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**EXPLORING THE IMPACT OF SHIPPING EMISSIONS ON PM LEVELS IN THE EASTERN
MEDITERRANEAN**

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Abstract: The study is addressing the issue of shipping emissions impact on the atmospheric chemical composition in the Eastern Mediterranean through a multi-scale spatial approach (regional versus local) and with the combination of pollutants' emission data, WRF-CAMx numerical modelling results and in situ chemical analysis data for January 2019. Emphasis is given on sulfur-containing species. There was a considerable % reduction in the monthly concentrations of SO₂ and sulfate over the greater part of the maritime areas of Greece when shipping emissions were not accounted for in model runs reaching 73% and 22% respectively. In Piraeus (Greece), one of the biggest European ports, the absolute concentration differences on a daily basis were up to -14 µgm⁻³ and -0.72 µgm⁻³, respectively. The period-mean sulfate concentration reductions when excluding shipping emissions in the Piraeus port area were approximately 20%, and on a daily basis they correlated significantly with shipping contributions to sulfate particles as calculated by PMF receptor modeling using chemical composition observations close to the port.

Key words: *Shipping, Emissions, WRF-CAMx, PMF, source apportionment, sulfate.*

INTRODUCTION

Shipping has an important role in the emissions and concentrations of air pollutants and this is an issue that needs to be examined considering also the expected increase in ship traffic in the coming years (Fink et al., 2023). The Eastern Mediterranean's key geographical position and the long coastline with numerous islands result in intense short-sea domestic shipping, which affects the particulate and gaseous pollutant concentrations at regional and local scales in addition to international shipping. Greece has the 10th longest coastline in the world (about 5% and 30% of the European and Mediterranean coastline, respectively). According to Eurostat (2019), the port of Piraeus in Greece was the 4th biggest port in Europe (after Rotterdam, Antwerp and Hamburg) regarding the number of containers and the 6th among European ports with respect to passengers. In addition, Piraeus is the busiest passenger and 2nd busiest container port in the Mediterranean. Observations at the air quality station of the Ministry of Environment and Energy (MEEN) in Piraeus reveal that the World Health Organization guideline for the number of days per year when the daily PM_{2.5} concentrations exceed 15µgm⁻³ is continuously violated.

To study the seasonal impact of ship emissions on the atmospheric particulate load in the Eastern Mediterranean with more focus on Greece, the modelling system comprised of the meteorological model

WRF and the photochemical model CAMx was applied following the zero-out approach for shipping emissions. At a local scale, particulate matter samples were collected near the port of Piraeus and were chemically analyzed to assess the impact of various aerosol sources including shipping, on air quality and climate-relevant compounds, in comparison also with WRF-CAMx results. The contribution of various sources, and especially shipping, to the emissions profile of specific pollutants in Piraeus was examined through the updated emissions inventory “Flexible Emission Inventory for Greece and the Greater Athens Area” (FEI-GREGAA).

METHODOLOGY

The modeling system WRF-CAMx was applied over two nested domains covering Europe and North Africa (in 18 km) and the Eastern Mediterranean (in 6 km). Simulations were performed for January 2019. WRF and CAMx models were driven by boundary conditions from the ERA5 reanalysis data and CAMS-IFS global model, respectively. Anthropogenic gaseous and particulate matter (PM) emissions were obtained from the most recent year of CAMS-REGv5 emissions database (Kuenen et al., 2021), including international and domestic shipping emissions calculated with the STEAM model (Johansson et al., 2017). A detailed emission inventory was prepared for the heating and road transportation (exhaust and non-exhaust) sectors, calculated on the basis of activity data and WRF meteorology (for heating emissions) (Liora et al., 2022). Natural emissions (sea salt, windblown dust and biogenic NMVOC) were calculated using the natural emissions model NEMO (Liora et al., 2015, Kontos et al., 2018). Two sets of WRF-CAMx runs were performed: a) base case (BCASE) and 2) zero-out shipping emissions in the maritime area of Greece (NOSHIP).

Fine aerosol chemical composition was concurrently measured in Piraeus during January 2019. The sampling site was located at a distance of approximately 150m to the east of the central terminals in the passenger port of Piraeus. More details regarding the location characteristics can be found in Stavroulas et al. (2021). Daily PM_{2.5} filter samples were collected using a low-volume reference-equivalent sampler. The quartz-fiber filters were analyzed for organic and elemental carbon by a Sunset analyzer, for major inorganic ions including sulfate (SO₄²⁻) by Ion Chromatography, and for trace elements including vanadium and nickel (the main shipping emission tracers) by ICP-MS. The chemical dataset was analyzed by PMF receptor modeling (Grivas et al., 2018) to apportion sources and atmospheric processes driving the PM_{2.5} daily variability. The analysis resulted in the identification of seven sources/processes including a shipping source, characterized by very high contributions to vanadium and nickel (associated with a diagnostic V/Ni ratio of approximately 2), presenting a large summertime enhancement due to the intensification of emissions in the passenger port, and linked to the prevalence of westerly winds transporting pollutants from the port zone (Grivas et al., 2022).

