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

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Feb 2024

This manuscript has been submitted for review in Environmental Research Letters. If accepted, the final version of this manuscript will be available via the ‘Peer-reviewed Publication DOI’ link on the right-hand side of the EarthArxiv web page for the paper

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, emissions of NOₓ, NH₃ and N₂O resulting from ammonia combustion could cause impacts on 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 NOₓ, NH₃ and N₂O emissions from ammonia-powered ships with two possible engine technologies (NH₃–H₂ vs pure NH₃ combustion) under three emission regulation scenarios (with corresponding assumptions in emission control technologies). We then use the GEOS-Chem High Performance global chemical transport model to simulate the air quality impacts of switching to ammonia-powered ships. We find that the tailpipe N₂O emissions from ammonia-powered ships have climate impacts equivalent to 5.8% of current shipping CO₂ emissions. Globally, switching to NH₃–H₂ engines avoids 33,100 (18900 to 47300, 95% confidence interval) mortalities annually, while the unburnt NH₃ emissions (82.0 Tg NH₃ yr⁻¹) from pure NH₃ engines could lead
to 595,100 additional mortalities annually under current legislation. Requiring NH₃ scrubbing within current Emission Control Areas leads to smaller improvements in public health outcomes (38,000 avoided mortalities for NH₃–H₂ and 554,200 additional mortalities for pure NH₃ annually, respectively), while extending both Tier III NOₓ standard and NH₃ scrubbing requirements globally leads to larger improvement in public health outcomes associated with a switch to ammonia-powered ships (79,100 and 21,100 avoided mortalities for NH₃–H₂ and pure NH₃ annually, respectively). 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.

**Keywords**: Ammonia, Shipping, Decarbonization, Air Quality

### 1. Introduction

Maritime shipping burns heavy fuel oil in large diesel engines for energy (propulsion, heat, and electricity), which leads to emissions of CO₂ and air pollutants. The main air pollutants emitted by the maritime transport sector include SOₓ (= SO₂+SO₄²⁻), NOₓ (= NO+NO₂), non-methane volatile organic compound (NMVOC), CO and carbonaceous aerosols. These are either components or precursors of particulate matter (PM) and ozone (O₃). Exposure to PM, particularly the fine PM (aerodynamic diameter < 2.5 μm, named PM₂.₅) 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 [6]. O₃ exposure exerts oxidative stress on the respiratory tract [27], which also leads to increased risk of cardiopulmonary diseases, and therefore another 1.04 – 1.24 millions of respiratory deaths in 2010 globally [23]. Shipping emissions are estimated to account for 2.7% of global energy-related CO₂ emissions and caused an estimated 84800 – 103000 annual premature deaths from PM₂.₅ exposure globally in 2015 [41].

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 [15]. The uses of alternative fuels (e.g. NH₃, H₂, methanol) and other energy solutions (e.g. electrification) are essential for reaching such a decarbonization goal [2]. NH₃ is one of the main candidates for alternative maritime fuels, and could represent up to 43% of the energy mix of shipping in 2050 [18]. Since NH₃ in mainly manufactured with H₂ and N₂ through Haber-Bosch Process, the carbon footprint of NH₃ production can be reduced by carbon capture (blue NH₃), or using renewable energy for N₂ and H₂ production and the synthesis process (green NH₃) [32].

Wolfram et al (2022) [37] summarized scientific concerns about the potential environmental impacts of using NH₃ as a marine fuel. NH₃ combustion may generate additional NOₓ and N₂O compared to other fuels [12]. NH₃ emission is one of the major source of global PM₂.₅ pollution [10] by neutralizing H₂SO₄ and HNO₃ in the
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atmosphere [19]. NH\textsubscript{3} emission leads to much higher PM\textsubscript{2.5} mortality costs per ton ($23000 – 66000) than SO\textsubscript{2} ($14000 – 24000) and NO\textsubscript{x} ($3800 – 14000) in the United States [11]. These show the potential danger of uncontrolled NH\textsubscript{3} emission via worsening PM\textsubscript{2.5} air quality. Emitted NO\textsubscript{x} and NH\textsubscript{3} would then deposit to Earth’s surface, causing damages to ecosystems (e.g. soil acidification and eutrophication) [28] and may lead to additional emission of N\textsubscript{2}O [37], 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.

2. Method

We use a bottom-up approach to estimate the global NO\textsubscript{x}, NH\textsubscript{3}, and N\textsubscript{2}O emissions from NH\textsubscript{3}-powered ships as a function of engine technologies, emission control strategies and policy under 6 scenarios, using result from ammonia engine experiments and ship Automatic Identification System (AIS) data. We then simulate the associated changes in O\textsubscript{3} and PM\textsubscript{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\textsubscript{3} and PM\textsubscript{2.5} on public health (expressed in annual premature mortalities) using concentration functions derived from epidemiological studies.

2.1. Scenarios

In all scenarios, we apply an AIS-based shipping emission model [42] 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. We choose the emission scenario with 0.5% cap on fuel sulphur content from Zhang et al (2021) [41] as our baseline. The “post-2020 NO\textsubscript{x} baseline” scenario imposes the most stringent IMO NO\textsubscript{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.

