Chapter 4

Air quality and climate in the Mediterranean region

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Abstract

Ambient air ranks number one among the natural resources vital to human beings, with an average individual daily need of 12 kg. Due to the specificities of the Mediterranean region (sunny, hot and dry climate; long-range transport converging over the basin), air pollution in reactive compounds over the Mediterranean is often higher than in most European inland regions. Climate change (increase in temperature and drought) and anthropogenic pressure (growing population) should significantly impact the regional air quality. As a result, Mediterranean inhabitants who are already regularly exposed to pollutant loads well above WHO air quality recommendations will be further exposed, resulting in an excess of premature deaths. Exposure monitoring and win-win strategies should be developed in the future both to improve air quality and develop a low carbon economy. The evolution of emissions under climate change is not always clear and much uncertainty remains around present emissions from large urban-industrial centers, although recent progress has been made on emissions of the different regional sources of pollutants. It has been established that the regional climate and water cycle are affected by atmospheric chemistry. By reducing solar radiation at the surface, aerosols reduce the yearly average precipitation in the Mediterranean by 10%, which is a major issue since water is already scarce. Aerosols could further reduce precipitations by reducing the size of cloud droplets or through the formation of cloud droplets and ice crystals. Moreover, recent *in situ* and model experiments indicate that anthropogenic nitrogen and desert dust phosphorus deposition in nutrient-depleted surface seawater favors phytoplankton development, which stimulates the sink of atmospheric CO_2 into marine sediments. But Saharan dust deposition by rain also stimulates bacterial growth, which reemits CO_2. The net effect of desert dust deposition at large scales needs to be established.

Résumé

L'air est sans aucun doute la ressource naturelle la plus essentielle à l'homme : chaque jour 12 kg d'air sont nécessaires à sa survie. Du fait des spécificités de la région méditerranéenne (climat ensoleillé, chaud et sec ; convergence de masses d'air d'horizons lointains), la pollution de l'air en espèces réactives y est souvent plus forte que dans la plupart de l'Europe continentale. Les changements climatiques (augmentation des sécheresses et de la température) et la pression démographique devraient dégrader encore la qualité de l'air. En conséquence, les habitants de la Méditerranée qui sont déjà régulièrement soumis à des niveaux de pollution bien au-dessus des recommandations de l'OMS devraient se trouver plus exposés encore, ce qui engendrera une surmortalité. Un meilleur suivi de l'exposition des habitants et des solutions « gagnant-gagnant » devraient être mises en place dans le but d'améliorer la qualité de l'air et de s'engager dans une économie décarbonée. Les conséquences des changements climatiques sur les émissions de polluants par les principales sources régionales ne sont pas toujours très claires. Il a été établi que le climat régional et le cycle de l'eau sont altérés par la chimie de l'atmosphère. En réduisant le flux solaire en surface, les aérosols réduisent les précipitations moyennes annuelles de 10 % en moyenne sur le bassin méditerranéen, réduisant un peu plus une ressource déjà rare. Les aérosols pourraient réduire plus encore les précipitations en réduisant la taille des gouttes d'eau dans les nuages ou en agissant sur la formation de cristaux de glaces. Par ailleurs, de récentes expériences indiquent que le dépôt atmosphérique d'azote et le phosphore issu des poussières désertiques à la surface des eaux pauvres en nutriments de la Méditerranée favorise le développement du phytoplancton activant par la même occasion l'absorption de CO_2 par l'océan. Cependant, il a aussi été observé que le dépôt de poussières favorise le développement de bactéries qui elles-mêmes rejettent du CO_2 du fait de la respiration. L'effet net du dépôt de ces poussières à grande échelle reste à établir.

Introduction

In this chapter, we consider relatively short-lived (<~1 month) particulate and gaseous tropospheric trace species that cause atmospheric pollution and have two-way interactions with climate. Emissions of long-lived greenhouse gases and their role, and evolution with climate change are dealt with elsewhere in this book.

Ambient air is one of our vital natural resources. Air quality in the Mediterranean region is generally poor, due to both particulate and gaseous pollution. For instance, particulate or ozone concentrations are generally higher in the

Mediterranean region than in most continental European regions, especially during the long dry and sunny summer season that characterizes the Mediterranean type of climate, (e.g. NABAT et al. 2013; DOCHE et al. 2014; MENUT et al. 2015). In addition, the Mediterranean region is a hot-spot for climate change (GIORGI and LIONELLO, 2008), whereas numerous two-way interactions take place between climate and air quality, which are not always well understood. In addition, the Mediterranean region is expected to undergo a major increase in population, especially the development of large urban centers on its eastern and southern sides (CIHEAM, 2009). Air quality is already very poor in such centers and has significant adverse effects on health (an average of more than 20 deaths per 100,000 inhabitants took place in 2008 in Egypt, Greece, Israel, Lebanon, Turkey; WHO, 2014). It is thus important to understand the impact of the expected climate change on atmospheric chemistry and the resulting surface air quality in the Mediterranean region (COLETTE et al. 2013).

