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Holistic evaluation of the environmental impacts of shipping in the sensitive region of Ria de Aveiro

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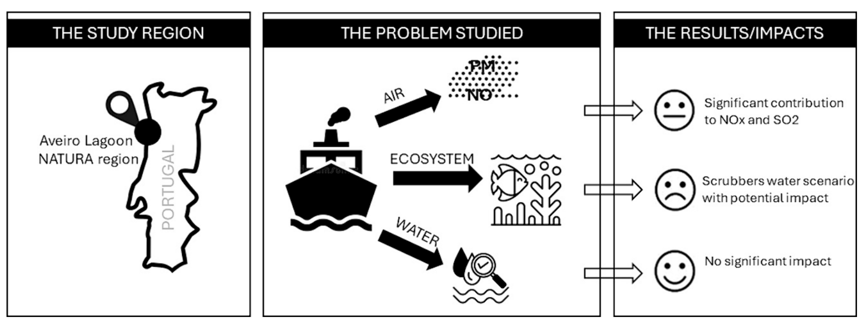
HIGHLIGHTS

- The impact of shipping on air, water and ecosystem is investigated.
- The sensitive area of Aveiro lagoon region in Portugal is the case study.
- Air/water quality modelling tools and ecotoxicological tests were performed.
- The increase of pollution is only significative for atmospheric pollutants.
- Ecotoxicological tests shown that scrubber waters could be a problem.

GRAPHICAL ABSTRACT



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ABSTRACT

Shipping activity can be a substantial source of pollution and impact on the environment, including air, water and ecosystems, as well as adverse health and climatic effects. Due to the distribution of maritime transport activity routes in the EU, a large portion of the population is exposed to shipping pollution throughout Europe.

The ongoing European project EMERGE aims to investigate and quantify these impacts over Europe, and in more detail, in specific case studies regions. The Aveiro lagoon region in Portugal is one of these case studies. This region is a Natura 2000 area, and also includes a medium-sized port. Both air quality and water modelling tools were applied to assess the impact of the emissions and discharges from shipping (to air and water) in the region in 2018. Additionally, ecotoxicological impacts were determined by bioassays to evaluate the impact of scrubber-water discharges on the most sensitive stages of marine invertebrates, and on the post-exposure feeding inhibition of crustacean and bivalve species.

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The results show that there was a substantial increase in atmospheric pollutant concentrations due to emissions attributed to shipping, which was most relevant for NOx and SO_2 (up to a 30 % shipping contribution). There was no significant degradation of the water quality, mainly as the ships operating in this area did not have scrubber equipment. The ecotoxicological tests were performed with three samples of scrubber water, including one artificial sample and two samples collected on-board ships. If scrubber water would have been discharged in this area, the results indicated that the majority of the tested species would be exposed to lowest observed effect concentration (LOEC) for the different scrubber-water samples, as well as to substantial concentrations of metals, PAHs, and alkylated PAHs.

1. Introduction

Maritime transport is one of the important transportation sectors in Europe that enables trade and contact between all European countries, with almost 90 % of the external freight trade being seaborne (Johansson et al., 2017). The shipping activities impact both the atmospheric and marine environments, including, in particular human health, climate and marine biota (Kukkonen et al., 2021; Byrnes and Dunn, 2020).

The growth of the shipping industry has contributed to the degradation of air pollution which has become an increasingly serious concern in coastal regions, for reasons of both environmental quality and human health (Endresen et al., 2003; Dalsøren et al., 2009; Corbett et al., 2007; Hassellöv et al., 2013; Serra and Fancello, 2020). Evidence for the importance of ship emissions has even been obtained from satellite observations such as GOME and SCIAMACHY (Richter et al., 2004). Studies have suggested that around 15 % and 4 %–9 % of all global anthropogenic emissions of NOx and SO_2, respectively, were from ocean-going ships (Beirle et al., 2004; Eyring et al., 2010). As most of the ship emissions occur within a few hundred kilometers of coastlines, they primarily contribute to air pollution in coastal areas, emitting carbon dioxide (CO_2), nitrogen oxides (NOx), sulphur dioxide (SO_2), carbon monoxide (CO), hydrocarbons and primary particulates, as well as secondary particulate precursors (Viana et al., 2014). However, these pollutants can be transported hundreds of kilometers downwind and impact a much broader region (Endresen et al., 2003; Derwent et al., 2005; Aksoyoglu et al., 2016).

Ship operations produce waste streams related to propulsion and engine operations, as well as crew and passenger activities. The waste streams related to propulsion and engine operations include bilge water from the machinery spaces, stern tube oil from the lubrication of the propeller shaft, scrubber wash water from exhaust gas cleaning systems (for the reduction of emissions of sulphur oxides into the atmosphere), ballast water from maintaining ship stability, biocides used in antifouling paints to prevent hull growth, cooling water, tank cleaning residuals, and also waste streams related to humans on board. These releases pose a significant potential risk to water quality and the marine environment (Stips et al., 2016). Nearshore waters are the most heavily trafficked, and these also hold the largest biodiversity. In general, the published data on waste streams from ships is scarce, both in terms of production rates and constituents. Most of the available data is based on different types of reports (e.g. from classification societies: ABS, 2018; HELCOM, 2014, 2018), which calls for a thorough discussion on data quality.

Currently, there is very limited quantitative information on the underlying phenomenon within the shipping sector and the overall environmental effects associated. Although there are already a large number of studies about the impact of shipping emissions, with and without control measures, on the air quality in different areas of the world and their importance in local communities near major ports and coastal areas (Viana et al., 2014; Zhang et al., 2017; Toscano, 2023), there is limited data on the overall environmental impacts derived from shipping activity. Since the atmospheric and marine environments are interconnected, taking a holistic approach is paramount when determining the total environmental load derived from shipping operations. The European EMERGE project seeks to answer some of these lingering questions (https://emerge-h2020.eu), aiming to investigate and quantify the general impacts on the marine environments in European waters, and the specific impacts in selected vulnerable coastal regions. EMERGE is one of the first research projects that simultaneously addresses emissions to the atmosphere and the marine environment, adding an important perspective to the ongoing discussion.

In this study, we present part of the results obtained in the EU-funded project EMERGE, "Evaluation, control and Mitigation of the EnviRonmental impacts of shippinG Emissions" (2020–2024, https://emerge-h2020.eu/). The EMERGE project focused on six selected case studies, including the Aveiro Lagoon region. This region is classified as a Natura 2000 area (network of protected areas covering Europe's most valuable and threatened species and habitats), and it also includes a medium-sized port. The impacts of shipping were assessed for air pollution, water pollution and the effects on the marine ecosystems in 2018. We used both mathematical modelling, experimental on-board sampling on a ship, laboratory analyses of water concentrations and toxicological analyses for several marine species. The present study is the first integrated study that addresses both marine and atmospheric pollution, and the impacts on marine biota attributed to shipping in this region. Such studies have been very scarce also internationally.

2. The EMERGE project & Aveiro case study

2.1. The EMERGE project

The EMERGE project addresses: (i) the impacts in general on the marine environments within the whole of Europe, and (ii) the specific impacts in selected vulnerable coastal regions. Besides the assessment purposes, the overarching aims of EMERGE have been to comprehensively quantify and evaluate the effects of a range of potential emission reduction solutions for shipping in Europe, and to develop more effective strategies and measures to reduce the environmental impacts of shipping (Fig. 1). The approach of studying both the European-scale water and air pollution, and the state of these case study regions in more detail, is unique and therefore facilitates new findings and conclusions. The transport of pollutants in air and water and their effects on ecosystems have been investigated using various types of numerical models within Europe. However, these have very rarely been combined as a holistic assessment (e.g. Zhang et al., 2017; Ytreberg et al., 2021).

