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Impact of shipping emissions on air pollution and pollutant deposition over the Barents Sea

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Abstract

Arctic warming leading to reduced summertime sea-ice is likely to lead to increased local shipping especially along the Northeast Passage near the northern coasts of Norway and Russia, which are shorter than the traditional southerly routes. Here, the regional chemistry-transport model WRF-Chem is used to examine the effects of shipping emissions on levels of air pollutants and deposition fluxes over the Barents Sea both for present-day and future conditions, based on a high growth scenario. Present-day shipping emissions are found to have already substantial effects on ozone concentrations, but limited effects on sulphate and nitrate aerosols. Predicted future changes in ozone are also important, particularly in regions with low nitrogen oxide concentrations, and results are sensitive to the way in which diversion shipping is distributed due to non-linear effects on photochemical ozone production. Whilst modest future increases in sulphate and nitrate aerosols are predicted,

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large enhancements in dry deposition of sulphur dioxide and wet deposition of nitrogen compounds to the Barents Sea are predicted. Such levels of future nitrogen deposition would represent a significant atmospheric source of oceanic nitrogen affecting sensitive marine ecosystems.

Keywords: Future shipping diversion routes, Regional chemistry-transport model, Aerosol deposition, Barents Sea

1. Introduction

The Arctic region is undergoing unprecedented warming leading to reductions in summer sea-ice extent (Box et al., 2019; Pörtner et al., 2019). This is opening up the possibility for increased Arctic shipping notably along the Northeast Passage, extending from the north coast of Norway along the north coast of Russia. As sea-ice declines further shipping may also increase along the Northwest Passage traversing northern Canada and the north coast of Alaska or even across the Arctic Ocean (Melia et al., 2016; Stephenson et al., 2018). Projections underline large changes in the duration of the ship-accessible season across the Canadian Arctic (Mudryk et al., 2021). Arctic warming may also lead to increased industrial activities, such as oil and gas extraction, with associated port development, urbanisation and shipping (Dalsøren et al., 2007; AACA, 2017; Schmale et al., 2018). Such development will likely increase the footprint of local anthropogenic emissions, including shipping, relative to pollutants transported from midlatitudes (Marelle et al., 2018). Shipping emissions are known to emit nitrogen oxides (NO_x) and volatile organic compounds (VOCs) producing ozone (O_3) and emissions of precursor trace gases (sulphur dioxide SO_2 , NO_x , VOCs) produc-

19 ing secondary aerosols, notably sulphate (SO_4^{2-}), nitrate (NO_3^-) and ammo-
 20 nium (NH_4^+), while black carbon (BC) is emitted directly (Corbett et al.,
 21 2010; Winther et al., 2014). Studies examining the effects of present-day
 22 shipping found substantial contributions to O_3 and fine particulate matter
 23 ($\text{PM}_{2.5}$, aerodynamic diameter less than $2.5\mu\text{m}$) concentrations, exceeding
 24 background levels in and around shipping lanes, for example, along the Nor-
 25 wegian coast (Marelle et al., 2016), in the Baltic Sea (Jonson et al., 2015; Karl
 26 et al., 2019b) and along the Canadian Northwest Passage (Aliabadi et al.,
 27 2016; Gong et al., 2018) as well as enhanced aerosols from cruise shipping
 28 in Svalbard (Eckhardt et al., 2015) during the summer months. In com-
 29 mon with air pollutants from other sources, ozone and $\text{PM}_{2.5}$ from shipping
 30 emissions can be harmful to human health causing chronic (e.g. respiratory
 31 and cardiovascular) disease and premature death (Im et al., 2018) even at
 32 low concentrations. Deposition of highly soluble pollutants containing acidic
 33 sulphur and nitrogen compounds can be damaging to vegetation and wa-
 34 ter bodies (lakes, oceans) and marine ecosystems. Nitrogen deposition can
 35 also lead to eutrophication and may affect the marine nitrogen cycle since
 36 nitrogen is a limiting nutrient for oceanic net primary productivity (NPP)
 37 (Duce et al., 2008; Randelhoff et al., 2020). While a recent study suggests,
 38 that atmospheric deposition to the Arctic ocean is not important compared
 39 to other fluxes (e.g. from rivers, coastal erosion) (Terhaar et al., 2021), it
 40 remains poorly quantified and may lead to enhanced NPP (Mills et al., 2018).

41 In the future, shipping along ice free Arctic routes is likely to increase.
 42 In particular, increased diversion of shipping from longer southerly routes
 43 along the shorter Northeast Passage, may increase local shipping emissions

44 and increase air pollution, both locally and regionally (Granier et al., 2006;
45 Winther et al., 2014). Modelling studies estimated that critical loads could
46 be exceeded along the coast of Norway (Dalsøren et al., 2007) whereas over
47 the Baltic Sea nitrogen deposition is likely to be reduced following the intro-
48 duction of a Nitrogen Emission Control Area (NECA) in 2021 (Karl et al.,
49 2019a). Hassellöv et al. (2013) suggested that acidic pollutants emitted from
50 shipping may contribute to regional pH reductions of the same order of mag-
51 nitude as those due to carbon dioxide (CO_2) in regions with dense shipping
52 traffic.

