Impacts of maritime shipping on air pollution along the U.S. East Coast

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Abstract.

Air pollution is considered a leading threat to human health in the U.S. and worldwide. An important source of air pollution in coastal areas is the globally increasing maritime shipping traffic. In this study, we take a high-resolution modeling approach to investigate the impacts of ship emissions on concentrations of various atmospheric pollutants, under the meteorological conditions and emissions of the year 2018. We utilize the Comprehensive Air Quality Model with extensions (CAMx) to simulate transport, diffusion, and chemical reactions, and the Weather Research and Forecasting (WRF) model to provide the meteorological inputs. We focus on four criteria pollutants – fine particulate matter with a diameter smaller than 2.5 μm ($PM_{2.5}$), nitrogen dioxide (NO_2), sulfur dioxide (SO_2), and ozone (O_3) – as well as nitrogen oxide (NO), and we calculate their concentrations in the presence and absence of the ship emissions along the U.S. East Coast, particularly in the proximity of major ports.

We find that ship emissions increase the $PM_{2.5}$ concentrations over the ocean and sparse areas inland. The 98-th percentile of the 24-hour average $PM_{2.5}$ concentrations (the "design value" used by the U.S. Environmental Protection Agency) increased by up to $3.2\,\mu\mathrm{g\,m^{-3}}$ in some coastal areas. In addition, ships contribute significantly to SO_2 concentrations, up to 95% over the Atlantic and up to 90% over land in coastal states, which represents a ~45 ppb increase in the SO_2 design values in some states. The 98-th percentile of the hourly NO_2 concentrations also increased by up to 15 ppb at the major ports and along the shore. In addition, we find that the impact of shipping emissions on O_3 concentrations is not uniform, meaning that ships affect ozone pollution in both positive and negative ways. Over the ocean, O_3 concentrations were significantly higher in the presence of ships, but in major coastal cities O_3 concentrations decreased in the presence of ships. Our simulation results show that ships emit significant amounts of fresh NO in the atmosphere, which then helps scavenge O_3 in VOC-limited areas, such as major ports. By contrast, over the ocean (NO_x-limited regime), enhanced NO_x concentrations due to ships contribute to the formation of O_3 and therefore enhance O_3 concentrations. Overall, due to the dominant southwesterly wind direction in the region, the impacts of ships on air pollutants mainly remain offshore. However, in coastal states near major ports, the impacts are significantly important.

1 Introduction

25 Globally, it is estimated that, in 2019, ambient air pollution, particularly particulate matter (PM) and ozone (O_3), was responsible for 4.5 million premature deaths worldwide (Fuller et al., 2022). This ranked air pollution as a leading risk factor in the Global Burden of Disease Study by the Institute for Health Metrics and Evaluation in 2019 (Murray and Lopez, 1996). Meanwhile, ship traffic is globally increasing and is becoming an important source of air pollution, especially in coastal areas (Corbett and Fischbeck, 1997; Eyring et al., 2010; Schnurr and Walker, 2019). Sea transport accounts for 80% of goods transported

30 worldwide (Schnurr and Walker, 2019), while recent studies estimate demand growth of almost 40% for seaborne trade by 2050 (Serra and Fancello, 2020). Marine vessels are important sources of air pollutants, emitting sulfur oxides (SO_x), nitrogen oxides ($NO_x = NO + NO_2$), particulate matter (PM), carbon monoxide (CO), volatile organic compounds (VOC), and carbon dioxide (CO_2) (Corbett et al., 2009; Eyring et al., 2007; Eyring, 2008; Smith et al., 2015). Low-grade marine fuel oil contains 3,500 times more sulfur than road diesel (Wan et al., 2016). Studies report fuel consumption of ocean-going ships between 200-290

35 million metric tons for the year 2000 (Corbett and Köhler, 2003; Endresen et al., 2007). Ships are responsible for about 15% of all global anthropogenic NO_x emissions and 4–9% of sulfur dioxide SO_2 emissions. In addition, oceangoing ships are estimated to emit 1.2–1.6 million metric tons (Tg) of PM annually (Corbett et al., 2009; Eyring et al., 2010; Viana et al., 2014).

About 70% of ship emissions occur within 400 km of the shore (Endresen et al., 1999, 2003). Thus, ships can be a major source of pollution in coastal areas and can impact human health. For instance, ship emissions in East Asia have caused

40 14,500–37,500 premature deaths in 2013, the amount of which had doubled since 2005 (Liu et al., 2016). Similarly, particulates emitted from ships cause 60,000 cardiopulmonary and lung cancer deaths each year worldwide (Corbett et al., 2009). Studies from different parts of the world like China show that shipping emissions increased the annual averaged $PM_{2.5}$ concentrations in the eastern coastal regions up to $5.2\,\mu g\,m^{-3}$, which was carried 900 km inland (Lv et al., 2018). In Europe, although the increase in $PM_{2.5}$ concentrations by ships is found to be small, their relative contribution is large because of the low background

45 $PM_{2.5}$ concentrations (Viana et al., 2009; Aksoyoglu et al., 2016).

Fine particulate matter ($PM_{2.5}$) is a harmful air pollutant that consists of microscopic particles with a diameter smaller than $2.5\,\mu m$. These particles can penetrate human lungs and even the bloodstream and cause serious health problems (U.S. Environmental Protection Agency (EPA), 2020b). NO_x are a group of highly reactive gases; although seven compounds are technically part of the NO_x family (NO, NO_2, nitrous oxide N_2O, dinitrogen dioxide N_2O_2, dinitrogen trioxide N_2O_3, dinitrogen

50 tetroxide N_2O_4, dinitrogen pentoxide N_2O_5), the most abundant are NO and NO_2, but only NO_2 is actually regulated in the U.S. NO_2 is harmful to humans by irritating the human respiratory system and to the environment by creating acid rain (U.S. Environmental Protection Agency (EPA), 2020a; Lin and McElroy, 2011); it is also a precursor to tropospheric ozone (O_3) formation, which has further negative impacts on human health (, EPA). Similarly, short-term exposure to SO_2 can harm the human respiratory system. These four pollutants – $PM_{2.5}$, SO_2, NO_2, and O_3 – are both primary (i.e., they can be directly emitted

55 into the atmosphere) and secondary (i.e., they can also form after chemical reactions in the atmosphere) pollutants. Here, we will focus on these four pollutants which are among the seven "criteria" pollutants that are regulated at the federal level by the

U.S. Environmental Protection Agency (EPA) via the National Ambient Air Quality Standards (NAAQS)(U.S. Environmental Protection Agency (EPA), 2022b).

Due to the complex nature of the atmosphere and its processes, such as chemical reactions, transport, and diffusion, high

60 concentrations of these pollutants are not necessarily found where their emissions are highest. Therefore, although the ship emissions are released in marine environments, the atmospheric conditions can play an essential role in transporting those pollutants, some of which are precursors for the formation of secondary EPA-regulated pollutants, like O_3.

