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Review

The Impact of Shipping on Air Quality in the Port Cities of the Mediterranean Area: A Review

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Abstract: Shipping emissions contribute significantly to air pollution at the local and global scales and will do so even more in the future because global maritime transport volumes are projected to increase. The Mediterranean Sea contains the major routes for short sea shipping within Europe and between Europe and East Asia. For this reason, concern about maritime emissions from Mediterranean harbours has been increasing on the EU and IMO (International Maritime Organization, London, UK) agenda, also supporting the implementation of a potential Mediterranean Emission Control Area (MedECA). Many studies are concerned with the impact of ship emissions in port cities. Studies of the contributions of ship emissions to air quality at the local scale include several monitoring and modelling techniques. This article presents a detailed review of the contributions of ship emissions of NO_2, SO_2, PM_{10}, and $PM_{2.5}$ on air quality in the main ports in the Mediterranean area. The review extracts and summarises information from published research. The results show a certain variability that suggests the necessity of harmonisation among methods and input data in order to compare results. The analysis illustrates the effects of this pollution source on air quality in urban areas, which could be useful for implementing effective mitigation strategies.

Keywords: air quality; Mediterranean area; ship emissions; NO_2; SO_2; PM

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1. Introduction

Maritime transport plays a fundamental role in the international transport of goods worldwide; in fact, sea transport accounts for 80% of goods transported, moving 10 billion (bn) tonnes of cargo annually [1]. Recent estimates foresee a growth in maritime transport activities of almost 40% for seaborne trade by 2050 [2]; consequently, greenhouse gas (i.e., GHG) emission levels in 2050 will rise to 90–150% of 2008 levels according to the fourth global IMO Greenhouse Gas (GHG) study [3]. With the growth of the shipping industry, air pollution from shipping has become an increasingly serious concern for both environmental quality and human health, especially in coastal regions [4–7]. The main pollutants emitted by maritime transport are primary and secondary particulate matter (PM_{10} and $PM_{2.5}$), black carbon (BC), sulphur dioxide (SO_2), nitrogen oxides (NO_x), non-methane volatile organic compounds (NMVOCs), and carbon dioxide (CO_2) [8–17]. Ocean-going vessels are responsible for approximately 15% of global anthropogenic NO_x emissions and 5–8% of global SO_x emissions [8,9]. Furthermore, it is estimated that nearly 70% of ships' emissions occur within 400 km of land [5]. As pollutants can be transported hundreds of kilometres towards the mainland, ships may contribute to air quality degradation in coastal areas, as well as inland areas [8]. The transport and dispersion of pollutants emitted by ship emissions in coastal areas are significantly influenced by several factors, including wind direction [18]. While the exact influence radius can vary, different studies have shown that the impact of shipping emissions can extend beyond the immediate port area and affect surrounding regions. Chen et al. [19] in their study have observed a downward trend in the contributions of shipping emissions to $PM_{2.5}$ as distance increased. The decline was steeper in closer proximity compared to farther distances. For instance, the annual average contribution at a distance of 200 km away from the coastline was rounded to 1.0%. Liu et al. [20] reported higher contributions of ship-contributed $PM_{2.5}$ in an urban area within a distance of 15 km from the coast. In East Asia, shipping contributes 16% of global shipping CO_2 and NO_x, respectively, and 19% of SO_2 [6]. In the Pearl River Delta (PRD) region in China, shipping emissions contribute 7% and 12%, respectively, of total $PM_{2.5}$ and O_3 [21]. Broome et al. [22] estimated that shipping in the Sydney Greater Metropolitan Region (GMR) contributes 5.7% of total $PM_{2.5}$. Crippa et al. [23] estimated that shipping accounted for 0.2% of Indian emissions, 0.3% of Chinese total emissions, 7.4% of African total emissions, 4.2% of North American total emissions, 2.6% of South American total emissions, 21.8% of Oceanian total emissions, and 4.4% of European total emissions. In Europe, shipping emissions contribute to 7–24% of NO_2 levels and to 1–14% of $PM_{2.5}$ levels in coastal zones [24]. Barregard et al. [25] estimated that shipping emissions accounted for over 50% of NO_2 in central parts of the Baltic Sea and for 20–50% in adjacent coastal areas. Tang et al. [26] estimated that regional shipping accounted for 11% of total $PM_{2.5}$ and 26% of total NO_2 in Gothenburg. Jonson et al. [27] estimated that shipping $PM_{2.5}$ contributions to total $PM_{2.5}$ ranged up to 15% for 12 European countries. Shipping emissions contributed to 45% of NO_x in the western part of the Mediterranean area [28]. The Mediterranean Sea contains one of the main sea routes connecting Europe and Asia. In addition to the North Sea, the Mediterranean Sea represents the region in Europe with the greatest contribution of ship emissions to gaseous pollutants [24]. The Mediterranean region stands out for significant growth in the number of ships calling at ports between 2011 and 2016 (Spain, 9.7%; Croatia, 20.1%; Malta, 14.8%; and Cyprus, 11.1% between 2015 and 2016) compared to northern ports (Belgium, −8.7%; Denmark, −23.2%; and Germany, −0.5%), also due to the relevant share (36%) of total cargo handled by EU28 ports in 2016 [29]. Several authors have studied the impact of shipping emissions on air quality [30–38] and on human health [29,39] in European coastal areas and port cities. Due to the known adverse effects of shipping emissions on human health and on the environment, the International Maritime Organization (IMO) regulates air pollution from shipping through Annex VI (Regulation for the Prevention of Air Pollution from Ships; [40]) of MARPOL (Marine Pollution Convention). The main regulations of Annex VI are regulations 13 and 14, regarding, respectively, NO_x and SO_x emissions from marine diesel engines. Regulation 13 limits NO_x emissions from all medium diesel engines (MDEs) installed on ships constructed on or after 1 January 2000. It also applies to engines of the same power that were subject to "major conversion" on or after 1 January 2000 [41]. The revision of Annex VI in 2008 significantly tightened the NO_x emissions allowed by introducing two additional limits of control which apply to MDEs installed on newer ships. These levels of control, known as Tiers, based on a ship's construction date, depend on an engine's rated speed. The current IMO limits for all engines constructed on or after 2011 are expressed by Tier II, while the Tier III standard is for new engines built since 2016 entering into any NECAs (NO_x Emission Control Areas). The latter standard reduces NO_x emissions by 80% compared to the Tier I limit. Regulation 14 of Annex VI controls the SO_x and PM emissions from ships and applies to all Marine Fuel Oils (MFOs) used on board ships. The sulphur content limits allowed in MFOs, expressed as a percentage of the mass fraction, have been progressively reduced in recent years. The current regulation 14 defines that, after 2020, the sulphur content of fuels used on board ships must be equal to or less than 0.5% (compared to 3.50% m/m (mass to mass)) for ships operating outside Emissions Control Areas. An ECA is a specific sea area, including port areas, designated by the IMO, in which more stringent emission regulations have been established. For the purpose of regulation 13, Tier III emission standards, and regulation 14 requirements on SO_x emissions limits, there are currently four designated ECAs: (i) the Baltic Sea Area, (ii) the North Sea Area, (iii) the North American Sea Area, and (iv) the US Caribbean Sea Area. The Convention for the Protection of the Marine Environment and the Coastal Region of the Mediterranean (Barcelona Convention) of the Conference of the Parties (COP 22) was presented by all the States bordering the Mediterranean, together with all the Member States of the European Union, and the European Commission designated the Mediterranean Sea as a whole as a Sulfur Oxide Emission Control Area (Med SO_x ECA) in accordance with regulation 14 of Annex VI of the MARPOL. The proposal was approved during the 78th Session of the Marine Environment Protection Committee (MEPC 78) of the International Maritime Organization (IMO), which met from 6 to 10 June 2022. The designation of the Mediterranean Sea as a SECA and/or NECA would socioeconomically benefit both the health and quality of life of European citizens and the economy. Studies indicate that the potential benefits of the ECA Med SO_x are considerable given the depth of cuts in sulphur oxides (SO_x) and particulate matter emissions that will result. Indeed, limiting the sulphur content in fuel oil used on board vessels operating within the Med SO_x ECA to 0.10% m/m—or one fifth of the current global legal limit—would result in a drop in SO_x emissions of 78.7%. Furthermore, the Med SO_x ECA would reduce particulate matter ($PM_{2.5}$) emissions by 23.7%. A recent report [42] tried to assess the feasibility and potential benefits of the implementation of a NECA or/and a SECA in the Mediterranean Sea. The health benefits (as in avoided premature deaths) of such implementations were calculated as increasing by more than an additional third compared to the impact of the 2020 sulphur regulation (with Algeria, Egypt, Italy, and Turkey as the main beneficiaries), estimated at nearly 1730 avoided premature deaths per year.

