

#### The Mediterranean Atmosphere Under Anthropogenic Pressures

Jean-François Doussin

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# The Mediterranean Region Under Anthropogenic Pressures

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Jean-François Doussin

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#### 1 Introduction

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The Mediterranean Sea is surrounded by three continents and stretches from the Atlantic Ocean to the Middle East. Considered the cradle of western civilization, it has been the major transportation route of people as early as 100,000 years BC (Strasser et al. 2010). Evidence has been found of an organized society structured as an extension of the Neolithic trend, from as early as the 8th millennium BC, with the rise of proto-urban centers on the Eastern shore (Langer 1972). While sometimes presented as a sea that separates Europe from Africa, the Mediterranean Sea has, on the contrary, mostly been a sea that relates and unites three continents, namely Europe, Asia, and Africa, a superhighway allowing the transportation of goods, technologies, and ideas. Since millennia, its shores have been an area of settlements and development.

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Chapter reviewed by **Paolo Prati** (Istituto Nazionale di Fisica Nucleare (INFN), Physics Dept., University of Genoa, Via Dodecaneso 33, I-16146 Genova, Italy), as part of the book Part I also reviewed by **Piero Lionello** (Univ. of Salento, and Centro Euro-Mediterraneo sui Cambiamenti Climatici (CMCC), Lecce, Italy).

J.-F. Doussin (⊠)

Laboratoire Interuniversitaire des Systèmes Atmosphériques (LISA), CNRS UMR 7583, Institut Pierre Simon Laplace, University Paris Est Créteil, University de Paris, Créteil, France

e-mail: jean-francois.doussin@lisa.u-pec.fr

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#### 2 Population, Density, and Urban Area

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Nowadays, the Mediterranean basin is surrounded by 22 countries, which together share a coastline of 46,000 km. These coasts are home to ca. 400 million people (UNEP/MAP Plan Bleu 2009) living across three continents: Africa, Asia, and Europe. It is still one of the world's busiest shipping routes, with about one-third of the world's total merchant shipping crossing the sea each year.

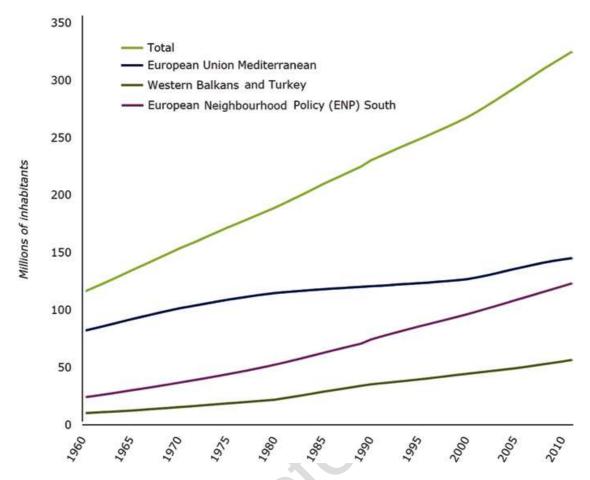
The total population of the Mediterranean countries grew from 281 million in 1970 to 419 million in 2000 and from 472 million in 2010 to 521 million in 2020 (United Nations 2019). The population is predicted to reach 572 million by 2030. Approximately one-third of the Mediterranean population is concentrated along its coastal regions, whereas more than half of the population resides in the coastal hydrological basins (Fig. 1). Based on the United Nations world population prospect and national population census, the population of the coastal areas grew from about 100 million in 1980 to 150 million in 2005, and it could reach 200 million by 2030 (United Nations 2017). The concentration of population in coastal zones is the highest on the western shore of the Adriatic Sea, the Middle East shore, and the Nile Delta. Overall, the population density in the coastal zone is higher in the southern Mediterranean countries. While the population share in the northern coastal area was 59% in 1970, a demographic transition is in progress, and the southern coastal area is expected to represent more than 62% in 2030 (Matteudi 2019). Interestingly, this is also where the variability of the population density in the coastal zone is highest, ranging from more than 1000 people km<sup>-2</sup> in the Nile Delta to fewer than 20 people km<sup>-2</sup> along parts of coastal Libya.

While the population in the north is almost stagnant, one observes a strong population growth in the southeast resulting in overexploitation of water, marine, and land resources, and also leading to increasingly problematic air quality issues in cities (EAA 2015). The southern and eastern countries of the Mediterranean are



**Fig. 1** Major urban areas and cities larger than 100,000 inhabitants. Circles: city data from JRC/GHS Urban Center Database 2015 (see Florczyk et al. 2019) superimposed over population density across the Mediterranean basin. (Data: Global Human Settlement Layer produced by the European Commission JRC and the CIESIN Columbia University, https://ghsl.jrc.ec.europa.eu/. Cartography: D Smith CASA UCL, https://luminocity3d.org/)

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**Fig. 2** The urban population growth in the Mediterranean area from 1960 to 2011. European Union Mediterranean Group: Cyprus, Spain, France, Greece, Croatia, Italy, Monaco, Malta, and Slovenia; western Balkans and Turkey Group: Albania, Bosnia and Herzegovina, Montenegro, and Turkey; European Neighbourhood Policy (ENP) South Group: Algeria, Egypt, Israel, Jordan, Lebanon, Libya, Morocco, Palestine, Syria, and Tunisia. (Figure provided by the European Environment Agency data visualization system based on data from the Mediterranean strategy for sustainable development follow-up provided by UNEP/MAP Plan Bleu; https://www.eea.europa.eu/ds\_resolveuid/B054CPK1NW)