Conversely, atmospheric pollution also affects the climate. The most obvious effect is global warming due to anthropogenic emissions of greenhouse gases (GHG), but anthropogenic aerosols and ozone, for instance, also perturb the Earth's radiative budget (IPCC, 2014). Up to now, future climate predictions have neglected feedback due to atmospheric composition, apart from that due to long lived GHGs. Atmospheric pollution, however, is also made up of many gaseous and particulate species that are more chemically active than GHGs and these short-lived species have various reciprocal interactions with climate, which must be accounted to better simulate their combined evolution. For instance, direct effects due to scattering and absorption of solar radiation by tropospheric anthropogenic aerosol particles compensate for and even cancel out the warming effect of GHG emissions in polluted regions at the regional scale (BERGAMO et al. 2008). But they are much more difficult to represent in climate models (i) because of the high temporal and spatial variability of their concentrations and optical properties whereas, in comparison, GHG vary very little in concentration (LE TREUT et al. 1998), and (ii) because to properly assess aerosol climatic impact, it is necessary to use coupled atmosphere-ocean dynamical models rather than fixed sea-surface temperature to account for a radiative impact that affects sea surface evaporation (NABAT et al. 2014). These reactive species form secondary products that are not directly emitted into the ambient air but control the concentration of fine particles in the background (QUEROL et al. 2009) and urban (EL HADDAD et al. 2011) Mediterranean air, they can be transported over long distances in the troposphere (LELIEVELD et al. 2002; RICAUD et al. submitted) and affect air quality at the surface (with profound impacts on human health) (KÜNZLI et al. 2000), perturb the radiative budget of the atmospheric column and modify cloud properties with significant consequences for the atmospheric water cycle and climate (NABAT et al. 2015), affect marine biogeochemistry (GUIEU et al. 2014a) and continental vegetation (PAOLETTI, 2006) through their deposition at the Earth's surface. There are thus many processes and different types of feedback to take into consideration when assessing interactions between air quality and climate and when modelling the present and future coupled air quality-climate system.

The present chapter is a contribution from the project ChArMEx (the Chemistry-Aerosol Mediterranean Experiment; http://charmex.lsce.ipsl.fr) of the multidisciplinary regional research program MISTRALS (Mediterranean Integrated Studies at Regional and Local Scales; http://mistrals-home.org) endorsed by ALLENVI. It summarizes current knowledge on the links (feedforward and feedback) between climate and the air resource in the Mediterranean region, on the impact of expected climate change and increasing anthropogenic pressure on that natural resource and its consequences for human health, highlighting on-going efforts and recommended research to overcome critical limitations of our present knowledge. In the following sub-chapter headed 'Sources of reactive species and source apportionment', we review emissions that affect the Mediterranean atmospheric environment and report source apportionment results in both coastal urban and background air. In a separate text box, we also present the regional emission database ECCAD/ChArMEx dedicated to the Mediterranean region. We describe the particular cases of large urban centers, aeolian erosion, and biogenic emissions of volatile organic carbonaceous species. In the sub-chapter headed 'High concentrations of aerosols and pollutant and greenhouse gases', we address the question of the high atmospheric loads of atmospheric pollutants in the Mediterranean region and our understanding of the reasons. We include a focus on secondary organic aerosols, a major component of fine particles in the ambient air and a challenge for atmospheric chemistry models because they are formed by complex chemical reaction chains from gaseous compounds emitted by both human activities and natural sources. The sub-chapter headed 'Atmospheric deposition to nutrient-depleted seawater' is dedicated to the deposition of aerosol particles to the oligotrophic Mediterranean Sea, a major pathway for the transfer of nutrients and contaminants, and its impact on surface marine ecosystems. The sub-chapter headed 'Impact of atmospheric chemistry on the regional climate' describes the impacts of atmospheric chemistry on climate and reviews recent results on their assessment. In the sub-chapter headed 'Impacts of air quality on health in the Mediterranean region', we report recent results on the detrimental effect of air pollution on human health. In the sub-chapter headed 'The (uncertain) future of air quality', we question our knowledge on the evolution of air quality in the coming decades. Finally, we summarize the main recent advances, open questions and related ongoing research and perspectives.