In this context, this paper aims to review the current knowledge of the impact of Mediterranean Sea harbours on air pollution due to ship emissions. The focus is to summarise and discuss the main findings from the available literature about the relative influence of shipping on atmospheric pollutants (gases and particulate matter) that could cause environmental and health issues. A deep state-of-the-art analysis of air quality near Mediterranean harbour areas is provided, although different contribution estimation methodologies have been applied. The paper is organised as follows. A description of the area study is reported in Section 2.1. The research methodology is described in Section 2.2, and the different approaches to assessing the contribution of ship emissions to air quality are described in Section 2.3. The results are reported in Section 3, and a discussion and conclusion are presented in Section 4.

2. Materials and Methods

2.1. Study Area

The Mediterranean Sea is located between three continents: Europe, Asia, and Africa. This explains the origin of the name, which in Latin means "in the middle of the lands". From a geographical point of view, the Mediterranean is an inland sea that is almost totally closed: in fact, it communicates with the Atlantic Ocean only, to the west, through the Strait of Gibraltar (the southernmost tip of Spain). It extends to about 2,505,000 km^2 (Figure 1); its maximum length, from west to east, is 3860 km (excluding, to the east, the foothills of the Black Sea and the Marmara Sea); and the maximum width from north to south is 1800 km. It has an average depth of 1430 m and a maximum depth—reached off the Peloponnese—of 5121 m. The Mediterranean Sea is bordered by 22 countries, overall accounting for more than 542 million inhabitants in 2020, or ~6–7% of the total world population. These values rank the Mediterranean basin among the most populous regions in the World, akin to the population density found in the Indian subcontinent or in the southeast of China. Moreover, the population is predicted to reach 657 million by 2050 [43].

The Mediterranean region's population is also concentrated near the coasts. The population of the coastal regions grew from 95 million in 1979 to 143 million in 2000 and could reach 174 million by 2025 [44]. Furthermore, the Mediterranean basin has experienced a rapid growth in urbanisation (urban population—towns with more than 10,000 inhabitants—increased by 1.9% per year during the period of 1970–2010, from 152 million to 315 million).

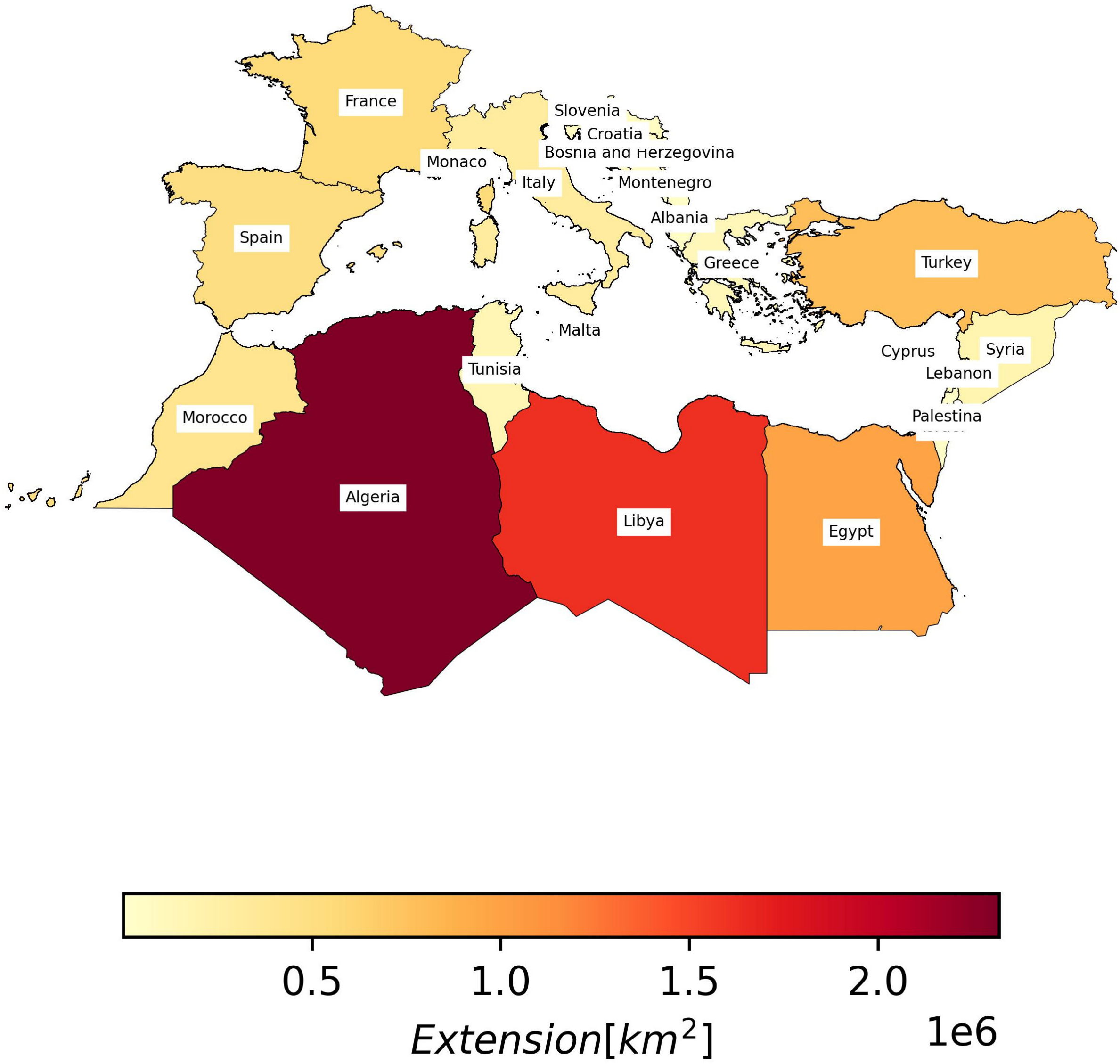

Figure 1. Extent of the countries in Mediterranean area.

The Mediterranean basin is characterised by complex meteorology, which favours polluted air masses' aging [45,46]. In the cold season, synoptic conditions characterised by the prevalence of westerly winds influence the atmospheric dynamics. On the contrary, in the warm season, recirculation of air masses on the western side of the basin [46,47] and the prevalent NE winds over the eastern side [48] play a dominant role.

From an atmospheric point of view, the Mediterranean basin is a crossroads of air masses coming from Europe, Asia, and Africa [49]. It is bounded to the north by the populated and highly industrialised area of southern Europe and to the south by northern Africa. For this reason, aerosol particle loading is therefore largely affected by natural and anthropogenic sources. The natural sources are Saharan dust [50–54], marine aerosols [55,56], and forest fires [57]. In particular, the most important natural source of atmospheric aerosols on a global scale is mineral dust from the Sahara Desert [58]. In fact, in Mediterranean countries, Saharan dust events occur in different seasons in the west (frequently in summer) and in the east (more concentrated in autumn and spring) [52–54] and are frequently responsible for elevated concentrations of particulate matter [58–62] with negative effects on health and the environment [62–66]. This phenomenon has been associated with increased mortality and respiratory symptoms and also affects climatic processes, soil formation, and nutrient cycles. Anthropogenic emissions are emitted by various urban activities (vehicular traffic, biomass burning, fossil fuel combustion, and cooking activities) [67–72], by industries, and by maritime traffic [24,73,74].