53 In this study, we focus on a region located north of Norway and Russia,
54 centered over the Barents Sea, but also including parts of neighboring seas
55 (Norwegian and Kara) and their coastal surroundings. This region is tra-
56 versed by international shipping travelling from Europe to Russia and Asia
57 along the Northeast Passage as well as local (fishing, cruise, passenger) ship-
58 ping (Silber and Adams, 2019) and where increased future traffic is predicted
59 (Corbett et al., 2010; Winther et al., 2014). There is also a high rate of ma-
60 rine productivity with substantial fishing stocks in this region as well as large
61 natural resources, such as oil and gas (AACA, 2017). Here, we examine the
62 impact of present-day and future shipping on atmospheric composition and
63 deposition of trace gases and aerosols in the Barents Sea region with potential
64 implications for human health and local ecosystems (Smedsrud et al., 2013).
65 We used the regional WRF-Chem model (Section 2.1) run with present-day
66 emissions and a high growth shipping scenario (Sections 2.2 and 2.3) to ex-
67 amine the potential future shipping impacts. Present-day and future scenario
68 model results are discussed in terms of impacts on concentrations and depo-

sition (Section 3) before providing concluding remarks (Section 4).

2. Experiment setup - model simulations

2.1. Model setup

For this work we used the regional Weather Research and Forecasting model with Chemistry (WRF-Chem) (Fast et al., 2006; Grell et al., 2005). We use the version 3.5.1 including updates described in Marelle et al. (2017) which has also been used to study Arctic atmospheric composition and climate effects (Marelle et al., 2015, 2016; Raut et al., 2017; Marelle et al., 2018). Details about the WRF-Chem model setup including boundary layer physics, radiation and surface schemes are provided in Raut et al. (2017); Marelle et al. (2018). In particular, this model version was evaluated against atmospheric composition observations in Marelle et al. (2017) and used to investigate the contribution of remote and local sources of pollutants, including shipping emissions, to Arctic aerosols and their climate impact (Marelle et al., 2018).

In order to estimate the impact of Arctic shipping in the Barents Sea region, the model was run on a parent domain covering a large part of the Northern Hemisphere with a horizontal resolution of $100 \times 100 \text{ km}^2$ and, via a one-way nest, over a region covering the Barents Sea and part of the Kara Sea, north of Norway and Russia with $20 \times 20 \text{ km}^2$ horizontal resolution (Fig. 1). Fifty vertical levels were used in both domains. The model was run from March to August for the present-day (year 2012) and the future (year 2050). In each case, the first 3 months were considered as spin-up and were not used in the analysis.

[Figure 1 about here.]

2.2. Emissions

The model was run with the same emissions as Marelle et al. (2018) since, as noted above, it was thoroughly validated and improved in terms of simulating Arctic atmospheric composition (Marelle et al., 2017). This includes ECLIPSEv5 (Evaluating the Climate and Air-Quality Impacts of Short-Lived Pollutants, version 5) anthropogenic emissions (Stohl et al., 2015; Klimont et al., 2017) and hourly wild fire emissions from the NCAR Fire Inventory (FINNv1) (Wiedinmyer et al., 2011; Jiang et al., 2012). Biogenic emissions were calculated online using MEGAN (Model of Emissions of Gases and Aerosols from Nature) (Guenther et al., 2006).

Shipping emissions below 60°N were taken from the Representative Concentration Pathways (RCP8.5) dataset (van Vuuren et al., 2011) used in ECLIPSEv5b. At latitudes above 60°N, shipping emissions from the Arctic-wide Winther et al. (2014) inventory based on activity data from real-time satellite Automatic Identification System (AIS) ship positioning data for 2012 (Jalkanen et al., 2012) were used. Nunes et al. (2017) note that the use of AIS data for reporting activities and movements of ships is the best current approach. Winther et al. (2014) also included an evaluation of present-day and future evolution in engine efficiency and took into account new sulfur fuel regulations (Jonson et al., 2015). Emissions from Winther et al. (2014) are higher compared to previous Arctic shipping inventories such as Corbett et al. (2010). This is because, for example, this more recent inventory includes emissions from fishing ships which represent close to 40% of Arctic

117 shipping emissions [Marelle et al. \(2016\)](#), and because marine traffic was larger
 118 in 2012 than in the past. More recent shipping inventories are now available,
 119 including updated emissions in [Winther et al. \(2017\)](#) based on 2012-2016 AIS
 120 data, and ECLIPSEv6b based on STEAM3 emissions using 2015 AIS data,
 121 as described in [Johansson et al. \(2017\)](#). However, ECLIPSEv6b shipping
 122 emissions do not include future diversion shipping. [Winther et al. \(2017\)](#)
 123 state that Arctic shipping emissions are -7% for SO_2 , $+4\%$ for NO_x and
 124 -55% for BC in 2012 compared to [Winther et al. \(2014\)](#).

125 Model simulations for 2050 used [Winther et al. \(2014\)](#) future emission sce-
 126 narios taking into account growth projections in shipping traffic for a variety
 127 of ship types developed by [Corbett et al. \(2010\)](#) and with the addition of fish-
 128 ing emissions. They also include diversion shipping through the Northeast
 129 Passage. In the future, reductions in Arctic sea-ice are predicted to open up
 130 sea routes which are shorter than traditional routes linking North America or
 131 Europe with Asia. Here, 5% of the global total shipping traffic was assumed
 132 to be diverted through the Arctic following [Corbett et al. \(2010\)](#). [Marelle](#)
 133 [et al. \(2018\)](#) evaluated the effects of emission increases from local Arctic
 134 sources ($> 60^\circ\text{N}$) in summer 2012 and 2050. They showed that shipping
 135 emissions from [Winther et al. \(2014\)](#) increase very strongly in summer 2050
 136 (e.g. by 1400% for BC, 1500% for NO_x) due to diversion shipping through
 137 the Arctic Ocean. The decrease in Arctic shipping emissions caused by re-
 138 duced sulphur fuel content is more than compensated for by the projected
 139 increase in traffic, and leads to total Arctic shipping SO_2 emissions increasing
 140 by 1200% compared to present-day (2012). Future shipping scenarios remain
 141 highly uncertain. Here, we focus on examination of potential responses by

142 using a high growth scenario (HGS) which predicts large increases in Arctic
143 shipping traffic.