The first main task of the EMERGE project was the refinement of a model to estimate shipping emissions to air and water for Europe, i.e., the STEAM model (Ship Traffic Emission Assessment Model; e.g., Johansson et al., 2017). This model is based on using the most detailed input data possible for the locations and movements of ships, the AIS - Automatic Identification System records, which is currently a state-of-the-art method in Europe and worldwide (e.g., Johansson et al., 2017; Schwarzkopf et al., 2021). The bioaccumulation by key marine organisms of the most important pollutants originating from shipping is investigated both (i) using bioaccumulation models (including aquatic food webs) and (ii) experimentally at the case study locations.

The investigation of the most appropriate range of emission reduction strategies for the main critical pollutants will consider the impacts on both water and air in an integrated manner. The associated costs for investments are also estimated, as well as the efficacy, suitability and cost-effectiveness of each abatement method, which will be compared with other alternative strategies.

The particular case study locations that have been selected can also be considered representative of their broader regions in many respects (Fig. 1). All these areas are also exposed to substantial discharges and emissions from shipping to both water and air. The main aims of the work in the case studies are: (i) to use and assess the methodologies that have been developed or refined in EMERGE in the selected regions, and (ii) to evaluate the efficiency of various abatement measures in selected vulnerable marine regions in Europe.

The case studies include measurements regarding the bio-accumulation of pollutants, and the ecological and biological state of these environments. The idea of the several case studies is also to them could be compared, after this individual study/analysis.

2.2. The case study of Aveiro Lagoon

All the five selected geographical case studies of the EMERGE project are in vulnerable environments, which represent sensitive areas, such as estuaries, straits and enclosed waters. The case study explored in this study was done in the region surrounding the city and Lagoon of Aveiro. The city of Aveiro is located on the Atlantic coast of Portugal. The site is located in a very sensitive nature region, classified as a specially protected area by the EU nature and biodiversity policy 'Natura 2000 Network'.

The Aveiro Port's strategic location – at the west coast of the Iberian Peninsula (IP) facing the Atlantic Ocean (40°39′ N, 8°45′ W) - and its favourable maritime and land accessibilities, allow this infrastructure to serve a major economic hinterland in Portugal and central Spain. This port is the most recent national infrastructure, showing a well-ordered and integrated area, benefiting from the natural protection from the coastal hazards bestowed by the sandy barriers. The port structure lies inside Ria de Aveiro, and the connection between the port and the ocean is carried out by a single navigation channel protected by the construction of two breakwaters (Fig. 2).

In this Atlantic area, there is no current legislation in place for a sulphur emission control area (SECA). However, the use of open loop scrubbers is allowed neither in Aveiro ports, nor along the port channel or at berth (moored), until the ship leaves the port area. Only scrubbers with a closed loop operation are allowed.

Since this work is aimed to evaluate the impact of shipping activity over this study area in 2018, no scrubbers were considered in the air and water impact assessment. However, the ecotoxicological tests were performed in order to evaluate the potential impacts of the use of open loop scrubbers in the future. This could occur if the regulations in place in 2018 would be revised to be more stringent, to protect the population from an exposure to poor air quality.

The year of 2018 was selected as the target year, as that was the most recent year with relevant data available. We also did not wish to select a year that would have been influenced by the restrictions caused by the COVID-19 pandemic.

The methodology followed in this study and present in detail in the next sections is summarized in Fig. 3, which includes the impacts on air quality, water quality and ecosystem.

3. Impact on air quality

An urban scale modelling approach was used for the air quality impact assessment study, focused on the main atmospheric pollutants. The impact of shipping activities on the air quality of the region was assessed using two different simulations: with (Run 1) and without (Run 2) shipping emissions.

3.1. Modelling system and setup

The URBan AIR model (URBAIR), an advanced Gaussian model that has been enhanced with a set of functionalities, particularly treating road traffic emissions and 3D urban elements (Borrego et al., 2016; Dias et al., 2018; see Fig. 4), has been applied to model the impact of shipping on air quality in Aveiro. As a second-generation Gaussian model, URBAIR dispersion parameters continuously vary with the atmospheric stability, calculating meteorological parameters such as atmospheric turbulence characteristics, mixing height, friction velocity, Monin-Obukov length and surface heat flux. Since topography and build-up structure characteristics have an important role in the dispersion of atmospheric pollutants in urban areas, URBAIR considers the spatial variation of the terrain surface elevation, land use and buildings 3D coordinates, as well as emission source locations and dimensions.

This air quality model has already been extensively applied and validated over Portuguese areas, in particular over this study region (Rafael et al., 2021; Oliveira et al., 2022).

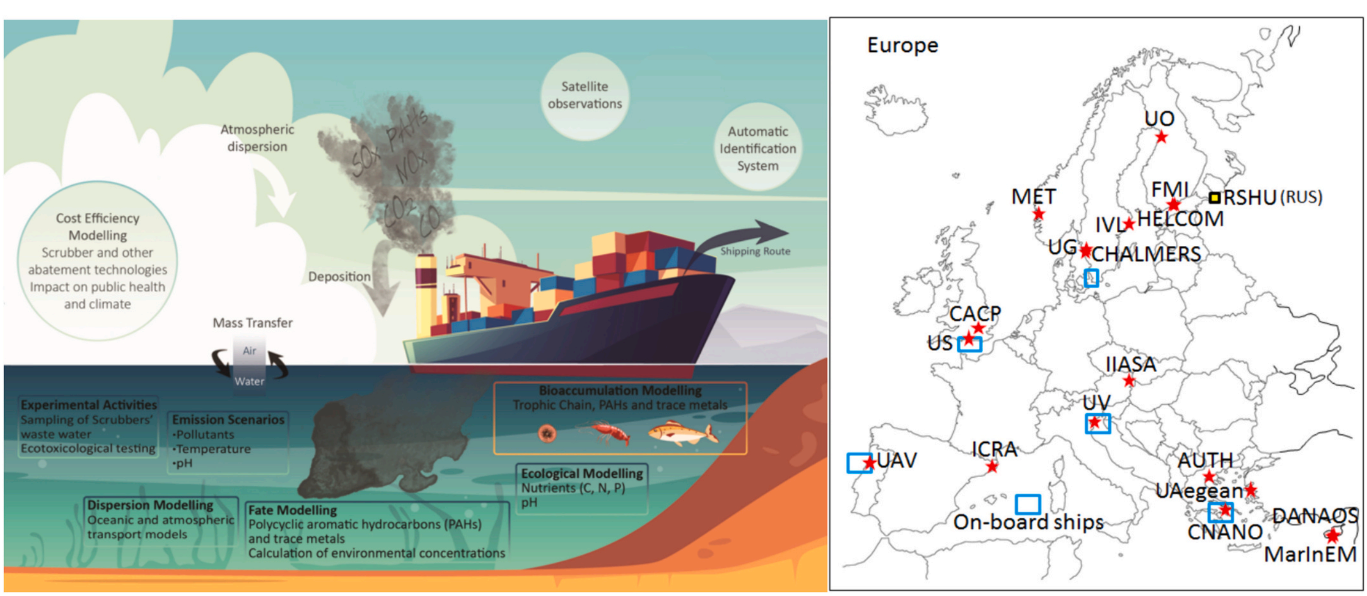

Fig. 1. A schematic diagram of the scope and main considerations of the EMERGE project (left) and the partners (red stars) and case study regions (blue rectangles, right).

