



Short communication

Nitrogen deposition from aviation emissions

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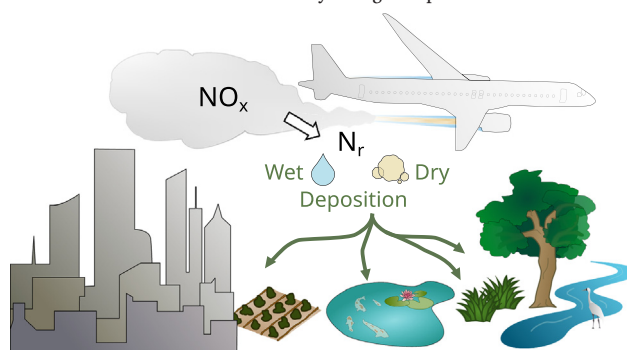


HIGHLIGHTS

- Atmospheric model estimates spatially-resolved nitrogen deposition from aviation.
- Aviation led to 1.39 Tg of N deposition globally in 2019, up 72 % from 2005.
- 0.7 % (Asia), 1.1 % (Europe), 1.6 % (North America) of N deposition is due to aviation.
- N deposition from aviation is spatially widespread, with 56 % occurring over water.

GRAPHICAL ABSTRACT

Aviation emissions lead to wet and dry nitrogen deposition over different land cover types.



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ABSTRACT

Excess nitrogen deposition from anthropogenic sources of atmospheric emissions, such as agriculture and transportation, can have negative effects on natural environments. Designing effective conservation efforts requires knowledge of the contribution of individual sectors. This study utilizes a global atmospheric chemistry-transport model to quantify, for the first time, the contribution of global aviation NO_x emissions to nitrogen deposition for 2005 and 2019. We find that aviation led to an additional 1.39 Tg of nitrogen deposited globally in 2019, up 72 % from 2005, with 67 % of each year's total occurring through wet deposition. In 2019, aviation was responsible for an average of 0.66 %, 1.13 %, and 1.61 % of modeled nitrogen deposition from all sources over Asia, Europe, and North America, respectively. These impacts are spatially widespread, with 56 % of deposition occurring over water. Emissions during the landing, taxi and takeoff (LTO) phases of flight are responsible for 8 % of aviation's nitrogen deposition impacts on average globally, and between 16 and 32 % over most land in regions with high aviation activity. Despite currently representing less than 1.2 % of nitrogen deposition globally, further growth of aviation emissions would result in increases in aviation's contribution to nitrogen deposition and associated critical loads.

1. Introduction

Multiple sectors of human activity affect the Earth's nitrogen cycle due to atmospheric release of reactive nitrogen (N_r) (Galloway et al., 2021). The increase in the use of fertilizers and dense livestock farming since the early 1900s has been essential in supporting food supply for a growing world population, but these activities also lead to the emission of reduced

N_r compounds, mainly NH_3 (Erisman et al., 2008). At the same time, burning fuel for transportation, energy generation, heating, and industrial processes leads to the emission of oxidized N_r compounds, such as NO_x ($\text{NO} + \text{NO}_2$). In addition to impacting the climate and air quality while in the atmosphere (Szopa et al., 2021), the deposition of these species leads to the degradation of water quality, causing higher toxicity to humans and animals, loss of biodiversity, and soil acidification (Galloway et al., 2021).

Conservation objectives for nitrogen-sensitive natural assets in nature reserves (e.g. Natura 2000 in Europe) have resulted in thresholds for the

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nitrogen load in such areas, but these are frequently exceeded. In 2017, in Europe ~64 % of ecosystem areas exceeded the critical loads for eutrophication (Fagerli et al., 2019). In hotspots, such as the Netherlands, where in 2019 the specified critical loads were exceeded in 118 out of the 162 Dutch Natura 2000 sites (Hazekamp, 2019), the resulting nitrogen problem is having increasing socioeconomic and political implications (Stokstad, 2019).

Although currently responsible for ~4.1 % of anthropogenic NO_x (O'Rourke et al., 2021; Quadros et al., 2022a), due to the growth rate of aviation forecast at ~3–4 % annually and to the lack of presently available NO_x reducing technologies for aircraft, global aviation NO_x emissions are estimated to continue increasing in the coming decades (International Civil Aviation Organization, 2021b; Fleming and de Lépinay, 2019; International Energy Agency, 2021; Quadros et al., 2022b). Part of the difficulty in reducing NO_x emissions are the decades-long trend in aircraft engine design of increasing gas temperature and pressure, which are beneficial for lowering fuel consumption but that promote NO_x formation, leading to tradeoffs that involve both climate and air quality impacts (Skowron et al., 2021; Miller et al., 2022). Aviation is also unique in terms of the altitude at which the emissions are released – the average cruise altitude of present-day civil aircraft is around 11 km. Out of the total of ~1.4 Tg N that was emitted by aircraft in 2019, ~1.3 Tg N were emitted at an altitude higher than 3000 ft. above ground, and primarily in the northern midlatitudes (Quadros et al., 2022a). The other N_r source at high altitudes is lightning NO_x which is estimated to be 2–8 Tg N per year (Schumann and Huntrieser, 2007), with more recent estimates at ~9 Tg N per year (Nault et al., 2017). The portion of flights below 3000 ft. is defined as the landing, taxi and takeoff (LTO) phases. While only approximately a tenth of total aircraft's NO_x is emitted in LTO, these emissions are largely within the Earth's planetary boundary layer, having a more localized effect and a larger air quality impact per mass than cruise emissions (Yim et al., 2015; Quadros et al., 2020). For non-CO₂ species, LTO emissions also constitute the focus of present environmental regulations for the aviation sector.

Recent research efforts have aimed to improve estimates of the climate and air quality impacts associated from aviation emissions (Skowron et al., 2021; Miller et al., 2022; Yim et al., 2015; Quadros et al., 2020; Lee et al., 2021). However, as we move towards setting sectoral budgets for controlling nitrogen emissions and associated loads in areas of ecological interest, there is also a need to extend this body of research by quantifying aviation emissions' contribution to nitrogen deposition. In this paper we present the first comprehensive quantification of aviation's global nitrogen deposition impacts. We use a 3D atmospheric chemistry-transport model and up-to-date aviation emissions inventories to quantify the aviation-attributable nitrogen deposition globally and over different land cover classes, both for LTO emissions and high altitude (cruise) emissions.