Here, we explore the impacts of ocean-going ship emissions on the air quality along the U.S. East Coast by utilizing the Comprehensive Air Quality Model with extensions (CAMx) for our simulations. For the first time, we use the most recent high

65 spatial (4 km) and temporal (hourly) resolution ship emission data from the EPA's National Emission Inventory (NEI). We also include the ship stack height to consider at what vertical layer the emissions are emitted into the atmosphere, to be able to account for stability and atmospheric impacts on the pollutants. We investigate the pollution concentrations in a control scenario based on the shipping activities in the year 2018. Then, we conduct another simulation for a hypothetical condition where we eliminate the ship emissions altogether while keeping everything else the same. The difference between the two scenarios gives

70 insights into the net contribution of the ships to air pollution.

Ozone pollution is one of the main focuses of this study. The rate of ozone production can be limited by the concentration of either VOC or NO_x and depends on the relative sources of hydroxyl radical (OH) and NO_x (Finlayson-Pitts and Pitts Jr, 1993). When the rate of OH production is greater than the rate of NO_x production, the rate of ozone production is NO_x-limited. In this situation, ozone concentrations are sensitive to NO_x emissions rather than VOC concentrations. In contrast, when the

75 rate of OH production is less than the rate of NO_x production, ozone production is VOC-limited. In this case, ozone is most effectively reduced by lowering VOC concentrations. NOx is generally higher where human mobility and transportation are higher (Archer et al., 2020) and While O_3 is generally NO_x-limited in rural areas and downwind suburban areas, in urban areas with high population and high traffic emissions O_3 is often VOC-limited (Seinfeld and Pandis, 1998). Motor vehicles are among the major sources of ozone pollution in the region through their NO_x and VOC emissions (Niemeier et al., 2006; Yao et al.,

80 2015; Zhang et al., 2014). However, the impact of ocean ship emissions along the East Coast of the United States is lacking in the literature and we fill this gap in this study. In locations that exceed the EPA ozone standards by only 2-3 ppb, like the small state of Delaware (Moghani et al., 2018), the ship contribution could be of even higher importance.

2   Methods

2.1   Setup of the WRF-CAMx modeling system

85 We take a modeling approach to explore the pollution concentration across the study domain. The models used in this study are the Weather Research and Forecasting (WRF) model, version 4.3, and the Comprehensive Air quality Model with extensions (CAMx) version 7.1 with the Carbon Bond version 6 revision 5 (CB6r5) chemical mechanism. WRF is developed at the National Center for Atmospheric Research (NCAR) (Skamarock et al., 2019) and is one of the most widely used numerical weather prediction models. CAMx is a modular, Eulerian, 3-dimensional photochemical air quality model (Ramboll Environment

90   and Health, 2020) that simulates the emission, production, advection, diffusion, chemical transformation, and removal of atmospheric pollutants at regional scales and is among the few that are recommended by the EPA for regulatory purposes (U.S. Environmental Protection Agency (EPA), 2022a). We use the WRF-CAMx modeling system to conduct simulations of two separate scenarios, based on the exact same setup and inputs: the first scenario includes the ship emissions (WithShips) while in the second hypothetical scenario we remove the ship emissions altogether (NoShips). The difference in pollution levels between

95   the two cases provides the net contribution of ship emissions to regional air quality.

CAMx requires input data to characterize meteorology and chemistry, initial and boundary conditions for all the modeling domains, and other environmental conditions such as the photolysis rates. Meteorology is an essential factor in the formation of many secondary pollutants, both directly and indirectly. Atmospheric stability plays a significant role in determining pollutant faith (Arya et al., 1999). In CAMx, the plume rise calculations for point sources including the CMV emissions depend on

100   meteorological conditions and atmospheric stability to determine what vertical layer the emissions are emitted in. In the summertime, various atmospheric stabilities have been found to be dominant over the Atlantic ocean depending on different locations (Golbazi et al., 2022; Golbazi and Archer, 2019; Archer et al., 2016). We use the WRF model to provide meteorological inputs to CAMx. The publicly available WRF-CAMx data processing program (Ramboll Environment and Health, 2020) is used to generate CAMx meteorological input files from WRF output files. Details on the WRF model setup are provided in Table 1. Our

105   period of study is the summer of 2018, selected to reflect the most recent emission inventory available (discussed next in Section 2.2). Photolysis rate inputs to CAMx were calculated using the Tropospheric Ultraviolet and Visible (TUV) radiative transfer and photolysis model (https://www2.acom.ucar.edu/modeling/tropospheric-ultraviolet-and-visible-tuv-radiation-model).

Table 1. Details of the WRF-CAMx model setup.

Simulation period	1 June – 30 August, 2018
Horizontal grid resolution	4 km
Vertical layers	35
Spin-up time	48 hours
WRF version 4.3	
Initial/boundary conditions	NAM reanalysis, 6-hourly, 12-km resolution
LSM	Noah-modified 21-category IGBP-MODIS
PBL Scheme	MYNN2
Shortwave radiation	RRTMG shortwave
Longtwave radiation	RRTMG scheme
SST update	NASA-JPL 1km resolution data
Grid size	400×400 grid cells
CAMx version 7.1	
Chemistry	Carbon bond 6 revision 5
Meteorological inputs	WRF model v4.3
Emission data	EPA/NEI 2018
CMV emissions	Inline point sources
Initial/boundary conditions	EPA 2018
Grid size	315×300 grid cells

The domain of this study covers the East Coast of the United States (Figure 1) and includes major cities and highly populated regions. Furthermore, it contains several major ports, which are found to experience high shipping traffic. The meteorological files have 400×400 horizontal grid points and 35 vertical layers covering the entire CAMx domain, which consists of 315×300 grid points, the same as the emission files. Details about the model configuration are discussed in Table 1. Both the WRF and CAMx models have a 4-km horizontal resolution, the same as the emission inventory, in order to avoid spatial interpolation of gridded emissions data. To minimize the impacts of the initial conditions on modeling results, we consider at least 48 hours of spin-up time for both models. Furthermore, as the areas of interest are far from the boundaries, the effects of boundary conditions on modeling results are expected to be minimal.