Large efforts have been made to reduce the greatest emission sources (industrial, power generation, etc.). These generated a relative increase in the weight of shipping emissions to total anthropogenic emissions. Viana et al. [29] applied a health impact assessment (HIA) to obtain an assessment of the health burden of shipping emissions across different Mediterranean coastal cities. They evaluated that, in the European Mediterranean coastal cities studied, exposure to shipping emissions could account for a sum of 432 premature deaths per year. The impact of this emission source is comparable in magnitude to the most typical urban source, vehicular traffic.

2.2. Literature Review

A literature survey was conducted to assess the contribution of ship emissions to the NO_2, SO_2, PM_{10}, and $PM_{2.5}$ concentrations in the urban areas of the port cities in the Mediterranean Sea. Scopus and Science Direct were selected as databases for the purposes of this study. In addition, the scientific search engine Google Scholar was used to perform the literature search. Keyword combinations were used in various orders and combined with the Boolean operators "AND", "OR", and "NOT" in order to optimise the search strategy. The definitive search string obtained was ("air quality" OR "air pollution") AND ("source apportionment" OR "receptor model" OR "dispersion model") AND ("maritime" OR "fuel oil" OR "ship emissions") AND "port".

The search was performed in such a way that all the keywords were present in the full text or metadata of the papers. A complementary technique using "bibliographic search", "citation tracking", "snowballing", or "pearl growing" was applied to identify more papers relevant to the current study [75,76]. A manual search in sources known to the research team and a general search on the Internet was also performed to deepen the study. Firstly, the papers identified in the narrow search were further filtered based on a screening of their title and abstract. Secondly, only peer-reviewed articles published in scientific journals in the English language were reviewed. Finally, for review inclusion, two eligibility criteria for the studies had to be met: (1) quantifying the contribution to air pollution concentration attributable to shipping emissions or port activities; (2) considering at least one port city located in the Mediterranean region. A total of 640 articles were identified with the initial search strategy. After the process of title and abstract screening, final full-text reading resulted in 32 studies that met the eligibility criteria. Figure 2 presents the compiled case studies, with a total of 34% located in Italy, 21% in Spain, 18% in Greece, 11% in Turkey, about 5% for both Croatia and Malta, and 3% in France and Cyprus.

2.3. Approach to Assess the Contribution of Ship Emissions to Air Quality

The assessment of the contribution of ship emissions is not an easy task, especially in areas where several sources co-exist such as industrial settlements, railway, and urban agglomerates and/or where there are complicated meteorological conditions.

Two different approaches have potential peculiarities for assessing the effects of ship emissions on air quality: (i) receptor models and (ii) air dispersion models. Both models have advantages and limitations [77]. Compared to measurements, numerical models have the advantage of providing outcomes on the whole studied territory with a certain resolution, but they need detailed input and reliable emissions information [78].

Conversely, for the receptor model approach, no detailed information on sources is needed; however, results can be obtained for limited periods of time and from a limited number of sites. Receptor model approaches can be used to estimate the contributions of shipping using statistical multivariate analysis on the chemical characterisation of data on PM and online high-resolution detection of pollutant concentration [20,79]. The first approach includes Positive Matrix Factorisation (PMF), principal component analysis (PCA), chemical mass balance (CMB), and multiple linear regression (MLR). In particular, chemical tracers of heavy oil combustion sources (including shipping) such as vanadium (V) and nickel (Ni) have been used to estimate the primary contribution to $PM_{2.5}$ [80,81]. This is especially because ratios of V/Ni values in the range of 2.5 to 3.5–4.0 [82,83] are considered typical combustion markers of ships' engines, while ratios <2 are commonly associated with the presence of Ni-rich atmospheric pollution sources [84]. Few studies are devoted to the characterisation of secondary aerosol contributions from ship emissions, even though they can be higher than the primary contributions [30]. Some studies were carried out using high-temporal-resolution measurements of particles and gaseous pollutants correlated with wind conditions and ship traffic [31,85–87].

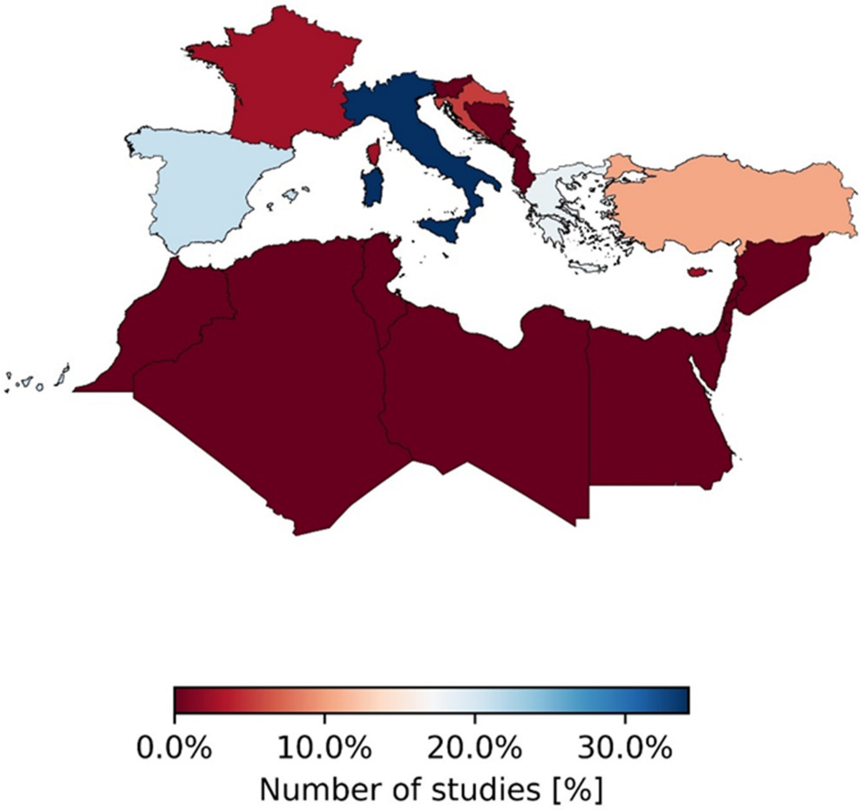

Figure 2. Location of the analysed published studies focusing on air quality over harbours.