2. Materials and methods

2.1. Method overview

A global atmospheric chemistry-transport model is used to estimate the amount and location of wet and dry nitrogen deposition due to aviation emissions. The model is run with and without aviation emissions, with the difference in output being attributed to aviation. Aviation emissions in this study consist of a nominal estimate of aircraft main engine and auxiliary power unit emissions of NO_x, SO_x (SO₂ + SO₄²⁻), CO, hydrocarbons, and black carbon. The analysis is performed for the years 2005 and 2019, with simulations using meteorology and both aviation and non-aviation emissions estimates specific to each year. Two additional scenarios are also simulated: 2019 with only the LTO portion of aviation emissions, and 2019 but with aviation emissions as they were in 2005.

2.2. Aviation emissions

Monthly-average civil aircraft emission rates in 2005 and 2019 are compared from inventories produced using the methods and data published by

Simone et al. (2013) and Quadros et al. (2022a). The emissions are allocated into a 3-D grid at the resolution of the atmospheric simulation. The 2005 inventory is created from a worldwide database of flight schedules produced by the company OAG, and the 2019 inventory uses a dataset provided by the company Flightradar24 listing all flights recorded by their network of telemetry receivers. Within those models, NO_x, CO, and hydrocarbon emissions throughout each flight are calculated from fuel burn and engine thrust setting using the Boeing Fuel Flow Method 2, described by Baughcum et al. (1996) and engine data from the ICAO Engine Emissions Databank (International Civil Aviation Organization, 2021a). Black carbon emissions use the same engine data source and the FOA 4.0 method (International Civil Aviation Organization, 2020). SO_x emissions are calculated with a constant emission index equivalent to a sulfur fuel content of 600 ppm. The resulting annual sums of nitrogen in aviation NO_x emissions is 0.82 (2005) and 1.41 Tg N (2019).

2.3. Atmospheric modeling

Global atmospheric chemistry and transport are simulated with the GEOS-Chem 13.3.3 model (The International GEOS-Chem User Community, 2021), using the stratospheric chemistry model developed by Eastham et al. (2014) and driven by meteorology from the MERRA-2 reanalysis product (Gelaro et al., 2017). Global simulations are performed at 2° latitude by 2.5° longitude resolution. For the 2019 scenarios, simulations are also performed using three 0.5° latitude by 0.625° longitude nested grids over Asia, Europe, and North America (Fig. S1), and those results are overlaid on the output from the coarser global simulation, which provides boundary conditions for the regional simulations, improving the allocation of deposition into the different land cover types in the regions with highest airport activity.

Nitrogen deposition is considered in the form of deposition of the following simulated species: NO₂, HNO₃, inorganic nitrates (NO₃⁻), inorganic nitrates on sea salt aerosol, NH₄⁺, NH₃, N₂O₅, and peroxyacetyl nitrate. Non-aviation anthropogenic emissions are from the CEDS v2 inventory, which includes the agriculture, energy, industry, surface transportation, residential, shipping, and waste sectors (O'Rourke et al., 2021). Soil NO_x, sea salt aerosols, and biogenic volatile organic compounds are calculated according to meteorological conditions (Weng et al., 2020). Lightning emissions are parametrized from modeled convection and corrected using satellite flash rate data (Murray et al., 2012). Further details on atmospheric modeling and nitrogen deposition with GEOS-Chem are included in Section S1 of the Supporting Information (SI).

2.4. Land cover and conservation areas

Average deposition fluxes are calculated for different land cover classes, according to the Copernicus global land cover product for 2019 (Buchhorn et al., 2020). Areas not covered by the Copernicus dataset are disregarded in the analysis – this consists mainly of all the areas north of 80°N and south of 60°S. From this 100 m resolution land cover data, the fraction of each grid cell that contains each class of land cover is calculated. Likewise, the percentage of each grid's area that lies in Natura 2000 sites, protected either through the Birds Directive or the Habitats Directive, is calculated from the 2019 definitions of the protected areas (European Environment Agency, n.d.). The analysis considering Natura 2000 sites is only performed for the scenarios that include the 0.5° × 0.625° resolution simulation over Europe.

3. Results and discussion

3.1. Quantification of aviation-attributable nitrogen deposition

Throughout 2019, aviation emissions led to additional 1.39 Tg N of nitrogen wet and dry deposition globally, which is 1.13 % of total modeled deposition. While certain areas might be net importers or exporters of N_r due to transport (Dentener et al., 2006; Schulte-Uebbing et al., 2022;

Zhang et al., 2012), the increase in global deposition is equal to 99.0 % of the amount of N_r mass released by aviation that year. In both 2005 and 2019, wet deposition accounts for approximately 67 % of total nitrogen deposition from aviation. Total aviation-attributable nitrogen deposition in 2019 is 72 % larger than in 2005 (0.81 Tg N), in the same proportion as aviation emissions, which increased an average of 4 % per year in this period. By contrast, global anthropogenic NO_x emissions in 2019, other than aviation, were only 7.5 % higher than in 2005. Given the long useful life remaining in the present fleet of conventionally powered aircraft, aviation emissions will likely continue to globally increase for decades if air traffic grows as projected (Quadros et al., 2022b), with the sector being responsible for a growing share of anthropogenic nitrogen deposition worldwide. The potential adoption of sustainable aviation fuels, currently being pursued to reduce net carbon emissions, is unlikely to lead to changes in aviation NO_x emissions (Hedges et al., 2019).

This deposition, however, does not occur uniformly over the globe's surface, nor does it match the spatial or speciated distribution of emissions. The highest fluxes of aviation-attributable nitrogen deposition are close to the regions of highest aviation activity (the coasts of North America, Europe, and Eastern Asia), but the high values extend out following the prevailing winds and in regions of heavy precipitation, as seen in the Atlantic, off the coast of North America, and in waters west of China and Japan (Fig. 1a). Although the absolute amount of aviation-attributable deposition is lower over the oceans than over the continents, the share of modeled nitrogen deposition that is due to aviation is higher there, where there are fewer other local sources of emission besides aircraft (Fig. 1b). It is expected that the percentage of nitrogen deposition that is due to non-land emissions, which also include oceanic NH_3 and shipping NO_x , be larger further away from continents (Paulot et al., 2013; Zhao et al., 2015). Considering only populated grid cells (i.e. including only land; Fig. S5) (Rose et al., 2020), aviation is responsible for 0.66 %, 1.13 %, and 1.61 % of all modeled nitrogen deposition averaged over Asia, Europe, and North America, respectively. Locally, this contribution can be up to 10 % in areas where other sources of nitrogen are less significant, such as in Greenland.