144 2.3. Simulations

145 To assess the effects of shipping emissions over the Barents Sea and nearby
146 maritime and coastal areas (nested domain in Fig. 1), four simulations were
147 performed: (i) BASE (base case simulation) using the present-day shipping
148 emissions for 2012; (ii) ZERO where present-day shipping emissions were
149 switched off to assess the present-day effect of shipping; (iii) HGS for 2050
150 using the high growth scenario detailed above and, (iv) HGS-WIDE where
151 diversion shipping in the Arctic was spread over a wider 60 km (3 model grid
152 box) shipping lane, instead of over 20 km (single model grid box) in HGS.
153 The HGS-WIDE scenario explores the sensitivity of model results to dilution
154 of future shipping emissions over a wider area. The coverage of future Arctic
155 shipping lanes is highly uncertain but likely to cover a wider geographic area
156 than considered in this study (Melia et al., 2016).

157 In order to investigate the impacts of present-day shipping in the Barents
158 Sea region, we compared the BASE and ZERO runs. The difference between
159 HGS or HGS-WIDE scenarios and the BASE run was used to examine the
160 impact of future shipping on O_3 , NO_3^- , SO_4^{2-} concentrations and nitrogen
161 (N) and sulphur (S) deposition.

162 The model version used in this study has already been evaluated against
163 aerosol and ozone observations in the Arctic during summer showing rea-
164 sonable performance (Raut et al., 2017; Marelle et al., 2017; Law et al.,
165 2017). In the Supplementary Material, results from the nested domain
166 are evaluated against observed concentrations of SO_4^{2-} , NO_3^- and O_3 and

167 wet and dry deposition fluxes from the European Monitoring and Evalua-
 168 tion Programme (EMEP) network over Europe obtained from EBAS (<http://ebas.nilu.no/>)
 169 at different sites Tustervatn, Mount Zeppelin, Birkenes,
 170 Pinega, Janiskoski, Pallas, Oulanka, Esrange (Fig. [.1](#)) in summer 2012. Time
 171 series and correlation plots between observations and model results for all
 172 selected stations illustrate reasonable model performance (Supplementary
 173 Material).

174 3. Results and discussion

175 3.1. Impacts of shipping on air pollutants in the Barents Sea region

176 3.1.1. Present-day impacts on pollutant concentrations

177 The monthly mean mixing ratios of O_3 and NO_x for summer 2012 are
 178 shown in Fig. [.2](#).

179 [Figure 2 about here.]

180 Figure [.2](#) shows monthly mean surface O_3 concentrations for summer 2012
 181 of around ~ 20 ppbv to 30 ppbv in the Barents Sea region. Lower values in the
 182 north of the domain are due to lower NO_x concentrations (< 0.1 ppbv) and
 183 efficient removal of O_3 by $O(^1D) + H_2O$ in summer, as well as dry deposition
 184 to the ocean. Figure [.2](#) also shows large contributions from present-day
 185 shipping of up to 9 ppbv O_3 locally, in particular where NO_x concentrations
 186 are low, north of $70^\circ N$. Lower O_3 enhancements are found in shipping lanes
 187 along the coast of Norway or between northern Norway and Svalbard where
 188 half the NO_x is from shipping emissions and reaches 1 ppbv locally. These
 189 results suggest that Arctic shipping is already a significant source of O_3

190 during summer when both shipping emissions and photochemistry are the
 191 highest. The enhancements predicted in this study are larger than those
 192 reported by [Ødemark et al. \(2012\)](#) (2 – 3 ppbv) or [Aksoyoglu et al. \(2016\)](#)
 193 ($< 3\%$) using lower shipping NO_x emissions than [Winther et al. \(2014\)](#), but
 194 are consistent with results from [Marelle et al. \(2018\)](#), suggesting that 15 to
 195 25% of O_3 is due to Arctic shipping activities over the Norwegian, Barents
 196 and Kara Seas (Fig. [.1](#)).

197 [Figure 3 about here.]