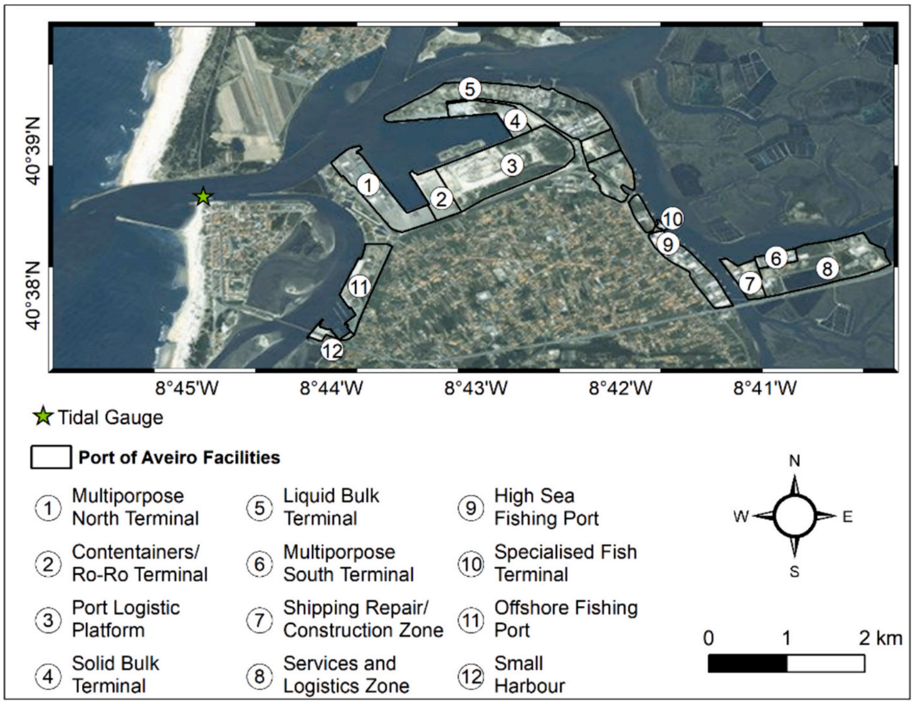

Fig. 2. The geography and the locations of the main terminal, platform and logistics installations in the Port of Aveiro (Ribeiro et al., 2021).

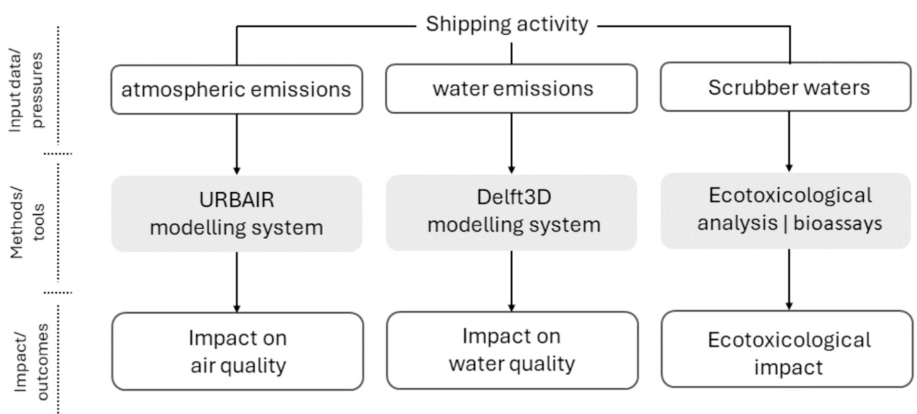

Fig. 3. The methodology followed in this study to achieve the impacts of shipping activity on air quality, water quality and ecosystem.

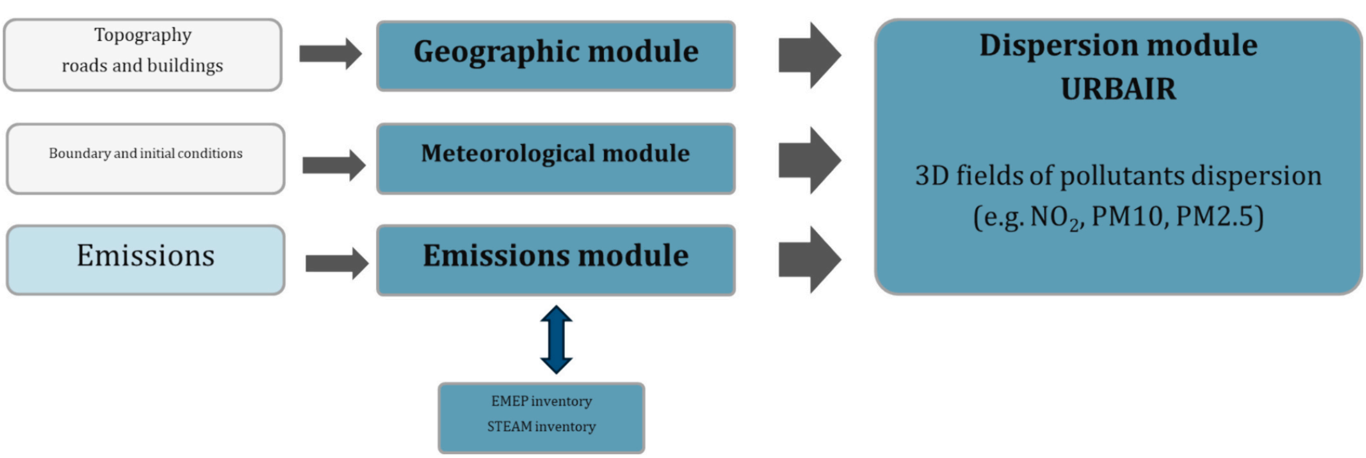

Fig. 4. A block diagram of the main components of the URBAIR modelling system.

The modelling system was applied for 2018, providing spatial distribution of NO_2, SO_2, PM10 and PM2.5 concentrations with 1 h of temporal resolution. The computational domain covers an area of about 36.5 km × 40 km (Fig. 5) over the Ria de Aveiro, with a horizontal resolution of 500 m. A dedicated high-resolution emission inventory of shipping was compiled using the STEAM model version 4.3. Ship traffic information is obtained from global AIS dataset by Orbcomm, which is combined with sea wave, sea current and wind data from Copernicus marine services and Copernicus climate data store. The spatial resolution of the ship emission inventory is 0.0060° x 0.0045° (lat, lon) and emissions are separated into two height layers (0-36 m, and 36 m or higher) based on the stack height of each ship. Temporal resolution of the inventory is 1 h. Other emission data were obtained from the EMEP emission inventory.

Initial meteorological conditions (with a temporal resolution of 1 h) were obtained from the meteorological model – the Weather Research and Forecasting Model (WRF), namely hourly surface data, as well as upper air soundings data. URBAIR meteorological pre-processor uses these data and generates hourly boundary layer parameters and multiple-level observations of wind speed and direction, temperature, and standard deviation of the fluctuating components of the wind.