Over land with high aviation activity, LTO emissions account for more than 16 % of total nitrogen deposition from aviation, even though globally LTO contributes to 7.8 % of aviation's nitrogen deposition impacts (Fig. 1c). Compared to full-flight emissions, a larger fraction of deposition due to LTO emissions occurs as dry deposition, 50 % (Fig. 2). This is expected due to the closer proximity of emissions to the ground, leading to more of the nitrogen to deposit through dry processes before water precipitation takes place.

The three simulations with full-flight emissions (2005, 2019, and 2005 aviation with 2019 background), in addition to having similar ratios of aviation-attributable wet and dry deposition, also show similar distributions of species deposited, once normalized by the extra amount of N_r emitted (Fig. 2). The proportionality of aviation-attributable total N_r mass deposited to NO_x emissions is a consequence of this species being the main responsible for the deposition and the linearity of their relationship, with the response to non- N_r emissions expected to be more nonlinear (Paulot et al., 2013). In all scenarios, most of the nitrogen is deposited as HNO_3 . A smaller fraction is deposited as inorganic nitrates, either aggregated on sea salt or dissolved in water precipitation. The aviation-induced increase in NH_4^+ deposition is matched by a decrease in NH_3 deposition, as the HNO_3 formed from NO_x released by aircraft reacts with NH_3 already present in the atmosphere, released by other sources, to form ammonium nitrate. To a lesser extent, this is also driven by the formation of ammonium sulfate from aviation emitted sulfur oxides. This also drives the difference in the speciated distribution of the nitrogen deposition of 2005 full-flight emissions in a 2005 and a 2019 atmosphere (two bottom rows in Fig. 2). Primarily driven by non-aviation emissions changes, the “potential” of NO_x emissions to form fine particulate matter ($PM_{2.5}$) increases from 2005 to 2019 (Quadros et al., 2020; Dedoussi et al., 2020; Dedoussi, 2021). Increases in the background concentration of NH_3 and lower emissions of NO_x from other sources can increase the sensitivity of ground level particulate matter to aviation emissions, affecting the form in which reactive nitrogen from aviation will deposit (Woody et al., 2011). Attribution to specific aviation emissions species was not evaluated in this study, but the speciation of nitrogen deposition obtained is consistent with NO_x being the main driver of these impacts, as is also suggested by previous findings that most of aviation-attributable

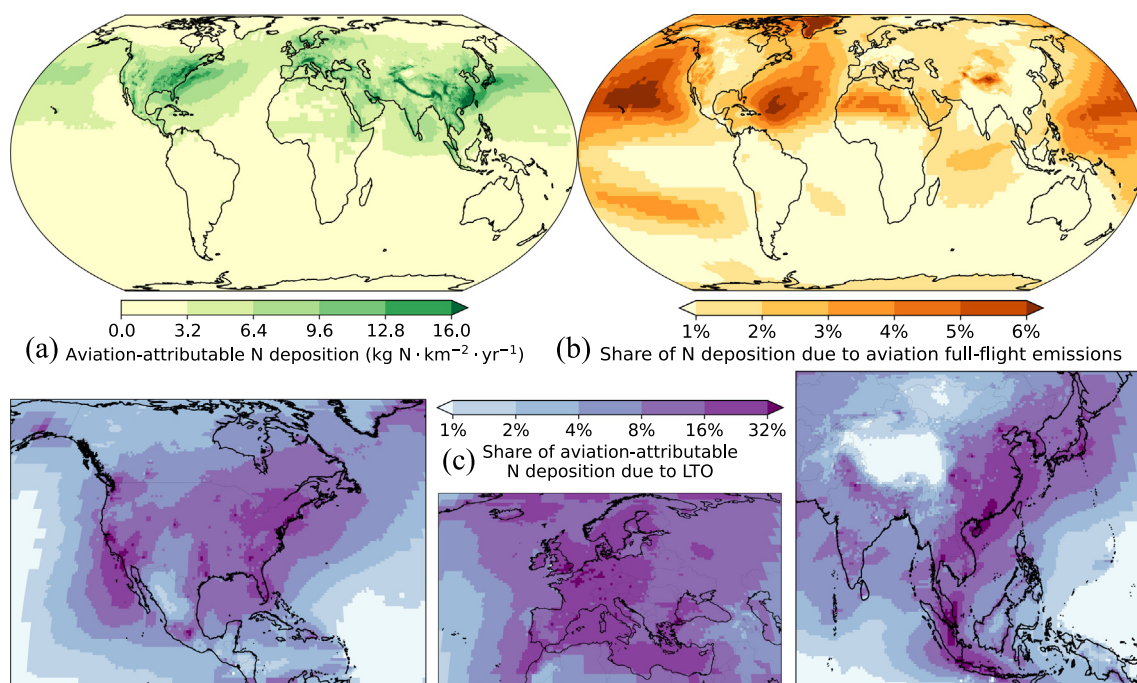


Fig. 1. Aviation-attributable total (wet + dry) nitrogen deposition in 2019: flux (a), relative to modeled deposition from all sources (b), LTO-attributable relative to deposition from full-flight emissions (c).