Modelling approaches have been adapted from the global/continental/national scale [7,88] to the local level [89,90] with many Gaussian models and an Eulerian model [91–93]. The most common and simplest one is a Gaussian-based model that assumes that the dispersion of air pollutants follows a Gaussian distribution. A steady-state Gaussian-based dispersion model such as the atmospheric dispersion modelling system (ADMS), which can simulate the effects of temporally and spatially variable meteorological conditions from point, line, area, or volume sources at a local scale (i.e., urban), has been applied [90,94]. Another popular steady-state Gaussian plume model, AERMOD, recommended by the United States Environmental Protection Agency (EPA), was also widely used by different groups [92,95] to evaluate the contribution of ships on PM emissions in harbour cities. In addition to the simple Gaussian plume models, some advanced, unsteady Gaussian puff models (such as CALPUFF), which can simulate the effects of time- and space-varying meteorological conditions on pollutant transport, transformation, and removal [96], have been widely used for simulating the dispersion of ship emissions [97–99]. Furthermore, a Lagrangian or Eulerian chemistry transport model (CTM) such as the comprehensive air quality model with extensions (CAMx), CAMx-PSAT (particulate matter source apportionment technology), SPRAY, or the flexible air quality regional model

(FARM) have received increasing attention [100–102]. These models simulate primary and secondary pollutant transport and concentrations at different scales. The spatial resolution is defined separately for the horizontal grid and vertical layers, and the model can be adapted to different meteorological models (i.e., WRF). Specific techniques such as particulate matter source-apportionment technology (PSAT) can be implemented in CAMx to provide source apportionment for primary and secondary particulate matter species. In this case, in order to estimate the contribution of shipping, an approach called the "zero-out method", using WRF-CAMX or FARM, computes the relative difference in concentration of investigated pollutants between two scenarios. In the first run, all natural and anthropogenic emission sources are included, while in the second one, shipping pollutant emissions are excluded. Then, the extrapolation of concentration values of specific grid cells indicates the impact of the shipping/harbour. Recently, integrated approaches have also been applied [20,77], combining high-temporal-resolution measurements, numerical simulations, emission inventories, and satellite imagery. Data with high temporal and spatial resolution from networks with dense air quality could significantly improve source attribution [103], especially for relatively short-lived species (i.e., NO_x) and, on the other hand, for pollutants transported on greater spatial (and temporal) scales (i.e., secondary particulates).

3. Results

The air quality of port cities is negatively affected by ship emissions. Emissions from maritime transport consist of primary and secondary particulate matter, mainly in the fine particle size fraction ($PM_{2.5}$), sulphur dioxide (SO_2), nitrogen oxides (NO_x), and non-methane volatile organic compounds (NMVOCs). The increase, in the last decade, of available studies that evaluate the impact of ship emissions shows the importance of this sector of activity and its impacts on quality. The literature review conducted in this document aims to provide an updated picture of scientific results on methodologies and estimates of the impact of maritime transport on local air quality in port cities of the Mediterranean Sea. All the studies selected in this literature review agree on the significant contribution of maritime and port activities in terms of atmospheric emissions and relative contributions of NO_2, SO_2, PM_{10}, and $PM_{2.5}$ to air quality.

Thirty-two studies were considered. A total of 30 ports were analysed, with 1 in Croatia, 3 in Cyprus, 1 in France, 4 in Greece, 8 in Italy, 2 in Malta, 6 in Spain, and 5 in Turkey (Figure 3).

Some studies also analysed multiple ports. For example, the APICE project studies the impact of maritime traffic emissions in five ports in the Mediterranean area (Barcelona, Genoa, Marseille, Venice, and Thessaloniki), and Merico et al. [104] studied the impact in four Adriatic ports (Venice, Patras, Brindisi, and Rijeka). Data from 32 papers were obtained. The most used approach is the receptor model; in fact, about 67% of the studies adopted this type of technique. The remaining 33% (about 10 papers) used modelling approaches. Different air dispersion models are used. Specifically, 50% use chemical transport Eulerian-type models (Chimere and CAMX), 45% use Gaussian-type models (ADMS, AERMOD, and CALPUFF), and the remaining 5% use the Lagrangian SPRAY model. The results of papers analysed are presented by country as follows: Italy, Greece, Croatia, Turkey, Cyprus, Malta, France, and Spain.

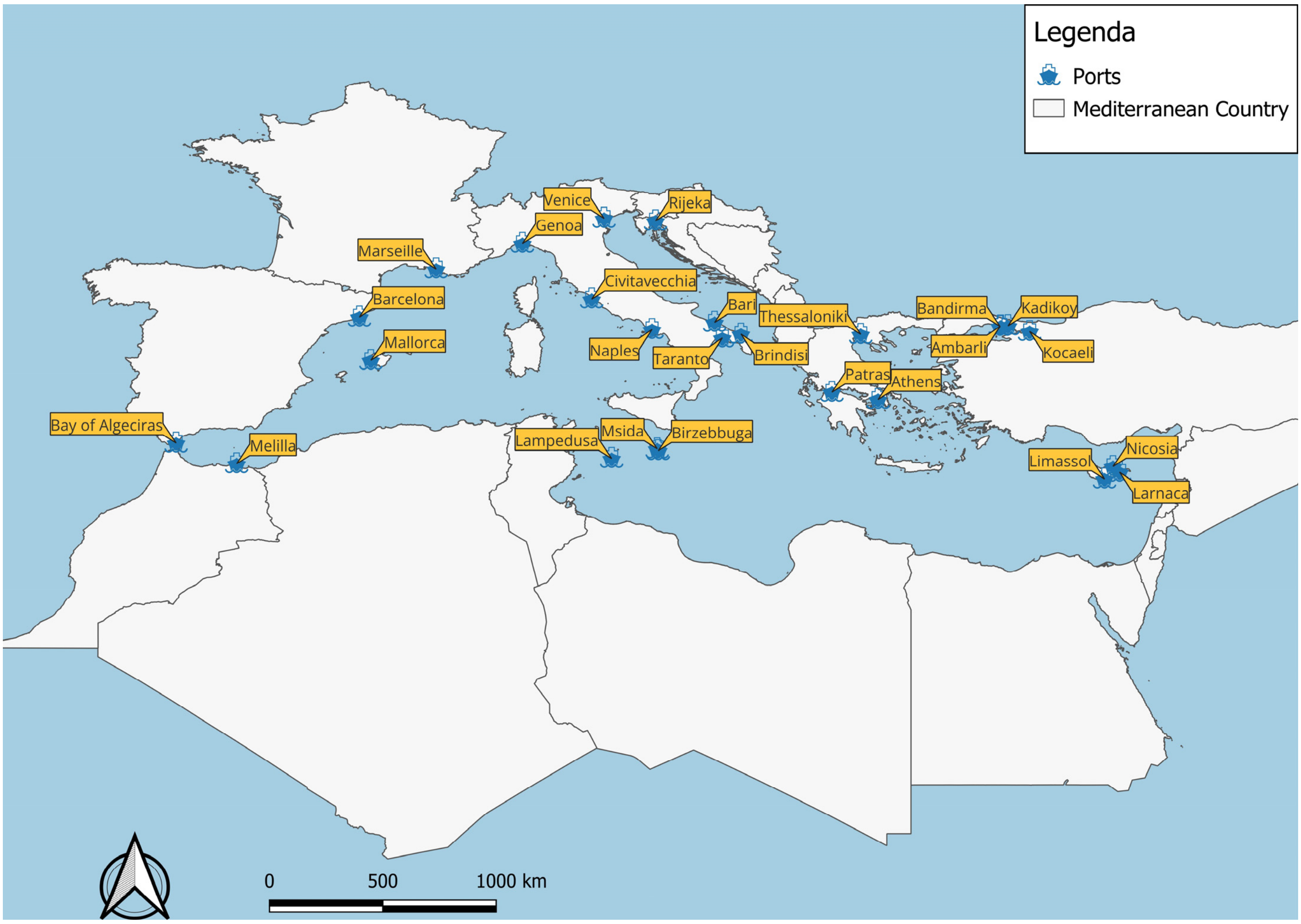

Figure 3. Location of the ports across the Mediterranean area.

3.1. Italy

Gariazzo et al. [102] evaluated the relative impact of harbour emissions on air quality in 2002, with respect to other emission sources located in the same area in the city of Taranto, Italy, with the 3D Lagrangian particle dispersion model SPRAY. The results show how harbour activities exhibit the second-largest contribution to SO_2 with 3–7% and 7–11% in winter and summer, respectively. On average, harbour activities contribute 7% for SO_2 and 9% for NO_x.