We consider the emissions from ammonia-powered ships with two types of engine technologies. The first type (“NH\textsubscript{3}-H\textsubscript{2}”) is proposed by Imhoff et al (2021) [14] based on the experimental data from Lhuillier et al (2020) [22]. Part of the NH\textsubscript{3} is transferred to a catalytic NH\textsubscript{3} cracker to generate H\textsubscript{2} as the pilot fuel, which improves the stability of NH\textsubscript{3} ignition and combustion. This leads to less unburnt NH\textsubscript{3}, but more NO\textsubscript{x} emissions compared to pure NH\textsubscript{3} combustion. By balancing the NH\textsubscript{3} and NO\textsubscript{x}
concentration in engine exhaust, both NO\textsubscript{x} and NH\textsubscript{3} emissions can be controlled by Selective Catalytic Reduction (SCR). The second type of engine technology considered is pure NH\textsubscript{3} combustion [26], which provides certain advantages over the NH\textsubscript{3}–H\textsubscript{2} technology described above (e.g. simpler design and lower NO\textsubscript{x} emissions (fig. 1)). The derivations of EF and load dependencies for the two types of engines, and a discussion about the uncertainty in engine technologies are given as Supplemental Information.

We acknowledge the uncertainty in ammonia engine designs. Our 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\textsubscript{x} and NH\textsubscript{3} emission management approaches: 1) with pure NH\textsubscript{3} engine having low NO\textsubscript{x} (currently regulated) and very high NH\textsubscript{3} (currently unregulated) emissions, versus 2) NH\textsubscript{3}–H\textsubscript{2} engine that strictly maintains the NO\textsubscript{x}/NH\textsubscript{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\textsubscript{x} EF are 32.7 g/kWh for NH\textsubscript{3}–H\textsubscript{2} and 7.08 g/kWh for pure NH\textsubscript{3} engines following the load corrections prescribed by IMO [17] (fig. 1). Current IMO guidelines [16] cap NO\textsubscript{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...
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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.

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) [29, 33, 3] 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.

2.2. Atmospheric Chemistry Modeling

We use version 13.4.1 of the GEOS-Chem High Performance model (GCHP, https://doi.org/10.5281/zenodo.4429193) [24, 7] 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 [9]. 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. O₃ is simulated from a coupled
O₃-NOₓ-VOCs-CO-halogen-aerosols chemical mechanism [30]. Anthropogenic emissions
are from Community Emission Data System [13] except the shipping sector. Biogenic
VOCs, soil NOₓ and sea salt aerosol emissions follow Weng et al (2020) [35] and dust
emissions follow Meng et al (2021) [25]. Formation of secondary inorganic aerosols are
simulated by the ISORROPIA II [8], which considers thermodynamic equilibrium of the
NH₄⁺-Na⁺-SO₄²⁻-NO₃⁻-Cl⁻-H₂O. PM₂.₅ 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 [21]. Ship plume chemistry
is parameterized by the PARANOX scheme [34], and the uncertainties in ammonia-
powered ship plume is explored with additional simulations presented in Supplemental
Information.

2.3. Health Outcome

We estimate the impacts of air quality changes on public health using the global gridded
population density data at 30 arc-second resolution from the Gridded Population of the
World version 4.11 [5]. O₃ and PM₂.₅ concentrations are taken as the area-weighted
averages from simulation grid cells overlapping with the 30 arc-second cell without any
interpolation. Country-level age distribution and baseline mortality rates are provided
by the World Health Organization (WHO) [36]. We estimate the risk of relative
mortality from chronic O₃ and PM₂.₅ 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 (ΔMort_{i}) due to some disease for that age group is then calculated for each
grid cell as:

\[ ΔMort_{i} = Mort_{base} \frac{RR_{i} - RR_{base}}{RR_{base}} \]  

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 O₃ that 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) O₃ concentration [31]. For PM₂.₅
we estimate RR for non-communicable diseases and lower respiratory infections using
the age-specific non-linear CRFs from the Global Exposure Mortality Model [4].

We estimate the median and 95% confidence interval of changes in mortalities due
to O₃ and PM₂.₅ for each scenario by performing 1,000 random draws of the CRF
parameters in a paired Monte-Carlo simulation.
Table 2. Modelled global total nitrogen-based air pollutants (in Tg yr\(^{-1}\)) and GHG emissions (in Tg CO\(_2\)\(_{eq}\)/yr) from different scenarios. CO\(_2\)\(_{eq}\) (equivalent amount of CO\(_2\) in terms of 100-year Global Warming Potential) is calculated as CO\(_2\) emissions + (N\(_2\)O emissions \times 273).