3.2. *The results of air quality modelling*

Fig. 6 shows the annual averages of NO_2, SO_2, and PM2.5 concentrations as simulated by the URBAIR model. These concentrations include the contributions of the emissions from all activity sectors in the EMEP emission inventory, together with the shipping emissions computed using the STEAM model (Fig. 6). Additionally, the annual average concentrations attributable only to shipping emissions from STEAM are also shown. The maps also include the annual averages of the four monitoring stations within the study area. These 4 monitoring stations represent very distinct environments and influences. The station of HRB is in the surroundings of the Port of Aveiro; the stations of ILH and ETR characterize a suburban background environment, and the station of AVE is in an urban environment with intensive traffic influence.

The spatial distributions of the annual average concentrations highlight the hot spots of concentrations which coincide with the region's most densely populated areas. The shipping sector is responsible for a fairly large contribution for some pollutants. In more detail, maximum annual average concentrations simulated within the domain reach 40, 51 and 37 $\mu g.m^{-3}$ for NO_2, SO_2 and PM2.5, respectively. An annual average maximum concentration of 2.5, 0.3 and 0.1 $\mu g.m^{-3}$ for NO_2, SO_2 and PM2.5 originates from shipping activities. This corresponds to a maximum contribution of shipping activities on an annual average basis around 34 % for NO_2, 31 % for SO_2 but below 5 % for PM2.5. These results are in agreement with the results and conclusions addressed at a previous national study by Russo et al. (2018).

The annual mean modelling results approximately agree with the annual mean observations (circles), except for the Aveiro monitoring site (AVE). This site is exposed by dense vehicular traffic influence, and the model grid resolution is not sufficiently fine to allow for these effects.

4. Impact on water quality

To assess the total impact of shipping on the marine environment it is essential to address the entire load of different stressors originating from different subsystem waste streams, along with an assessment of the load of species reaching the marine environment through atmospheric deposition of the shipping air pollutants. This was done using a numerical modelling system that has been previously applied and tested over this specific study area (Dias et al., 2021).

4.1. *Modelling system and setup*

The pollutant's fate in the water was simulated by applying a transport model based on the Delft3D model suite (hydrodynamic Delft3D-FLOW and Delft3D-WAQ water quality modules) (Deltares, 2021; Deltares, 2014). The flow model was used to predict hydrodynamics, while the water quality module allows to evaluate trace metals (Cadmium – Cd and Lead - Pb).

The model was set up with a curvilinear orthogonal grid with 193 × 458 cells resulting in a solution computed in 18,330 elements. Bathymetric data from a general survey performed by the Hydrographic Institute of the Portuguese Navy (HIPV) in 1987/88, data collected in 2011 by Polis Litoral Ria de Aveiro (PLRA) in the main channels of the lagoon, and in 2020 by Aveiro Port Administration (APA) and University of Aveiro (Cavalinhos et al., 2020) in the lagoon inlet and port area,

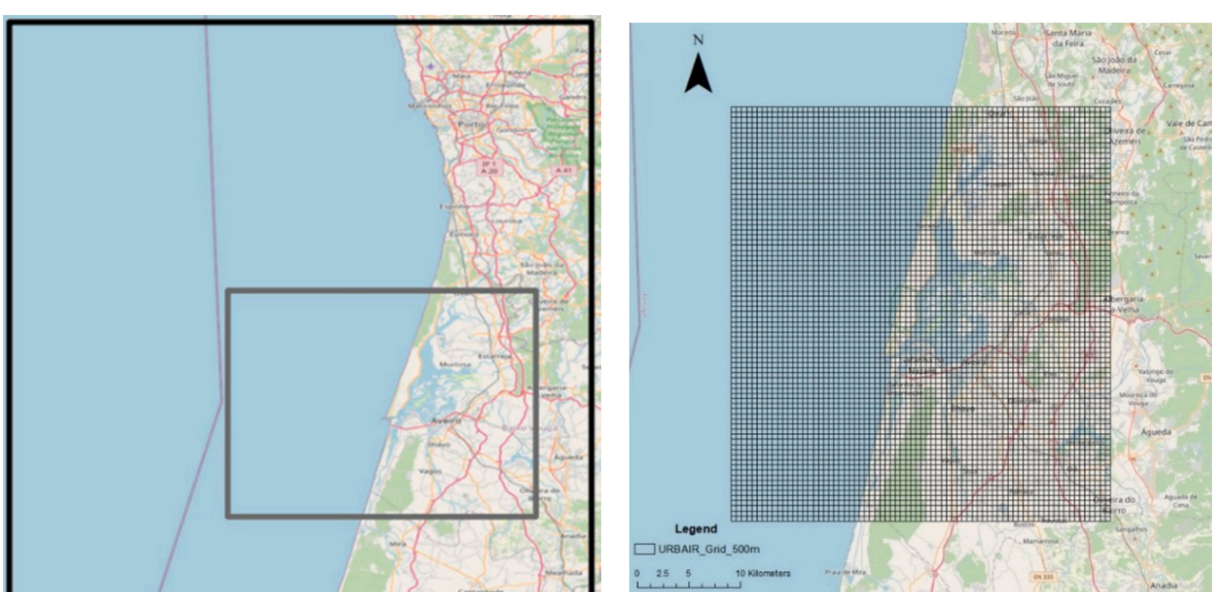

Fig. 5. a-b. The modelling domain of the Aveiro case study. The panel (b) on the right-hand side illustrates the area of the smaller rectangle in the left-hand side panel (a).