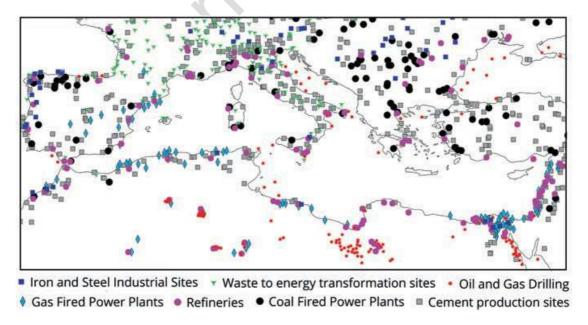
rapidly urbanizing (Fig. 2), with almost all of the future population growth projected to be in the cities—while urbanization rates in the north are quasi-constant at moderate values. Coastal areas historically rich in natural resources are now providing opportunities for economic activities such as tourism, oil and gas extraction, and maritime transport that tend to be located in these areas (EEA 2015). As a consequence, around 40% of the total coastal zone is estimated to be under some form of artificial land cover (concrete, asphalt...). Close to 100% of the population in the coastal region resides in urban localities. The Mediterranean catchment area includes 39 cities or agglomerations that are expected to be over one million inhabitants in 2030, and already more than 200 cities are larger than 100,000 people (see Fig. 1). The issue is further amplified by the increase in the fraction of urban population. Hence the urbanization phenomenon is not only due to the general increase of the population but also to a concentration process in cities and mega-cities. Mediterranean coastal areas are threatened by coastal development that modifies the

coastline through the construction of buildings and infrastructure needed to sustain residential, tourism, commercial, and transport activities. Coastal man-made infrastructures can damage landscapes in the long term, habitats and biodiversity, and shoreline configuration by disrupting sediment transport. These densely urbanized areas are also intense emission "point sources" for atmospheric pollutants that tend to accumulate and react over the basin atmosphere (see Giorgi and Raffaele this volume; Dayan this volume).

#### 3 Industry

When considering the impact of human activity on the atmosphere, large industrial installations fall upon the primary sources to be considered. While being quasi-point sources at the regional scale, their intensity often allows them to affect atmospheric composition at a very large scale.

Being both densely populated areas and the terminal of many marine routes, the Mediterranean coastal regions include several large industrial sites that often gather over a few square-kilometers installations from the chemistry sector, oil and gas terminal, power production, steel production, cement production... (e.g., Fos-Berre area near Marseille in France, Genoa area or Taranto in Italy, or Cairo in Egypt; Fig. 3). The western Mediterranean basin is especially concerned by these high industrial emission areas. These activities often involve combustion processes and emitting CO, CO<sub>2</sub>, NO<sub>x</sub>, unburnt volatile organic species, and combustion particles, but they also involve direct emission of hydrocarbons (HC) from tank leakages.



**Fig. 3** Major industrial sites in the Mediterranean region. (Plotted data from the mixed sources: www.carbonbrief.org, www.industryabout.com, www.powerplantmaps.com)

Some of these industries also involve releases of chemical compounds (HC, NOx, SO<sub>2</sub>, or CO<sub>2</sub>) produced in their processes.

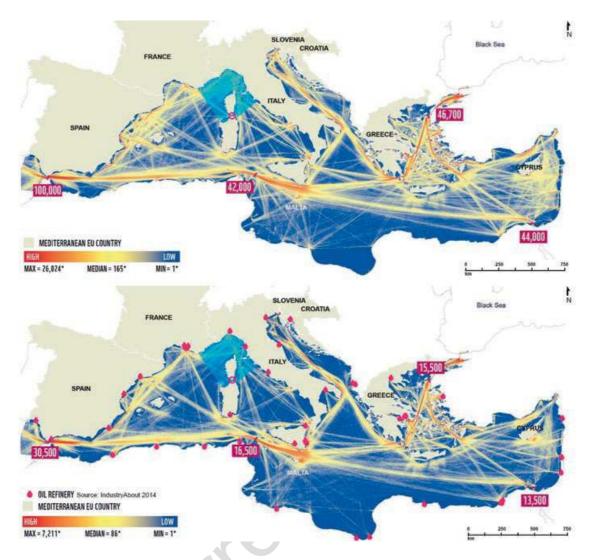
As the formation of secondary atmospheric pollutants such as ozone, photooxidants, and secondary organic aerosol (SOA) is extremely sensitive to the  $NO_x/HC$  ratio, the dual nature of the industrial emission has diverse effects at diverse scales. Locally, where the ozone and photooxidants production is generally limited by the availability of volatile organic compounds (VOCs), the VOCs that they emit allow a sustained high efficiency of secondary pollutants production in the atmosphere. For example, in the Fos-Berre region near Marseille, France, where lies a very large industrial complex, a record ozone rate production has been recorded: the study of Lasry et al. (2005) reports an unforeseen ozone episode in the outflow of the Fos-Berre industrial area where concentration as high as 450  $\mu$ g m<sup>-3</sup> and ozone production rates larger than 130  $\mu$ g m<sup>-3</sup> h<sup>-1</sup>. They demonstrate that it could be attributed to the combination of breeze dynamics and industrial release of reactive VOCs such as acetylene. At the regional scale, particularly offshore, ozone production is generally  $NO_x$  limited. The  $NO_x$  emission by these large industrial sites contributes to the general nitrogen oxide burden and hence to large-scale atmospheric chemistry.