<table>
<thead>
<tr>
<th>Scenario</th>
<th>NO(_x) (Tg/yr)</th>
<th>NH(_3) (Tg/yr)</th>
<th>CO(<em>2)(</em>{eq}) (Tg/yr)</th>
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<tr>
<td>Baseline</td>
<td>17.2</td>
<td>0.004</td>
<td>867</td>
</tr>
<tr>
<td>Post-2020 NO(_x) baseline</td>
<td>3.59</td>
<td>0.004</td>
<td>867</td>
</tr>
<tr>
<td>[NH(_3)-H(<em>2)](</em>{2020})</td>
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<td>2.51</td>
<td>50.2</td>
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<td>2.21</td>
<td>50.2</td>
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<tr>
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<td>0.125</td>
<td>50.2</td>
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<tr>
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<td>6.84</td>
<td>82.0</td>
<td>50.2</td>
</tr>
<tr>
<td>[Pure NH(<em>3)](</em>{NH3, ECA, LIM})</td>
<td>6.84</td>
<td>71.7</td>
<td>50.2</td>
</tr>
<tr>
<td>[Pure NH(<em>3)](</em>{GLOB, LIM})</td>
<td>0.762</td>
<td>3.92</td>
<td>50.2</td>
</tr>
</tbody>
</table>

Figure 2. Spatial pattern of annual total NO\(_x\) emissions (kg m\(^{-2}\) yr\(^{-1}\)) under different scenarios.

3. Result

3.1. Modelled Shipping Emissions

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\textsubscript{x} emissions than NH\textsubscript{3}-H\textsubscript{2} engines. If the Tier III NO\textsubscript{x} regulations is enforced globally (“GLOB\textsubscript{LIM}”), the NO\textsubscript{x} emission of fossil fuel (3.6 Tg NO\textsubscript{x}/yr) and NH\textsubscript{3}-H\textsubscript{2} (4.4 Tg NO\textsubscript{x}/yr) engines are similar, while pure NH\textsubscript{3} engines (0.8 Tg NO\textsubscript{x}/yr) produce the lowest NO\textsubscript{x} emissions.

Figure 3 shows the spatial distribution of modelled NH\textsubscript{3} emissions under different technology and policy scenarios. Under current regulations (“2020”), switching to NH\textsubscript{3}-H\textsubscript{2} engines leads to 2.5 Tg/yr NH\textsubscript{3} emissions, while switching to pure NH\textsubscript{3} engines leads to NH\textsubscript{3} emissions (82.0 Tg/yr) that are 32.8 times higher than that from NH\textsubscript{3}-H\textsubscript{2} engines. For pure NH\textsubscript{3} engines, SCR can only remove 7% of NH\textsubscript{3} from engine exhaust, leading to high tailpipe NH\textsubscript{3} emissions. In the “NH\textsubscript{3}-ECA\textsubscript{LIM}” scenario, which requires NH\textsubscript{3} scrubbing over ECA (mostly North American coast and northern Europe), global NH\textsubscript{3} emissions reduce by 12% for both NH\textsubscript{3}-H\textsubscript{2} (2.2 Tg/yr) and pure NH\textsubscript{3} (71.7 Tg/yr ) engines. In the “GLOB\textsubscript{LIM}” scenario, with both SCR and NH\textsubscript{3} scrubbing are required globally, NH\textsubscript{3} emissions fall to 0.1 Tg/yr for NH\textsubscript{3}-H\textsubscript{2} engines and 3.9 Tg/yr for pure NH\textsubscript{3} engines.

Table 2 also shows the long-lived GHG emissions from each scenario, given as the equivalent amount of CO\textsubscript{2} (CO\textsubscript{2,e}) in terms of 100-year Global Warming Potential (GWP100) using a conversion factor of 273 from N\textsubscript{2}O emission to CO\textsubscript{2,e} (Smith et al 2021). CO\textsubscript{2,e} from the baseline scenario does not include GHG other than CO\textsubscript{2} (mainly CH\textsubscript{4} and N\textsubscript{2}O), which contribute to less than 3% of global shipping CO\textsubscript{2,e} during 2013 – 2015 (Olmer et al 2017). We find that the tailpipe CO\textsubscript{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\textsubscript{2}O emissions” from reactive nitrogen deposition (Wolfram et al 2022) is not a problem for NH\textsubscript{3}-H\textsubscript{2} engine as the total reactive nitrogen emissions are lower than current fleets. For pure NH\textsubscript{3} engine, the net climate effects from nitrogen deposition are likely to be smaller than reduction in tailpipe GHG emissions (817.2 Tg CO\textsubscript{2,e}/yr) from switching to ammonia-powered ships, showing the potential of blue and green ammonia as a climate-friendly shipping
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Figure 4. Changes in annual mean MDA8 O₃ concentration (ΔO₃, ppb) for different ammonia-powered ship scenarios

fuel, though considerable uncertainties exist on how CO₂ uptake and N₂O emissions respond to nitrogen deposition. This analysis, however, does not fully consider the life cycle GHG emissions (e.g., energy, methane slip) of NH₃ production.