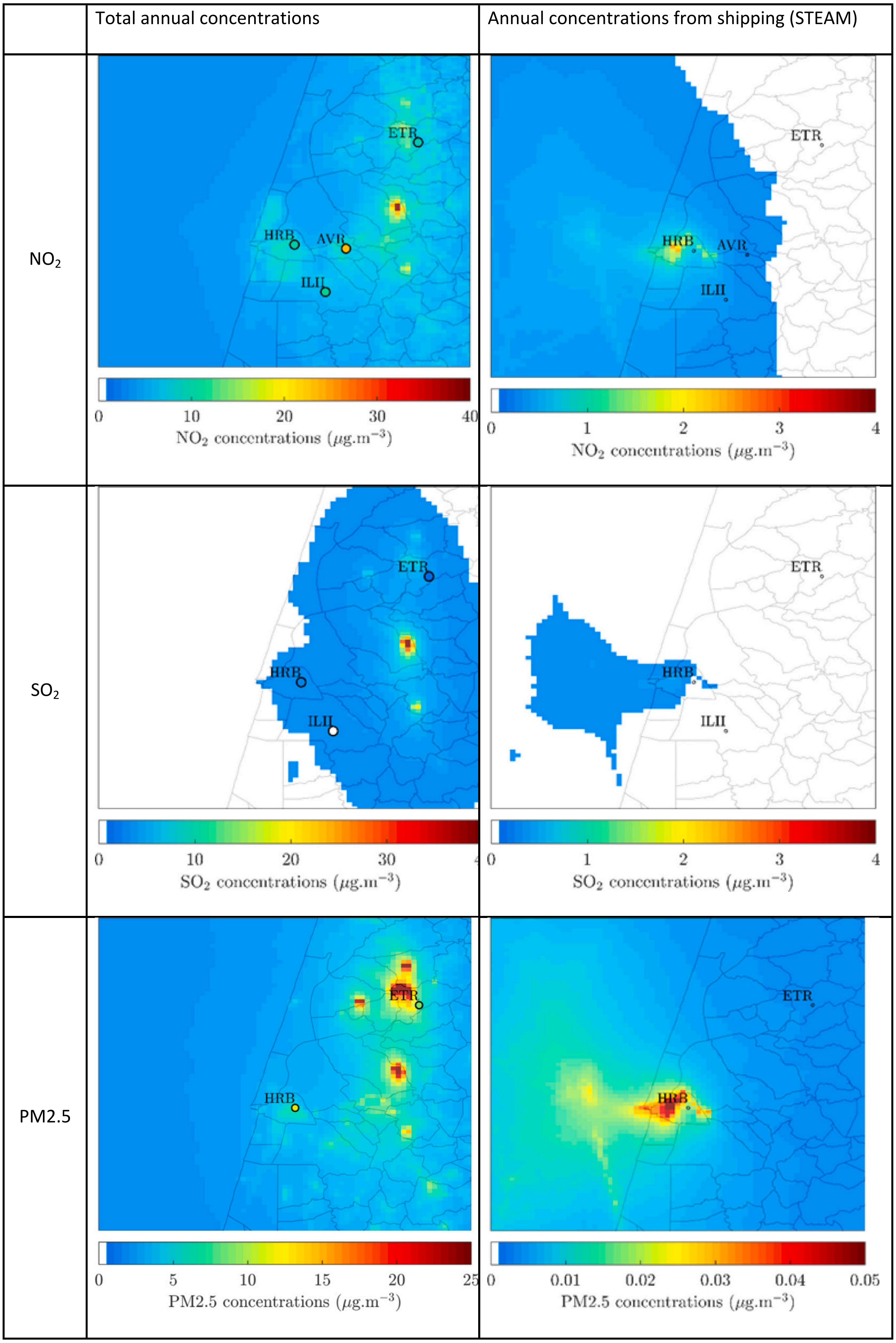

Fig. 6. NO_2, SO_2 and PM2.5 annual average concentrations modeled for the Aveiro case study. The dots represent the annual averages of the monitored concentrations at the air quality monitoring stations. The left-side maps show the total annual averaged concentrations, considering all the emission sectors, including shipping, while the right-side maps show the annual averaged concentrations associated with emissions from shipping (STEAM inventory).

were used to construct the numerical bathymetry of the lagoon. Data were interpolated for the grid through a triangular interpolation (Fig. 7). Because of the shallowness of the lagoon and the low daily freshwater input when compared with the total tidal prism, the vertical gradients in the lagoon can be neglected when compared to the longitudinal gradients (Vaz et al., 2009). Therefore, a single vertical layer was considered in the scope of this work.

The open ocean boundary was forced with harmonic constants (amplitude and phase of 19 harmonic constituents: M2, S2, ETA2, K2, L2, LABDA2, NU2, N2, MU2, 2N2, FI1, K1, P1, O1, Q1, SIGMA1, 2Q1, MSF, SSA) obtained from the harmonic analysis performed to observations at a tide gauge (RA1) in 2002/2003 (Fig. 7). A correction factor was considered taking into account the distance between the boundary and the tidal gauge (Lencart e Silva et al., 2013; Vaz et al., 2005). Temporal series of salinity, water temperature, dissolved oxygen, chlorophyll and nutrient concentration from the model Iberian Biscay-Ireland Regional Seas from the Marine Copernicus database (daily temporal resolution) was also imposed at the open ocean boundary. Regarding trace metals and PAHs, no data is available in the open ocean, therefore emissions from sources other than shipping will not be considered.

For the five main lagoon rivers (Fig. 7), inflow, water temperature, nutrient concentration and dissolved oxygen predictions are derived from the watershed model Soil and Water Integrated Model (SWIM) (Stefanova et al., 2015). Continuous data regarding Cd and Pb concentrations for rivers are not available and therefore residual concentrations were imposed according to a few specific measurements carried out in 2000 by Sistema Nacional de Informação dos Recursos Hídricos (SNIRH) and according to a monitoring report by MONITAR, published in 2019 (MONITAR, 2019) as part of POLIS Litoral Ria de Aveiro, which evaluated the ecological physical and chemical status of Ria de Aveiro waters. Results showed that the chemical status of most of the locations analyzed is Excellent to Good, presenting values below the quantification limits for the different substances, including Cd and Pb.

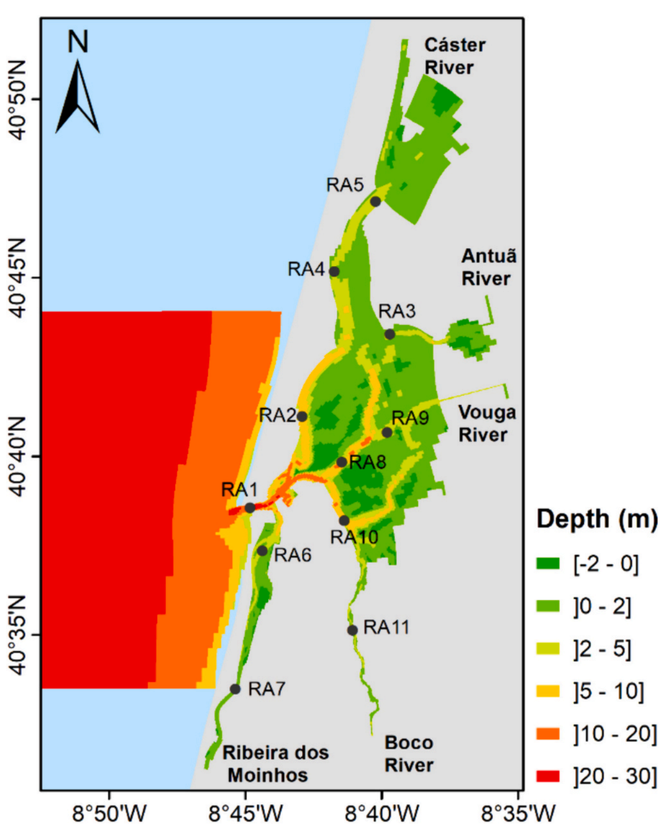

Fig. 7. Numerical bathymetry of the Ria de Aveiro with the location of the main channels, rivers and sampling stations.

For the atmospheric interface, the "absolute flux, net solar radiation" heat flux model was used. This model specifically accounts for the combined net solar radiation (short wave) and net atmospheric radiation (long wave). Additionally, the model computes terms related to heat losses resulting from evaporation, back radiation, and convection (Deltares, 2021). It was applied considering surface air temperature, relative humidity, and net solar radiation data to calculate heat losses from convection, evaporation, and back radiation. Also, wind intensity and direction were applied at the surface boundary. Atmospheric data (with a temporal resolution of 3 h) was obtained from the Weather Research and Forecasting Model (WRF) developed for the study region and also used for the meteorological forcing of the air quality modelling. Atmospheric deposition sources of Pb and Cd predicted by SILAM (System for Integrated modeLling of Atmospheric coMposition), with a spatial resolution of 5 km, were also considered.

A computational time step of 0.5 min for the FLOW module and 2 min for the WAQ were chosen to guarantee the stability and accuracy of numerical results. In addition, the background horizontal eddy viscosity and diffusivity were set to 0.5 and 10 m^2s^{-1}, respectively.