198 Figure [.3](#) shows monthly mean surface summer aerosol concentrations and
 199 the impact of present-day shipping emissions. Along the Northeast Passage,
 200 average SO_4^{2-} concentrations are $\sim 0.4\mu\text{g m}^{-3}$ with 2 – 10% ($\sim 0.04\mu\text{g m}^{-3}$)
 201 from present-day shipping activities spread over a larger area than the ship-
 202 ping tracks. Over land, surface SO_4^{2-} concentrations are also affected by ship-
 203 ping emissions over northern Scandinavia and Russia. The impact of shipping
 204 on surface NO_3^- concentrations is larger, ranging from $0.08\text{--}0.15\mu\text{g m}^{-3}$ in the
 205 western Barents Sea and reaching $0.2\mu\text{g m}^{-3}$ along the west coast of Norway
 206 (Fig. [.3](#)). This represents a $\sim 20\%$ enhancement in NO_3^- due to present-day
 207 shipping. Smaller contributions from shipping are found over land, of simi-
 208 lar magnitude to SO_4^{2-} ($\sim 0.04\mu\text{g m}^{-3}$). These results are comparable, but
 209 slightly larger, than previous studies. [Jonson et al. \(2015\)](#) estimated increases
 210 of the order of 2 – 5% in SO_4^{2-} due to shipping in the Southern Norwegian
 211 coast, an area comparable to this study. [Karl et al. \(2019b\)](#) estimated an av-
 212 erage contribution from shipping to $\text{PM}_{2.5}$ (including SO_4^{2-} , NO_3^-) in coastal
 213 land areas around the Baltic Sea in the range 3.1 – 5.7% using three different

214 chemistry transport models. Differences between studies may be ascribed to
 215 different regions, some where sulphur and nitrogen emission reduction regu-
 216 lations already apply, but also to differences in model aerosol treatments, in
 217 particular with respect to inorganic aerosol formation. Our results suggest
 218 that shipping activities could already be having an influence on levels of air
 219 pollutants over the Barents Sea region.

220 3.1.2. Future impacts on pollutant concentrations

221 Figure [.4](#) shows the contribution of future shipping emissions on O_3 , NO_3^-
 222 and SO_4^{2-} concentrations in summer 2050. Outside the main shipping lane
 223 (Northeast Passage), a large impact on O_3 is found in particular due to the
 224 inclusion of diversion shipping in the future scenario (HGS and HGS-WIDE).
 225 Enhancements in O_3 mixing ratios up to 5.5 ppbv are predicted in low NO_x
 226 regions, especially in the Kara Sea (Fig. [.4](#)). Along the diversion route, O_3
 227 may increase or decrease depending on the local chemistry regime (low- NO_x
 228 or high- NO_x , as already noted for the present-day). Where NO_x emissions are
 229 large, O_3 titration by the reaction with NO occurs along the main shipping
 230 lane leading to lower predicted future O_3 . Notable differences are found
 231 depending on how the diversion route is included in the future simulations.
 232 The effect of widening the diversion route (HGS-WIDE) is twofold and highly
 233 non-linear. First, the region where O_3 titration occurs north of Norway is
 234 spread over a wider area but decreases in O_3 are lower (1 ppbv in HGS-WIDE
 235 compared to 5 ppbv in HGS). Second, O_3 production is increased in low NO_x
 236 regions in the Kara Sea due to the introduction of diversion shipping NO_x
 237 over a wider area in HGS-WIDE. Finally, although local shipping emissions
 238 have a local titration effect, diversion emissions as a whole may increase O_3

239 levels over most of the Arctic since, due to the lifetime of O_3 (several days)
 240 in summer, O_3 produced from shipping emissions can be transported away
 241 from the shipping lanes. These results highlight the sensitivity of Arctic O_3
 242 to the way in which diversion shipping, in particular, is introduced in models.

243 [Figure 4 about here.]

244 Figure .4 also shows differences in NO_3^- and SO_4^{2-} concentrations between
 245 the HGS scenario and the BASE simulation. Increased shipping emissions,
 246 largely due to traffic diversion, are responsible for moderate increases in
 247 SO_4^{2-} concentrations ($\sim +37\%$) relative to the present-day ($0.4\mu\text{g m}^{-3}$ in the
 248 coastal areas (Fig. .3) and up to $0.15\mu\text{g m}^{-3}$ over the whole domain. This
 249 is because HGS SO_2 emissions do not increase by much due to implemented
 250 sulphur emission mitigation. In contrast, large increases (by around 100%),
 251 in the range $0.5 - 0.7\mu\text{g m}^{-3}$, are predicted for NO_3^- aerosols in the region of
 252 the diversion route. As a consequence, future shipping emissions may lead,
 253 on average, to a doubling of NO_3^- concentrations over the Barents Sea region.

254 3.2. Impacts of shipping on pollutant deposition in the Barents Sea region

255 3.2.1. Present-day impacts on wet and dry deposition

256 Figure .5 shows total (wet and dry) N and S deposition from the BASE
 257 simulation and the absolute differences between BASE simulation and the
 258 ZERO run with emissions switched off for July and August. Here, total N
 259 deposition includes wet and dry deposition of NO_3^- , NH_4^+ aerosols, nitric acid
 260 (HNO_3), ammonia (NH_3) and minor gaseous N species. Total S deposition
 261 includes wet deposition of SO_4^{2-} aerosols, sulphuric acid (H_2SO_4) and dry
 262 deposition of SO_2 . The model predicts that most of the present-day total N

263 deposition is over land ($0.6 - 1.2 \text{ mgN/m}^2/\text{day}$). The amount of N deposited
 264 over the ocean is smaller, with an average value of $0.3 \text{ mgN/m}^2/\text{day}$ and a
 265 maximum of $0.7 \text{ mgN/m}^2/\text{day}$ over the Barents Sea. The contribution from
 266 wet deposition fluxes due to present-day shipping presents a spotty distribu-
 267 tion because it closely follows modelled precipitation patterns. The largest
 268 values are found over the western part of the Barents Sea ($0.03 \text{ mgN/m}^2/\text{day}$
 269 on average, +10% contribution) and reaching $0.05 \text{ mgN/m}^2/\text{day}$ locally.
 270 Dry deposition fluxes due to shipping have a smoother distribution, with
 271 values of $\sim 0.01 \text{ mgN/m}^2/\text{day}$ over the Barents Sea (+3% contribution).
 272 Higher contributions are estimated along the Norwegian coast ($\sim 0.02 - 0.03$
 273 $\text{mgN/m}^2/\text{day}$, +10%) where NO_x emissions are at maximum. These results
 274 suggest that shipping already plays a role in the deposition of nitrogen.