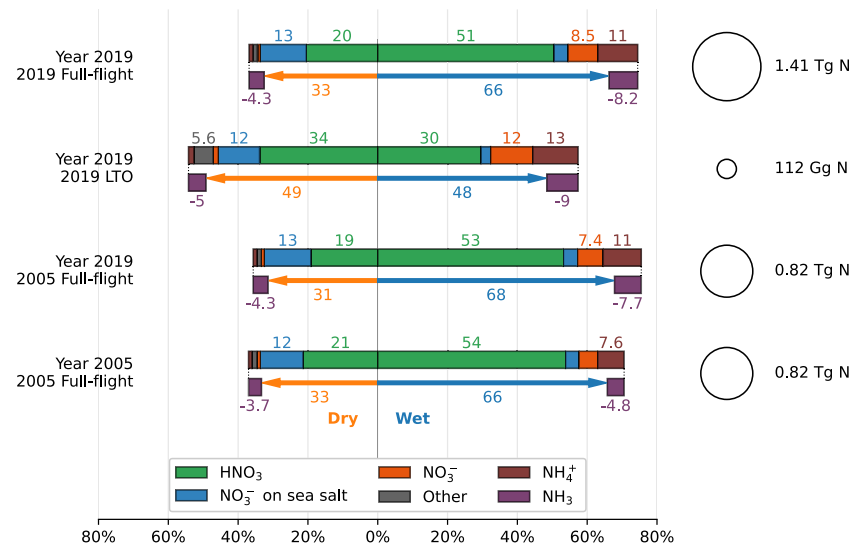


Fig. 2. Mass of aviation-attributable nitrogen deposited globally, per species, relative to the nitrogen mass emitted by aviation as NO_x , which is indicated by the quantities on the right.

ground level particulate matter is due to its NO_x emissions (Prashanth et al., 2022; Grobler et al., 2019).

3.2. Aviation-attributable nitrogen deposition per land cover and on Natura 2000 areas

Environmental policies aim to reduce excessive nitrogen deposition over areas with ecosystems sensitive to it, such as Natura 2000 sites, and

therefore a quantification of aviation's impacts also requires an estimate of how much nitrogen is deposited on vulnerable areas. Due to the extensive area covered by flights and their altitude, with most emissions released above the planetary boundary layer, aircraft emissions tend to have more spatially widespread impacts than emissions by sources closer to the ground. In our 2019 simulation, 38 % of nitrogen from all sources is deposited over water, while 43 % and 56 % of nitrogen attributable to LTO and full-flight emissions, respectively, do so (Table 1). Within

Table 1

Average nitrogen deposition flux over different land cover classes from all sources in 2019 and from aviation emissions for the four scenarios considered. Percentage values under deposition fluxes represent the proportion of nitrogen mass of each emissions scenario deposited in each land cover class. Deposition over Natura 2000 sites is only evaluated for the finer resolution simulations.

Land cover class	Area (10^{-6} km^2) % of total area considered	Average nitrogen deposition flux ($\text{kg N} \cdot \text{km}^{-2} \cdot \text{yr}^{-1}$) % total mass deposited				
		All sources in 2019	LTO emissions in 2019	Full-flight emissions in 2019	2005 full-flight emissions in 2019	Full-flight emissions in 2005
Urban / built up	1.2	1397	1.95	9.15	5.14	5.54
	0.3 %	1.4 %	2.1 %	0.8 %	0.8 %	0.8 %
Cultivated and managed vegetation	14.9	1169	0.84	6.52	3.77	3.93
	3.2 %	14.3 %	11.5 %	7.0 %	6.9 %	7.2 %
Closed forest	36.4	703	0.61	5.04	2.76	2.84
	7.7 %	21.0 %	20.5 %	13.2 %	12.4 %	12.8 %
Open forest	13.2	703	0.56	4.69	2.66	2.75
	2.8 %	7.6 %	6.8 %	4.5 %	4.3 %	4.5 %
Shrubs	11.2	365	0.24	3.22	1.95	1.95
	2.4 %	3.4 %	2.5 %	2.6 %	2.7 %	2.7 %
Herbaceous vegetation	28.4	400	0.26	3.81	2.18	2.1
	6.0 %	9.3 %	6.8 %	7.8 %	7.7 %	7.4 %
Herbaceous wetland	2.3	422	0.34	3.35	1.97	1.96
	0.5 %	0.8 %	0.7 %	0.6 %	0.6 %	0.6 %
Moss and lichen	1.4	48	0.06	1.36	0.76	0.69
	0.3 %	0.1 %	0.1 %	0.1 %	0.1 %	0.1 %
Bare / sparse vegetation	20.3	232	0.3	4.6	2.68	2.48
	4.3 %	3.9 %	5.6 %	6.8 %	6.7 %	6.2 %
Snow and ice	2	98	0.11	2.43	1.28	1.15
	0.4 %	0.2 %	0.2 %	0.3 %	0.3 %	0.3 %
Permanent water bodies	3.2	457	0.49	4.45	2.72	2.88
	0.7 %	1.2 %	1.4 %	1.0 %	1.1 %	1.1 %
Open sea	338.5	134	0.13	2.26	1.35	1.34
	71.6 %	37.1 %	41.7 %	55.3 %	56.4 %	56.3 %
All land classes	131.2	574	0.47	4.61	2.62	2.62
	27.8 %	61.7 %	56.9 %	43.7 %	42.5 %	42.6 %
All water classes	341.7	137	0.14	2.28	1.36	1.36
	72.2 %	38.3 %	43.1 %	56.3 %	57.5 %	57.4 %
Natura 2000	1.2	705	1.38	7.40	n/a	n/a
	0.2 %	0.70 %	1.54 %	0.65 %		

land areas, LTO impacts are more concentrated on urban areas (2.1 % of global deposition due to LTO emissions) than impacts from full-flight emissions (0.8 %) and from all sources (1.4 %). The global average of aviation-attributable nitrogen deposition flux over land are 0.46 % (2005) and 0.80 % (2019) of the fluxes from all sources in those areas. These global values are lower than the averages for regions with higher aviation activity, such as 1.61 % over populated grid cells in North America in 2019. Despite the global sum of aviation-attributable nitrogen deposition increasing by 72 % between 2005 and 2019, and despite different growth rates of aviation emissions per region, the fraction of deposition over each land cover class remained similar.