Regarding shipping emissions from open/closed loop scrubbers, trace metals (Cd and Pb) from the ChemicalDrift estimated by the Norwegian Meteorological Institute were used as ocean boundary conditions (Aghito et al., 2023). Additionally, the mass of wastes discharged directly from vessels to the water column from five waste streams: open/closed loop scrubber, grey, black and ballast water provided by Potiris et al. (2023) were included in the model. Discharge rates from ships were modeled using the STEAM model version 4.1 with temporal resolution of 1 h and spatial resolution of 0.0025° x 0.0025°. More detailed description of the modelling of discharges from shipping can be found in Jalkanen et al. (2021). Atmospheric deposition from shipping of Cd and Pb predicted by SILAM model is considered.

To investigate and quantify the potential impacts of shipping emissions to water on the aquatic environment of Ria of Aveiro in terms of trace metals, two simulations were carried out, differing in the atmospheric data imposed. For Run 1, the atmospheric deposition values for Cd and Pb encompass contributions from both shipping activities and other non-shipping-related sources. On the other hand, Run 2 only considers the atmospheric deposition contribution from shipping. For both simulations, open ocean boundary data of metals was provided by ChemicalDrift and mass fluxes from the five waste streams by Potiris et al. (2023), as previously referred.

Water level, temperature and salinity were validated through the comparison of in situ data with model predictions in Dias et al. (2021). Generally, a good fit was found between model predictions and observations.

For both trace metals (Cd and Pb) a field campaign was carried out between October 17 and October 19, 2022, and water was collected at 11 stations throughout the lagoon (Fig. 7). The water analysis resulted in concentrations below the equipment detection limit for all metals.

4.2. Water quality modelling results

In Fig. 8, the annual average and maximum concentrations of cadmium (Cd) and lead (Pb) are presented for Run 1 and Run 2. In general, considering Run 1, model results show that the highest average concentrations of Cd and Pb occur in the upstream regions, with values higher than 4×10^{-5} and 1×10^{-3} μg.L^{-1} in the head of the S. Jacinto channel (northern channel), respectively. In the head of Mira and Ílhavo channels (southern channels), the annual average concentrations are approximately 3×10^{-5} μg.L^{-1} for Cd and higher than 0.9×10^{-3} μg.L^{-1} for Pb. In the central region, the annual average Cd and Pb concentrations are between 0.6 and 1.8×10^{-5} μg.L^{-1} and 0.1 and 0.6×10^{-3} μg.L^{-1}, respectively. Maximum concentrations of Cd and Pb are observed in the intertidal regions with values higher than 1.7×10^{-3} μg.L^{-1} and 3.2×10^{-2} μg.L^{-1}, respectively. In the channels of the central region, the

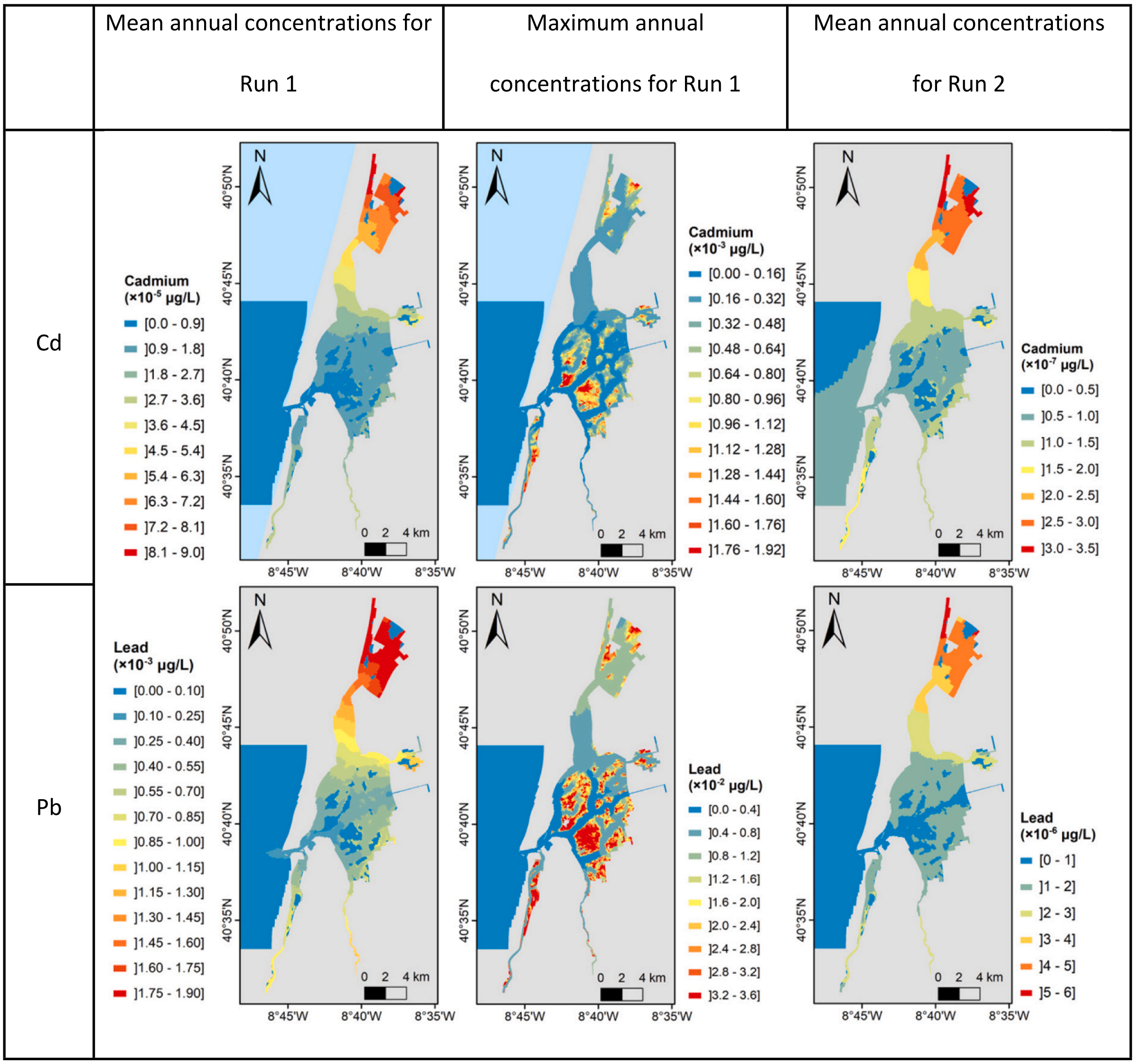

Fig. 8. Predicted annual average and maximum concentrations (μg/L) of Cd and Pb for Run 1 and annual average concentrations (μg/L) of Cd and Pb for Run 2.

maximum concentration reached is lower than 0.16×10^{-3} μg.L^{-1} and 0.4×10^{-2} μg.L^{-1}.

These mean annual concentrations of Cd and Pb remain notably lower than the environmental quality standards defined for Portuguese transition waters which are 0.15 μg.L^{-1} and 7.2 μg.L^{-1} (Decree-law n.° 218/2015, Annex II of the n. ° 1 Article 1°), respectively. The maximum concentration allowed for Cd is set at 0.9 μg.L^{-1} and according to model results the maximum concentration predicted is significantly below this limit for all the lagoon. For Pb, the maximum threshold is not applicable. Therefore, Cd and Pb meet the regulatory thresholds, indicating a relatively lower environmental impact and safe conditions for aquatic ecosystems and human activities.