275 In the BASE simulation, total S deposition in the domain is quite low (0.3
 276 $\text{mgS/m}^2/\text{day}$), with nevertheless higher values along the northern coast of
 277 Russia and the Timan-Pechora Basin in northern Siberia (Fig. [1](#)) ($0.5 - 0.7$
 278 $\text{mgS/m}^2/\text{day}$). Specific spots over land are due to high local SO_2 emissions
 279 in Russia from metal smelters (up to $2 \text{ mgS/m}^2/\text{day}$). Shipping has an
 280 important effect on dry deposition of pollutants as they emit close to the
 281 surface and in the boundary layer, whereas wet deposition is highly non-
 282 linear and also sensitive to pollution aloft. Wet deposition of S due to ship-
 283 ping can only be detected along the western coast of Norway ($0.02 - 0.04$
 284 $\text{mgS/m}^2/\text{day}$, +10% contribution). In contrast, dry S deposition due to ship-
 285 ping has considerable influence along the Norwegian coast contributing on
 286 average $\sim 0.1 \text{ mgS/m}^2/\text{day}$ ($\sim +150\%$) and, to a lesser extent, along shipping
 287 lanes between northern Norway and the Svalbard archipelago ($0.03 - 0.04$

288 $\text{mgS}/\text{m}^2/\text{day}$, +10%).

289 [Figure 5 about here.]

290 3.2.2. Future impacts on pollutant deposition

291 Figure .6 shows the absolute differences in July-August average dry and
292 wet deposition of N and S between the future scenario (HGS) and the BASE
293 run. The influence of diversion shipping can clearly be seen. An interesting
294 result of this study is that dry and wet N deposition contribute almost equally
295 to absolute enhancements of future N deposition ($\sim +10$ Gg over the ocean
296 in July-August). Values as high as $0.13 - 0.2 \text{ mgN}/\text{m}^2/\text{day}$ (+25 – 50%)
297 are estimated for dry N deposition in the proximity of the shipping lanes,
298 but are spread over a larger area, reaching land, and expanding toward the
299 Timan-Pechora Basin and the west Siberian Plain covered by tundra. The
300 distribution of wet N deposition is even more spread out affecting almost
301 similarly ($0.15 \text{ mgN}/\text{m}^2/\text{day}$, +50%) most of the Nordic waters (Norwegian,
302 Barents and Kara Seas) but also the coastal areas of northern Russia and
303 to a lesser extent northern Norway. Over the ocean, we estimate the total
304 amount of N deposited in July-August 2050 to be ~ 56.5 Gg N from all
305 sources. This represents an increase of +22% in deposition due to future
306 shipping compared to present-day. Extrapolation of our estimate to ice-
307 free regions over the entire Arctic Ocean (May to September) would suggest
308 a much larger potential contribution than previous estimates such as that
309 from Lamarque et al. (2013) who estimated annual total future atmospheric
310 N deposition from all sources of ~ 100 Gg/year over the ice-free Arctic Ocean.
311 A recent study by Terhaar et al. (2021) concluded, based on the results of

312 [Lamarque et al. \(2013\)](#), that present-day atmospheric N deposition is not
313 important compared to other sources like riverine import (~ 1 Tg/year) and
314 coastal erosion (~ 1.6 Tg/year). In contrast, our results imply that the
315 future deposition of atmospheric N due to increased shipping emissions in
316 the Arctic may have a substantial effect ecological impact, especially since
317 the shipping lanes coincide with high primary productivity in this nitrogen
318 poor marine ecosystem ([Baker et al., 2017](#); [Tuerena et al., 2021](#)).

319 Future emissions also lead to large increases in dry S deposition fluxes
320 (~ 1 mgS/m²/day) in the vicinity of the shipping lanes. This corresponds to
321 an enhancement of +114% of total S deposited over the ocean compared to
322 present-day. Over land, substantial increases in dry S deposition fluxes only
323 occur in hotspots associated with higher future SO₂ emissions due to the en-
324 ergy and industrial sectors in the ECLIPSE inventory. Enhancements in wet
325 S deposition due to future shipping show similar spatial distributions as wet
326 N deposition since they are also linked to simulated clouds and precipitation,
327 but of slightly smaller magnitude (~ 0.1 mgS/m²/day). Over the ocean, the
328 total amount of S deposited in July-August 2050 is estimated to be ~ 43 Gg,
329 representing an increase of +37% compared to present-day deposition.

330 Ozone deposition is also enhanced in the south of the Novaya Zemlya
331 archipelago, along the coastal region of the west Siberian Plain with values
332 reaching 3 mg/m²/day (+15%, not shown). Future shipping emissions con-
333 tribute to 21% (5.8 Gg N/month) and 34% (7.3 Gg S/month) to the total
334 amount of N and S deposited over the Ocean in summer 2050. Without a
335 plan to define the Barents Sea region and other Arctic seas as nitrogen or
336 sulphur Emission Control Areas (NECAs/SECAs) in the foreseeable future,

337 the deposition fluxes of N and S due to shipping will greatly increase in the
338 warming Arctic.