2.2 Emission data

For emission inputs, we use the most recent emission inventory developed by the EPA, which includes the period May 1st, 2018 to October 1st, 2018 (U.S. Environmental Protection Agency (EPA), 2017) at 4-km horizontal grid resolution and hourly temporal resolution. The emissions are distributed on a 315×300 grid, which covers the entire East Coast of the U.S. (Figure 1),
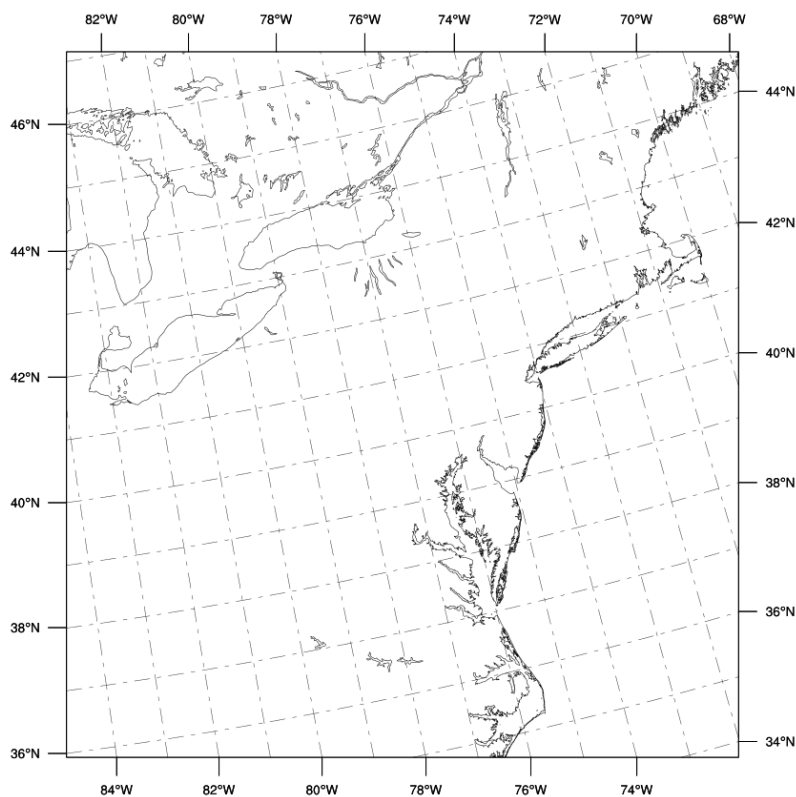
Figure 1. The study domain with 315×300 grid cells and 4-km horizontal grid resolution.

120 with 35 layers vertically. Emissions are treated in two basic ways within CAMx: gridded 2D emissions that are released into each grid cell of the modeling domain near the surface (i.e., "area sources", such as traffic or residential heating) and stack-specific "point sources", where each stack is assigned unique coordinates and parameters (i.e., smokestacks or ship chimneys). For inline point source emissions, CAMx computes the plume rise using stack parameters and the hourly emissions for each emission sector.

125 In the 2018 NEI data, the gridded 2D emissions are merged, meaning that they are provided as one set of surface emissions that include all emission sectors. On the other hand, the elevated point sources are provided for each potential sector separately. For ship emissions, we use the emission data for the Commercial Marine Vessels (CMV) sector, which includes Category 1, 2 (small engine), and 3 (large engine) ships. CAMx computes the time-varying buoyant plume rise using stack parameters and the hourly emissions for each emissions sector, including CMV. Unlike previous EPA data sets, the CMV emissions in 2018 are

130 at a one-hour temporal resolution. The initial and boundary conditions for this study are also provided by the EPA.

3 Results

We study the impacts of ship emissions on the concentrations of four criteria pollutants: $PM_{2.5}$, SO_2, NO_2, and O_3. We calculate the time-average concentrations over different time periods depending on the pollutant as follows, to match the EPA national standards: one hour for SO_2 and NO_2; 8 hours for O_3; and 24 hours for $PM_{2.5}$. We analyze every pollutant from two perspectives:

135   1) from a regulatory perspective, thus calculating the statistics that are as close as possible to the EPA design value for each pollutant (Table 2), and 2) from a worst-case perspective, thus calculating the maximum contribution of ships to each pollutant over the entire three-month study period.

Table 2. Design values for criteria pollutants (U.S. Environmental Protection Agency (EPA), 2022b). For attainment purposes, the design values should be calculated by taking the average of the various percentiles over the past three years; since only one year was simulated in this study, the 3-year average could not be calculated and therefore only the actual percentiles were used.

Pollutant	Design value	Threshold for attainment
O_3	4-th highest 8-hr averaged daily maximum	70 ppb
SO_2	99-th percentile 1-hr daily maximum	75 ppb
NO_2	98-th percentile 1-hr daily maximum	100 ppb
$PM_{2.5}$	98-th percentile 24-hr average	$35\,\mu g\,m^{-3}$

To calculate the maximum contribution, we first find the differences between the two cases (WithShips minus NoShips) at every grid cell, averaged over the relevant time interval, which depends on the pollutant (Table 2); then, we find the maximum

140   difference through the 3 months at every grid cell as follows:

$$\max(\Delta P_{i,j}) = \max_{t \in [1...n]} (P_{i,j}^{WithShips}(t) - P_{i,j}^{NoShips}(t)) \tag{1}$$

where n is the number of data on the 1-, 8-, or 24-hr averaged pollutant P concentration values (the exact time averaging window depends on the pollutant, see Table 2) over the 3-month period of study, $P^{WithShips}$ and $P^{NoShips}$ are pollutant P concentrations with and without the ships, respectively, and i and j correspond to the model grid cell indices.

145   Although the maximum contribution from Eq. 1 is not valuable in terms of reaching or maintaining the EPA attainment for states, it is essential to understand the importance of maritime shipping on air quality, physically and statistically. A summary of the design values defined here for each pollutant to represent the EPA standards and the threshold for attainment are presented in Table 2. The defined design values follow the same criteria as defined by the EPA (U.S. Environmental Protection Agency (EPA), 2022b) but only for the time period of this study. For the remainder of the article, we will assume that the design values

150   defined in Table 2 are serving the purpose of analyzing the pollution from a regulatory perspective and are the same as the EPA standards for those pollutants for the time period of this study.

We find that the concentration of carbon monoxide (CO) remains unchanged in presence of ships (not shown), suggesting that the CMV sector has minimal impact on the CO concentrations in the region. As such, we do not discuss the CO results in the rest of this study.

3.1 Fine particulate matter (PM$_{2.5}$)

The PM$_{2.5}$ species used in this study are those included in the CAMx model output (Ramboll Environment and Health, 2020). The EPA requires that the 3-year average of the 98-th percentile of the daily mean PM$_{2.5}$ concentrations should not exceed 35 µg m^{-3}. Here, we calculated the 98-th percentile of the 24-h averaged PM$_{2.5}$ concentrations at every grid cell during the simulation period in both scenarios, WithShips, and NoShips.