Industrial activities also induce direct emission of semi-volatile persistent organic pollutants (POP) (e.g., Esposito et al. 2012) as well as heavy metals, which are found both in the airborne particulate matter and in the sediments (Trifuoggi et al. 2017). Further, both the refinery and cement industry often involve large SO<sub>2</sub> emissions that will later contribute to the secondary sulfate aerosol formation and fine particulate burden (Esposito et al. 2012).

### 4 Ship Traffic

While being highly interlinked with industrial activities, ship traffic delivering freight and raw material has a specific impact on the Mediterranean atmosphere due to the distributed nature of its emissions and the weak regulation of ship emissions in international waters. In a quite deleterious way, they worsen the effect of industrial areas and megacities by emitting reactive species in a relatively pristine area: the offshore marine atmosphere.

Being a route for exchanges of manufactured products between Europe and Asia through the Suez Canal and for the supply of gas and oil to Europe from the Gulf countries, the Mediterranean Sea is amongst the world's busiest waterways (Piante and Ody 2015). Intra-Mediterranean fluxes illustrated in Fig. 4 account for less than a quarter of the total Mediterranean maritime traffic (REMPEC 2008) that is largely dominated by international fluxes. Following a general global trend, vessel activity in the Mediterranean Sea has recorded a rise of 58% in transit capacity, combined with an increased size of vessels by 30% between 1997 and 2014 (UNEP/MAP-Plan Bleu 2014). It is expected that shipping in the Mediterranean basins will increase in the coming years, both in the number of routes and traffic intensity. Maritime traffic towards and from EU Mediterranean ports will be influenced by the



**Fig. 4** *Top* density of cargo traffic as extrapolated from the density of automatic identification system (AIS) signals from cargo ships in 2014 (total number of tracks: 13,144; color scale: logarithmic; AIS data http://www.navama.com/). *Bottom* density of oil tanker traffic as extrapolated from the density of AIS signals from oil tankers in 2014 (total number of tracks: 38,897; color scale: logarithmic); oil refineries are indicated by red droplets. The blue area and red circle in the northwestern Mediterranean indicate particularly sensitive areas. (Reprinted from Piante and Ody (2015), © WWF-France)

doubling of the Suez Canal that will allow a proportional traffic increase, but also by key drivers such as weak oil refining capacity outlook for Europe, a changing energy mix, the global demand for Liquified Natural Gas (LNG) as a fuel for maritime shipping, etc. (UNEP/MAP-Plan Bleu 2014).

Fortunately, the Mediterranean Sea is not totally without regulation with respect to atmospheric ship emissions. In 1997, the International Marine Organisation (IMO) through its Marine Pollution Convention (MARPOL) has defined various emission limit values for atmospheric pollutants emitted by shipping activities and forbidden releases of substances likely to affect the stratospheric ozone layer. In October 2016, IMO decided to lower the global sulfur limit in marine fuels to 0.5% by 2020.

The impact of ship emissions on the Mediterranean atmosphere is discussed in a recent study aiming to evaluate the feasibility of emission control over the Mediterranean Sea (Rouïl et al. 2019). Indeed, as Appendix 3 of Annex VI of the MARPOL Convention gives the possibility to define Emission Control Areas (ECA) where sulfur oxides (SO<sub>x</sub>) and nitrogen oxides (NO<sub>x</sub>) emitted by maritime traffic should be reduced, they consider the benefit of implementing even stricter emission control over the Mediterranean Sea. Their findings demonstrate that controlling ship emission alone, by further reducing the sulfur content of fuels and equipping ship engines with depolluting devices compliant with TIER III cleaner technologies (Wik and Niemi 2016), would lead to a substantial air pollutant concentration reduction. Through a careful ship emission inventory and an advanced modeling approach, they calculated that such a regulation would lead to a reduction by up to 11% in annual average fine particulate matter (PM<sub>2.5</sub>) concentration, 70% in nitrogen dioxide (NO<sub>2</sub>), and 11% in ozone (O<sub>3</sub>) over the coastal areas. Rouïl et al. (2019) conclude that such a regulation would avoid over 6200 premature deaths per year, mainly in Italy, Algeria, Egypt, and Greece. Being limited to coastal areas, these results are relevant to air quality but do not allow further evaluation of these emission impacts on the regional climate (see the two other Chapters of this book by Mallet et al. (2021) and Nabat et al. (2021) for interactions between air pollution and regional climate).

#### 5 Off-Shore Gas and Oil Production

Among the potential sources of atmospheric pollution located away from the seashore, one must cite offshore extraction of fossil fuels even if, so far, the Mediterranean region has been a relatively small producer of offshore oil and gas. Total offshore oil and gas production in the Mediterranean region was estimated in 2011 at 87 million tons of oil equivalent (TOE), including 19 million TOE for crude oil and 68 million TOE for natural gas. Nevertheless, the oil and gas industry is clearly looking at offshore developments as they represent 9400 million TOE, equivalent to 4.6% of world oil reserves (Piante and Ody 2015).