Emission data for the area of Piraeus were used in this study taken from the emission inventory FEI-GREGAA (Fameli and Assimakopoulos, 2016) updated for the year 2019 according to the EMEP/EEA Emission Inventory Guidebook 2019 methodology for all anthropogenic sources (stationary combustion sources, transportation (road, navigation and aviation), agriculture and industry) and on the basis of official national and European sources. New emission factors were calculated especially for the road transport and aviation sectors. Regarding navigation, pollutant emissions were calculated for liquid bulk tankers, dry bulk carriers, container ships, specialised carrier ships, cruise ships and passenger ships. The engine type was diesel and the fuel was HFO, MDO and MGO. The calculated total annual emissions were disaggregated to a spatial resolution of 1 km² on an hourly basis. The spatial distribution of SO_x emissions at the port of Piraeus is presented in Figure 1 depicting the increased SO_x emissions at the locations of the entrance and exit routes of the port.

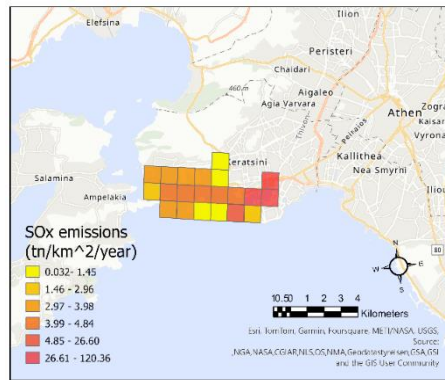


Figure 1. Gridded SO_x shipping emissions at the port of Piraeus including movement from both commercial and passenger ships (reference year 2019).

RESULTS AND DISCUSSION

Despite the effort to reduce shipping emissions, especially over the last decade, the shipping sector is the major source of nitrogen oxides and sulfur oxides (SO_x) emissions, and an important source of PM_{2.5} emissions in Greece. Shipping was responsible for 60% and 90% of SO_x and sulfate anthropogenic emissions, respectively, in January 2019 (Figure 2). This contribution was more evident in the Greater Piraeus Area where, according to the FEI-GREGAA emission database, shipping had a 81% share to SO_x anthropogenic emissions in 2019 (four times higher than the emissions from residential heating).

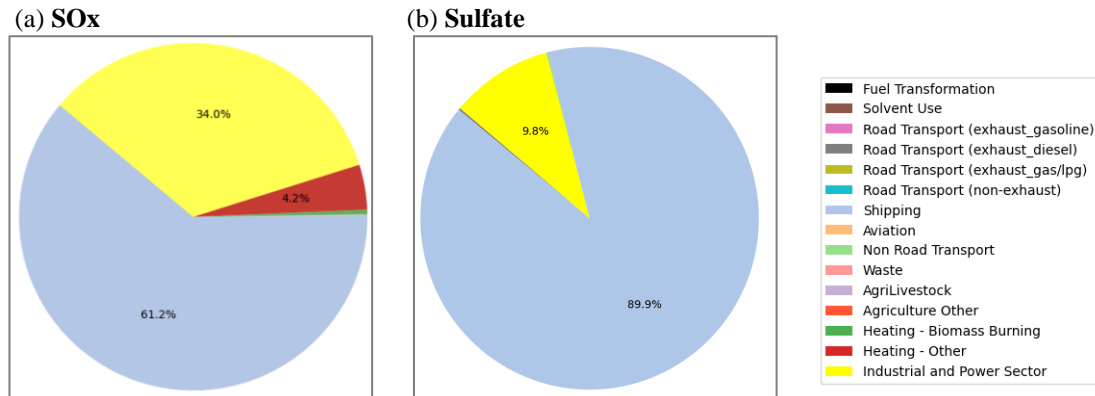


Figure 2. Anthropogenic source contribution to SO_x (a) and sulfate (b) emissions on a national scale for Greece in January 2019.

Figure 3 depicts the impact of shipping on monthly SO₂ and sulfate atmospheric levels in January 2019. There is a considerable percentage reduction in the monthly concentrations of SO₂ over the greater part of the maritime areas of Greece when shipping emissions are not accounted for in model runs, reaching 73%. In terms of absolute difference (not shown here), the impact is more evident over Piraeus and the maritime areas to the south (maximum decrease 4.4 μgm⁻³), where shipping emissions have a higher density. Similar results are obtained for sulfate concentrations, but the shipping impact is less pronounced (maximum percentage and absolute reductions equal to 22% and 0.3 μgm⁻³ respectively).