We find that PM$_{2.5}$ levels stayed below 35 µg m^{-3} across most of the domain and that only two locations, i.e., Manhattan, NY, and Easton, PA (Figure 2a), crossed the 35 µg m^{-3} maximum allowed concentrations and therefore were in non-attainment based on the design value defined in this study in Table 2. From a policy perspective, the CMV sector increases PM$_{2.5}$ levels up to 3.2 µg m^{-3} in Manhattan, NY, and up to 2 µg m^{-3} elsewhere (Figure 2c). This is while the percent contribution to PM$_{2.5}$ concentrations remains below 27% across the domain (Figure 2d). In a worst-case scenario, however, the maximum contribution of the ships to PM$_{2.5}$ concentrations within the 3 months is significantly high across the domain but due to the dominant southwesterly wind direction in the region (Golbazi et al., 2022), it mostly remains over the Atlantic Ocean (Figure 2b). The maximum impact on PM$_{2.5}$ during the 3 months reaches as high as 8 µg m^{-3}. Across the domain, the highest impacts are found offshore of MD and VA and in the Chesapeake Bay, DE. Over the land, the highest impacts are in Manhattan, NY, CT, and coastal MA.

Figure 2. PM$_{2.5}$ concentration results: a) 98-th percentile of the 24-hr average PM$_{2.5}$ concentrations with ships (withShips); b) maximum contribution of the ships during the 3-month period (Eq. 1); c) difference between the two scenarios (WithShips minus NoShips) from a regulatory perspective, meaning the changes to the 98-th percentile PM$_{2.5}$ concentrations; and d) percent contribution of the ships to 98-th percentile of the 24-hr average PM$_{2.5}$ concentrations.

3.2 Sulfur dioxide (SO$_2$)

The SO$_2$ design value is defined as the 99-th percentile daily maximum SO$_2$ concentrations in the simulation period, which should not exceed 75 ppb (Table 2). Here we calculated the 99-th percentile of daily maximum SO$_2$ concentrations at every grid cell over the simulation period in the two scenarios, i.e., WithShips and NoShips. Then, we subtracted these two cases from one other (WithShips minus NoShips) to obtain the net effect of the maritime shipping sector.

Our results show that ships have a significantly high impact on SO$_2$ concentrations, up to 95% and 90% over the Atlantic Ocean and inland, respectively (Figure 3d). This suggests that the CMV sector is one of the highest contributors to SO$_2$ levels in the region. The increase in the SO$_2$ design value by ships remains mainly offshore and around the major shipping routes (Figure 3c). However, it reaches the interior of land in major ports and some parts of the coastal states. Over the simulation period, the contribution of the ships to the 99-th percentile daily SO$_2$ maxima is up to 45 ppb, with the highest impact in Baltimore, Maryland (MD), and Norfolk, Virginia (VA), and parts of New Jersey and Long Island (Figure 3c). We note that, however, the SO$_2$ design value in the region remained below 75 ppb in all states (Figure 3a). It is worth mentioning that the locations with the highest SO$_2$ concentrations are the ones highly impacted by the ships.

In addition, we calculated the 3-month maximum contributions of the ships to SO$_2$ concentrations, which indicates the worst-case scenario at every grid cell (figure 3b). Although the increase in SO$_2$ design value was mainly offshore, the maximum contribution of the ships to SO$_2$ showed a different pattern, with a maximum increase of up to ~185 ppb at a few grid cells around Norfolk, VA. We note that an occasional and short-term spike of high concentrations of SO$_2$, as we report here for Norfolk, is not necessarily associated with a strong health impact.
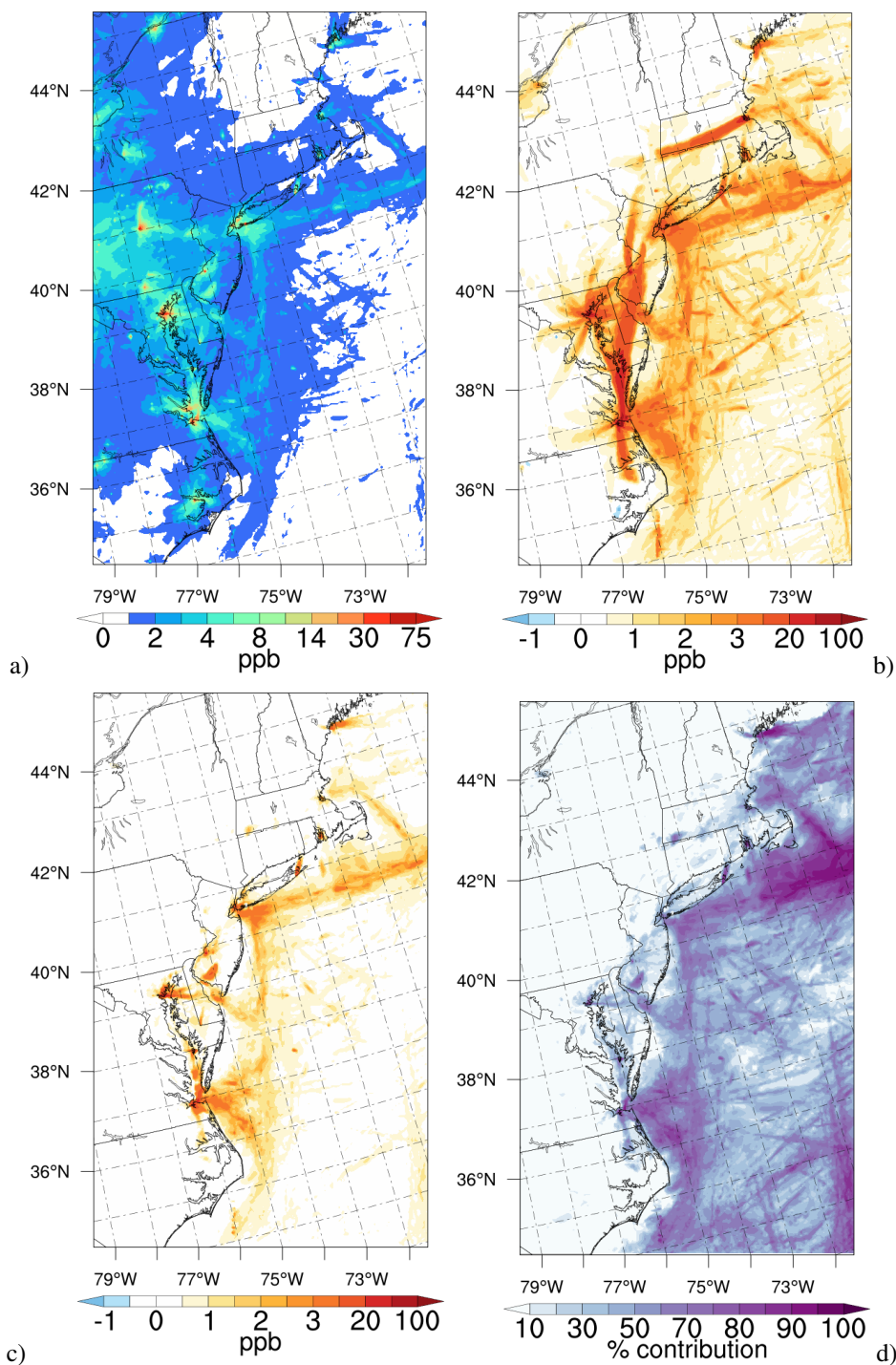
Figure 3. SO$_2$ concentration results: a) 99-th percentile of daily 1-hour maximum SO$_2$ concentrations with ships (WithShips); b) maximum contribution of the ships during the simulation period (Eq. 1); c) differences between the 99-th percentile daily maximum between the two scenarios (WithShips minus NoShips); and d) percent contribution of the ships to the 99-th percentile of the daily SO$_2$ maximum.