339 [Figure 6 about here.]

340 4. Conclusions

341 The results of our study suggest that Arctic shipping is already affect-
342 ing atmospheric composition in the Barents Sea region, and in particular
343 ozone and nitrate aerosol concentrations. Using a high growth scenario for
344 future shipping which also includes diversion shipping, substantial increases
345 in ozone are predicted, especially in areas where NO_x concentrations are low.
346 Results are very sensitive to the way in which diversion shipping is included
347 in the model and points to the need for more accurate determination of pos-
348 sible routes along the Northeast Passage and together with high resolution
349 modelling. Increases in ozone and aerosol concentrations, in particular along
350 coastal areas, has implications for background levels of these air pollutants
351 which are damaging to human health. We also predict large total nitrogen
352 deposition over the Barents Sea region due to future shipping based on the
353 high growth scenario. This may be an upper estimate but is nevertheless
354 higher than one prior estimate and suggests that atmospheric nitrogen input
355 to the Arctic Ocean may increase considerably in the future. It may already
356 be important, in contrast to what has already been published. This has im-
357 plications for marine primary productivity, in particular in the Barents Sea
358 which is a nitrogen-poor in terms of oceanic nutrients. We also predict signif-
359 icant dry sulphur deposition in the vicinity of shipping lanes. Such increases

360 in deposition of acidic pollutants from shipping emissions may have the po-
361 tential to increase ocean acidity locally. Deposition of nitrogen and sulphur
362 over coastal areas (forest, tundra) may also affect land-based ecosystems.
363 Finally, ozone deposition is also enhanced along the eastern coasts of the
364 Barents Sea and inland. These findings should be considered in discussions
365 about possible implementation of regulations to limit shipping emissions in
366 the fragile Arctic environment.

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373 Dynamique).

374 **Competing interests**

375 The authors declare that they have no conflict of interest.

376 **Author statement**

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651		(in ppbv) (a), nitrate aerosol concentrations (in $\mu g m^{-3}$) (c)	
652		and sulphate aerosol concentrations (in $\mu g m^{-3}$) (d) between	
653		the HGS scenario and the BASE run. Panel (b) shows the	
654		absolute difference in monthly mean surface O_3 mixing ratios	
655		between the HGS-WIDE scenario and the BASE run.	33
656	.5	Monthly mean deposition fluxes of total nitrogen (in $mgN/m^2/day$)	
657		(a) and total sulphur ($mgS/m^2/day$) (b) simulated for July-	
658		August 2012 and the contribution of present-day shipping to	
659		wet (c,d) and dry (e,f) deposition fluxes of total nitrogen (c,e)	
660		and total sulphur (d,f) from differences between simulations	
661		with (BASE) and without (ZERO) shipping emissions.	34
662	.6	Absolute differences in monthly mean dry (a,c) and wet (b,d)	
663		deposition fluxes of total nitrogen (in $mgN/m^2/day$) (a,b) and	
664		total sulphur (in $mgS/m^2/day$) (c,d) between the HGS sce-	
665		nario and the BASE run.	35

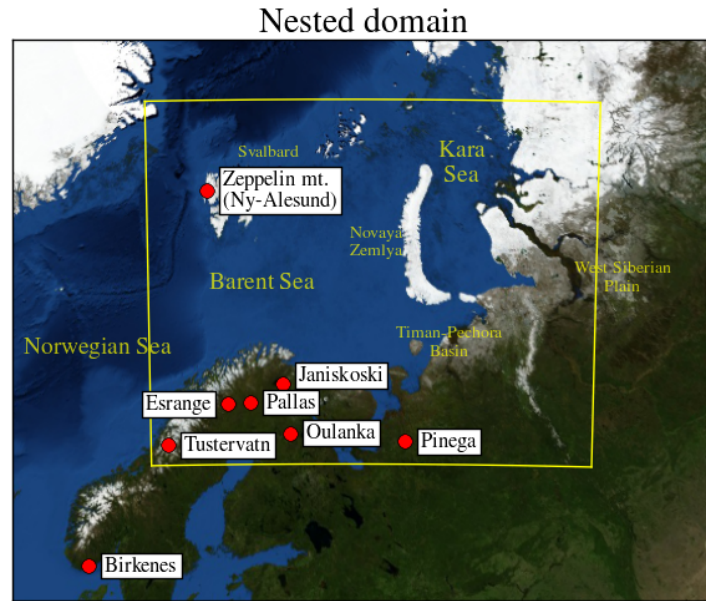


Figure .1: Domains used in the WRF-Chem simulations. The large domain covers most of the Northern Hemisphere in a polar stereographic grid, and the nested domain is centered on the Barents Sea region. Stations used for model evaluation, as well as seas and land regions referred to in the text are also reported on the map.