The results of the simulations only considering the shipping emissions (Run 2 – Fig. 8) suggest that the atmospheric deposition is the main source of Cd and Pb and that the shipping contribution remains negligible in comparison to other contributing factors (Fig. 8). Comparing the results between both simulations, the concentrations of Cd and Pb in Run 2 are 98–99 % lower when compared to Run 1. This suggests that shipping emission data used to force the model show low concentrations of these trace metals. These low concentrations occur since only closed-loop operation is allowed in the port of Aveiro throughout the entire duration of a ship's presence, including its entry, transit within the port channel, berthing, and departure.

5. Ecotoxicological impacts

The use of scrubbers has facilitated a reduction in pollutant emissions into the atmosphere. This has transferred this problem to seawater, but the impact of these SW (scrubber waters) on marine chemistry, biodiversity and geochemical processes is not well known (Koski et al., 2017, Teuchies et al., 2020, Thor et al., 2021, Picone et al., 2023, Genitsaris et al., 2023, etc). These studies have reported an increase in the mortality of marine zooplankton (Jönander et al., 2023), a delay in larval development and a reduced feeding attributed to the synergistic effects of metals, PAHS, alkylated-PAHs and other constituents of scrubber-water discharge.

To bridge this knowledge gap, this study aims to assess the ecotoxicological effects of SW on selected estuarine marine species, which are important in the case study area. The selected species included the mussel *M. galloprovincialis*, the sea urchin *P. lividus*, the polychaeta *S. alveolata* and the small crustacean *Artemia* sp.

Three scrubber water samples (called here as SWA, SWB and SWC) were produced with Atlantic seawater from opened loop systems; the SWA was artificially produced from a pilot system at Chalmers University of Technology, Sweden, and SWB and SWC were produced on board two different ships (DANAOS Shipping co. LTD). The SWs were employed in ecotoxicological testing using a gradient of SW concentrations (0, 0.001, 0.01, 0.1, 1.0, 10.0 %). Several parameters were assessed, including fertilization, larval development, and post-exposure feeding inhibition. These parameters, linked to reproduction and feeding, serve as ecotoxicological measures, which connect individual physiological responses to effects at the population and community levels (Ré et al., 2020; Re et al., 2021). Furthermore, they have ecological significance and sensitivity, and are linked to essential ecosystem functions (Agostinho et al., 2012). Additionally, these tests can discern the impacts of contaminants in both the dissolved and particulate fractions of the environment.

The scrubber-waters were analyzed for chemical compounds, namely metals (analyzed using a SeaFAST pre-concentration device and an Agilent 7900 ICP-MS), the 16 US EPA priority PAHs and 25 representative alkylated PAHs (analyzed using a methodology based on headspace solid-phase microextraction (HS-SPME), equipped with a polydimethylsiloxane (PDMS, 100 μm) fiber coupled to gas chromatography-tandem mass spectrometry (GC–MS/MS)) (Picone et al., 2023). While the sample SWC exhibited the highest concentrations of PAHs and alkylated PAHs in both dissolved and particulate fractions, the sample SWA had the highest metal concentrations, including Cr, Mn, Fe, Co, Ni, and Cu. PAHs and metals affect the growth, feeding and reproduction of marine biota, can accumulate through food chains reaching humans, and are persistent in the marine environment.

The predictive laboratory bioassays indicated that the sea urchin larval development test was more sensitive than the fertilization test (Fig. 9), with NOEC (No observed effect concentration) values of 0.01 % for the sample SWC and NOEC<0.001 % for the samples SWA and SWB.

For SWC the LOEC (lowest observed effect concentration) was equal

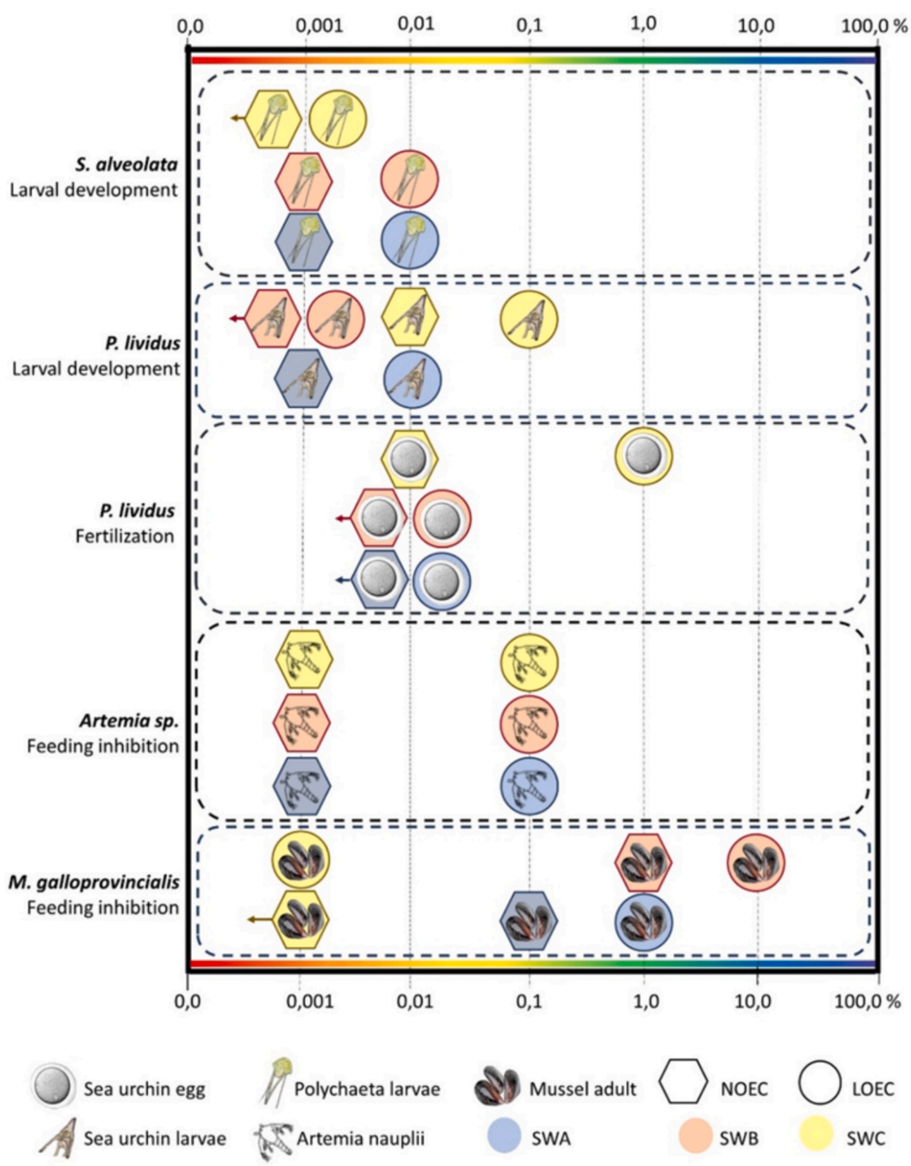

Fig. 9. NOEC (No observed effect concentration ⬡) and LOEC (Lowest observed effect concentration ◯) values obtained with three different scrubber water samples (marked as SWA ⬡ or ◯; SWB ⬡ or ◯ and SWC ⬡ or ◯) in a: fertilization success bioassay with the sea urchin *lividus;* larval development bioassay with the sea urchin *P. lividus;* polychaeta *S. alveolata and* post-exposure feeding inhibition bioassay with *Artemia* sp. nauplii and mussel adults *M galloprovincialis.*

to 1.0 % while for SWA and SWB the LOEC was 0.01 %. In the sea urchin larval development test, the values obtained were 0.001 % $\geq$ NOEC $\geq$0.01 % and 0.001 % $\geq$ LOEC $\geq$0.1 % (see Fig. 9). This assay indicates a heightened sensitivity for SWB and comparatively lower sensitivity for SWC, consistently aligning with findings from the fertilization bioassay involving the same species.