Until 2011, 94% of Mediterranean offshore oil reserves were held by Libya, Algeria, and Egypt, and only very limited quantities were available in EU Mediterranean countries (UNEP/MAP-Plan Bleu 2014). Italy operates approximately 100 offshore installations, mainly for gas extraction and exploration. These facilities are located in the Adriatic Sea, the Ionian Sea, and the Sicily Channel. Spain also has two installations in the Mediterranean Sea (EMSA 2014). Until 2015, no active offshore installations were reported by the Cypriot, French, Maltese, and Slovenian sectors, but this situation is rapidly changing. In Greece, the average production of 2014 was 75,900 TOE of crude oil and 4870 TOE of natural gas. Since then, a new investment program has been implemented in order to increase the current crude oil annual production up to one million TOE. To this end, the Greek authorities have leased new blocks in western Greece: one onshore in

Ioannina and two offshore in the Gulf of Patraikos and in Katakolon (January 2015). They also invited applications for bids for 20 blocks in the Ionian Sea and south of Crete (July 2015) (Papailias and Mavroidis 2018).

For several years, the far eastern basin (in particular, Cyprus and Israel coasts) has been the subject of an intense investigation to identify potentially hydrocarbonrich off-shore areas (Deloitte 2018). In addition to Libya, Algeria, and Egypt, off-shore oil and gas activities also take place in the waters of other North African states and the non-EU Member States around the Mediterranean Sea, such as Israel and Tunisia. In March 2013, production started at the Israeli offshore Tamar gas field, the first ultradeep field (below 1500-m depth) in the Mediterranean. In the last 15 years, oil production has remained relatively stable, whereas gas production has steadily increased with a doubling of gas production in the Mediterranean region between 2000 and 2009 (Papathanassiou et al. 2013). With respect to the atmospheric environment, the consequences of such development lie in the multiplication of sources from "controlled" burns at extraction platforms, which emit both NO<sub>x</sub> and unburnt VOCs, as well as soot and SO<sub>2</sub> leading after oxidation to sulfate particles (Papailias and Mavroidis 2018). Indirectly, this also implies the development of additional ship traffic.

#### 6 Tourism

According to EEA (2016), the Mediterranean Sea attracts millions of visitors every year. It is the world's leading tourism destination as it accounts for about one-third of all global tourism. In 2014, it accounted for more than 300 million international tourist arrivals, to which domestic tourism must be added. International tourist arrivals have grown from 58 million in 1970 to nearly 314 million in 2014, with a forecast of 500 million by 2030. About 50% of these arrivals are in coastal areas. Tourism is a major pillar of Mediterranean economies, offering consistent employment (11.5% of total employment in 2014) and economic growth (11.3% of regional GDP). In the Mediterranean basin, tourism is vital for many countries: considering exclusively the coastal areas' economy, tourism represents over 70% in terms of production value and gross value added. It is a major pillar of several countries' economies, such as Spain, Italy, Greece, or Turkey. The interlinkage between touristic activities and air pollution is dual: on the one hand, they contribute to air pollution in particular with the car traffic and aircraft-related emissions, and on the other, they require large infrastructures that are often highly demanding in energy through leisure activity and air conditioning. Further, these large infrastructures are leading to the urbanization of the coastlines bringing originally semirural areas to the emission levels of urban centers.

Tourist transport by car causes the largest impact on air quality (EEA 2015), whereas air transport accounts for the largest share of tourism-related greenhouse gas emissions (80% in 2000) in the EU-25. On the northern shore of the Mediterranean, rail, coaches, and ferries account for only 20% of all tourism trips

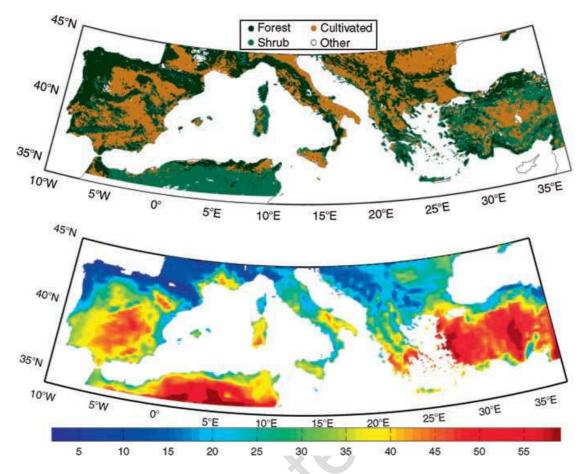
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and are responsible for a very small percentage of environmental impacts. Between 2005 and 2013, motor vehicles (private or rented) were the main means of transport (65.0%) used by tourists, followed by air and railway transport (15.3% and 11.4%, respectively). Not surprisingly, air transport was the preferred means of transport for outbound trips (53.2%), while for domestic trips, EU residents traveled mainly by motor vehicles (76.4%), followed by rail (13.6%) (EEA 2015). Obviously, the situation is different for islands as air transport accounted for more than 90% of all intra-EU inbound flows (94% to Malta and 91% to Cyprus (91%), for example). While little or no data concerning the southern shore of the Mediterranean are available, one can certainly consider that the share of ground-based motor vehicles (cars and coaches) for inbound tourist transportation is even larger than in the north due to the lack of railroad infrastructure.