Sources of reactive species and source apportionment

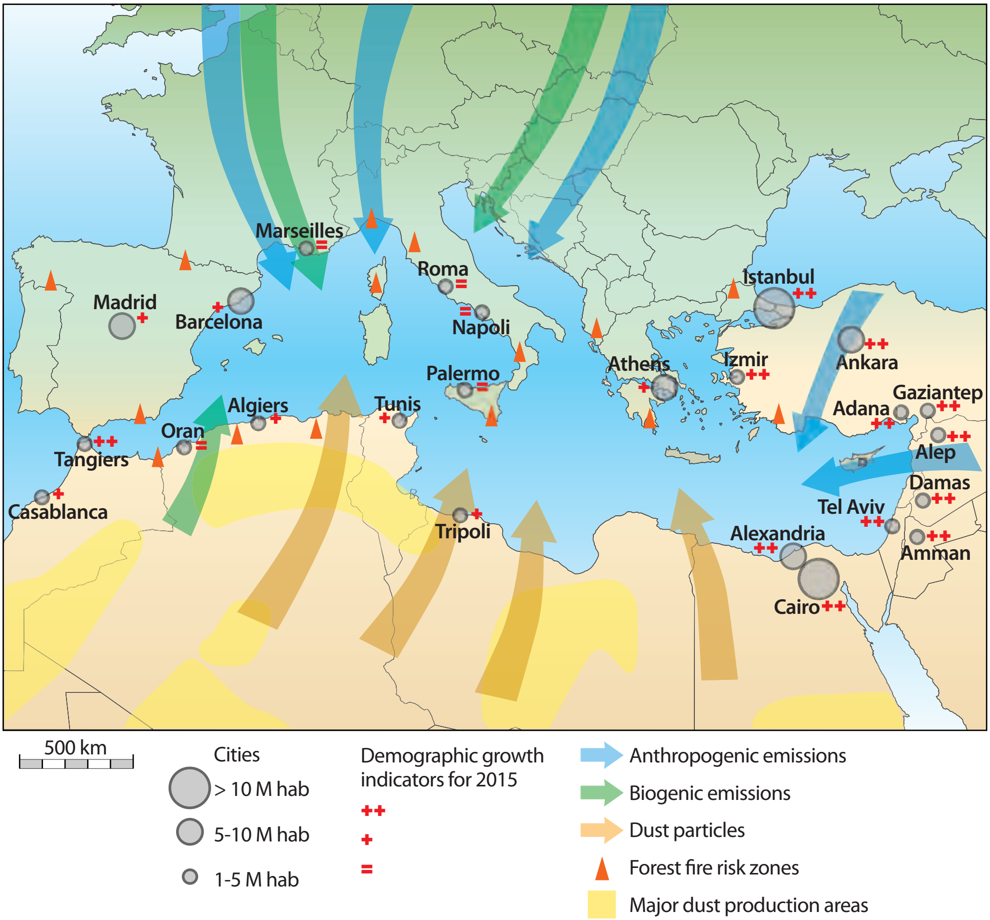

Figure 1
Variety of continental sources impacting the Mediterranean basin.

Natural and anthropogenic emissions of gaseous and particulate pollutants are key factors in air quality degradation and climate change. As indicated by the Latin origin of the name Mediterranean ("the middle of the lands"), the Mediterranean basin is a receptor of anthropogenic emissions from eastern Europe and surrounding coastal urban areas combined with wind-driven dust from the Sahara and Arabian deserts, biogenic emissions from the surrounding vegetation, and sea salt [KANAKIDOU et al. 2011; Fig. 1].

Natural sources

Natural sources play a key role in the exchange of compounds between the Earth's surface, the oceans and the atmosphere. The species emitted are characterized by high diversity and potentially high chemical reactivity. Consequently they have direct and indirect impacts on climate and air quality.

Marine sources

The sea surface produces airborne particles that contribute to the aerosol load, Earth's albedo, climate, and air quality in marine environments. There are large gaps in our knowledge of marine emissions, which, in turn, are responsible for large uncertainty on our future climate (CARSLAW et al. 2013). *Marine primary aerosols* are produced by bubble bursting processes, mostly under the influence of wind-driven wave breaking mechanisms. Primary marine organic particles consist of microbiological organisms (including viruses and bacteria), biological debris, exudates and by-products. *Marine secondary aerosols* are formed by condensation of gas-phase species emitted from the seawater. Secondary organic aerosol (SOA) particles are expected to result from the atmospheric oxidation of biologically driven emissions of volatile organic compounds (VOC), which have not yet been clearly identified. Among the formation pathways of secondary aerosol particles, nucleation is the process responsible for the formation of new nanoscale particles (as opposed to the process of condensation onto pre-existing particles). Triggered by photochemical processes, new particle formation (NPF) takes place as an "event" that lasts several hours, during which the concentration of the clusters of nanoparticles increases to high levels by nucleation. These clusters grow rapidly to a few nanometers in size when they can be detected. NPF is expected to generate a large number of aerosols, which, in turn, can affect climate by influencing cloud radiative processes (SPRACKLEN et al. 2006). Questions are still open concerning the conditions that favor the occurrence of NPF and particularly the type and the origin of precursors. How are VOC emissions and nucleation events influenced by