3.3 Nitrogen dioxide (NO₂)

NO_2 is a precursor to tropospheric ozone (O_3) formation, which has further negative impacts on human health (, EPA). NO_2
190 is generally directly emitted into the atmosphere from emission sources, including ships. We find that ships cause a significant
increase in the 98-th percentile of daily maximum NO_2 concentrations, up to 34 ppb, but only at a few locations along the coast
and in coastal states with major ports (Figure 4c), suggesting that, except for states with large ports, ships do not significantly
impact the state compliance with the EPA standards. However, in NC, VA, DE, NY, and CT the shipping impacts reach beyond
15 ppb from a regulatory standpoint. Among these states, while NC remains in attainment with regulations (below 100 ppb),
195 it experiences up to 80% contribution from the ships to its NO_2 concentrations. On the other hand, NY is the only state in
the study domain that exceeds the 100 ppb standard for NO_2 concentrations, and shipping contribution to its non-attainment is
20-25%.

The maximum contribution of the ships to NO_2 concentrations, which is illustrated in Figure 4b, shows that, in a worst-case
scenario, ships contributed to up to ~50 ppb of NO_2 along the coast and 75 ppb over the Atlantic, which is significantly high
200 compared to 100 ppb standard. The 3-month highest impact happens near the major ports and shipping routes but stretches to
the land and over the ocean.

3.4 Ozone (O₃)

Tropospheric ozone is formed by both naturally occurring and anthropogenic sources. Ozone is not emitted directly into the air,
but, in the presence of sunlight, it is created by its precursors: VOC and NO_x. The rate of ozone production can be limited by
205 either VOCs or NO_x. As a result, a specific location can be either VOC-limited or NO_x-limited. The rate of production/destruction
of O_3 in the atmosphere is different in either of these regimes. We will further discuss this matter later in this section.

We use 8-hour average ozone concentrations for our analysis to maintain consistency with the EPA standards (U.S. Environ-
mental Protection Agency (EPA), 2022b). We calculated the 8-hr averaged ozone values by averaging consecutive eight hours
of O_3 outputs at each hour of the day and storing it at the start hour (Cohen et al., 1999). For instance, O_3 at 11:00 in a day
210 indicates the time-averaged O_3 concentrations between hours 11:00 and 19:00 in that day. Hereafter, we will refer to the 8-hr
averaged O_3 concentrations simply as O_3 concentrations.

Ambient ozone concentrations are directly affected by temperature, solar radiation, wind speed, and other meteorological
factors. Since O_3 production is a photochemical reaction, its peak concentrations are found during the daytime when tropospheric
ultraviolet radiation in the atmosphere is highest. Since the focus of this study is the daily high episodes of O_3 that are associated
215 with adverse health impacts, we limit our analysis of the maximum impacts to only daytime hours. To select the daytime hours,
we considered the 8-hr averaged O_3 daily profiles in 10 different locations along the coast from which 4 selected locations are
shown in Figure 5. The lowest concentrations of O_3 are associated with hours 00:00 to 08:00 UTC (20:00 to 04:00 local time).
We eliminated these hours from our analysis to only focus on concentrations during the high episodes. Therefore, we select the
hours with peak O_3 concentrations during the 24-h period i.e., 09:00 to 23:00 UTC (05:00 to 19:00 local time).
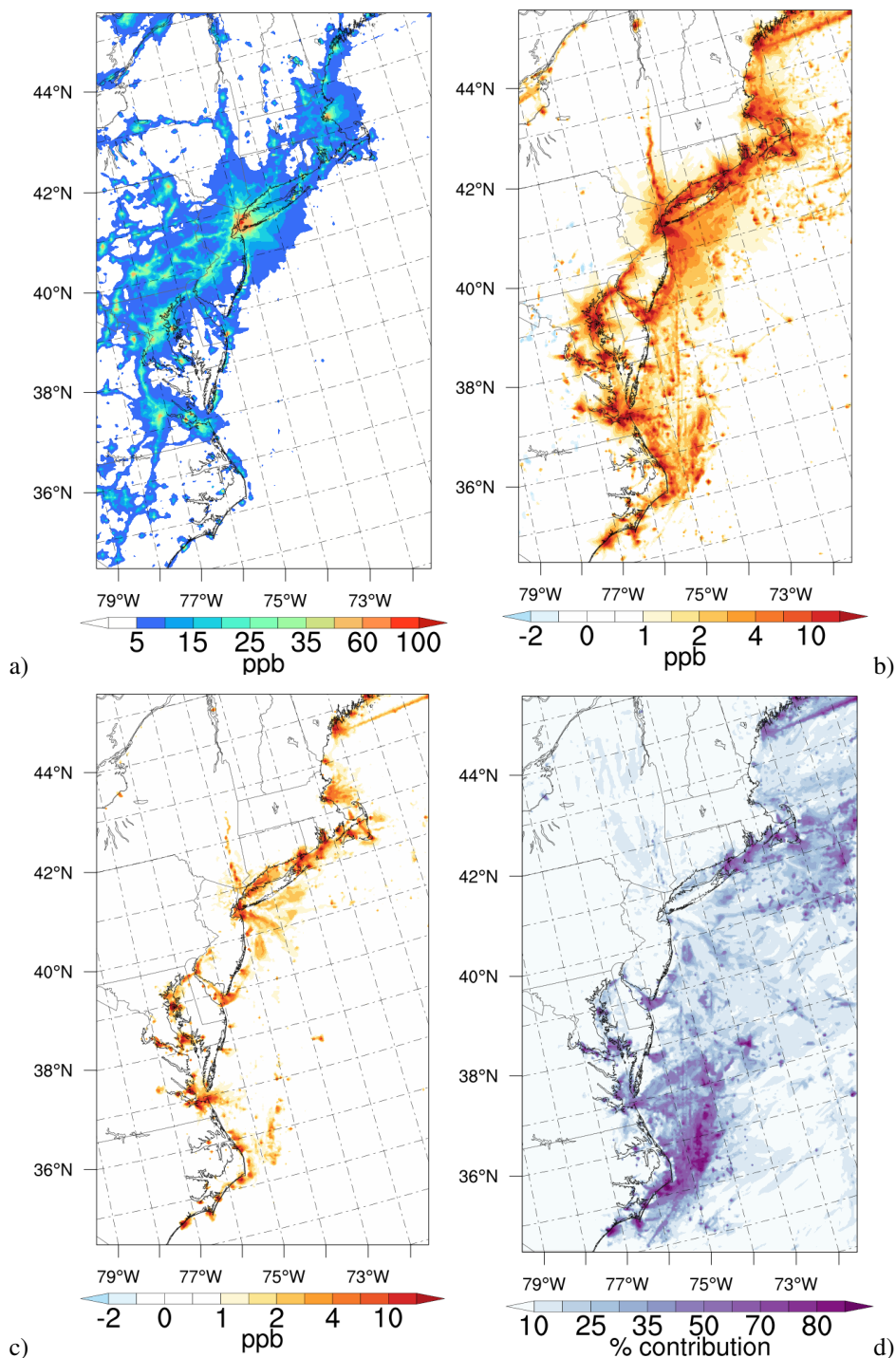
Figure 4. NO₂ concentration results: a) 98-th percentile of 1-hour daily maximum NO₂ concentrations with ships (WithShips); b) maximum contribution of the ships during the simulation period (Eq. 1); c) differences between the 98-th percentile of the 1-hour daily maximum between the two scenarios (WithShips minus NoShips); and d) percent contribution of the ships to the 98-th percentile of the daily NO₂ maximum.