In the last decade, an intensely emitting mode of transport has strongly developed: tourist cruise ships. In 2012, there were 207 cruise ships active in the Mediterranean with a capacity of 249,000 passengers and an annual total of 5.7 million passengers and 28.7 million visits to its ports (Carić and Mackelworth 2014). The European market has grown by 162% over the 10 years from 2002 to 2012, and despite the economic downturn, European cruising is expected to reach ten million passengers by 2020 (ECC 2012). Tourist cruise emissions must not only be considered as offshore emissions contributing to the general background of air pollution over both the western and the eastern basin but also as being significant in ports when maneuvering or because of hoteling activities. Emission estimates when cruising the Mediterranean sea are scarce in the literature, while the in-port emission is well documented (see APICE project; http://www.apice-project.eu, last access 1 Aug. 2021; Carić and Mackelworth 2014; Papaefthimiou et al. 2016; Murena et al. 2018; Ruiz-Guerra et al. 2019). As an example, for the Greek ports only, the emissions from international cruise ships in 2013 were estimated (Papaefthimiou et al. 2016), NO<sub>x</sub> being found dominant (2488 tons), followed by SO<sub>2</sub> and PM<sub>2.5</sub> (995 and 121 tons respectively), while the total emissions of greenhouse gases (GHG) were 124,768 tons of CO<sub>2</sub>-equivalent. Emissions during cruise ship hoteling in harbors significantly outweighed their emissions during maneuvering (89.2% vs. 10.8% of total).

7 Wildfires

When discussing the anthropization of the Mediterranean atmosphere, one has to consider the occurrence of wildfires as a significant contributor to atmospheric composition. Indeed, even if the fire has always been part of the natural forest life cycle and can even contribute to a form of regeneration, it must be recognized here that it has become an anthropogenic phenomenon. From the analysis of data in the European Fire Database on fire causality during the 2006–2010 period, Ganteaume et al. (2013) have shown that, among the 71% of forest fires in the Mediterranean



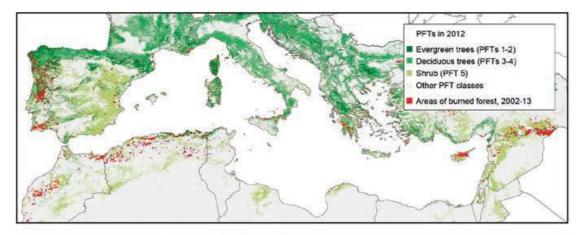
**Fig. 5** *Top* Geographical distribution of the three main vegetation types as derived from 1-km resolution Global Land Cover 2000 (GLC2000) dataset as derived from SPOT-4 VEGETATION. *Bottom* Geographical distribution of mean values of the Fire Weather Index over July–August 2007 to 2009. (Reprinted from DaCamara et al. (2014))

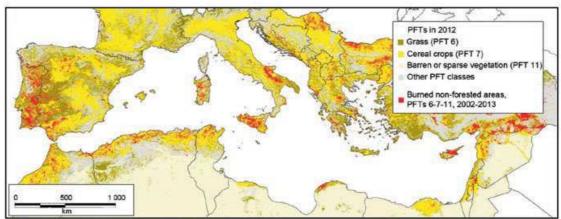
countries for which the cause was identified, more than 95% of fires were directly or indirectly caused by human agents.

In Europe, the Mediterranean gathers the large majority of forest fires: on average, 85% of the annual burned area (San-Miguel-Ayanz and Camia 2010). Because of the characteristics of the Mediterranean climate (summer dryness, occurrence of strong winds linked with a marked orography) and the nature of its vegetation, the Mediterranean area is naturally prone to wildfires, as one can verify by observing the summer average of the Fire Weather Index (Fig. 5), a parameter that combines both weather, vegetation and vegetation stress components to assess the risk of fire outbreak. As seen in Fig. 5, the hilly regions surrounding the western and eastern Mediterranean basins are especially at risk.

In addition to these risks, the main drivers of fire ignition are strongly interconnected to the evolution of the human society and, particularly, its use of forest resources. Socioeconomic developments in recent decades have led to changes such as enhanced mobility, development of tourism, and recreational activities, increasing the number of visitors to high fire-risk areas such as the Mediterranean forest or dry lands. Additionally, in many rural areas of Mediterranean Europe, fire is a

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**Fig. 6** Land cover types in 2012 (classified as plant functional types, PFT) and wildfires as detected from 2002 to 2013 from MODIS (Moderate Resolution Imaging Spectroradiometer) satellite-derived data, in the Mediterranean region. (Reprinted and adapted from Figs. 4 and 7 by Darques (2016))

traditional and long-established management tool. Further, the traditional rural socioeconomic systems that once characterized the Mediterranean region collapsed in the last few decades, resulting in a rural exodus in the northern part of the Mediterranean basin, such as in Spain, Italy, or Greece leading to desertification, which favors wildfires (see e.g., Hill et al. 2008). On the other hand, in the eastern regions, the overexploitation of natural resources (Turkey, Lebanon, or Cyprus) has led to land degradation, which is also increasing the risk of fire (Marquina 2004). Consequently, as shown by the 11-year record of fires by the MODIS satellite (Fig. 6), the shore of the Mediterranean basin encounters regular outbreaks of wildfires during the dry season.