Box 1
ECCAD/ChArMEx regional Mediterranean emission database

The production of the ChArMEx state-of-the-art specific regional emission inventory for the preceding decade began in 2011 using the most recent literature, starting with yearly anthropogenic fossil and biofuel emissions in southern Europe from TNO (Kuenen et al. 2011; Liousse et al. 2014) completed by estimates for northern Africa (Assamoi & Liousse, 2010; Liousse et al. 2014). It was further completed with key emissions from agricultural and forest biomass burning (Turquety et al., 2014), soils for dust (Callot et al. 2000) and for NO_x (Yienger & Levy 1995), aircraft (Riahi et al. 2007), shipping, volcanoes (Andres & Kasgnoc, 1998), sea surface (Schwier et al. 2015), and vegetation (Guenther et al. 2006). The domain of interest extends from 10°N (tropical Africa) to 70°N (northern Europe) and from 20°W (Iceland) to 50°E (Caspian Sea) sometimes with a spatial resolution of 10 km. Fig. 2 summarizes the content of the database.

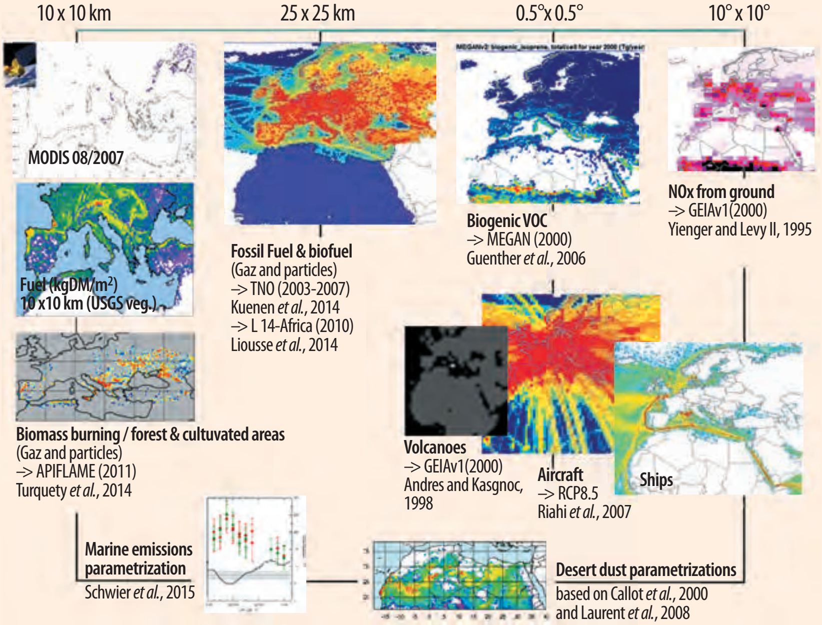

Figure 2
The different emission subsets and emission parameterizations available in the ECCAD/ChArMEx regional emission inventory (http://www.aeris-data.fr/redirect/eccad/ChArMEx).

water composition (clean or polluted seawaters), biological composition and activities? In the framework of ChArMEx, marine aerosol emissions to the atmosphere have recently been studied in two research projects, MEDSEA (MEDiterranean Sea Acidification) and SAM (Sources of marine Aerosol in the Mediterranean atmosphere). Both used mesocosms, i.e. semi-opened chambers containing natural seawater and an atmospheric headspace. Results showed a clear correlation between the level of seawater chlorophyll *a* (Chl-*a*), measured as the standard proxy for phytoplankton biomass, and the amount of organic compounds in airborne particles produced during the wave breaking process (Schwier et al. 2015). This result is in agreement with parametrizations obtained in the Atlantic Ocean combining Chl-*a* levels measured by satellite and the organic fraction of ambient marine aerosol measured at receptor sites (Rinaldi et al. 2013). Seawater biology also influences gas phase emissions, and the number of particles formed from some of these gas phase species by nucleation. SAM experiments showed that some VOC emissions increase during phytoplankton blooms. In parallel, several nucleation events were seen to be initiated from seawater emissions both in the mesocosms and in laboratory experiments. The experiments also identified iodine species that trigger the formation of new particle clusters after they reach a threshold concentration, and excluded the usually suspected precursor dimethyl sulfide (DMS) in the northwestern Mediterranean region investigated (Sellegri et al. 2016). Another result revealed that emitters of iodine species are not linked to Chl-*a* as expected, but to other biological tracers. Furthermore biologically driven emissions of amines appear to contribute strongly to early growth of the cluster in the 1-10 nm size range. These findings advance our understanding and our ability to model the complex climate feedback loop that involves temperature, biological populations in the seawater and marine aerosol emissions.