Throughout 2019, aviation led to an average increase of 7.40 kg N·km⁻² in nitrogen deposition on Natura 2000 conservation sites, with 1.38 kg N·km⁻² being due to LTO emissions (Table 1). Aviation is responsible for an average of 1.0 % of nitrogen deposition from all sources in those areas. The ratios between the average aviation-attributable nitrogen deposition fluxes over Natura 2000 sites on land and the averages over all European land of members of the European Union are 0.973 for full-flight emissions and 0.941 for LTO emissions, meaning that the impacts are only marginally smaller in Natura 2000 regions than in other areas of the EU. Besides the spatially widespread nature of nitrogen deposition caused by aviation, this may also be partially attributed to the spatial coarseness of the model used (with grid cells in the region having sides of roughly between 25 and 55 km), which does not resolve small features such as the borders of these conservation areas. This analysis is made considering global aviation emissions, but future work could highlight emissions from specific sources or locations that have the highest impact on ecologically vulnerable areas (Paulot et al., 2013).

3.3. Limitations

This work focuses on the impacts associated with inorganic nitrogen deposition, as this has been shown to be responsible for over 85 % of anthropogenic nitrogen deposition impacts (Kanakidou et al., 2016), although organic nitrogen deposition may be growing (Sun et al., 2016). The presented results are inherently limited by GEOS-Chem's ability to reproduce observed nitrogen deposition values. We have evaluated GEOS-Chem against available wet and dry deposition measurements for 2005 and 2019 from North America, Europe, and Asia (Figs. S18–S24 and Tables S1–S7) (National Atmospheric Deposition Program, n.d.; U.S. Environmental Protection Agency, n.d.; Hjellbrekke and Fjæraa, 2007; Hjellbrekke, 2021; Network Center for EANET, 2006; Network Center for EANET, 2020). We found model biases between –20 % and +29 % for wet deposition. For the three datasets that also included dry deposition estimates we found dry deposition biases of +7 % (U.S. 2005), 0 % (U.S. 2019), and +104 % (Japan 2019), and total (wet + dry) deposition biases of +4 % (U.S. 2005), 0 % (U.S. 2019), and +51 % (Japan 2019). Combining all measurements for 2019, the model shows a +2.5 % bias in total deposition. The inclusion of higher resolution nested simulations did not reduce this bias compared to measurements (the bias of just the coarse resolution global simulation is +2.2 %), but increasing the resolution had some effect on the distribution of aviation-attributable nitrogen deposition per land cover class, most notably increasing deposition on urban areas due to LTO emissions by 13 % (Table S10). While we choose not to incorporate these biases in the numbers presented here, in line with other work (Ackerman et al., 2019), we note that we may be overestimating or underestimating nitrogen deposition in different regions. These biases could be carried over in the aviation-attributable nitrogen deposition estimates, for which there are no measurements to validate against. However, we do not expect this to influence the relative results on the contribution of high-altitude emissions to nitrogen deposition. Only impacts associated with aircraft activity are evaluated, including the use of the auxiliary power units at the airports, and we do not account for the use of ground support equipment or other airport-related operations. As the amount of aviation-attributable nitrogen deposition is directly proportional to NO_x

emissions, the magnitude of this impact depends on the accuracy of emission estimation. The 12 % higher top-down estimate of global fuel burn based on jet fuel sale data, which also includes military uses, compared to the bottom-up estimate used here is indicative of the combined uncertainty in fuel burn estimation (Quadros et al., 2022a). The accuracy in the fleet-average NO_x emission index is dependent on the uncertainty in assigning the correct engine model to each aircraft, and in the uncertainty in the emission measurements for each engine and the difference between them and performance in operation, including inaccuracies in the correction for atmospheric conditions. While we estimate the nitrogen deposition fluxes over Natura 2000 sites, we highlight that the grid coarseness as well as the emission assumptions of our simulations may not be able to resolve these appropriately, and future studies could further characterize more local impacts using higher fidelity modeling. Finally, future work could address the ecosystem implications of the aviation-attributable nitrogen deposition, given that the different nitrogen forms deposited will affect ecosystems in different ways and magnitudes (Ackerman et al., 2019; Liu et al., 2018; Kanakidou et al., 2018).

3.4. Policy implications

Overall, we find that aviation NO_x emissions contribute 1.39 Tg N (or 1.13 %) of global nitrogen deposition. LTO emissions are responsible for 7.8 % of those global impacts, but regionally this can be up to 32 % for areas with dense air traffic (specifically landing and takeoff operations), such as Europe and North America. The majority (73–75 %) of deposited nitrogen mass associated with aviation emissions is in the form of HNO₃, with aviation emissions resulting in a reduction in the mass of NH₃ deposition, and an increase in the mass of deposited NH₄⁺. A larger portion of deposition from full-flight (55 %) and LTO (42 %) occur over open seas than the all-source average in the model (37 %), suggesting a relatively lower impact intensity of emissions from aviation due to their spatial distribution. The non-local reach of aviation's nitrogen deposition impacts observed in the results, exemplified by similar deposition rates in Natura 2000 sites compared to adjacent areas, as well as the transnational nature of air travel, pose challenges in the implementation of local policies aiming to protect vulnerable areas. Present aviation NO_x emissions controls, such as those by ICAO CAEP, focus on the LTO phase and are primarily motivated by controlling local air quality around airports. Multiple modeling studies indicate that high altitude emissions, relative to LTO emissions, contribute 3–4 times more to aviation's impacts on global air quality and human health, in line with what is also found for nitrogen deposition in this work. Efforts to control nitrogen deposition on a global scale would benefit from addressing full-flight aviation emissions. However, we note that similarly with air quality impacts, the nitrogen deposition improvements will not necessarily be close to airports. Even surface level emissions result in impacts far from their emission location (Dedoussi et al., 2020; Liang et al., 2018). This is already acknowledged in a policy context, as significant nitrogen is imported from country to country.

CRediT authorship contribution statement

ID conceptualized the study, FQ and ML performed the model simulations and the analysis, FQ visualized the results, and wrote the initial draft, together with ID. All authors reviewed the manuscript.

Data availability

The nitrogen deposition rate data resulting from the simulations are available under the following data repository DOI: <https://doi.org/10.4121/20359683>.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Additional description of atmospheric modeling methods (Section S1), geographic distribution of aviation emissions (Figs. S1–S3), gridded Natura 2000 areas (Fig. S4), map of populated grid cells in select regions (Fig. S5), aviation-attributable N deposition fluxes per deposition process and scenario (Figs. S6–S11), ratio of 2005 aviation-attributable and all-source N deposition per scenario (Figs. S12 and S13), all-source N deposition per process and scenario (Figs. S14–S17), comparison of model to measured nitrogen deposition rates (Section S2, Figs. S18–S24 and Tables S1–S7), summary of scenarios tested (Table S8), mass of aviation-attributable N deposited over each land cover class (Table S9), grid resolution effect on deposition rates per land cover class (Table S10). Supplementary data to this article can be found online at <https://doi.org/10.1016/j.scitotenv.2022.159855>.