3.2. Impacts on Air Quality

Figure 4 shows the modelled global changes in annual mean MDA8 O₃ due to converting current fleet to ammonia-powered ships with different technology and policy options. Generally, the lower NOₓ emissions from ammonia-powered ships reduce annual mean MDA8 O₃. Under all scenarios, global population-weighted average MDA8 O₃ decreases (-0.27 ppb for [NH₃-H₂]₂₀₂₀, -1.13 ppb for [Pure NH₃]₂₀₂₀, -0.37 ppbv for [Pure NH₃]GLOB_LIM). The greatest reductions in population-weighted O₃ 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 NOₓ-saturated coasts near northern China, northern Europe, and Persian Gulf, local increases in surface O₃ are simulated, especially under the scenarios with greater NOₓ reductions ([NH₃-H₂]₂₀₂₀ and [Pure NH₃]GLOB_LIM). Over North Sea, the NOₓ-saturation leads to further increases in MDA8 O₃ as NOₓ emissions become lower, increasing the population-weighted O₃ from 1 ppb under [Pure NH₃]₂₀₂₀ to up to 1.5 ppb under [Pure NH₃]GLOB_LIM over the Netherlands. Over East Asia, population-weighted MDA8 O₃ decreases by 2.4 ppb under the scenario with least NOₓ reduction ([Pure NH₃]₂₀₂₀), but increases by 0.2 ppb under [Pure NH₃]GLOB_LIM and [NH₃-H₂]₂₀₂₀ as NOₓ emissions become lower. This shows the importance of local chemical environment in controlling the response of O₃ pollution to marine NOₓ control.

In addition, we find substantial sensitivity of O₃ response to assumptions in ship plume chemistry (mainly NOₓ lifetime, see Supplemental Material), which could be a major source of uncertainties. This shows the importance of understanding the plume chemistry of NH₃ ship in capturing the O₃ response.

Figure 5 shows the modelled changes in annual mean surface PM₂.₅. Under [NH₃-H₂]₂₀₂₀, population-weighted PM₂.₅ 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 PM₂.₅ (4.82 µg m⁻³) is low. PM₂.₅
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$_3$-H$_2$]$_{NH_3,ECA,LIM}$, 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$_3$-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 [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 [Pure NH$_3$,NH$_3$,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 [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 ([Pure NH$_3$]$_{GLOB,LIM}$), the spatial pattern of PM$_{2.5}$ changes largely resembles that from [NH$_3$-H$_2$]$_{2020}$ due to comparable combined NO$_x$+NH$_3$ emissions (4.7 Tg/yr for [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.

**Figure 5.** Changes in annual mean PM$_{2.5}$ concentration (∆PM$_{2.5}$, µg m$^{-3}$) for all ammonia-powered ships scenarios.
Table 3. Estimated changes in annual global mortality attributable to PM$_{2.5}$ ($\Delta M_{\text{PM}_{2.5}}$), O$_3$ ($\Delta M_{\text{O}_3}$), and their sum ($\Delta M_{\text{total}}$) from each scenario. Parentheses indicates 95% confidence interval (CI) of the estimates from 1000 Monte-Carlo simulations.

<table>
<thead>
<tr>
<th>Scenario</th>
<th>$\Delta M_{\text{PM}_{2.5}}$</th>
<th>$\Delta M_{\text{O}_3}$</th>
<th>$\Delta M_{\text{total}}$</th>
</tr>
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<tbody>
<tr>
<td>[NH$_3$–H$<em>2$]$</em>{2020}$</td>
<td>-16,900 (24,000; 10,000)</td>
<td>-16,200 (-23,300; 9,000)</td>
<td>-33,100 (-47,300; 18,900)</td>
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<td>-15,900 (-23,000; 7,000)</td>
<td>-38,000 (-52,000; 23,100)</td>
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<td>-12,600 (19,900; 5,200)</td>
<td>-79,100 (-98,700; 59,600)</td>
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<td>+668,100 (+542,600; 797,300)</td>
<td>-73,100 (-94,600; 51,100)</td>
<td>+595,100 (+448,000; 746,200)</td>
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<tr>
<td>Pure NH$<em>3$$</em>{\text{NH}_3\text{-ECA LIM}}$</td>
<td>+623,900 (+504,000; 747,300)</td>
<td>-69,700 (-90,300; 48,700)</td>
<td>+554,200 (+413,700; 698,600)</td>
</tr>
<tr>
<td>Pure NH$<em>3$$</em>{\text{GLOB LIM}}$</td>
<td>+1,200 (-10,200; 12,700)</td>
<td>-22,400 (-31,600; 13,000)</td>
<td>-21,100 (-41,800; 300)</td>
</tr>
<tr>
<td>Post-2020 NO$_x$ baseline</td>
<td>-46,200 (-54,800; 37,700)</td>
<td>-13,000 (-21,100; 4800)</td>
<td>-59,100 (-75,900; 42,500)</td>
</tr>
</tbody>
</table>

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).