Emissions from vegetation

Ecosystems are a notable source of a wide variety of reactive volatile organic compounds (VOCs), biogenic VOCs (BVOCs), such as isoprene, monoterpenes, methanol, and many others. BVOC emissions largely dominate anthropogenic VOC emissions at the global scale. BVOCs play a key role in atmospheric chemical processes particularly in the ozone cycle and in the formation of secondary organic aerosols (SOA). Due to high temperatures, high levels of solar radiation, and high biodiversity, to which BVOC emissions are very sensitive, the Mediterranean region is a major source of BVOCs (Owen et al. 2001), and has a significant impact on ozone and aerosol formation (Sartelet et al. 2012). Models of the correct meteorological and chemical conditions that are able to reproduce the diurnal variation in isoprene emissions (Guenther et al. 2012) are needed to understand the underlying atmospheric photochemistry.

Large uncertainties persist in the composition, distribution and levels of BVOC emissions. Both experimental and modeling studies are thus crucial to improve our knowledge, especially in the context of climate change that is projected to severely impact the Mediterranean region. As part of the ChArMEx project, a thorough case study was performed at the OHP Oak Observatory (Haute Provence, France) in a coppice of downy oak, a widely represented tree species in the Mediterranean area. Measurements from the branch (Genard-Zielinski et al. 2015) to the canopy scale confirmed high isoprene emissions (up to almost 10 mg m^{-2} h^{-1}), significant methanol emissions (up to 0.63 mg m^{-2} h^{-1}), and negligible monoterpene emissions (Kalogridis et al. 2014). The isoprene degradation within the canopy was found to be very low (<3%) due to the low level of NO_x and the low canopy height (Kalogridis et al. 2014). Measurements showed that isoprene emissions increased with an increase in radiation and air temperature, and latent heat flux was also shown to be a useful parameter to explain variations in isoprene emissions (Baghi, 2013). A study of the potential impacts of climate change on water resources (namely a 30% water deficit, as foreseen for the year 2100) suggests a significant increase in isoprene emissions in the future, irrespective of the warming scenario (Genard-Zielinski et al. submitted).

Atmospheric chemistry is being modeled to simulate the fate of BVOC emissions on a typical hot sunny day (July 3) of the 2014 ChArMEx airborne experiment above the OHP. Preliminary results show that strong diurnal emissions of BVOC lead to a clear SOA formation event.

Aeolian erosion/soil dust emissions

Dust emission results from the erosion of soil by wind (Bagnold, 1941; Gillette, 1981), which mainly occurs in the arid and semi-arid regions of the Earth, the Sahara desert and its fringes being considered as the main source region in the world. The resulting aerosols both scatter and absorb solar and Earth radiations, which affect the Earth's radiative budget. When deposited on the Earth's surface, mineral dust contributes to the input of growth-limiting macro and micro nutrients to oceanic surface waters (see the dedicated sub-chapter hereafter) and terrestrial ecosystems.

In North African countries, the rapid increase in population has led to a growing demand for agricultural products. As a result, the pressure on natural resources is increasing steadily with the expansion of cultivated areas stimulated by the introduction of modern plowing techniques. Beginning in the 1960s, the disc plow pulled by powerful tractors has progressively replaced the traditional mold board plow pulled by draft animals, which affects dust emissions.

As can be seen in Fig. 3, wind erosion is more than one order of magnitude greater on land tilled with a disc plow than in fields tilled with a mold board plow, land prepared with a tiller being between the two. These results strongly suggest that new tillage techniques such as using a disc plow drastically increase soil erosion by wind in agricultural fields with loose soils. They also confirm that traditional tillage tools like the mold board plows are the most suitable tillage tools to preserve soils in semi-arid agricultural regions. Finally, these results suggest that dust emissions from North African countries are increasing because of changes in land use management rather than because of the extension of the cultivated area. For example, Yoshioka et al. (2005) suggest that this type of land preparation could now be responsible for up to 25% of North-African dust emissions.

A monitoring station was recently set up close to Medenine (southern Tunisia; http://193.95.22.108/) for the long term monitoring of the surface atmospheric concentration and atmospheric column load of soil dust, and of the dust deposition flux.