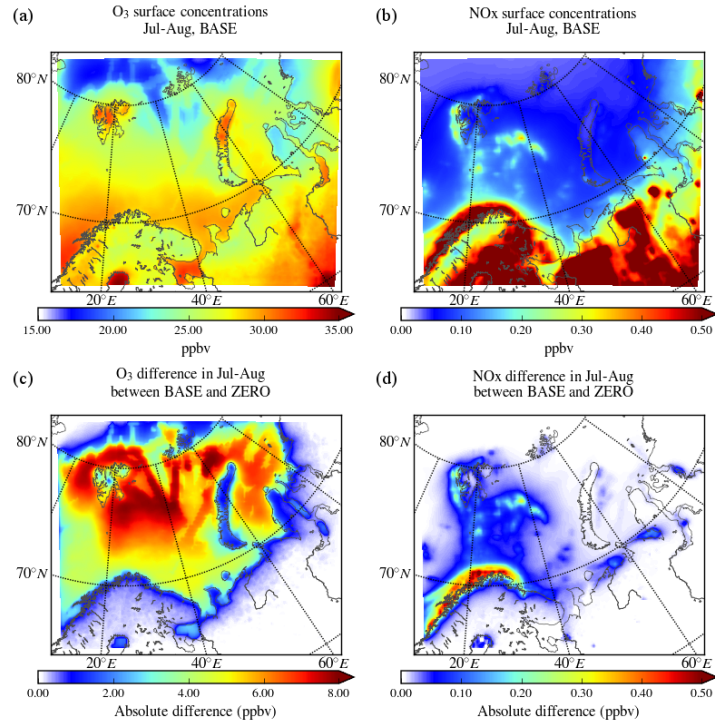


Figure 2: Monthly mean O₃ (a) and NO_x (b) mixing ratios simulated for July-August 2012 (BASE run) and the contribution of present-day shipping (BASE - ZERO) on O₃ (c) and NO_x (d) mixing ratios (in ppbv).

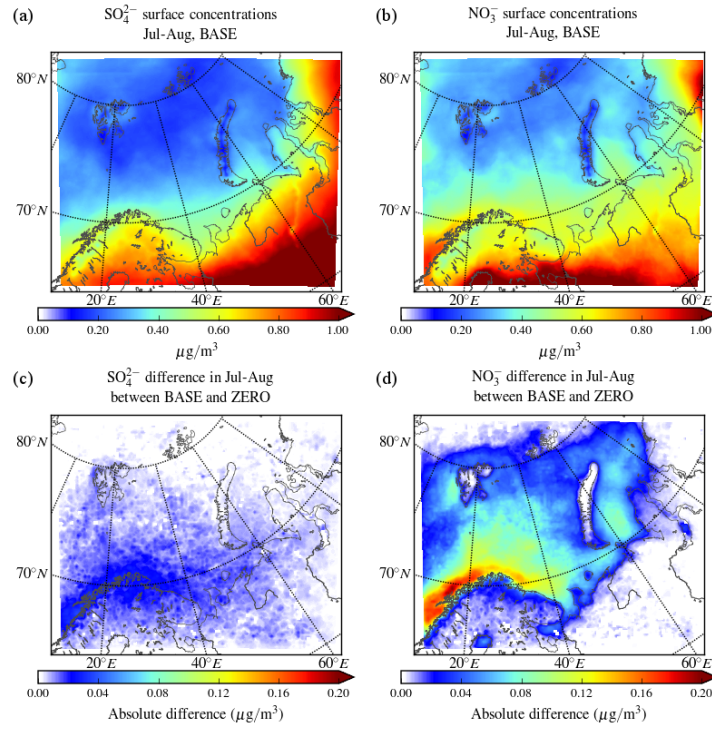


Figure .3: Monthly mean sulphate (a) and nitrate (b) aerosol concentrations predicted by the model for July-August 2012 (BASE run) and absolute differences in present-day sulphate (b) and nitrate (d) concentrations between simulations with (BASE) and without (ZERO) shipping emissions (in $\mu\text{g m}^{-3}$).

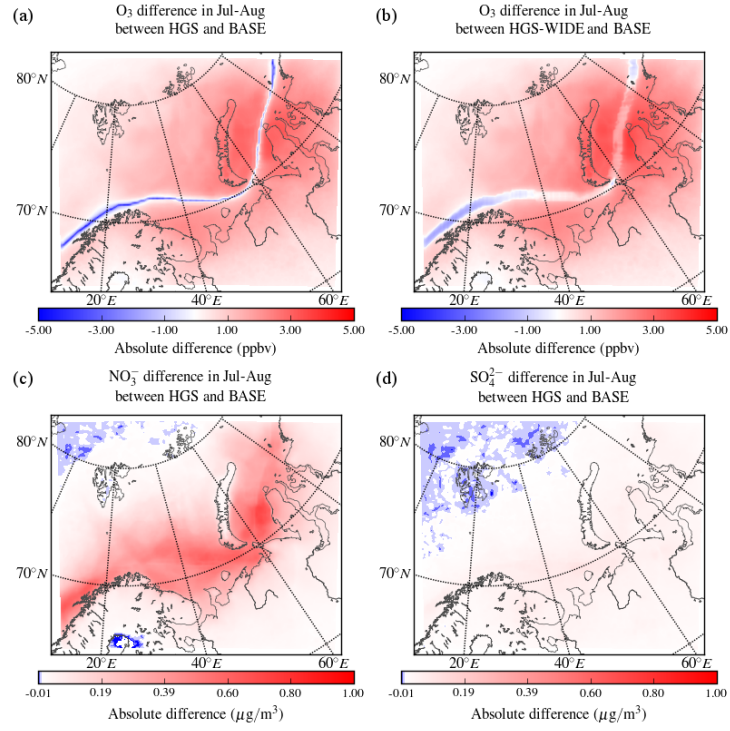


Figure 4: Absolute differences in monthly mean surface O₃ mixing ratios (in ppbv) (a), nitrate aerosol concentrations (in $\mu\text{g m}^{-3}$) (c) and sulphate aerosol concentrations (in $\mu\text{g m}^{-3}$) (d) between the HGS scenario and the BASE run. Panel (b) shows the absolute difference in monthly mean surface O₃ mixing ratios between the HGS-WIDE scenario and the BASE run.