When focusing in the grid cells representing the Piraeus passenger port and the container/cargo terminals, the percentage differences induced in the concentrations of SO₂ and sulfate by the elimination of shipping emissions are approximately -50% and -20%, respectively. The absolute differences on a daily basis are up to -14 μgm⁻³ and -0.72 μgm⁻³, respectively (i.e. up to 3.2 and 3.6 times higher than the respective monthly change) (Figure 4). The average value of PMF-resolved daily shipping contributions to fine aerosol sulfate determined in situ is relatively comparable (15.3%). The daily modeled sulfate concentration changes due

to zeroing-out of shipping emissions have statistically significant ($p < 0.1$) negative correlation with the daily source-apportioned net ($\mu\text{g m}^{-3}$) shipping contributions to sulfate (Figure 5).

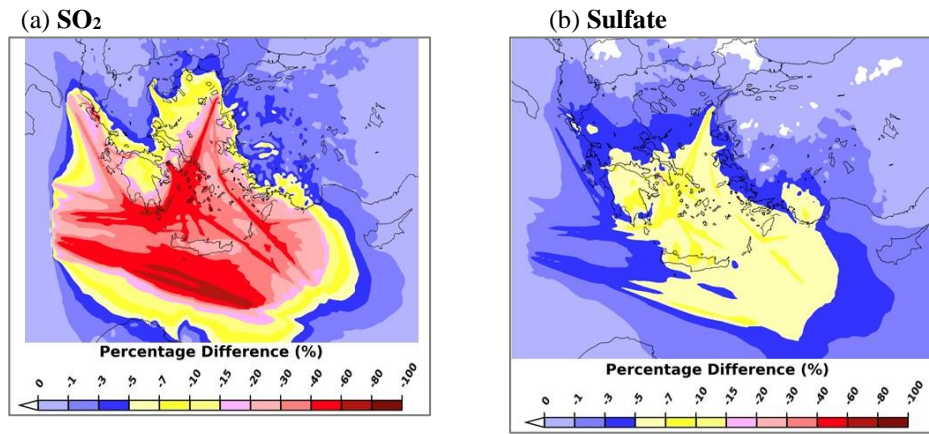


Figure 3. Percentage differences in monthly concentrations between NOSHIP and BCASE scenarios for SO₂ (a) and Sulfate (b) (January 2019).

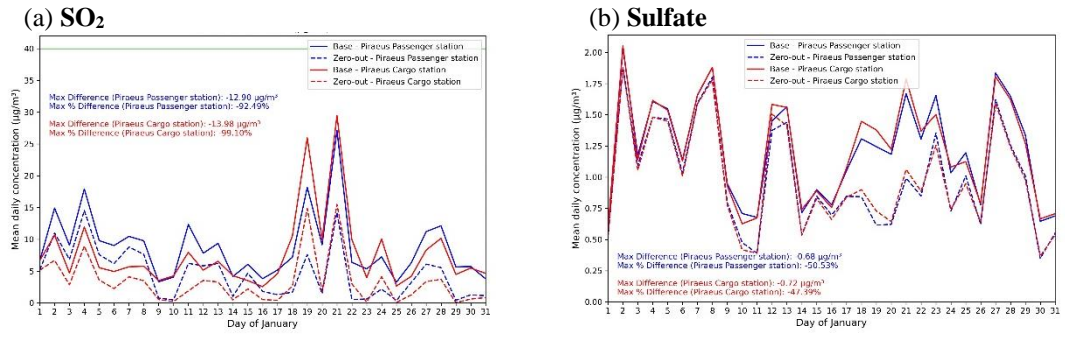


Figure 4. Shipping contribution to SO₂ (a) and Sulfate (b) simulated daily concentrations in January 2019.

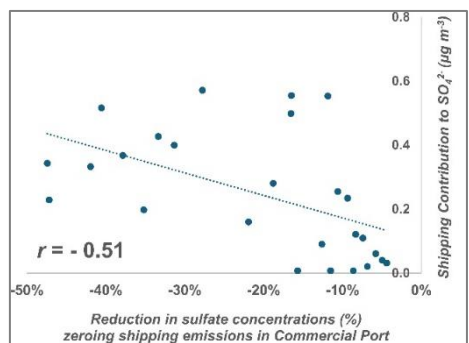


Figure 5. Scatterplot with fitted ordinary least squares line displaying the negative association between the daily PMF-calculated shipping contribution to sulfate concentrations in PM_{2.5} measured in situ in Piraeus, and the daily fractional changes in modeled sulfate concentrations in the grid cell corresponding to the Piraeus commercial port after zeroing-out shipping emissions in January 2019.

CONCLUSIONS

The important contribution of shipping emissions in the determination of SO₂ and sulfate atmospheric levels in the Eastern Mediterranean was quantified in regional and local scales with the synergy of pollutants'

emission data, numerical modelling results and chemometric source apportionment based on in situ aerosol composition. To cut down SO₂ and PM_{2.5} emissions with beneficial results for air quality and human health, the Mediterranean Sea has been designated as an Emission Control Area for sulfur oxides meaning that as of 1 May 2025 ships will be required to use marine fuel with 0.1% sulfur content. Future studies should account for the new regulations when studying the impact on atmospheric environment of ship emissions, considering also the climate extensions given the cooling direct and indirect effect of sulfate aerosols from shipping.

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