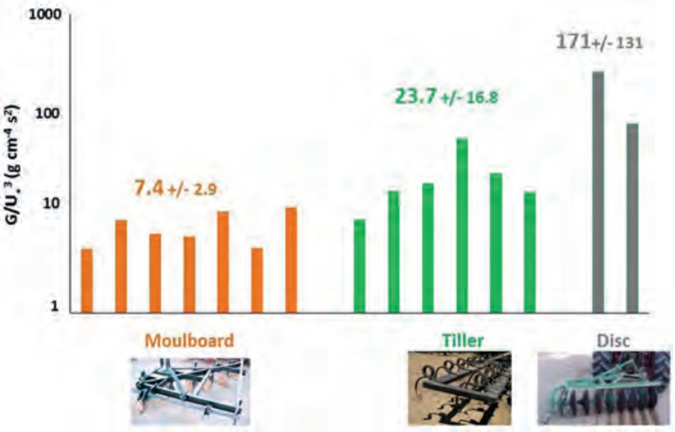

Figure 3
Normalized horizontal flux (G) of sediments eroded by wind in fields prepared by three different types of plow used in Tunisia (adapted from Labiadh *et al. 2013).*

Source apportionment

Urban areas

The coastal areas of the Mediterranean include megacities like Cairo (12 million inhabitants), Istanbul (12 M), Athens Great Area (5 M), and Barcelona (5 M). All these cities are subject to heavy gaseous and particulate pollution. The population of these regions will continue to increase, especially in the eastern part of the basin, leading to a higher anthropogenic pressure in a context of climate change.

Satellite images of nitrogen dioxide columns from SCIAMACHY identified coastal urban areas in the Middle East as hot-spots of pollution in the region (Lelieveld et al. 2009). Global emission inventories all agree on a marked increase in anthropogenic emissions of major pollutants (NO_x, VOC and $PM_{2.5}$) in the Middle East Area (MEA), in contrast to what is observed in post-industrialized regions like Europe and the USA. In the coming decade, anthropogenic emissions from MEA are projected to be even larger than those from Europe and the USA (SALAMEH et al. submitted).

Anthropogenic emission inventories provide key input data for the atmospheric models used for the prediction of air quality and for the study of the most efficient regulations to reduce air pollution. The quality of emission inventories from rapidly growing megacities in the southern and eastern Mediterranean is of concern because local emission data are sparse. A new road traffic emission inventory was built for Algiers, where road traffic is a major source of atmospheric pollution. It was validated by comparing high resolution (4 km) simulations with the regional atmospheric chemistry and transport model CHIMERE and observed air quality measurements of NOx and CO (RAHAL et al. 2014). Some highly resolved inventories have also recently been developed at the regional scale for Beirut (WAKED et al. 2012) and Istanbul (MARKAKIS et al. 2012), but their uncertainties are unknown. As part of the TRANSEMED initiative supported by the MISTRALS/ENVIMED program, a source-receptor methodology was developed for emission inventory evaluation. The approach consists in combining existing and newly collected observations and complementary source-receptor approaches (i.e., urban enhancement emission ratios, multivariate models like positive matrix factorization, PMF) in large urban areas like Beirut (Lebanon), Istanbul (Turkey), and more recently Athens (Greece) and in the near future, Cairo (Egypt). Very detailed databases of ambient and near-source observations are being built with a focus on the composition of gaseous organic carbon. The results recently obtained for Beirut (SALAMEH et al. 2014, 2015, 2016) showed (i) the extremely high levels of pollution for organics, (ii) the dominant effect of traffic emissions on concentrations of VOC, (iii) the poor spatial variability of speciated NMHC traffic emissions regardless of the region, and (iv) the high uncertainty and discrepancies between large scale emission inventories compared to observational constraints and local scale inventories (see also ABDALLAH et al. 2016).

Source apportionment at the regional scale

Sources impacting the Mediterranean basin can be apportioned at regional scale using models that exhaustively account for all the processes (emission intensity, chemistry, air mass transport) that occur between the sources and the receptor zones. This approach has been applied for the summer (JJA) 2012 period at a resolution of 50 km using the chemistry-transport model CHIMERE dynamically forced by the model WRF (REA et al. 2015). The contributions from different sources of particles and gaseous precursors to both the surface particulate air quality ($PM_{2.5}$ and PM_{10}) and the aerosol optical depth (a proxy of the aerosol concentration in the vertical column) were determined by eliminating particle sources one by one (anthropogenic, fire, soil dust, vegetation, sea), and comparing the results with the reference simulation with all the sources activated. Results showed that desert dust had the most influence on surface PM concentrations in the Mediterranean basin (up to 86% of PM_{10}) followed by anthropogenic aerosols (up to 75% of $PM_{2.5}$) in Western Europe. Sea salts also had a significant influence (up to 29% of PM_{10}) in Atlantic and Mediterranean coastal regions.