3.3. Health Impacts

Table 3 shows the changes in annual global mortality attributable to O$_3$ ($\Delta M_{\text{O}_3}$) and PM$_{2.5}$ ($\Delta M_{\text{PM}_{2.5}}$) 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 $\Delta M_{\text{PM}_{2.5}}$ (-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$_2$]$_{\text{GLOB LIM}},$ -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 by 33,100, attributable to both changes in PM$_{2.5}$ (51%) and O$_3$ (49%). 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), causing a net effect of 595,100 increased mortalities. This is mostly due to the increased mortality over East Asia (+468,400; 79% of $\Delta M_{\text{total}}$). Since current ECA are mostly over North America and northern Europe, additional NH$_3$ emissions control over current ECA
(“NH<sub>3</sub>_ECA_LIM”) only provides marginal benefits in terms of mortalities (4,900 (13%) for NH<sub>3</sub>–H<sub>2</sub> engines and 40,900 (7%) for pure NH<sub>3</sub> engines) since most of the increases in PM<sub>2.5</sub> occur over East Asia, North Africa, Southeast Asia and Mediterranean region. In contrast, when both Tier III NO<sub>x</sub> and NH<sub>3</sub> emission controls are extended globally (“GLOB_LIM”), the negative impacts of pure NH<sub>3</sub> engines on PM<sub>2.5</sub> can be largely mitigated, leading a net reduction in mortalities (-21,100). For NH<sub>3</sub>–H<sub>2</sub> engines, the low NH<sub>3</sub> emissions, and therefore global reduction in PM<sub>2.5</sub> level, lead to substantial reduction in mortalities (-79,100) equivalent to 76% of mortalities attributable to current shipping emissions.

4. Discussion

Using blue and green NH<sub>3</sub> 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<sub>2</sub>O emissions, air pollution, eutrophication, soil acidification) of such large additional reactive nitrogen production and emission into the Earth System [1, 37]. Despite the uncertainties in the engine 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<sub>2</sub>O 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<sub>3</sub> 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<sub>x</sub> and NH<sub>3</sub> concentration from engine exhaust to allow simultaneous reduction of NO<sub>x</sub> and NH<sub>3</sub> 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<sub>3</sub> and PM<sub>2.5</sub> levels. If the engines are tuned to have lower NO<sub>x</sub> emissions than NH<sub>3</sub>–H<sub>2</sub> combustion, which is more compatible with current NO<sub>x</sub>–focused regulatory framework, the unburnt NH<sub>3</sub> emission, if unmitigated, can lead to large increases in PM<sub>2.5</sub>, and consequently 595,100 additional global premature mortalities annually. Imposing NH<sub>3</sub> emission regulation over current ECA only mitigates 7% of the increases in annual mortalities from pure NH<sub>3</sub> engines, since the largest negative impacts are expected over East Asia, which is not currently part of any ECAs. Extending stringent control of NO<sub>x</sub> and NH<sub>3</sub> emissions to the globe provides substantial air quality benefits. 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%) NH₃ removal strategies, priority should be given towards developing and enforcing working NH₃ emission regulations. Our additional simulations (see Supplemental Information) shows that these conclusions are not affected by the assumptions in plume chemistry, though better understanding plume chemistry of ammonia-powered ships could help better evaluate the O₃ impacts.

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 NH₃ 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 [40]. However, NH₃ combustion generates more H₂O than diesel combustions (see Supplemental Information), which limits the efficacy of SCR [20, 38]. Excessive tailpipe N₂O emissions can result from mistuned SCR and ammonia oxidation systems [39], 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 NH₃ to be a climate-friendly shipping fuel despite the concern on tailpipe N₂O emission, and highlights the adverse effects of unburnt NH₃ emissions on PM₂.₅ air quality, which can be mitigated by emission control measures feasible under current technology. Minimizing tailpipe NOₓ and NH₃ emission through engine design, emission control technologies and 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. Further studies are required to understand other environmental impacts (e.g. NH₃ leakage, GHG emissions from NH₃ production) of using NH₃ as shipping fuel from a life-cycle perspective.

5. Acknowledgement

This research was supported by a grant from the MIT Climate and Sustainability Consortium. The MERRA-2 data used in this study/project 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 (IENV) and Environmental Central Facility (ENVF) of the Hong Kong University of Science and Technology (HKUST).

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Supplemental Information

Additional details in deriving EF for the pure NH₃ and NH₃–H₂ engines

Fig. S1 Emission factors (a – c) and indicated specific fuel consumption (ISFC, d) as the function of engine load for pure NH₃ drivetrain. Dots indicate the raw data from (Mounaïm-Rousselle et al 2022), and lines indicate the generalized additive model fitting.

The engine EF for pure NH₃ engines follow the experiment result of Mounaïm-Rousselle et al (2022) corrected by the same drivetrain mechanical efficiency (92.5%) implied by Imhoff et al (2021), with a generalized additive model to derive the continuous load curves (Fig. S1a – 1c). We do not extrapolate the EF and ISFC beyond engine load < 20% due to the lack of data.

The engine EF of NH₃–H₂ is significantly more complicated due to the possibility of varying NH₃:H₂ ratio to achieve different efficiency, engine stability and emission profile (Mercier et al 2022, Lhuillier et al 2020). We assume the drivetrain EF before any treatment presented by Imhoff et al (2021) (29.4 g NOₓ/kWh and 16.7 g NH₃/kWh) are representative of the emissions at full engine load.

Fig. S2 Engine exhaust NH₃ and NOₓ concentration as a function of engine load and hydrogen fraction in the fuel mix from an NH₃–H₂ engine test (Mounaïm-Rousselle et al 2021)
The data from Mounaïm-Rousselle et al (2021) (Fig. S2) suggest that as engine load decreases and H$_2$ fraction is held constant, NH$_3$ EF increases while NO$_x$ EF decreases. However, the introduction of additional H$_2$ as load decreases, which serves as the combustion promoter to improve engine stability and performance, partially offsetting the trends in NO$_x$ and NH$_3$ EF.