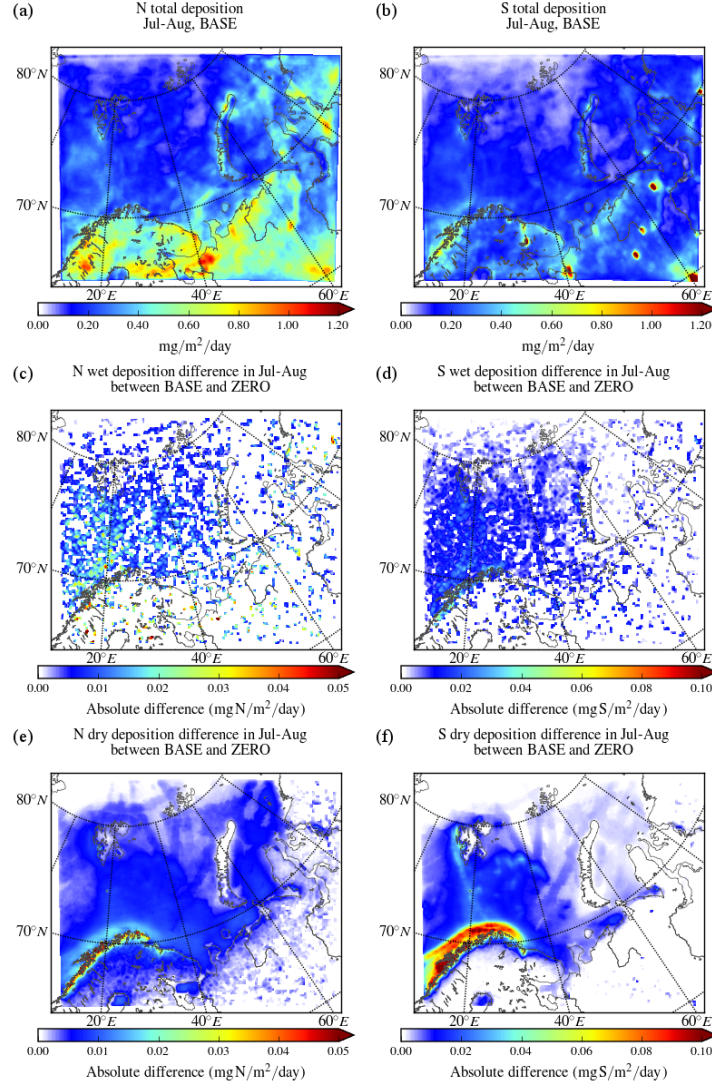


Figure .5: Monthly mean deposition fluxes of total nitrogen (in $\text{mgN/m}^2/\text{day}$) (a) and total sulphur ($\text{mgS/m}^2/\text{day}$) (b) simulated for July-August 2012 and the contribution of present-day shipping to wet (c,d) and dry (e,f) deposition fluxes of total nitrogen (c,e) and total sulphur (d,f) from differences between simulations with (BASE) and without (ZERO) shipping emissions.

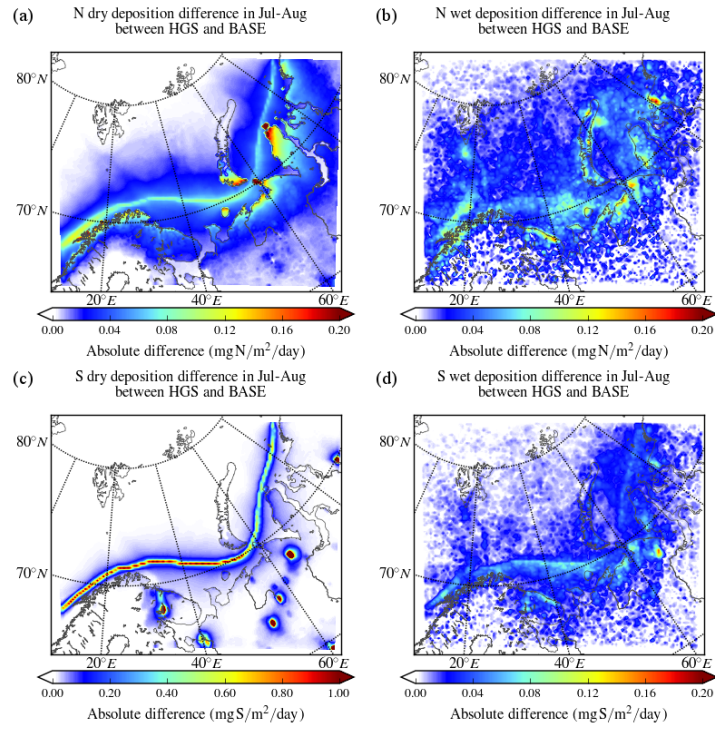


Figure .6: Absolute differences in monthly mean dry (a,c) and wet (b,d) deposition fluxes of total nitrogen (in $\text{mgN/m}^2/\text{day}$) (a,b) and total sulphur (in $\text{mgS/m}^2/\text{day}$) (c,d) between the HGS scenario and the BASE run.