In the Apulia region of the Adriatic Coast, Merico et al. [86], in summer 2012 and summer 2014, estimated the contributions of maritime emissions to atmospheric concentrations of NO_2, SO_2, and $PM_{2.5}$, in Brindisi harbour, considering manoeuvring (arrival and departure of ships) and hotelling phases (including loading/unloading activities). In port areas, maritime emissions gave contributions of 55% for SO_2, 55% for NO_2, and 10% for $PM_{2.5}$.

In Brindisi, Merico et al. [104] reported the contribution of shipping emissions using the WRF-CAMx model for July (summer) and January (winter) 2012. They found that in the port area, the contribution to NO_2 concentration is between 16.7 and 32.5%, the contribution to SO_2 is 23.5–46.3%, the contribution to PM_{10} is 3.9–3.7%, and the contribution to $PM_{2.5}$ is 5.0–4.7%. On average, the contribution is 25% for NO_2, 35% for SO_2, 4% for PM_{10}, and 5% for $PM_{2.5}$. The contribution for particulate matter is also studied with two different methodologies, Positive Matrix Factorisation (PMF) and statistical analysis. Using PMF, the contributions of ships to the urban concentration of $PM_{2.5}$ and PM_{10} are 2.8% and 2.1%, respectively. With a statistical analysis of high temporal resolution, they obtained a contribution in the port area of 7.4% for $PM_{2.5}$, 5.8% for PM_{10}, and 26% for particle number concentration (PNC).

Merico et al. [90], using the ADMS-5 dispersion model, simulated the dispersion of harbour emissions in Bari, thereby estimating their impact on gaseous and particulate matter in seven monitoring sites (where one is in the harbour) in the year 2018. In the port area, they observed that the contribution of yearly average concentrations due to shipping emissions was 80.6% for SO_2, 40% for NO_2, 6.7% for PM_{10}, and 11.8% for $PM_{2.5}$. In contrast, the contribution in the urban area was in the range of 17.9–20.8% for SO_2, 3.7–36.8% for NO_2, 1.3–5.0% for PM_{10}, and 0.19–1.48% for $PM_{2.5}$ as a function of the distance from the port of the receptors. Using the CALPUFF model, Murena et al. [99] assessed the impact of cruise ship emissions on air quality in the urban area of Naples in 2016. The yearly average contribution of NO_x was 2.47%. Additionally, in Naples, Toscano et al. [105], using the CALPUFF dispersion model, simulated the dispersion of shipping emissions, thereby estimating their impact on gaseous and particulate matter for the year 2018. They estimated that the contributions of ship emissions in urban areas as a function of the distance to the harbour range from 5 to 64% for NO_2 and 1–92% for SO_2. For PM_{10}, the contributions are in the range of 1–11%. Gobbi et al. [106] studied the influence of the port of Civitavecchia on air quality for three years (2013–2016). In the port area, the analysis of the three-year record indicates a contribution of 33% to PM_{10}, 43% to NO_2, and 60% to SO_2. These contributions decrease to 19% for PM_{10}, 25% for NO_2, and 43% for SO_2 if the city centre is considered.

The contribution of Venice port has been studied by several authors in different years and with different methodologies. Contini et al. [87] assessed the direct contribution of ships on particulate matter concentration in the urban area during different campaigns from June to September 2007 in one site and from March to October in two other sites. From June to September, the contribution was 2–7% for PM_{10} and 3–8% for $PM_{2.5}$, while from March to October, the contribution was 1% and 2% for PM_{10} and $PM_{2.5}$, respectively. On average, the contribution was 4% for both PM_{10} and $PM_{2.5}$. Contini et al. [107] performed an analysis of the primary contribution of tourist ship traffic emissions to $PM_{2.5}$ concentrations in an urban background site of Venice. Measurements were taken in the summer periods of 2007, 2009, and 2012 in similar meteorological and micrometeorological conditions. The estimated contribution of tourist ship traffic to primary $PM_{2.5}$ decreased from 2007 (7% ± 1%) to 2009 (5% ± 1%) and 2012 (3.5% ± 1%). Gregoris et al. [108] quantified the impact of maritime traffic on various pollutants in urban areas, such as particulate matter, using different state-of-the-art methodologies, starting from data collected between 2007 and 2013. The contribution of ship traffic to primary PM_{10}, according to data from 2007 to 2013, was 1.9–2.5%. The contribution to primary $PM_{2.5}$ was in the range of 2.4–3.3%.

Merico et al. [104], with the WRF-CAMx model chain, estimated that, for the year 2010, ships' emissions contributed to NO_2 at 2.8% in winter and 9.1% in summer; SO_2 at 5.2% (winter) and 16.5% (summer); PM_{10} at 1.2% and 2.3% for winter and summer, respectively; and $PM_{2.5}$ at 1.2% (winter) and 2.6% (summer). In the same study, during summer 2012, with Positive Matrix Factorisation, the authors estimated a contribution to particulate matter concentration level in the urban area of 3.0% for $PM_{2.5}$, 2.3% for PM_{10}, and, with high temporal resolution, of 3.5% and 2.7% for $PM_{2.5}$ and PM_{10}, respectively. Merico et al. [109], with measurement campaigns performed from 6 September 2018 to 27 November 2018, estimated that the relative contribution of shipping to PM_{10} and $PM_{2.5}$ in ports was about 2%. Becagli et al. [110] conducted measurements of aerosol chemical composition during the years 2004–2008 to identify the influence of ship emissions on aerosol particles in the urban area of the island of Lampedusa, south of the Sicily channel. This source contributed, on average, 22% of the $PM_{2.5}$ mass (26% in summer and 16% in winter). In Genoa, PMF and CAMx-PSAT analysis was performed to apportion the $PM_{2.5}$ sources in the urban area [111]. Using PMF, the authors found that ship emissions contributed 9–13% of the $PM_{2.5}$ level during spring and summer 2011, and with CAMx-PSAT found that the maritime contribution to $PM_{2.5}$ concentrations varied between 14 and 16%.

3.2. Greece

Tolis et al. [112] studied the mass concentration and chemical characterisation of atmospheric $PM_{2.5}$ in the city of Thessaloniki during a one-year period from June 2011 to May 2012. The mean concentration for the whole sampling period was 37.7 ± 15.7 μg/m^3. Minimum and maximum values were 12.9 and 116 μg/m^3, respectively. In addition, the port area on average exhibited higher particle concentration levels of 66.0 μg/m^3 than the city centre area, revealing the strong influence of the port activities on the area's air quality. Saraga et al. [113] analysed the data of Tolis et al. [112] and identified the chemical fingerprints of potential $PM_{2.5}$ sources and estimated their contributions to Thessaloniki port-city's air quality. For this purpose, the authors applied a Positive Matrix Factorisation model at two sampling sites: the port and the city centre. They found that the shipping emissions contributed 13.4% of the total PM mass concentration measured at Thessaloniki's port, while in urban areas the contribution was 9.4%. For the same site, a source apportionment for $PM_{2.5}$ was evaluated by CAMx-PSAT for both a summer period (June–August 2011) and a late autumn period (15 November–15 December 2011). Maritime and harbour activities present a rather small contribution to the average $PM_{2.5}$ levels: 1–2.2% in the urban site and 2.8–5.8% in the harbour. Progiou et al. [114] aimed to assess the contribution of ship emissions to air quality at Piraeus port from 1 October 2017 to 30 September 2018. Simulations were conducted with the air dispersion model AERMOD. Maximum hourly NO_2 concentration and 24 h mean concentration due to port operation were 78 and 25 μg/m^3, respectively, located mainly in the passenger port. The contributions of port activities to the hourly maxima and 24 h mean concentrations were 91.8% and 71.4%, respectively. SO_2 and PM_{10} maximum hourly concentrations reached 25 and 30 μg/m^3, respectively. Manousakas et al. [115] used PMF to show that shipping emissions accounted for 10% of total $PM_{2.5}$ in the urban area of Patras during the year 2011. Merico et al. [104], with the WRF-CAMx model chain, estimated the contribution of ship emissions to air quality in the harbour of Patras in summer (July 2012) and in winter (January 2012). They found contributions of 14.6–22.5% for NO_2, 8.8–24.7% for SO_2, 2.1–2.5% for PM_{10}, and 2.6–3.4% for $PM_{2.5}$. With high temporal resolution in summer 2013 and winter 2014, the campaign's ship emissions accounted for 3.8% of $PM_{2.5}$ and 3.3% of PM_{10}. Diapouli et al. [116] assessed the contribution of ship emissions to PM_{10} and $PM_{2.5}$ concentration levels in the urban area of Athens, Greece, during an intensive monitoring campaign conducted in 2011–2012. The contributions of shipping emissions estimated with PMF for PM_{10} were in the range of 5–6% in suburban and urban areas, respectively. Regarding $PM_{2.5}$, the contribution was 4% (0.5 μg/m^3) in the suburban site and 6% (1.0 μg/m^3) in urban areas.