Another approach to apportioning sources at regional scale uses receptor oriented methods focusing on the chemical composition of pollutants measured at a representative receptor site. Two intensive observation campaigns were conducted as part of the ChArMEx research program in Corsica (2013) and in Cyprus (2015). Statistical analysis of the chemical composition of both gaseous and particulate pollutants measured at Cap Corsica combined with the residence time analysis of air mass trajectories pointed to the contribution of anthropogenic sources located in regions characterized by intense anthropogenic activities (e.g. the Po valley and south-eastern France, both located in the north-western Mediterranean basin). In addition to primary (i.e. directly emitted) volatile organic compounds (VOCs) emitted by biogenic sources (BVOCs), a group of secondary pollutants composed of first-generation oxidation products of BVOCs was also identified, while another group was characterized by more oxidized VOCs (OVOCs) of both biogenic and anthropogenic origin. The combined analysis of VOC and aerosol compositions in PM_1 showed that during periods under a dominant biogenic influence, aerosol composition was dominated by the secondary organic fraction, whereas during periods of long range transport of anthropogenic emissions, the relative contributions of inorganic and organic fractions were the same. These results underline the importance of considering the roles of both anthropogenic and natural emissions in particulate pollution. The same approach is being applied using the observations acquired in Cyprus as representative of the eastern part of the Mediterranean basin.

Sub-chapter 1.4.2

High atmospheric concentrations of aerosols, greenhouse gases and other pollutants

Evidence is growing for the deterioration of air quality over the Mediterranean basin. In this sub-chapter, we review the current situation over this semi-enclosed basin. We describe the spatio-temporal variabilities of the pollutants, greenhouse gases (GHGs) and aerosols, observed and modeled over the Mediterranean basin and how they help trace physical-chemical processes at regional and global scales through long-range transport. In a separate text box (see below), we focus in particular on secondary organic aerosols (SOA), a major component of fine particles, for which data and simulations were particularly scarce and uncertain especially over the western Mediterranean basin before ChArMEx. The Mediterranean basin is located in a transition zone between subtropical and mid-latitude climate regimes (LIONELLO, 2012), and is highly sensitive to climate change. In terms of sources of anthropogenic pollution, the basin is located at the intersection of three continents, Europe, Africa and Asia.

Satellites and models (e.g., LELIEVELD et al. 2002; NABAT et al. 2013) together with campaigns such as the Mediterranean Intensive Oxidant Study (MINOS) (LADSTÄTTER-WEISSENMAYER et al. 2003; SCHEEREN et al. 2003) show that, during the warm and dry Mediterranean summer season, air pollution above the Mediterranean often exceeds that observed over most parts of Europe. This is due to the convergence of European, African and Asian polluted air masses, to the absence of rain to clean up the atmosphere, and to the high insolation, which favors the formation of secondary pollutants like ultrafine particles or ozone. Natural aerosol pollution could originate from sources such as the African and Arabian deserts, active volcanoes, vegetation, or the sea surface. Pollutants, GHGs and aerosols originating from Asia can be trapped in the Asian monsoon and entrained to the upper troposphere before being redirected towards the eastern Mediterranean basin via the Asian monsoon anticyclone, where they accumulate and are subject to subsidence (RICAUD et al. 2014).

Continental sources including industrial and densely populated coastal areas (Kanakidou et al. 2011; Im and Kanakidou, 2012) or forest fires (Cristofanelli et al. 2013) affect the ozone (O_3) and carbon monoxide (CO) budgets with which methane emissions (CH_4) interplay through complex reactions with nitrogen oxides (NO_x) (Dentener et al. 2005), although the impacts of the respective source types are still not fully understood. Polluted air masses affecting the Mediterranean basin may originate from Europe (e.g., Pace et al. 2006), Asia (e.g., Lelieveld et al. 2002; Randel and Park, 2006), Africa (e.g. Ziv et al. 2004; Liu et al. 2009) and even North America (e.g., Formenti et al. 2002; Christoudias et al. 2012).

The transport and dispersion conditions of atmospheric pollutants over the western and eastern Mediterranean basin differ, as illustrated in Fig. 4, which shows the contrasted vertical air motion in the western and eastern parts of the basin in summer. The subsiding air aloft induced by the descending branch of the Hadley global circulation cell affecting the eastern basin, and the depth of the Persian Trough (an extension of the Indian monsoon), control the spatio-temporal distribution of the boundary layer (BL) height during summer. The resulting shallow mixed layer and weak zonal flow, leads to poor ventilation