The past 10–15 years have shown a relative stagnation of the total burned area over the Mediterranean (Darques 2016) and even a small decrease in Mediterranean EU countries (de Rigo et al. 2017). Recently, Turco et al. (2018) provided an estimation of the future trend in wildfire occurrence over the Mediterranean in the context of climate change. They conclude that the observed trend towards warmer and drier conditions in southern Europe would possibly lead to an increased risk of large fires.

TROPOMI tropospheric NO2, April 2018

NO2 tropospheric column gridded mean, TROPOMI sensor (1e-6 mol m^-2)

**Fig. 7** Nitrogen dioxide column over the Mediterranean region averaged over April 2018, derived from Sentinel-5P/TROPOMI data (http://www.tropomi.eu, last access 01 Aug. 2021). (Reproduced after https://eoportal.org/documents/163813/4638036/Sentinel5PStatus2018-2017\_AutoA.jpeg (last access 01 Aug. 2021), © modified Copernicus Sentinel data (2018) processed by KNMI)

Under 1.5–3 °C global warming scenarios, the summer burned area in Mediterranean Europe would increase in the future from ca. 40% to 100%.

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Wildfires are leading to a massive injection of reactive matter in the atmosphere. This comprises not only soot, ashes, and CO<sub>2</sub> but also CO, NO<sub>y</sub>, and a large variety of non-methane organic compounds (NMOC). For example, Alves et al. (2010) have analyzed gaseous and particulate samples from smoke from prescribed burnings of a shrub-dominated forest with some pine trees in Lousã Mountain, Portugal. Under a very strong predominance of flaming combustion, gaseous compounds, such as CO<sub>2</sub>, acetylene, and propene, were found with emission factors higher than those proposed for savannah and tropical forest fires. The emission factors of species that are favored by the smoldering phase (e.g., CO and CH<sub>4</sub>) were below the values reported in the literature for biomass burning in other ecosystems. Approximately 50% of the particulate mass was carbonaceous in nature, with a clear dominance of organic carbon. The organic carbon-to-elemental carbon ratios up to 300, or even higher, measured in the present study largely exceeded those reported for fires in savannah and tropical forests. For Portugal, Rosa et al. (2011) report that pyrogenic emissions represent 1–9% of the total greenhouse gas emission from the various economic sectors.

In fact, biomass burning (BB) is the second largest source of trace gases and the largest source of primary fine carbonaceous particles in the global troposphere (Akagi et al. 2011). Further, large NMOC emission strongly influences secondary organic aerosol (SOA) and ozone formation. This contributes to the large-scale impact of forest fire plumes over several days after their emission. These impacts involve the degradation of air quality, especially close to the fire themselves, and are

particularly detrimental when the plume trajectory reaches densely populated areas. For example, Di Carlo et al. (2015) sampled air masses from a site located 30 km downwind of a large forest fire in the Abruzzo region (Italy) and observed a massive increase in  $NO_2$  mixing ratio and  $PM_{2.5}$  concentration. Concerning secondary chemistry, by comparing observations and simulations on different days with and without a fire plume during a case study, they deduced an impact of fire emissions on the  $O_3$  mixing ratios of about 10%.

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Several studies have also been performed on the impact of biomass burning on air quality in Greece. The contribution of forest fire pollutant emissions to the total anthropogenic and natural emissions has been estimated in detail by Lazaridis et al. (2008) for a specific period during July 2000 when widespread forest fires occurred in the Greek mainland (269 fires recorded). Mesoscale air quality modeling was used to quantify the contribution of forest fire emissions to the air pollution levels in Greece. Forest fire emissions were found to be the largest contributor to air pollution in regions tens of kilometers away from the fire source. The wildfire emissions were calculated to cause an increase in the average PM<sub>10</sub> concentration, organic aerosol mass, and gaseous concentration of several pollutants, among them CO, NO<sub>x</sub>, and NH<sub>3</sub>. An average contribution of 50% to the PM<sub>10</sub> concentration over the region around the burnt area and downwind of the fire source (up to approximately 500 km) was calculated with a maximum of 80%. The average contribution of wildfires to CO was also 50%. The large wildfire events that struck Peloponnese during the summer of 2007 have also allowed a series of case studies (Liu et al. 2009; Athanasopoulou et al. 2014; Majdi et al. 2019a, b). They have shown that in rural areas impacted by fires, gaseous chemistry resembles that in urban environments and led to nitrogen dioxide and ozone exceedances in several cities in proximity to and downwind the fire spots. Due to the atmospheric dynamics, the influence was found significant in Athens from the smoke plume originating from Evia (45% of total surface PM<sub>10</sub>) and small (5%) from the fires in Peloponnese. Overall, during these events, fire emissions would contribute up to 90% of surface PM<sub>2.5</sub> concentrations in the fire regions (Balkans and Greece), with a significant regional impact associated with long-range transport. From a detailed analysis of the 2009 fire events over Peloponnese, Amiridis et al. (2012) also pointed out that fires burning near urban environments may produce particularly dense pollution plumes. They carry a mixture of anthropogenic and biomass burning pollutants, resulting in chemical processes different from those within a pure biomass burning or pure anthropogenic pollution environment and impacting very large areas.