3.3. Croatia

Merico et al. [104,109] estimated the contribution of ship emissions to air quality in the port area of Rijeka with various approaches and during different periods. With the WRF-CAMx model chain, the contribution estimated for NO_2 was in the range of 9.7–21.9%, 2.4–4.1% for SO_2, 1.0–2.0% for PM_{10}, and 1.1–2.2% for $PM_{2.5}$. Using PMF, in 2012, the authors found a contribution of 1.1% for $PM_{2.5}$ and 0.8% for PM_{10}, and in 2013–2014, they found contributions of 0.5% and 0.3% for $PM_{2.5}$ and PM_{10}, respectively. During the campaign of 28 March 2019–13 May 2019, the relative contributions were similar for $PM_{2.5}$ and PM_{10}, at <0.2%.

3.4. Turkey

Deniz and Kilic [117] used the CALPUFF modelling program to observe dispersions of the calculated emissions from ships in Ambarli Port in 2005. They reported that the maximum values of SO_2 and NO_x concentrations modelled in a 2 km range from the port exceeded 100 μg/m^3 for NO_x and 55 μg/m^3 for SO_2.

Kuzu et al. [118] employed air quality modelling using AERMOD to study the effect of ship emissions on air quality in Bandirma district and identified the most impacted areas in 2018. The contributions of NO_x, SO_2, and PM_{10} emissions from vessels had a share of 18, 36, and 1%, respectively, of the measured concentrations in urban areas.

Ekmekçioğlu et al. [119] calculated ship emissions arriving at Turkey's Kocaeli and Ambarlı ports for a year between 1 September 2017 and 1 September 2018. They determined the effect of ship emissions using AERMOD and compared their results with the results of the air quality measurement station. The authors estimated the contribution of ship emissions on air quality in urban areas for NO_x to be 19% and >100% for Ambarli port and Kocaeli port, respectively. For SO_2 and PM_{10}, the contributions for both ports were >100 and 5%, respectively. The contribution of greater than 100% is an unrealistic result. This indicates that the model overestimated the impact of ship emissions, probably due to overestimation in the calculation of emissions [119]. Another possible reason is that the model underestimated the dispersive properties of the atmosphere in some conditions [105].

Ünlügençoğlu and Alarçin [120] performed real-time measurement of air quality in terms of $PM_{2.5}$, PM_{10}, SO_2, and NO_2 emissions for the Port of Ambarli from June to August 2017. Additionally, real-time measurements were received from the air quality monitoring station in the Avcılar and Kadıköy urban districts of Istanbul. Average NO_2 emission values of Avcılar, the Port of Ambarlı, and Kadıköy districts were 14.7 $\mu g/m^3$, 62.6 $\mu g/m^3$, and 46.7 $\mu g/m^3$, respectively. Average SO_2 emission values for the Port of Ambarlı, Avcılar, and Kadıköy districts were 3.6 $\mu g/m^3$, 2.6 $\mu g/m^3$, and 2.1 $\mu g/m^3$, respectively. For PM_{10} and $PM_{2.5}$, the concentrations were 51.3 and 32.8 $\mu g/m^3$ for the Port of Ambarlı, 38.1 and 19.4 for Avcılar, and 31.0 and 25.3 $\mu g/m^3$ for Kadıköy.

3.5. Cyprus

Achilleos et al. [121] collected $PM_{2.5}$ and PM_{10} samples in different cities in Cyprus between January 2012 and January 2013 to conduct a source apportionment analysis. Using PMF, they found that the contribution of ship emissions to $PM_{2.5}$ concentration in urban areas was 13% (1.7 $\mu g/m^3$) in Larnaca, 10% (1.3 $\mu g/m^3$) in Limassol, 8% (1.2 $\mu g/m^3$) in Nicosia, and 6% (0.7 $\mu g/m^3$) in Paphos.

3.6. Malta

Scerri et al. [122] collected $PM_{2.5}$ samples in 2016 at the traffic station in Msida and applied Positive Matrix Factorisation (PMF), finding that shipping contributes 5% of the $PM_{2.5}$ levels in urban areas (7.5 $\mu g/m^3$). Camilleri et al. [123] used Positive Matrix Factorisation (PMF) to identify and quantify the main natural and anthropogenic sources of $PM_{2.5}$ at an urban background site in Birzebbuga village located in the south-eastern part of Malta from June 2018 to June 2019. They found that shipping emissions contributed 10% to $PM_{2.5}$ air quality concentrations, with an average concentration of 0.6 $\mu g/m^3$.

3.7. France

In the framework of the APICE project, a long-term monitoring campaign (July 2011–July 2012) was carried out at an urban background site in Marseille [124] to investigate air quality and the relative contribution of pollution sources to PM levels. On an annual basis, shipping emissions contributed significantly to fine aerosol mass, with a contribution of 18% (3.5 ± 2.3 $\mu g/m^3$) of $PM_{2.5}$.

Source apportionment for $PM_{2.5}$ was evaluated by both CHIMERE and CAMx, using a zero-out modelling and tracer approach (PSAT), respectively, for both winter, February 2011, and summer, August 2011, periods. Focusing on maritime contributions, the contribution at the port site was between 7 and 10% of the $PM_{2.5}$ concentration. At the urban background site, the maritime contributions were lower and ranged between 7% and 9% of the $PM_{2.5}$ concentrations.

3.8. Spain

Viana et al. [82] used the PMF model to detect the impact of shipping contributions to urban PM levels in Melilla during a monitoring campaign carried out between January

2007 and April 2008. For PM_{10}, the mean contribution of shipping emissions accounted for 2.5 μg/m^3 (6% of the mean annual PM_{10}). Regarding $PM_{2.5}$, shipping accounted for 2.6 μg/m^3 (14% of the mean annual $PM_{2.5}$). Pandolfi et al. [125] collected multi-year (2003–2007) ambient speciated PM_{10} and $PM_{2.5}$ data at four strategic urban sampling locations around the Bay of Algeciras and used a PMF model to identify major PM sources with particular attention paid to the quantification of total shipping emissions. Primary direct contributions from shipping in the Bay of Algeciras were estimated at 1.4–2.6 μg/m^3 (3–7%) for PM_{10} and 1.2–2.3 μg/m^3 (5–10%) for $PM_{2.5}$.

Pey et al. [126] discussed the results of an intensive sampling campaign (7 November 2011 to 5 January 2012) in the harbour area of Barcelona to identify sources of emissions and quantify their contributions to PM_{10} in the vicinity of the harbour of Barcelona, with a special focus on primary shipping emissions. Using the PMF method, they calculated that ship emissions contribute 2.7% to PM_{10}.