We assume that the NH$_3$–H$_2$ engines can adaptively increase the fraction of H$_2$ input to preserve the NO$_x$ and NH$_3$ concentration in the engine exhaust as engine load lowers. Assuming the air–fuel ratio stays relatively constant, NO$_x$ and NH$_3$ EF can be approximated as a function of indicated specific fuel consumption (ISFC) (see Eq. S1 – S3) which indicates engine efficiency because:

Emission factor of pollutant i (EF$_i$) can be expressed as the mass of pollutant emitted per unit energy output, which is equivalent to pollutant mass flow rate ($\rho_i$) per unit power output (P):

$$EF_i = \frac{\rho_i}{P} \quad (S1)$$

Power output can be expressed in terms of indicated specific fuel consumption (ISFC, fuel mass flow per unit power output):

$$P = \frac{\rho_f}{ISFC} \quad (S2)$$

Where $\rho_f$ is the fuel mass flow rate. This converts the expression of EF$_i$ into:

$$EF_i = \frac{\rho_i}{\rho_f} ISFC = \frac{(\rho_e)}{\rho_f} m_i C_i ISFC \quad (S3)$$

Where $\rho_e$ is the exhaust mass flow rate, $m_e$ and $m_i$ are the molar mass of the exhaust mixture and i, respectively, and $C_i$ is the concentration of i in exhaust.

Now we examine equation S3 as a function of engine load. Assuming the air–fuel ratio remains constant, $\frac{\rho_e}{\rho_f} = $ constant by conservation of mass. As most of the combustion mixture consists of inert dinitrogen gas from the air, $m_e$ is also relatively stable. Therefore, if $C_i$ remains relatively constant over a range of load, EF is mostly a function of ISFC. As indicated by Fig. S2, introducing additional H$_2$ counteracts the trending of decreasing NO$_x$ and increasing NH$_3$ concentrations as load lowers, leading to relatively stable C$_{NH3}$ and C$_{NOx}$ as load changes. Therefore, we assume that the EF of NH$_3$–H$_2$ is solely a function of ISFC.

Since there is no information about the load-dependence of ISFC for NH$_3$–H$_2$ engines, we assume that it takes similar shape as that of pure NH$_3$ engines (Fig. S1d). Though the addition of H$_2$ increases combustion and thermodynamic efficiency, extra energy would be required to crack more H$_2$ into NH$_3$, which lowers the overall efficiency as engine load decreases. Similarly, the N$_2$O EF and load curve are assumed to be the same between pure NH$_3$ and NH$_3$-H$_2$ engines, due to lack of direct measurements for NH$_3$-H$_2$ engines.
Engine emissions are also influenced by engine size and speed, which cannot be directly accounted for in this study due to lack of experimental data. For both pure NH$_3$ and NH$_3$–H$_2$ engines, a 24% of EF penalty is added to ships with lengths under 100m to account for the lower thermodynamic efficiency from smaller engines, which is consistent with Imhoff et al. (2021).

We rely on experimental data from small fast four-stroke engines, which is different from the slow large two-stroke engines that are typical for large commercial vessels (Anantharaman et al 2015), since there is no published experimental data for large two-stroke marine engines that uses ammonia as the major energy source. Large two-stroke engines have been shown to have lower unburnt methane (fuel) slip than the smaller four-stroke engines when operating with liquefied petroleum gas (Pavlenko et al 2020). Thus, our study likely provides an upper bound of NH$_3$ emissions from ammonia-powered ships.
Effects of turning off PARANOX

Fig. S3 Differences in modelled responses in annual mean MDA8 O\textsubscript{3} and PM\textsubscript{2.5} under emissions of [Pure NH\textsubscript{3}]\textsubscript{2020} when PARANOX is turned off, calculated as ([Pure NH\textsubscript{3}]\textsuperscript{No_PARANOX} \textminus Baseline\textsuperscript{No_PARANOX}) \textminus ([Pure NH\textsubscript{3}]\textsubscript{2020} \textminus Baseline)

The model result in the main text is performed with PARANOX turned on, which parameterizes the chemical evolution in a typical fossil fuel powered ship plume before it is blended into the background atmosphere. In practice, turning on PARANOX reduces NO\textsubscript{x} lifetime by promoting NO\textsubscript{x} loss to HNO\textsubscript{3}, which is a terminal NO\textsubscript{x} sink that deposits rapidly and a PM\textsubscript{2.5} precursor, affecting both O\textsubscript{3} and PM\textsubscript{2.5} sensitivity to precursor emissions. Due to distinct chemical composition (e.g. presence of NH\textsubscript{3}, absence of SO\textsubscript{x}, NMVOC and carbonaceous aerosols), the NO\textsubscript{x} lifetime within an ammonia-powered ship plume is highly uncertain.

To briefly explore how might the uncertainty in plume chemistry affect our result, we perform another set of simulations ([Pure NH\textsubscript{3}]\textsubscript{2020} and baseline) with PARANOX turned off ([Pure NH\textsubscript{3}]\textsuperscript{NO_PARANOX} and baseline\textsuperscript{NO_PARANOX}), which means the ship plume is immediately blended into background atmosphere. While being seemingly less realistic than turning PARANOX on, this configuration can be interpreted as simply assuming a longer NO\textsubscript{x} lifetime than that in our standard simulations.