Voluntary agricultural practices also contribute to the fire emission burden over the Mediterranean basin. Among those, one counts the use of prescribed burning to minimize the risk of wildfire and clear surfaces for pastoral activities (e.g., Favez et al. 2008; Fernandes et al. 2013). Other contributions come from regular open fire to get rid of agricultural residues, such as on the coast of Spain, where biomass residues originating from orange-tree plantations and rice fields are burnt in the open air every year during March–April and September–October. In Valencia (Spain), for example, Viana et al. (2008) have shown that open burning of rice field residues increased daily  $PM_{10}$  concentrations on a regional scale by 10– $15~\mu g~m^{-3}$  on

average, with a maximum of 30  $\mu$ g m<sup>-3</sup> on peak episodic days impacting an area of ca 18,000 ha and leading to the exposure of ca. 2.5 million inhabitants.

The impact of wildfire on the atmospheric environment is not limited to air quality reduction. By contributing to the aerosol burden at a larger scale, wildfires exhibit a significant climate impact at the basin scale (see for review the Chapter by Mallet et al. 2021).

## 8 Anthropization of the Mediterranean Region as Detected from Space Measurements

Aside from the bottom-up approach that we have followed so far in the present chapter, which consists of identifying sources, their related strength and temporal/ spatial distribution, another strategy becomes available to assess the anthropization of the Mediterranean Sea: the top-down use of satellite remote sensing. Indeed, the past 20 years have seen the development of several satellite-borne instruments such as GOMOS, MIPAS, and SCIAMACHY on board of ENVISAT (launched in 2002), IASI and GOME2 on board of the MetOp series (launched in 2006, 2012 and 2018), or TROPOMI on board of Sentinel-5P (launched in 2017) that exhibit powerful capabilities for the monitoring of both particulate and gaseous species of air quality interest. These sensors are often limited to the retrieval of the total atmospheric column of the relevant species (Doche et al. 2014)—even if recent improvements in the combination of complementary signals allow measurements in the lowermost layers of the troposphere (Cuesta et al. 2013). Nevertheless, the total retrieved column is generally useful to detect the regional pattern of the emission of those species and often reveals their anthropogenic nature. It has become a very powerful tool to explore the chemical background of the basin, especially where no or little in-situ measurements were available.

#### 8.1 Nitrogen Dioxide

Among various other pollutants, the TROPOMI instrument on board Sentinel-5P can deliver tropospheric nitrogen dioxide ( $NO_2$ ) column measurements with a resolution of up to  $7 \times 3.5$  km<sup>2</sup>. As shown in Fig. 7, TROPOMI provides a striking view of the anthropization of the Mediterranean Sea. Indeed,  $NO_2$  is mostly produced during combustion and so can be directly related to human activities, including both ground transportation (cars and trucks) and ship traffic, but also industrial activities, electricity production, and residential heating.  $NO_2$  has only limited natural sources, its atmospheric lifetime toward oxidation is only of the order of a few days, and its vertical profile is such that one can consider the pattern drawn by the tropospheric

column density as a trustful reflection of the lowermost troposphere concentrations.

Figure 7 reveals the strong footprint on the atmospheric environment of major cities such as Madrid, Barcelona, Marseille, Turin and the Po Valley, Rome, Naples, Athens, Izmir, Istanbul, Ankara and the Iskenderun Bay on the northern side of the basin, Algiers, Tunis, Tripoli, Cairo and the Nile Delta on the southern coasts, Beirut and Israel on the eastern shore. Urban centers and population densities provided in Fig. 1 can almost perfectly be superimposed on this remote sensing product revealing how Megacities act as hot spots of air pollution all around the Mediterranean (Kanakidou et al. 2011). In North Africa and the Middle East, the background level seems generally lower than in the North, which is probably due to the lower level of distributed infrastructures, but the urban centers seem much more intense emitters than in the northern shore. Here the lack of regulation coupled with a more difficult access to the latest technologies as well as the greater use of individual power production systems to the detriment of common and more effective infrastructures probably explains this situation (e.g., see Waked et al. (2015) for Beirut).

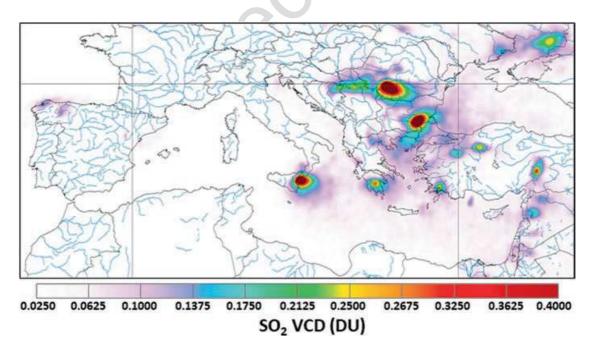
Major waterways can also be detected, demonstrating how the high NO<sub>2</sub> emission rate of cargo ships coupled with the high density of the major routes across the Mediterranean Sea (see Fig. 4) are strongly impacting the nitrogen dioxide background even far away from the urban centres. The major ground transportation networks are also well visible. Major roads, railways, freeways, industrial sites, and power generation installations often gather in the same valleys. It is hence remarkable to observe how the Po valley is a source of nitrogen oxides for the Adriatic Sea, and the Rhone valley pollution is spread towards Corsica and the Ligurian Sea following the mistral pathways.