Pey et al. [127] identified and quantified natural and anthropogenic PM sources at the suburban insular site of Castillo de Bellver, in Mallorca (Spain, Western Mediterranean). Simultaneous PM_{10} and $PM_{2.5}$ daily samples were collected for almost one and a half years between January 2004 and July 2005. Harbour emissions were estimated by PCA to be 1.2 μg/m^3 for PM_{10} and 1.0 μg/m^3 for $PM_{2.5}$, with a contribution of 4.1% and 5% for PM_{10} and $PM_{2.5}$, respectively.

In the APICE project (http://www.apice-project.eu/, accessed on 10 April 2023), the contributions of ships' emissions of different sources to particulate matter concentrations were highlighted by the CHIMERE Chemical Transport Model. The mean contribution calculated for $PM_{2.5}$ among the three sites varied from 17% at the urban sites to 28% at the world trade centre and 54% inside the port area during summertime, whereas in winter, these contributions decreased to 5% at the urban site, 23.2 at the world trade centre, and 38% at the port. Very similar results were recorded for PM_{10} source apportionment (between 16% and 52% in summer and between 7% and 41% in winter).

Perez et al. [128] measured PM_{10} and $PM_{2.5}$ from 5th February to 31st December 2011 in the port area and at an urban background site in Barcelona to evaluate the impact of harbour activities. The mean fuel oil combustion concentrations were 2.9 μg/m^3 and 2.4 μg/m^3 for PM_{10} and $PM_{2.5}$, respectively, at the port area and 1.0 μg/m^3 for both PM_{10} and $PM_{2.5}$ in the urban area. The results evidenced the contribution of shipping emissions to PM inside the port of 9% and 17%, for PM_{10} and $PM_{2.5}$, respectively. In the urban area of Barcelona, the contribution of fuel oil combustion was 5% and 6% for PM_{10} and $PM_{2.5}$, respectively.

In the sea areas along the main shipping routes, especially in the Strait of Gibraltar and in the Mediterranean Sea, Nunes et al. [129] quantified the impacts of shipping emissions on the ambient air quality using the EMEP/MSC-W model in 2015. They found a contribution of more than 90% for NO_2, 80% for SO_2, between 20% and 35% for PM_{10}, and 25–50% for $PM_{2.5}$. Clemente et al. [130] calculated the contribution of port-related activities to PM_{10} levels at the port–city boundary of Alicante using the PMF method. PM_{10} samples were collected between March 2017 and February 2018 at an air quality monitoring station located at the perimeter of the harbour of Alicante. Shipping emissions accounted for 6% of the average PM_{10} mass concentration (1.51 μg/m^3). Guiterrez et al. [131] estimated the contributions of NO_x, SO_x, PM_{10}, and $PM_{2.5}$ on air quality in the Strait of Gibraltar (Spain) using the Ship's Energy and Emissions Model (SENEM) and the CALPUFF model in 2017. The stations that resulted in the most emissions from maritime traffic were located in the Bay of Algeciras. In terms of annual averages, the contributions of maritime traffic were 9 μg/m^3 for NO_2 with a contribution of about 28%, 3 μg/m^3 for SO_x with a percentage of 25%, 0.4 μg/m^3 for PM_{10} with a percentage of 1.5%, and 0.2 μg/m^3 for $PM_{2.5}$ with a percentage of 2.2%.

4. Discussion and Conclusions

The selected studies indicate large variability in the contributions of port emissions to particulate matter and gaseous concentrations in urban areas between distinct countries.

Particulate matter (PM_{10} and $PM_{2.5}$) was the most studied pollutant, and the contributions of ship emissions in terms of minimum and maximum concentration values measured in urban areas for each country are reported in Figures 4 and 5. The contribution of shipping emissions to PM_{10} ambient air concentration in Croatia varied from a minimum of 1.0% to 2.0% [104], both measured in the urban area of Rijeka. In Greece, the contribution varied between an estimated 3.0% in Patras [104] and 6% in Athens [116]. In Italy, the minimum contribution of 1% was estimated in Naples and Venice [87,105] and the maximum was 11% (Naples) [105]. In Spain, it varied between 3% in the Bay of Algeciras [125] and 16% in Barcelona (APICE, 2012). In Turkey, the contribution of ship emissions to PM_{10} concentration varied between 1% in Bandirma [118] and 5% in Ambarli and Kocaceli [119].

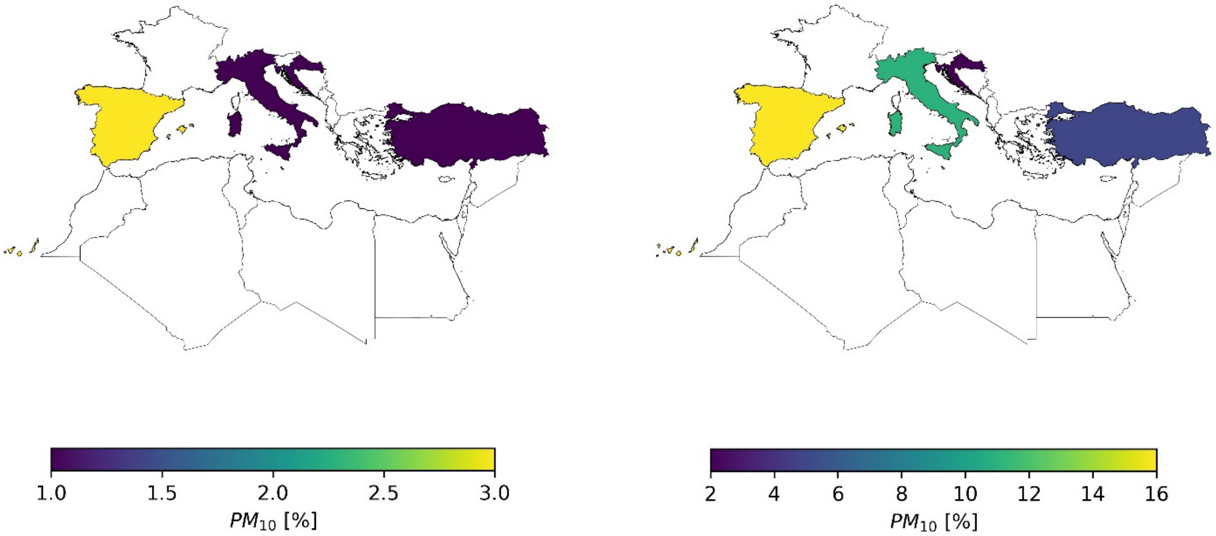

Figure 4. Minimum (**left**) and maximum (**right**) PM_{10} concentration values measured for each Mediterranean case study during the corresponding sampling period.

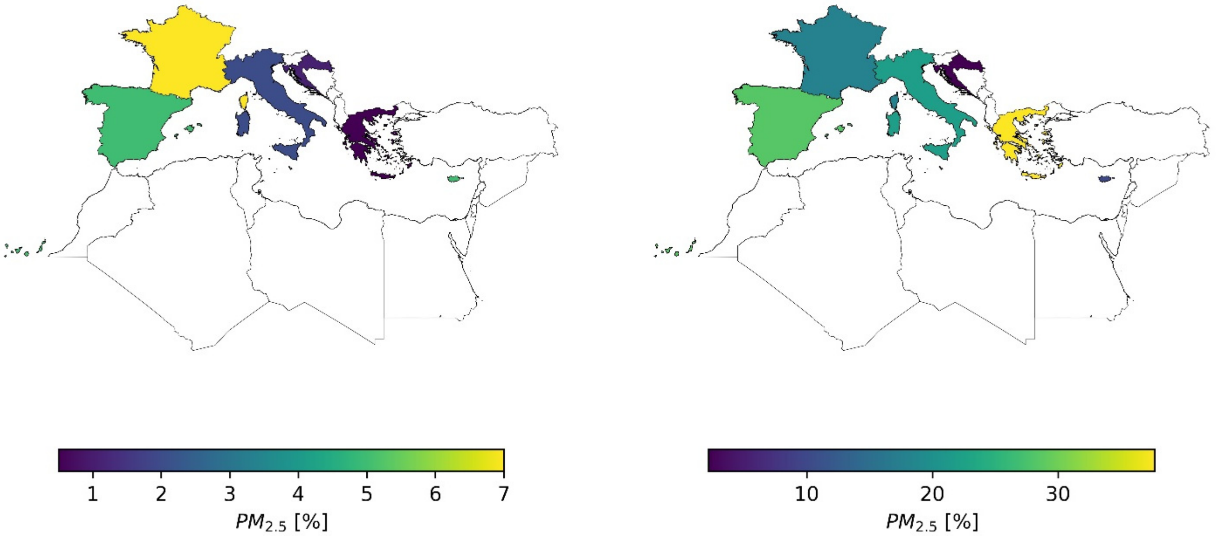

Figure 5. Minimum (**left**) and maximum (**right**) $PM_{2.5}$ concentration values measured for each Mediterranean case study, during the corresponding sampling period.