220   We find that although ship emissions contribute to O_3 enhancement in the region, they reduce O_3 at some urban locations. We detect a significant increase or decrease in O_3 concentrations in presence of the ships, depending on whether the location was NO_x or VOC limited.
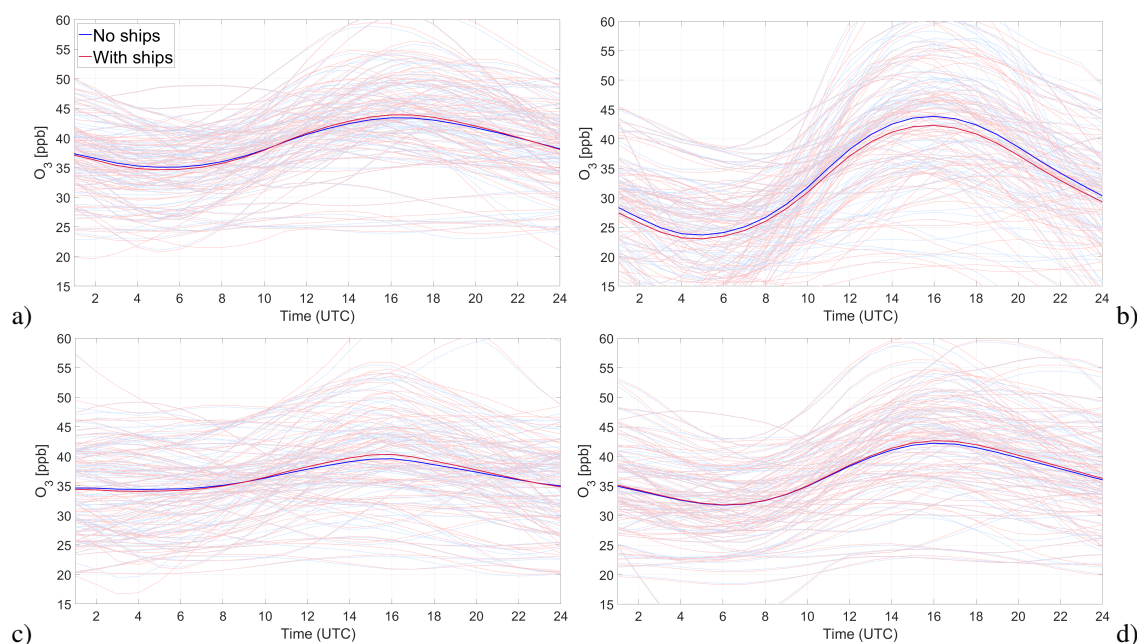
Figure 5. Eight-hour average O_3 concentration time series at four different locations along the East Coast from June 1st to August 30st, 2018: a) Atlantic City, NJ; b) Manhattan, NY; c) Cape Cod, MA; and d) Providence, RI. The thin lines represent the daily time series and the thick lines are the 3-month average profiles with red lines for WithShips and blue lines for the NoShips simulations.

The maximum contribution of the ships to the O_3 levels over the entire 3-month period is illustrated in Figure 6c as a worst-case scenario. Over the ocean, the maximum increase is large, up to 8.6 ppb. However, the pollution increase remained

225   primarily offshore and did not significantly impact the coastal areas. O_3 increased by 4-5 ppb at most in parts of North Carolina, Baltimore, MD, parts of CT, and MA. Otherwise, the maximum increase over the land was up to 3.5 ppb. It's important to note that the maximum impact is not necessarily at the time when high O_3 episodes (from a regulatory perspective) are found. Despite the O_3 increase over the Atlantic, at the shore where the major ports are built, the maximum contribution of the ships is negative, suggesting that O_3 was destructed in presence of the ships. This is due to the complex nature of atmospheric chemistry,

230   where the fresh NO emissions from the ships scavenge O_3 and reduce its concentrations in urban VOC-limited areas.

While Figure 6b is important to understand the worst-case scenario of the shipping impact on O_3 pollution in the region, it does not help to measure the impact of these changes on state compliance with EPA standards. This is because a high impact on O_3 in the worst-case scenario may not correspond to the time of the day when O_3 daily maxima occurred. Therefore, to study the impacts of ship emissions from a policy perspective, it is necessary to explore the impacts on the O_3 design value at every

