) *Geoscientific Model Development*

Imhoff T B, Gkantonas S and Mastorakos E 2021 Analysing the performance of ammonia powertrains in the marine environment *Energies* **14**

International Maritime Organization 2018 *INITIAL IMO STRATEGY ON REDUCTION OF GHG EMISSIONS FROM SHIPS Contents*

International Maritime Organization 2017 Nitrogen Oxides (NOx) – Regulation 13 Online: https://www.imo.org/en/OurWork/Environment/Pages/Nitrogen-oxides-(NOx)-–-Regulation-13.aspx

International Maritime Organization 2008 *Technical Code on Control of Emission of Nitrogen Oxides from Marine Diesel Engines*

IRENA 2021 *A pathway to decarbonise the shipping sector by 2050* (International Renewable Energy Agency)

Jacob D J 1999 *Introduction to Atmospheric Chemistry* (Princeton University Press)

Kuta K, Przybyła G, Kurzydym D and Żmudka Z 2023 Experimental and numerical investigation of dual-fuel CI ammonia engine emissions and after-treatment with V2O5/SiO2–TiO2 SCR *Fuel* **334** 126523

Latimer R N C and Martin R V 2019 Interpretation of measured aerosol mass scattering efficiency over North America using a chemical transport model *Atmospheric Chemistry and Physics* **19**

Lhuillier C, Brequigny P, Contino F and Mounaïm-Rousselle C 2020 Experimental study on ammonia/hydrogen/air combustion in spark ignition engine conditions *Fuel* **269**

Malley C S, Henze D K, Kuylenstierna J C I, Vallack H W, Davila Y, Anenberg S C, Turner M C and Ashmore M R 2017 Updated Global Estimates of Respiratory Mortality in Adults ≥30Years of Age Attributable to Long-Term Ozone Exposure *Environ Health Perspect* **125** 087021

Martin R V, Eastham S D, Bindle L, Lundgren E W, Clune T L, Keller C A, Downs W, Zhang D, Lucchesi R A, Sulprizio M P, Yantosca R M, Li Y, Estrada L, Putman W M, Auer B M, Trayanov A L, Pawson S and Jacob D J 2022 Improved advection, resolution, performance, and community access in the new generation (version 13) of the high-performance GEOS-Chem global atmospheric chemistry model (GCHP) *Geoscientific Model Development* **15**

Melse R W and Ogink N W M 2005 Air scrubbing techniques for ammonia and odor reduction at livestock operations: Review of on-farm research in the Netherlands *Transactions of the American Society of Agricultural Engineers* **48**

Meng J, Martin R V, Ginoux P, Hammer M, Sulprizio M P, Ridley D A and Van Donkelaar A 2021 Grid-independent high-resolution dust emissions (v1.0) for chemical transport models: Application to GEOS-Chem (12.5.0) *Geoscientific Model Development* **14**

Mounaïm-Rousselle C, Mercier A, Brequigny P, Dumand C, Bouriot J and Houillé S 2022 Performance of ammonia fuel in a spark assisted compression Ignition engine *International Journal of Engine Research* **23**

Nuvolone D, Petri D and Voller F 2018 The effects of ozone on human health *Environmental Science and Pollution Research* **25**

Olmer N, Comer B, Roy B, Mao X and Rutherford D 2017 *Greenhouse gas emissions from global shipping, 2013–2015 Detailed Methodology* (Washington, DC, USA: International Council on Clean Transportation)

Payne R J, Dise N B, Field C D, Dore A J, Caporn S J M and Stevens C J 2017 Nitrogen deposition and plant biodiversity: past, present, and future *Frontiers in Ecology and the Environment* **15** 431–6

Rathod S, Edwards M R, Roy C, Warnecke L, Rafaj P, Kiesewetter G and Klimont Z 2023 Air quality and health effects of a transition to ammonia–fueled shipping in Singapore *Environ. Res.: Health* **1** 041002

Sherwen T, Schmidt J A, Evans M J, Carpenter L J, Großmann K, Eastham S D, Jacob D J, Dix B, Koenig T K, Sinreich R, Ortega I, Volkamer R, Saiz-Lopez A, Prados-Roman C, Mahajan A S and Ordóñez C 2016 Global impacts of tropospheric halogens (Cl, Br, I) on oxidants and composition in GEOS-Chem *Atmospheric Chemistry and Physics* **16**

Smith C, Nicholls Z R J, Armour K, Collins W, Forster P, Meinshausen M, Palmer M D and Watanabe M 2021 The earth's energy budget, climate feedbacks, and climate sensitivity supplementary material *Climate change* 1850–2005

Turner M C, Jerrett M, Pope C A, Krewski D, Gapstur S M, Diver W R, Beckerman B S, Marshall J D, Su J, Crouse D L and Burnett R T 2016 Long-Term Ozone Exposure and Mortality in a Large Prospective Study *American journal of respiratory and critical care medicine* **193**

Valera-Medina A, Amer-Hatem F, Azad A K, Dedoussi I C, De Joannon M, Fernandes R X, Glarborg P, Hashemi H, He X, Mashruk S, McGowan J, Mounaim-Rouselle C, Ortiz-Prado A, Ortiz-Valera A, Rossetti I, Shu B, Yehia M, Xiao H and Costa M 2021 Review on Ammonia as a Potential Fuel: From Synthesis to Economics *Energy Fuels* **35** 6964–7029

Van der Heyden C, Demeyer P and Volcke E I P 2015 Mitigating emissions from pig and poultry housingfacilities through air scrubbers and biofilters: State-of-the-art andperspectives *Biosystems Engineering* **134**

Vinken G C M, Boersma K F, Jacob D J and Meijer E W 2011 Accounting for non-linear chemistry of ship plumes in the GEOS-Chem global chemistry transport model *Atmospheric Chemistry and Physics* **11**

Weng H, Lin J, Martin R, Millet D B, Jaeglé L, Ridley D, Keller C, Li C, Du M and Meng J 2020 Global high-resolution emissions of soil NOx, sea salt aerosols, and biogenic volatile organic compounds *Scientific Data* **7**

WHO 2018 WHO methods and data sources for country-level causes of death 2000-2016 Online: https://terrance.who.int/mediacentre/data/ghe/healthinfo/Deaths/GHE2016_COD_methods.pdf

Wolfram P, Kyle P, Zhang X, Gkantonas S and Smith S 2022 Using ammonia as a shipping fuel could disturb the nitrogen cycle *Nature Energy* **7** 1112–4

Xiang P, Liu J, Zhao W, Ji Q, Ao C, Wang X, Sun P, Wang X and Li Z 2024 Experimental investigation on gas emission characteristics of ammonia/diesel dual-fuel engine equipped with DOC + SCR aftertreatment *Fuel* **359** 130496

Yates M, Martín J A, Martín-Luengo M Á, Suárez S and Blanco J 2005 N2O formation in the ammonia oxidation and in the SCR process with V2O5-WO3 catalysts *Catalysis Today* vol 107–108

Zhang G, Yan H, Li T, Zhu Y, Zhou S, Feng Y and Zhou W 2021a Relation analysis on emission control and economic cost of SCR system for marine diesels *Science of the Total Environment* **788**

Zhang Y, Eastham S D, Lau A K H, Fung J C H and Selin N E 2021b Global air quality and health impacts of domestic and international shipping *Environmental Research Letters* **16** 084055

Zhang Y, Fung J C H, Chan J W M and Lau A K H 2019 The significance of incorporating unidentified vessels into AIS-based ship emission inventory *Atmospheric Environment* **203**

DO NOT CITE, COPY OR DISTRIBUTE