Regarding $PM_{2.5}$, in Croatia, the contribution varied between 1.1% and 2.2% [104], both measured in Rijeka. In Cyprus, the percentage contribution varied between 6% measured in Nicosia and 13% calculated in Larnaca [121]. In France, the contribution of ship emissions to $PM_{2.5}$ concentration in urban areas varied between 7 and 18% (APICE, 2012). In Greece, it varied between 1% (Thessaloniki) (APICE, 2012) and 10% estimated in Patras [115]. In Italy, it varied between an estimated 2% in Venice [87] and 22% in Lampedusa [110]. In Malta, the contribution was in the range from 5% (Msida) [122] to 10% (Birzebbuga) [123]. In Spain, the $PM_{2.5}$ contribution varied from 5% in both Palma de Mallorca [127] and Barcelona [125] and an estimated 28.4% in summer in Barcelona (APICE, 2012).

The contributions of NO_2 and SO_2 ship emissions to an ambient air concentration of both pollutants were generally higher than that of particulate matter concentration (Figures 6 and 7).

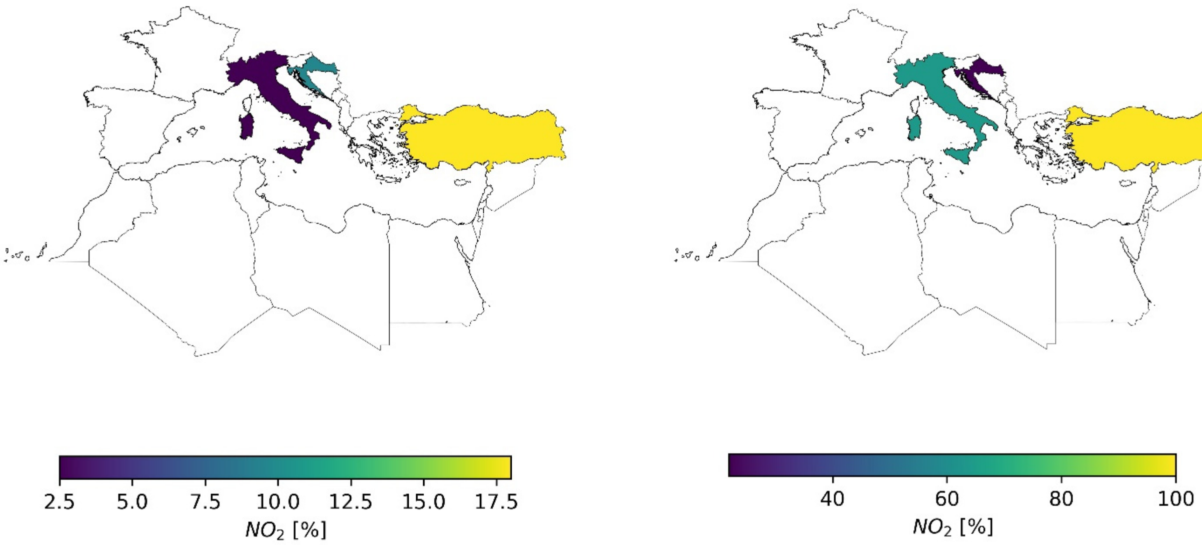

Figure 6. Minimum (**left**) and maximum (**right**) NO_2 concentration values measured for each Mediterranean case study during the corresponding sampling period.

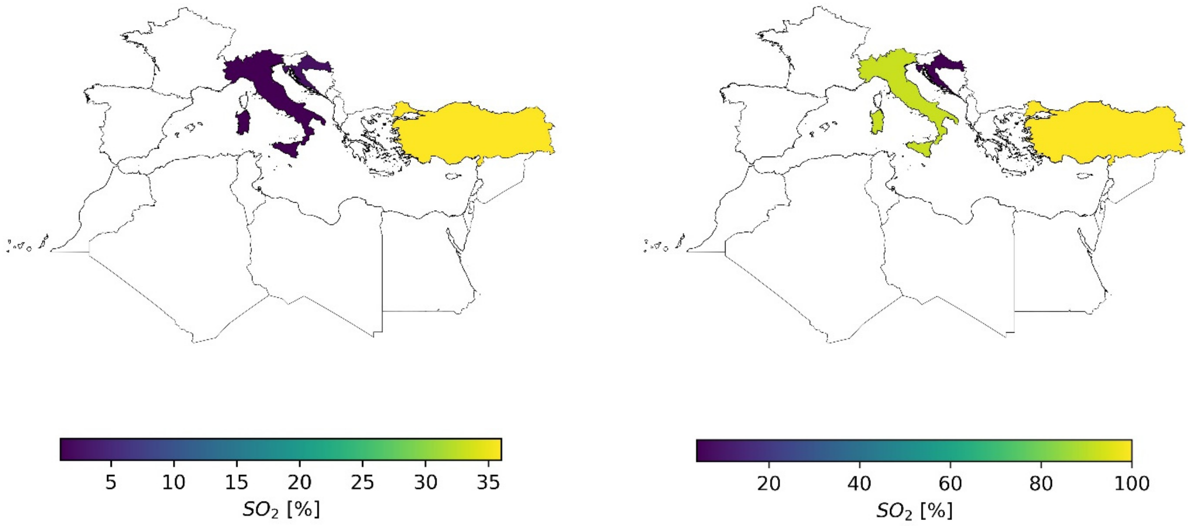

Figure 7. Minimum (**left**) and maximum (**right**) SO_2 concentration values measured for each Mediterranean case study during the corresponding sampling period.

The contribution in the urban area of Croatia of NO_2 varied between 9.7% and about 22% [104] and between 2.4% and 4.1% for SO_2 [104]. In Italy, the percentage contribution for both NO_2 and SO_2 ranged from a minimum of 1% in the urban area of Naples [99] to a maximum of 64% for NO_2 and 92% for SO_2, also in the urban area of Naples [105]. In Turkey, the contribution of shipping emissions to NO_2 concentrations in urban areas ranged from 19% in Ambarli to >100% in Kocaeli [119], and the contribution of SO_2 varied from a minimum of 36% in Bandirma [118] to a maximum of 100% in Kocaeli and Ambarli [119].

The relevance of the maritime transport sector to air pollutant emissions and its impact on air quality and human exposure, in particular on urban port areas, is evident. The designation of the Med SO_x ECA is therefore necessary, as similar mitigation strategies have already demonstrated their effectiveness in other parts of the European territory with a decrease in SO_2 concentrations in various ports and the consequent reduction in particulate matter.

It will therefore be necessary to carry out a joint study to verify the benefits of adopting this type of policy in terms of its impact on health.

Furthermore, it is important to note that the current literature primarily focuses on European ports, with a lack of research specifically addressing shipping emissions in African ports. This research gap presents an opportunity for future studies to provide a more comprehensive understanding of the impact of shipping emissions in the Mediterranean region.

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