The larval development bioassay with the polychaeta *S. alveolata* presented values of NOEC = 0.001 % and 0.001 % $\geq$ LOEC $\geq$0.01 %. These values were lower than those obtained with *P. lividus* in the same type of bioassay. Thus, the larval development test with the polychaeta *S. alveolata*, revealed that SWC was the most toxic with a LOEC of 0.001 %.

Likewise, the post-exposure feeding inhibition bioassay with *M. galloprovincialis* also showed to be more sensitive to SWC with a NOEC <0.001 % and LOEC = 0.001 %. However, for SWA the values of NOEC and LOEC (0.01 % and 1.0 %, respectively) were much higher than for SWC. The mussel showed to be less sensitive to SWB with a NOEC = 1.0 % and LOEC = 10.0 %. Conversely, *Artemia* sp., in the post-exposure feeding inhibition bioassay, showed similar values for all three scrubber-waters tested (NOEC = 0.001 % and LOEC = 0.1 %) (Fig. 9).

These findings align with the concentrations of metals, PAHs, and alkylated PAHs analyzed in the three scrubber-waters. It is crucial to emphasize that mixtures of substances are widely acknowledged to induce, at a minimum, additive toxicity and occasionally, synergistic responses (Cedergreen, 2014). Even if each individual substance in the SW is below its respective NOEC, the presence of a mixture can results in significant toxicity (Walter et al., 2002). Empirical support for this additive toxicity is evident in a study involving a mixture of several PAHs, including those found in scrubber-water, which adversely affected the small crustacean zooplankter *Oithona davisae* (Barata et al., 2005).

In summary, these results from the ecotoxicity assays clearly demonstrate the high toxicity of SW for the marine and estuarine species under investigation. These responses are closely linked to the differing contaminant concentrations presented in the three types of scrubber water samples.

Moreover, this study has revealed clear responses among parameters and species; post-feeding inhibition was proved to be an exceptionally sensitive parameter. This emphasizes the importance of selecting appropriate species, their developmental stages and toxicological measures to achieve reliable results.

The NOEC and LOEC values obtained in this study were remarkably low (NOEC <0.001 % and LOEC = 0.001 %). In numerous ports, where only ships equipped with closed-loop scrubber systems are permitted, SW discharges into the open sea could potentially be a problem.

6. Discussion and conclusions

This study is the first holistic evaluation on the impacts of shipping activities on the environment in the sensitive and vulnerable area of the Aveiro Lagoon, presenting a methodological approach valid for each compartment that can be reproduced by other case studies. Both the impacts on air and water were evaluated using various numerical modelling systems, and ecotoxicological tests were performed for three collected samples of scrubber-water. In the case of both air and water, the modelling tools allowed simulating the contribution of each emission source category on the final environmental state. Regarding the ecotoxicological impacts, the most critical pollution scenario – that of the discharged scrubber-waters – was evaluated using water samples obtained directly from ships equipped and operating with scrubbers. Bioassays were determined for marine invertebrates, crustacean and bivalve species.

The modelling results showed that the ongoing shipping activities were responsible for a maximum concentration of 30 % of atmospheric pollutants in case of NO_2, SO_2 and PM2.5. However, the impacts of shipping on the water quality – regarding the concentrations of heavy metals (Cd and Pb) – were found to be non-significant in 2018.

Although this study found no impacts of shipping on the marine ecosystems in this region in 2028, shipping activity would probably cause effects on marine ecosystems, if scrubber-water would be discharged in this area. The ecotoxicological bioassays performed on the most sensitive stages of marine invertebrates, and on the post-exposure feeding inhibition of crustacean and bivalve species revealed lowest observed effect concentration (LOEC) for the different scrubber-waters, as well as regarding the concentrations of metals, PAHs, and alkylated PAHs.

Regarding the sampling, processing and analysis of scrubber water and the ecotoxicological analyses, the methodologies should be harmonised in the future. In particular, a harmonisation or at least good practise guidelines would be needed for the selection of key marine species of biota and their developmental stages to be used in toxicological analyses. This will be needed to avoid potentially conflicting or contradictory results, which would have been caused by widely differing methodologies.

In summary, the results highlight the need of more research on the impact of scrubber-waters discharges in this region and in other vulnerable regions worldwide. The abatement of the emissions by scrubber technologies can result in substantially increased discharges to the sea. Considering only the emissions to air or the discharges to sea can therefore lead to entirely misleading results and misinformed policy decisions. This study has highlighted the need for integrated, holistic assessments of the environmental impacts of shipping. Although EU efforts towards reaching environmental goals for shipping have been advancing with intermediate policies, such as GHG emission reporting for large ships and the recent inclusion of maritime emissions in the EU Emissions Trading System (ETS), there are no integrated policies in the shipping sector that have a holistic approach to the issues of environmental pollution. Effective policy measures could be defined and implemented that are not only focused on the impacts on air or water, but the entire ecosystem, considering a balance of the impacts to multiple types of environments. This would reduce the risk associated with policies that focus on one sub-system such as atmospheric pollutant emissions potentially having detrimental effects on, for example, water quality.

The main limitations of this study are related to the fact that different compounds/pollutants were analyzed in distinct environmental compartments, and no common species were investigated among them. Future work should focus on this integration, with more scenario data to test different fuels and ship configurations. Finally, these impacts should also be quantified under future climate change scenarios, since efforts towards reducing pollutant emissions from the shipping sector could be countered by the effects of climate change.

CRediT authorship contribution statement

A. Monteiro: Writing – original draft, Project administration, Methodology, Investigation, Funding acquisition, Formal analysis, Conceptualization. **V. Rodrigues:** Writing – original draft, Investigation, Formal analysis, Data curation. **A. Picado:** Writing – original draft, Methodology, Investigation, Formal analysis, Data curation. **J.M. Dias:** Supervision, Project administration, Conceptualization. **N. Abrantes:** Project administration, Methodology, Investigation, Formal analysis, Data curation. **A. Ré:** Writing – original draft, Methodology, Investigation, Formal analysis, Data curation. **M. Rosa:** Data curation. **M. Russo:** Writing – review & editing, Methodology, Investigation. **A. Barreirinha:** Software, Data curation. **M. Potiris:** Software, Investigation, Data curation. **M. Aghito:** Writing – review & editing, Software. **R. Hänninen** Data curation, Writing – review & editing. **E. Majamäki:** Writing – review & editing, Investigation, Data curation. **T. Grönholm:** Investigation, Data curation. **U. Alyuz:** Writing – original draft, Data curation. **R. Sokhi:** Supervision, Project administration, Methodology. **J. Kukkonen:** Writing – review & editing, Project administration, Methodology, Conceptualization. **J.-P. Jalkanen:** Supervision, Project

administration, Investigation, Conceptualization.

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.

Data availability

Data will be made available on request.

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