Figure S3 shows the difference in modelled O\textsubscript{3} and PM\textsubscript{2.5} responses when PARANOX is turned off for [Pure NH\textsubscript{3}]\textsubscript{2020}. PARANOX simulates plume chemistry by converting NO\textsubscript{x} emissions into O\textsubscript{3} and HNO\textsubscript{3} before releasing them into model grid cells. When PARANOX is turned off, the NO\textsubscript{x} concentration differences between the two scenarios are amplified. Therefore, the modelled O\textsubscript{3} reduction is stronger (more negative) globally by up to 2.8 ppbv, particularly over East Asia, Mediterranean Basin, Red Sea, and Persian Gulf, which significantly impacts ΔM\textsubscript{O3} (-100,000 versus -73,100 with PARANOX on). Overall, turning off PARANOX does not significantly affect ΔM\textsubscript{PM2.5} in global scale (+666,520 versus +668,100 with PARANOX on). However, significant local differences in modelled PM\textsubscript{2.5} responses of up to 2 µg m\textsuperscript{-3} exist over East Asia and northern Europe, which may be attributable to differences in other NO\textsubscript{x} sources near the shore and interactions with concomitant SO\textsubscript{x} reductions. The resulting ΔM\textsubscript{total} with PARANOX off (+566,600) is well within the 95\% CI (+448,000; +746,200) of that with PARANOX on. This suggests that the uncertainty in plume chemistry is much more likely to affect modelled O\textsubscript{3} than PM\textsubscript{2.5} response. While such uncertainty does not affect our main findings (1. Newer engines have lower NO\textsubscript{x} emissions, which generally benefits O\textsubscript{3} air quality; 2. unburnt NH\textsubscript{3} emission worsens PM\textsubscript{2.5} air quality unless it is tightly controlled).
Rough estimate of climate effects from nitrogen deposition

Wolfram et al (2022) raise concern about “secondary N₂O emissions” from reactive nitrogen (NH₄⁺ + NH₃ + NOₓ + Other oxidized nitrogen species derived from NOₓ) deposition. This is not a concern NH₃-H₂ engines, since most of the reactive nitrogen from NH₃-H₂ engines is removed by SCR, which converts NOₓ and NH₃ into non-reactive forms of nitrogen (N₂ and to lesser extent N₂O). The total reactive nitrogen emissions (and therefore deposition) (3.4 TgN/yr) from NH₃-H₂ engines are lower than that from current fleet (5.3 TgN/yr).

For pure NH₃ engine, most reactive nitrogen emissions are removed through scrubbing, which does not convert reactive nitrogen back to non-reactive forms of nitrogen, leading to large amount of reactive nitrogen entering the Earth System (60 – 70 TgN/yr). While rigorous evaluations of the impacts of nitrogen deposition on global GHG balance are beyond the scope of this paper, we use scenarios with highest total reactive nitrogen emissions (Pure NH₃) for to briefly discuss about the potential impacts and range of uncertainties of reactive nitrogen deposition on GHG emissions.

Over the ocean, the fraction of ammonium being converted into N₂O vary from 0.01% under typical condition to up to 2% under oxygen depletion (Babbin et al 2020). This implies that nitrogen deposition over oxygen minimum zones (OMZ) can have disproportionate impact on N₂O emission. Under the scenario with the largest increases in reactive nitrogen emissions (Pure NH₃), we find a total increase of 5.0 TgN/yr in nitrogen deposition over OMZ (defined as grid cells with O₂ concentration < 20 µM at the most anoxic depth (Paulmier and Ruiz-Pino 2009), with maps of ocean oxygen levels provided by World Ocean Atlas 2018 Garcia et al (2019)). If 1% and 0.01% of the depositing reactive nitrogen were converted into N₂O within and outside OMZ, respectively, this would lead to 45.7 Tg CO₂.e/yr (88% of tailpipe GHG emission from ammonia-powered fleet) increase in GHG emission. Such increases may be offset by increase in global ocean carbon sinks (Jickells et al 2017).

We also compare the contemporary effect of nitrogen deposition on GHG balance over land. Yang et al (2021) estimate that global nitrogen deposition leads to N₂O emission of 0.89 Tg/yr (= 243 Tg CO₂.e/yr) over cropland, which is comparable to net climate effect of nitrogen deposition over natural ecosystems (500 Tg CO₂.e/yr sequestered) (Xiao et al 2023). However, another recent study suggests a much smaller increase in forest carbon sequestration (150 Tg CO₂/yr) (Schulte-Uebbing et al 2022) due to nitrogen deposition. The net effects of nitrogen deposition on GHG balance over land is therefore highly uncertain. However, it is likely that the net effect is smaller than the reduction in tailpipe GHG emissions from switching to ammonia powered ships (817.2 Tg CO₂.e/yr). Despite this, with other adverse environmental effects from excessive nitrogen deposition (e.g. eutrophication, soil acidification, biodiversity loss) (Payne et al 2017, Baessler et al 2019), large amount of reactive nitrogen entering the Earth System is undesirable.
Sensitivity of PM$_{2.5}$ to ammonia emission under presence of sea salt