References

- Ackerman, D., Millet, D.B., Chen, X., 2019. Global estimates of inorganic nitrogen deposition across four decades. *Glob. Biogeochem. Cycles* 33 (1), 100–107. <https://doi.org/10.1029/2018GB005990>.
- Baughcum, S.L., Tritz, T.G., Henderson, S.C., Pickett, D.C., 1996. *Scheduled Civil Aircraft Emission Inventories for 1992: Database Development And Analysis*; NASA CR-4700. Langley Research Center, p. 205.
- Buchhorn, M., Smets, B., Bertels, L., Roo, B.D., Lesiv, M., Tsendbazar, N.-E., Herold, M., Fritz, S., 2020. Copernicus Global Land Service: Land Cover 100m: Collection 3: Epoch 2019: Globe. <https://doi.org/10.5281/zenodo.3939050>.
- Dedoussi, I.C., 2021. Implications of future atmospheric composition in decision-making for sustainable aviation. *Environ. Res. Lett.* 16 (3), 031002. <https://doi.org/10.1088/1748-9326/abe74d>.
- Dedoussi, I.C., Eastham, S.D., Monier, E., Barrett, S.R.H., 2020. Premature mortality related to United States cross-state air pollution. *Nature* 578 (7794), 261–265. <https://doi.org/10.1038/s41586-020-1983-8>.
- Dentener, F., Drevet, J., Lamarque, J.F., Bey, I., Eickhout, B., Fiore, A.M., Hauglustaine, D., Horowitz, L.W., Krol, M., Kulshrestha, U.C., Lawrence, M., Galy-Lacaux, C., Rast, S., Shindell, D., Stevenson, D., Van Noije, T., Atherton, C., Bell, N., Bergman, D., Butler, T., Cofala, J., Collins, B., Doherty, R., Ellingsen, K., Galloway, J., Gauss, M., Montanaro, V., Müller, J.F., Pitari, G., Rodriguez, J., Sanderson, M., Solomon, F., Strahan, S., Schultz, M., Sudo, K., Szopa, S., Wild, O., 2006. Nitrogen and sulfur deposition on regional and global scales: a multimodel evaluation. *Glob. Biogeochem. Cycles* 20 (4). <https://doi.org/10.1029/2005GB002672>.
- Eastham, S.D., Weisenstein, D.K., Barrett, S.R.H., 2014. Development and evaluation of the unified tropospheric-stratospheric chemistry extension (UCX) for the global chemistry-transport model GEOS-Chem. *Atmos. Environ.* 89, 52–63. <https://doi.org/10.1016/j.atmosenv.2014.02.001>.
- Erisman, J.W., Sutton, M.A., Galloway, J., Klimont, Z., Winiwarter, W., 2008. How a century of ammonia synthesis changed the world. *Nat. Geosci.* 1 (10), 636–639. <https://doi.org/10.1038/ngeo325>.
- European Environment Agency, n.. Natura 2000 End 2019 - Shapefile. https://www.eea.europa.eu/ds_resolveuid/2cf5c8d7d248489f94f238067f6d59da. (Accessed 7 July 2022).
- Fagerli, H., Tsyro, S., Jonson, J.E., Nyfiri, Á., Gauss, Simpson, D., Wind, P., Benetictow, A., Klein, H., Mortier, A., Aas, W., Hjellbrekke, A.-G., Solberg, S., Platt, S.M., Yttri, K.E., Tørseth, K., Gaisbauer, S., Mareckova, K., Matthews, B., Schindlbacher, S., Sosa, C., Tista, M., Ullrich, B., Wankmüller, R., Scheuschner, T., Bergström, R., Johanson, L., Jalkanen, J.-P., Metzger, S., van der Gon, H.A.C.D., Kuenen, J.J.P., Visschedijk, A.J.H., Barregård, L., Molnár, P., Stockfelt, L., 2019. *Transboundary Particulate Matter, Photo-oxidants, Acidifying And Eutrophying Components; EMEP Report 1/2019*. METEOROLOGISK INSTITUTT Norwegian Meteorological Institute.
- Fleming, G.G., de Lépinay, I., 2019. Environmental trends in aviation to 2050. ICAO Environmental Report 2019: Aviation And Environment – Destination Green the Next Chapter. International Civil Aviation Organization, Montreal, pp. 17–23.
- Galloway, J.N., Bleeker, A., Erisman, J.W., 2021. The human creation and use of reactive nitrogen: a global and regional perspective. *Annu. Rev. Environ. Resour.* 46 (1), 255–288. <https://doi.org/10.1146/annurev-enviro-012420-045120>.
- 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., Zhao, B., 2017. The modern-era retrospective analysis for research and applications, version 2 (MERRA-2). *J. Clim.* 30 (14), 5419–5454. <https://doi.org/10.1175/JCLI-D-16-0758.1>.
- Grobler, C., Wolfe, P.J., Dasadhikari, K., Dedoussi, I.C., Allroggen, F., Speth, R.L., Eastham, S.D., Agarwal, A., Staples, M.D., Sabnis, J., Barrett, S.R.H., 2019. Marginal climate and air quality costs of aviation emissions. *Environ. Res. Lett.* 14 (11), 114031. <https://doi.org/10.1088/1748-9326/ab4942>.
- Hazekamp, A., 2019. Question for Written Answer E-003873/2019 to the Commission. Subject: Pollution of Nature Reserves in Europe Due to Excess Nitrogen Deposition.
- Hedges, C.J., Sundstrom, L.L., Navarrete, J., Vagnerova, H., Delaney, E.P., Barnes, N., Corroero, J., Purcell, A., Hadley, R.R., Krugler, L.A., Lu, E.C., Stash, A.G., Syed, S.A., Cuddy, T., Gillette, W., Gerencer, C., 2019. *Alternative Jet Fuels Emissions - Quantification Methods Creation And Validation Report; Airport Cooperative Research Program (ACRP); ACRP 02-80*. Booz Allen Hamilton, McLean, Virginia, p. 122.
- Hjellbrekke, A.-G., 2021. Data Report 2019 Particulate Matter, Carbonaceous And Inorganic Compounds; EMEP/CCC-Report 1/2021; EMEP Co-operative Programme for Monitoring And Evaluation of the Long-range Transmission of Air Pollutants in Europe: Kjeller, Norway, p. 146.