Combined with the close sea nature of the Mediterranean and its peculiar aerology (Dayan this volume), all these features contribute to a significant nitrogen oxides background over the Mediterranean basin, twice as high as the one found in the eastern part of the Atlantic Ocean (Fig. 7). With respect to the atmospheric chemistry of the Mediterranean area, this high background is not without consequence. Nitrogen oxides (NO<sub>x</sub>) photo-interconversion (Finlayson-Pitts and Pitts 2000) is, indeed, the propagating force that drives the radical oxidation of pollutants during their atmospheric transit leading to oxidized species, secondary aerosols, and ozone. Being bathed with these anthropogenic emissions of nitrogen oxides and naturally gifted with sunlight, the Mediterranean area is exhibiting a very active photochemistry that governs the fate of many atmospheric pollutants such as volatile organic species of both biogenic and anthropogenic origins or SO<sub>2</sub>, leading to high ozone (e.g., Butković et al. 1990), and secondary organic, nitrate, and sulfate aerosol (e.g., Simó et al. 1991; Kostenidou et al. 2015; Drugé et al. 2019) concentrations.

#### 8.2 Sulfur Dioxide

Sulfur dioxide is an irritating gas that is well placed among the list of regulated species by air quality regulation. It is also a very efficient precursor of particles: its oxidation by atmospheric oxidants such as OH radicals and ozone leads to sulfuric acid that has a sufficiently low vapor pressure to condense and form secondary sulfate over preexisting particles or even to form new ultrafine particles during nucleation events (Sellegri 2021). As a strong acid, it also contributes to the acidification of precipitation (Avila and Rodà 2002) and is prone to react with atmospheric bases such as ammonia to form new particles (Koçak et al. 2004). SO<sub>2</sub> emissions are strongly regulated in Western Europe by controlling the sulfur content of fuels which is much less the case in eastern Europe, the Middle East, and North Africa, as well as in offshore areas (see above *Ship traffic* Sect. 4).

Instruments such as TROPOMI, IASI, or GOME-2 have the ability to retrieve sulfur dioxide vertical columns. Here we focus on reanalysing 2004–2009 OMI data on-board the AURA satellite by Theys et al. (2015). Results presented in Fig. 8 illustrate very well the anthropization of the Mediterranean basin. If volcanic sulfur dioxide emission from Mount Etna in Sicily is well visible, this long-term average brings evidence that non-volcanic sources are also significant, if not major, for the SO<sub>2</sub> contamination of the eastern basin. It illustrates how the mid-troposphere and boundary layer circulation patterns described in Lelieveld et al. (2002) are efficient in bringing towards the Mediterranean area the pollution emitted in central and eastern Europe. Sulfur emissions from the Danube valley and Balkans are then



**Fig. 8** Vertical column density (VCD) of  $SO_2$  in Dobson unit (DU), as retrieved from space by OMI/AURA, averaged for the 2004–2009 period on a  $0.25^{\circ} \times 0.25^{\circ}$  grid, and showing emission hot spots over Europe. Cloudy pixels and measurements affected by strong volcanic eruptions are excluded from the analysis. (Reprinted from Fig. 6 in Theys et al. (2015))

recirculated from the southern Black Sea area and feed the Aegean and Levantine areas with sulfur dioxide. Beyond the fact that these emissions lead to the formation of particulate sulfate, SO<sub>2</sub> can be seen here as a tracer, and one should consider that many other co-emitted reactive species and contaminants are transported along the same routes. Local industries nevertheless can have a major impact, as demonstrated by the clear detection of the lignite-based Megalopolis power generation center in the central Peloponnese (Fig. 8).

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9 Conclusion 458

We have reviewed here in a non-exhaustive way the major sectors responsible for the main anthropic pollutant sources in the Mediterranean area. This overview is not only intending to provide starting points for a more targeted and thorough evaluation of anthropic sources, it also depicts a critical panorama of the human activities over the Mediterranean. Indeed, in addition to exhibiting a very peculiar geographical pattern (see Sect. 1.2), in addition of being an interface between Africa and Europe and between Asia and the Atlantic, in addition of being a tipping point area in the context of climate change, the Mediterranean area is under the pressure of unprecedented anthropic forcings that strongly impact the composition of its atmospheres. All sectors of human activities seem to have reached such levels of development-and sometimes of non-regulation-that they present acute impacts on the atmospheric environment. Considering the relatively small size of the basin and the mix of recirculation processes, this variety of sources leads to a strong complexity of the Mediterranean background atmosphere. Moreover, the extrapolation of temporal trajectories of the various emission sources is a dreadful scenario: population, tourism, ground transportation, sea transportation, and fossil fuel extraction are expected to increase in the coming years, preparing the Mediterranean atmosphere to be the place of a future silent environmental disaster.

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