Under polluted environments, NH$_3$ mostly forms secondary inorganic aerosols with H$_2$SO$_4$ and HNO$_3$, which are the oxidation products of NO$_x$ and SO$_x$:

\[
2\text{NH}_3 + \text{H}_2\text{SO}_4 \rightarrow (\text{NH}_4)_2\text{SO}_4 \quad \text{(R1)}
\]
\[
\text{NH}_3 + \text{HNO}_3 \leftrightarrow \text{NH}_4\text{NO}_3 \quad \text{(R2)}
\]

Therefore, if the changes in ammonium aerosols were only caused by interactions with H$_2$SO$_4$ and HNO$_3$, the changes in ammonium, sulphate and nitrate aerosols should follow the stoichiometry prescribed by R1 and R2. In contrast, if the changes in ammonium, sulphate and nitrate aerosols were not stoichiometric, that could indicate alternative aerosol formation pathways that alters the sensitivity of secondary inorganic PM$_{2.5}$ level to precursor emissions, which is highly possible over the ocean due to presence of ion-rich sea spray aerosols.

To examine the potential impacts of such alternative aerosol formation pathways, we calculate the deviation of changes in ammonium PM$_{2.5}$ mass from the stoichiometric conditions of the NH$_3$–H$_2$SO$_4$–HNO$_3$ system ($\Delta M^*_\text{NH}_4$) from [NH$_3$–H$_2$]$_{2020}$ run, which is shown in Figure S4:

\[
\Delta M^*_\text{NH}_4 = 1.1 \rho m_\text{NH}_4 \left( \Delta C_{\text{NH}_4} - 2\Delta C_{\text{SO}_4} - \Delta C_{\text{NO}_3} \right) \quad \text{(S4)}
\]

Where $\rho$ is the molar density of air, $m_\text{NH}_4$ is molar mass of NH$_3$, $\Delta C_i$ are the concentration changes of the corresponding secondary inorganic aerosol species. The factor 1.1 is included to match the standard conditions of PM$_{2.5}$ measurements used in the main manuscript.

Fig. S4. Deviation of changes in ammonium PM$_{2.5}$ mass from stoichiometric conditions ($\Delta M^*_\text{NH}_4$) as calculated by Equation S4, and changes in PM$_{2.5}$ mass from sea salt chloride ($\Delta M_{\text{Cl}}$)

We find that $\Delta M^*_\text{NH}_4$ is always positive, with largest value of 0.7 µg m$^{-3}$ around the Arabian Peninsula. While studying the detailed chemical interactions between NH$_3$ and other atmospheric acids and ions is beyond the scope of this paper, one possible explanation of positive $\Delta M^*_\text{NH}_4$ is aerosol formation with other anions within sea salt aerosols (e.g. chloride as shown in Figure S4). Also, fine sea spray aerosols are inherently acidic (Angle et al 2021), which could react with NH$_3$ independently. This shows the existence of extra sensitivity of PM$_{2.5}$ to NH$_3$ in marine environment that is independent to SO$_x$ and NO$_x$ emissions. Thus, controlling coastal and marine NO$_x$ and SO$_x$ alone could not eliminate the sensitivity of PM$_{2.5}$ to NH$_3$ emissions.
Excessive Water Vapor generated from Ammonia Combustion

The mass of water vapor generated per unit energy released from complete combustion of fuel i (\(m_{w,i}\)) can be calculated as:

\[
m_{w,i} = \frac{1}{LHV_i} \left( \frac{M_w}{M_i} \right) \lambda_{w,i} \quad (S4)
\]

where \(LHV_i\) = the Lower Heating Value (LHV) of fuel i, \(M_w\) = molar mass of water, \(M_i\) = molar mass of fuel i, \(\lambda_{w,i}\) = moles of water formed per moles of fuel i under complete combustion.

Using lower heating values at 25 °C of diesel (43.4 MJ/kg) (Linstrom 1997) and ammonia (18.8 MJ/kg) (Valera-Medina et al 2018), and taking the average chemical formula of diesel to be \(\text{C}_{12}\text{H}_{23}\) (Date and Date 2011), \(m_{w,diesel} = 28.6\, \text{g/MJ}\) and \(m_{w,ammonia} = 84.5\, \text{g/MJ}\). This implies for a given amount of energy released, complete combustion of ammonia generates two times more water vapor than diesel.

Another equation (S5) can be derived from equation S4 to explain the difference between \(m_{w,diesel}\) and \(m_{w,ammonia}\):

\[
\frac{m_{w,ammonia}}{m_{w,diesel}} = \frac{LHV_{diesel}}{LHV_{ammonia}} \left( \frac{M_{ammonia}}{M_{diesel}} \frac{\lambda_{w,diesel}}{\lambda_{w,ammonia}} \right) = \frac{LHV_{diesel}}{LHV_{ammonia}} R \quad (S5)
\]

\(R = 1.27\), while \(\frac{LHV_{diesel}}{LHV_{ammonia}} = 2.31\). Therefore, the excessive water vapor generated from ammonia combustion is mainly attributable to its low LHV.


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