- Hjellbrekke, A.-G., Fjæraa, A.M., 2007. Data Report 2005 Acidifying And Eutrophying Compounds And Particulate Matter; EMEP/CCC-Report 1/2007; EMEP Co-operative Programme for Monitoring And Evaluation of the Long-range Transmission of Air Pollutants in Europe: Kjeller, Norway, p. 123.
- International Civil Aviation Organization, 2020. Doc 9889, *Airport Air Quality Manual Second Edition*. International Civil Aviation Organization.
- International Civil Aviation Organization, 2021k. ICAO Aircraft Engine Emissions Databank. <https://www.easa.europa.eu/domains/environment/icao-aircraft-engine-emissions-databank>, Issue 28C. (Accessed 20 July 2021).
- International Civil Aviation Organization, 2021. Post-COVID-19 Forecasts Scenarios. <https://www.icao.int/sustainability/Pages/Post-Covid-Forecasts-Scenarios.aspx>. (Accessed 10 November 2021).
- International Energy Agency, 2021. Technology needs in long-distance transport. *Energy Technology Perspectives 2020*. IEA, pp. 250–309. <https://www.iea.org/reports/energy-technology-perspectives-2020>. (Accessed 6 August 2022).
- Kanakidou, M., Myriokefalitakis, S., Daskalakis, N., Fanourgakis, G., Nenes, A., Baker, A.R., Tsigaridis, K., Mihalopoulos, N., 2016. Past, present, and future atmospheric nitrogen deposition. *J. Atmos. Sci.* 73 (5), 2039–2047. <https://doi.org/10.1175/JAS-D-15-0278.1>.
- Kanakidou, M., Myriokefalitakis, S., Tsigaridis, K., 2018. Aerosols in atmospheric chemistry and biogeochemical cycles of nutrients. *Environ. Res. Lett.* 13 (6), 063004. <https://doi.org/10.1088/1748-9326/aabcb>.
- Lee, D.S., Fahey, D.W., Skowron, A., Allen, M.R., Burkhardt, U., Chen, Q., Doherty, S.J., Freeman, S., Forster, P.M., Fuglestedt, J., Gettelman, A., León, R.R.D., Lim, L.L., Lund, M.T., Millar, R.J., Owen, B., Penner, J.E., Pitari, G., Prather, M.J., Sausen, R., Wilcox, L.J., 2021. The contribution of global aviation to anthropogenic climate forcing for 2000 to 2018. *Atmos. Environ.* 244, 117834. <https://doi.org/10.1016/j.atmosenv.2020.117834>.
- Liang, C.-K., West, J.J., Silva, R.A., Bian, H., Chin, M., Davila, Y., Dentener, F.J., Emmmons, L., Flemming, J., Folberth, G., Henze, D., Im, U., Jonson, J.E., Keating, T.J., Kucsera, T., Lenzen, A., Lin, M., Lund, M.T., Pan, X., Park, R.J., Pierce, R.B., Sekiya, T., Sudo, K., Takemura, T., 2018. HTAP2 multi-model estimates of premature human mortality due to intercontinental transport of air pollution and emission sectors. *Atmos. Chem. Phys.* 18 (14), 10497–10520. <https://doi.org/10.5194/acp-18-10497-2018>.
- Liu, X.-Y., Koba, K., Koyama, L.A., Hobbie, S.E., Weiss, M.S., Inagaki, Y., Shaver, G.R., Giblin, A.E., Hobara, S., Nadelhoffer, K.J., Sommerkorn, M., Rastetter, E.B., Kling, G.W., Laundre, J.A., Yano, Y., Makabe, A., Yano, M., Liu, C.-Q., 2018. Nitrate is an important nitrogen source for Arctic tundra plants. *Proc. Natl. Acad. Sci.* 115 (13), 3398–3403. <https://doi.org/10.1073/pnas.1715382115>.
- Miller, C.J., Prashanth, P., Allroggen, F., Grobler, C., Sabnis, J.S., Speth, R.L., Barrett, S.R.H., 2022. An environmental cost basis for regulating aviation NOx emissions. *Environ. Res. Commun.* 4 (5), 055002. <https://doi.org/10.1088/2515-7620/ac6938>.
- Murray, L.T., Jacob, D.J., Logan, J.A., Hudman, R.C., Koshak, W.J., 2012. Optimized regional and interannual variability of lightning in a global chemical transport model constrained by LIS/OTD satellite data. *J. Geophys. Res. Atmos.* 117 (D20). <https://doi.org/10.1029/2012JD017934>.
- National Atmospheric Deposition Program, .. National Trends Network. <https://nadp.slh.wisc.edu/networks/national-trends-network/>. (Accessed 12 July 2022).
- Nault, B.A., Laughner, J.L., Wooldridge, P.J., Crounse, J.D., Dibb, J., Diskin, G., Peischl, J., Podolske, J.R., Pollack, I.B., Ryerson, T.B., Scheuer, E., Wennberg, P.O., Cohen, R.C., 2017. Lightning NOx emissions: reconciling measured and modeled estimates with updated NOx chemistry. *Geophys. Res. Lett.* 44 (18), 9479–9488. <https://doi.org/10.1002/2017GL074436>.
- Network Center for EANET, 2006. Data Report on the Acid Deposition in the East Asian Region 2005. Network Center for EANET, p. 255. <https://monitoring.eanet.asia/document/public/download?cd=139>. (Accessed 12 July 2022).
- Network Center for EANET, 2020. Data Report 2019. Network Center for EANET, p. 332. <https://monitoring.eanet.asia/document/public/download?cd=242>. (Accessed 12 July 2022).
- O'Rourke, P., Smith, S., Mott, A., Ahsan, H., McDuffie, E., Crippa, M., Klimont, Z., McDonald, B., Wang, S., Nicholson, M., Hoesly, R., Feng, L., 2021. CEDS V.2021_04_21 Gridded Emissions Data. <https://doi.org/10.25584/PNNLDATAHUB/1779095>.
- Paulot, F., Jacob, D.J., Henze, D.K., 2013. Sources and processes contributing to nitrogen deposition: an adjoint model analysis applied to biodiversity hotspots worldwide. *Environ. Sci. Technol.* 47 (7), 3226–3233. <https://doi.org/10.1021/es3027727>.
- Prashanth, P., Eastham, S.D., Speth, R.L., Barrett, S.R.H., 2022. Aerosol formation pathways from aviation emissions. *Environ. Res. Commun.* 4 (2), 021002. <https://doi.org/10.1088/2515-7620/ac5229>.