235   grid cell. For O_3, the EPA defines this standard as the 4-th highest 8-hr averaged O_3 daily maxima, averaged over a 3-year period
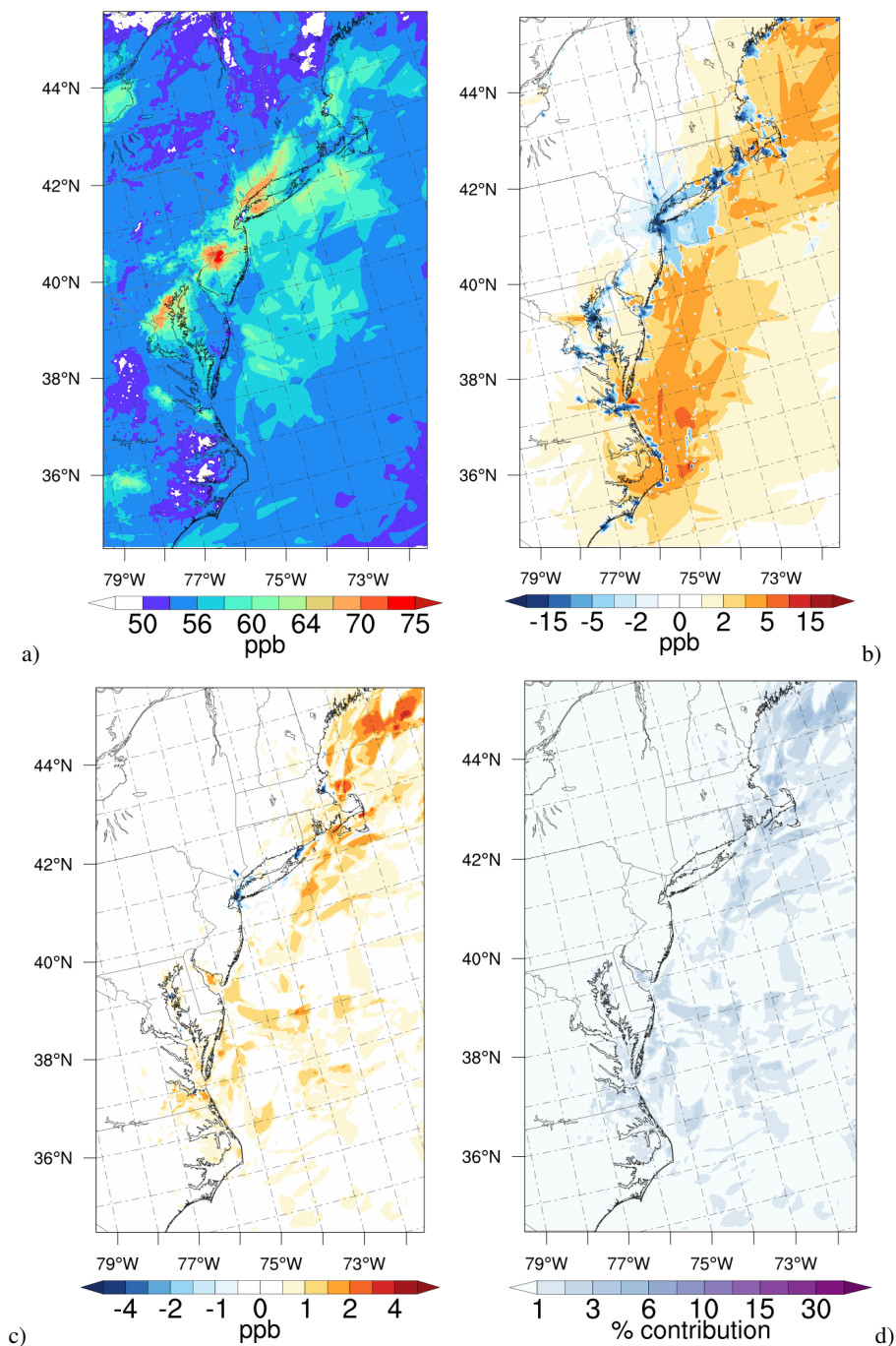
Figure 6. O$_3$ concentration results: a) 4-th highest 8-hour daily maximum O$_3$ concentrations with ships (WithShips); b) maximum contribution of the ships during the 3-month simulation period (Eq. 1); c) differences between the 4-th highest 8-hour daily maximum between the two scenarios (WithShips minus NoShips); and d) percent contribution of the ships to the 4-th highest daily O$_3$ maximum.

which should not exceed 70 ppb (U.S. Environmental Protection Agency (EPA), 2022b). Since we do not have data for three consecutive years, we focus on the 4-th highest daily maximum in our study period, the summer of 2018. Hereafter, we will refer to this value as O_3 design value in this study. We assume that this value represents the 4th highest daily maximum in the year 2018 since the highest O_3 episodes are expected to occur during the summer period. Thus, the regions with higher than 70

240 ppb O_3 concentrations in 6a are most susceptible to being in non-attainment with the EPA standards and therefore the impacts of the ships are of higher significance in those regions. Out of all states in the domain, NY, NJ, and MD are the only states that exceed the 70 ppb standard and are likely to be in non-attainment. All other states stay in attainment with the standards defined in Table 2 in either scenario.

From a policy perspective, O_3 design values increased in presence of the ships by up to 3.5 over the Atlantic Ocean. However,

245 we find a reduction (up to 6.5 ppb) in O_3 concentrations at major ports along the East Coast (Figure 6c), where fresh NO is emitted by the ships into the atmosphere in VOC-limited regions (Figure 7). In most parts, the major impact of the ships remains offshore away from the coastal areas. However, in some regions in MA, RI, CT, ME, VA, NC, and MD ships contribute to O_3 increase at the coast from which, only MD is likely to be in non-attainment. The highest increase in O_3 design value inland is found in VA and NC and is up to 2.5 ppb, while we note that in NY, the 4-th highest daily maximum is decreased by 4 ppb in

250 presence of the ships for the reasons discussed later in this section. However, the decrease in O_3 values remains in Manhattan, NY, and is not associated with the parts of Long Island (NY) where O_3 exceeds the standards.

In the atmosphere, the formation or destruction of ozone depends on the concentrations of both NO_x and VOC and the ratio between them (VOC/NOx). Transportation usually is associated with high NO_x emissions, therefore O_3 is generally NO_x-limited in rural areas and VOC-limited in urban areas, with low and high traffic densities, respectively.

255 In the VOC-limited regions, high concentrations of freshly emitted NO locally scavenge O_3 and lead to the formation of NO_2. Close to the emission sources, this titration process can be considered an ozone sink. In addition, high NO_2 concentrations deflect the initial oxidation step of VOC by forming other products such as nitric acid (HNO_3), which slows down the formation of O_3 (National Research Council, 1992; Beck et al., 1998). Because of these reactions, an increase in NO leads to a decrease in O_3 at VOC-limitted regions.

260 In CAMx, the VOC-limited regime is defined when the rate of change of hydrogen peroxide (H_2O_2) is lower compared to the rate of change of HNO_3. A higher $\Delta H_2O_2/\Delta HNO_3$ ratio indicates a NO_x-limited regime, while a lower $\Delta H_2O_2/\Delta HNO_3$ ratio corresponds to a VOC-limited regime. There are other indicators for determining the NO_x/VOC-limited regimes that are discussed in the literature (Li et al., 2022). However, here we use the $\Delta H_2O_2/\Delta HNO_3$ ratio as is used in the CAMx model (Ramboll Environment and Health, 2020).