- Quadros, F.D.A., Snellen, M., Dedoussi, I.C., 2020. Regional sensitivities of air quality and human health impacts to aviation emissions. *Environ. Res. Lett.* 15 (10), 105013. <https://doi.org/10.1088/1748-9326/abb2c5>.
- Quadros, F.D.A., Snellen, M., Sun, J., Dedoussi, I.C., 2022. Global civil aviation emissions estimates for 2017–2020 using ADS-B data. *J. Aircr.*, 1–11 <https://doi.org/10.2514/1.C036763>.
- Quadros, F.D.A., Snellen, M., Dedoussi, I.C., 2022. Recent and projected trends in global civil aviation fleet average NOx emissions indices. *AIAA SCITECH 2022 Forum*; AIAA SciTech Forum; American Institute of Aeronautics and Astronautics: San Diego, CA & Virtual <https://doi.org/10.2514/6.2022-2051> p AIAA 2022-2051.
- Rose, A., McKee, J., Sims, K., Bright, E., Reith, A., Urban, M., 2020. LandScan Global 2019. <https://doi.org/10.48690/1524214>.
- Schulte-Uebbing, L.F., Beusen, A.H.W., Bouwman, A.F., de Vries, W., 2022. From planetary to regional boundaries for agricultural nitrogen pollution. *Nature* 610 (7932), 507–512. <https://doi.org/10.1038/s41586-022-05158-2>.
- Schumann, U., Huntrieser, H., 2007. The global lightning-induced nitrogen oxides source. *Atmos. Chem. Phys.* 7 (14), 3823–3907. <https://doi.org/10.5194/acp-7-3823-2007>.
- Simone, N.W., Stettler, M.E.J., Barrett, S.R.H., 2013. Rapid estimation of global civil aviation emissions with uncertainty quantification. *Transp. Res. Part D: Transp. Environ.* 25, 33–41. <https://doi.org/10.1016/j.trd.2013.07.001>.
- Skowron, A., Lee, D.S., De León, R.R., Lim, L.L., Owen, B., 2021. Greater fuel efficiency is potentially preferable to reducing NOx emissions for aviation's climate impacts. *Nat. Commun.* 12 (1), 564. <https://doi.org/10.1038/s41467-020-20771-3>.
- Stokstad, E., 2019. Nitrogen crisis threatens Dutch environment—and economy. *Science* 366 (6470), 1180–1181. <https://doi.org/10.1126/science.366.6470.1180>.
- Sun, J., Fu, J.S., Huang, K., 2016. Organic nitrates and other oxidized nitrogen compounds contribute significantly to the total nitrogen depositions in the United States. *Proc. Natl. Acad. Sci.* 113 (31), E4433–E4434. <https://doi.org/10.1073/pnas.1608717113>.
- Szopa, S., Naik, V., Adhikary, B., Artaxo, P., Bernsten, T., Collins, W.D., Fuzzi, S., Gallardo, A., Kiendler-Scharr, A., Klimont, Z., Liao, H., Unger, N., Zanis, P., 2021. *Short-Lived Climate Forcers. Climate Change 2021: The Physical Science Basis. Contribution of Working Group I to the Sixth Assessment Report of the Intergovernmental Panel on Climate Change*. Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA, pp. 817–922.
- The International GEOS-Chem User Community, 2021. Geoschem/GCClassic: GEOS-Chem 13.3.3. <https://doi.org/10.5281/zenodo.5748260>.
- U.S. Environmental Protection Agency, n.. CASTNET - download data. <https://java.epa.gov/castnet/clearsession.do>. (Accessed 12 July 2022).
- Weng, H., Lin, J., Martin, R., Millet, D.B., Jaeglé, L., Ridley, D., Keller, C., Li, C., Du, M., Meng, J., 2020. Global high-resolution emissions of soil NOx, sea salt aerosols, and biogenic volatile organic compounds. *Sci. Data* 7 (1), 148. <https://doi.org/10.1038/s41597-020-0488-5>.
- Woody, M., Haeng Baek, B., Adelman, Z., Omary, M., Fat Lam, Y., Jason West, J., Arunachalam, S., 2011. An assessment of aviation's contribution to current and future fine particulate matter in the United States. *Atmos. Environ.* 45 (20), 3424–3433. <https://doi.org/10.1016/j.atmosenv.2011.03.041>.
- Yim, S.H.L., Lee, G.L., Lee, I.H., Allroggen, F., Ashok, A., Caiazzo, F., Eastham, S.D., Malina, R., Barrett, S.R.H., 2015. Global, regional and local health impacts of civil aviation emissions. *Environ. Res. Lett.* 10 (3), 034001. <https://doi.org/10.1088/1748-9326/10/3/034001>.
- Zhang, L., Jacob, D.J., Knipping, E.M., Kumar, N., Munger, J.W., Carouge, C.C., van Donkelaar, A., Wang, Y.X., Chen, D., 2012. Nitrogen deposition to the United States: distribution, sources, and processes. *Atmos. Chem. Phys.* 12 (10), 4539–4554. <https://doi.org/10.5194/acp-12-4539-2012>.
- Zhao, Y., Zhang, L., Pan, Y., Wang, Y., Paulot, F., Henze, D.K., 2015. Atmospheric nitrogen deposition to the northwestern Pacific: seasonal variation and source attribution. *Atmos. Chem. Phys.* 15 (18), 10905–10924. <https://doi.org/10.5194/acp-15-10905-2015>.