265 To understand the formation/destruction of ozone in presence of the ships in our study domain, we calculated the frequency of the VOC-limited regime based on the ratio at every grid cell. Figure 7a illustrates the percentage of the times that a VOC-limited regime occurred at every cell, which is the highest along the coast and in major cities. This indicates that O_3 may be affected by titration when ship emissions are present. We find that the NO concentrations increase along the coast where we detect a decrease in O_3 concentrations (Figure 7b) and a VOC-limited regime (Figure 7c).

270   This finding does not necessarily mean that ships help create better air quality since a reduction in O_3 is due to a significant increase in other important air pollutants i.e., NO_x concentrations.
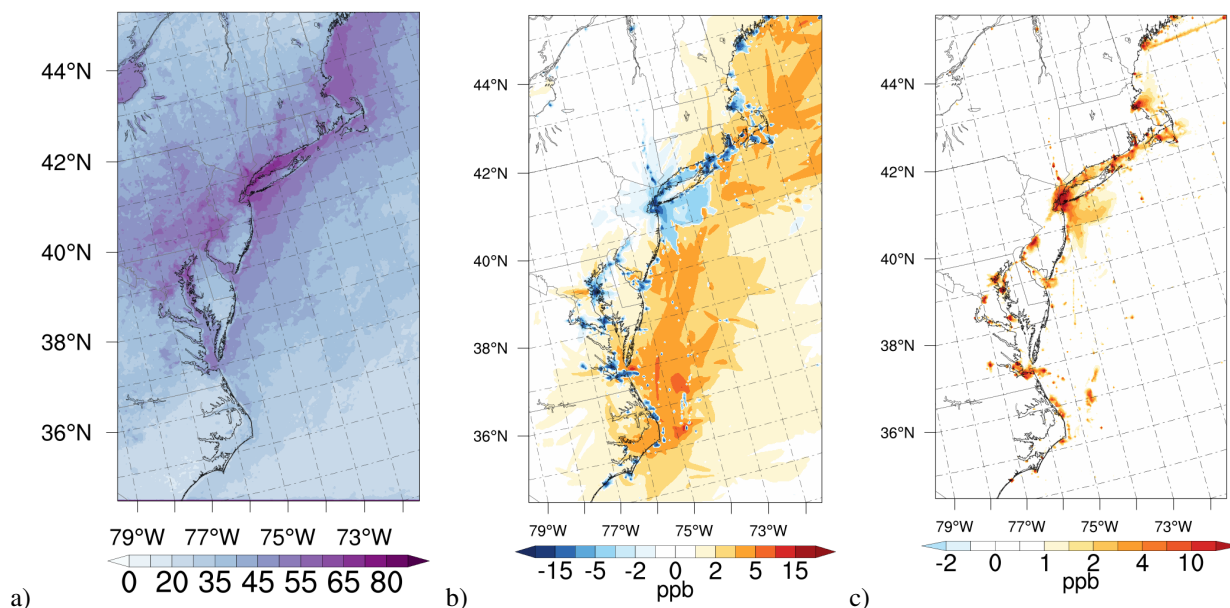
Figure 7. VOC- versus NO_x-limited regime determination: a) percentage of the times when H_2O_2/HNO_3 < 0.35 as determined in CAMx model, which is an indication of a VOC-limited regime; b) maximum contribution of ships to O_3 pollution (Figure 6c); and c) same as in b) but for NO.

4   Conclusions

Ships emit significant amounts of pollutants within 400 km of the shores. Here, we studied the ocean-going ship emissions on the air quality of the U.S. East Coast. We utilized the WRF-CAMx modeling system to simulate the pollution concentrations in

275   the presence and absence of shipping activities along the East Coast and at the major ports. We used the WRF model to provide the meteorological inputs for the CAMx air quality model for the year 2018, on which the most recent EPA/NEI emission inventory is based. We particularly focused on $PM_{2.5}$, SO_2, NO_2, and O_3. Overall, we studied the outcomes on every pollutant from two perspectives: 1) from the EPA perspective concerning the national concentration standards for each specific pollutant, and 2) the maximum contribution of ships to that pollutant over the 3 months.

280   We find that shipping increases the $PM_{2.5}$ concentrations across the domain. the 98-th percentile daily average $PM_{2.5}$ levels increased by $3.2\,\mu g\,m^{-3}$ over the ocean and in some coastal areas. However, in a worst-case scenario, ships contribute up to approximately $8.0\,\mu g\,m^{-3}$ to $PM_{2.5}$ concentrations, only over the Atlantic off the coast of MD, and VA. In addition, we find that ships have a significantly high impact, up to 95% and 90%, on the SO_2 concentrations over the Atlantic and inland, respectively. This suggests that the CMV sector is one of the highest contributors to SO_2 levels in the region. The shipping

285   contribution to SO_2 levels was up to 45 ppb over coastal regions. Ship emissions also impacted the NO_2 design value by up

to 34 ppb. In addition, our simulation results show that the impact of ship emissions on O_3 concentrations is not uniform, meaning that maritime shipping affects ozone pollution in both positive and negative ways. Although over the ocean O_3 concentrations increase significantly in the presence of ships (up to 8.6 ppb), in coastal areas with major cities and major ports O_3 concentrations decrease by up to ~6.5 ppb. To understand the reasons behind the O_3 reduction in the presence of ships, we

290 analyzed the $\Delta H_2O_2/\Delta HNO_3$ ratio in the region, which is used to determine NO_x- or VOC-limited ozone production, as well as changes to NO concentrations, since they play a significant role in O_3 formation and destruction. We found that ships emit significant amounts of fresh NO in the atmosphere, which then helps scavenge O_3 in VOC-limited regimes. As a result, with higher NO concentrations in the atmosphere produced by ship emissions, O_3 is destructed in major cities and urban areas. By contrast, over the ocean (a NO_x-limited regime), excessive NO_x emissions due to the ships contribute to the formation of O_3

295 and therefore an enhancement in O_3 concentrations. It is important to note that the destruction of O_3 by ship emissions in major cities does not necessarily mean that the ships create better air quality because a decrease in O_3 is a consequence of a significant increase in other pollutants, like NO. Overall, the majority of the time, due to the dominant southwesterly wind direction in the region, the impacts on different pollutants remained spatially confined offshore. However, in some coastal areas near the major ports, the impacts were significant.

300 **Data availability**

The data for the model setup is available in the GitHub repository at:

https://github.com/golbazimaryam/ShippingEmissionsAndAirQuality

Author contribution

M. Golbazi contributed to the design of the study, preparing the manuscript, setting up and carrying out simulations, and

305 analysis. C.L. Archer contributed to the design of the project, preparing the manuscript and editing, as well as data analysis.

Competing interests

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

Acknowledgements. *

Partial funding for this research came from the University of Delaware (UD) Graduate College Doctoral Fellowship and from the Delaware

310 Natural Resources and Environmental Control (DNREC, award no. 18A00378). The simulations were conducted on the UD Caviness and NCAR Cheyenne high-performance computer clusters.

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