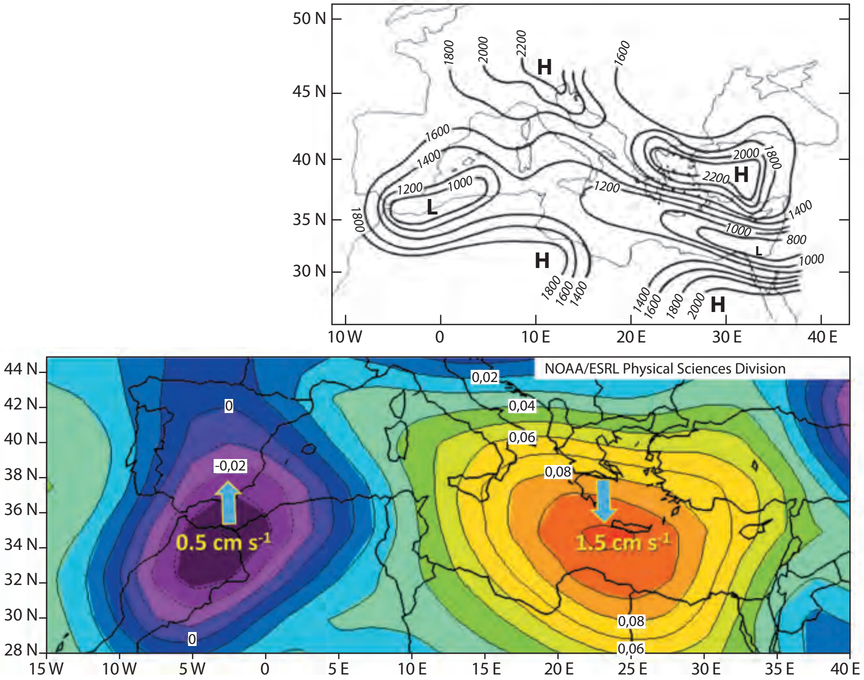

Figure 4

Top panel: June to August 1948-2015 average of the NCEP/NCAR model omega (Pa s^{-1}) at 500 hPa (about 5.5 km altitude) showing a maximum downward air motion of about 1.5 cm s^{-1} over Crete in the eastern Mediterranean, contributed by the descending branch of the African and Asian monsoon, and a maximum upward motion of about 0.5 cm s^{-1} over the westernmost Mediterranean basin. Bottom panel: spatial distribution of the summer mixed layer height over the Mediterranean (from Dayan et al. 1996).

rates, inhibiting the efficient dispersion of the pollutants. Several studies pointed to specific local (e.g. ventilation rates) and regional peculiarities (long-range transport) that enhance the building up of pollutant concentrations (WANGER et al. 2000; MATVEV et al. 2002; EREL et al. 2007; RUDICH et al. 2008; DRORI et al. 2012) over the eastern Mediterranean basin.

Considering the long-range transport climatology that characterizes the Mediterranean basin, two different regimes can be observed: 1) From fall to spring with predominance in winter, and from the boundary layer to the upper troposphere, air masses mostly come from either Europe or the eastern Atlantic Ocean. 2) In summer, the origin of air masses affected by long-range transport over the eastern and western Mediterranean basin is distinct, more complex and altitude dependent. In the lower troposphere over the western basin, convective cells develop within the boundary layer with mostly air masses coming from Europe, northern Africa and eastern Atlantic Ocean. The coasts and mountains surrounding the western basin favor the development of mesoscale recirculations in summer that lead to the formation of ozone- and aerosol-rich layers above coastal areas and the sea (e.g. MILLÁN et al. 2000). Over the eastern basin, the air masses originate from four major source regions: (i) west-north-west long fetch of maritime European air masses all year round, (ii) north-west flow originating in south-eastern Europe (etesian winds) in summer, (iii) south-east flow from the Arabian Peninsula in the fall, and (iv) south-west flow along the North African coast most frequent in late winter and spring (DAYAN, 1986). In the mid-troposphere, whatever the season, air masses in both parts of the basin mainly come from the west. In summer, upper tropospheric air masses in the western basin mainly come from the west, but in the eastern basin, they also come from North Africa and the Arabian Peninsula (ZIV et al. 2004; LIU et al. 2009), and even farther away, from Asia (LELIEVELD et al. 2002).

Several airborne campaigns have recently been conducted above the Mediterranean basin as part of the ChArMEx program: TRansport and Air QuAlity (TRAQA) in 2012, Aerosol Direct Radiative Impact on the regional climate in the MEDiterranean region (ADRIMED) and Secondary Aerosol Formation in the MEDiterranean (SAFMED) in 2013, and SAFMED+ and Gradient in Longitude of Atmospheric constituents in the Mediterranean basin (GLAM) in 2014. They addressed different processes that impact pollutants, GHGs and aerosols: air quality, radiative impact of aerosols, secondary aerosol formation, and long range transport. Combined with spaceborne and modeling studies, airborne *in situ* measurements highlighted the strong pollutant, GHG, and aerosol gradients between the western and eastern Mediterranean basin in summer from the mid-to-upper troposphere. Maxima in ozone, carbon monoxide, methane, nitrous oxide are commonly observed in the eastern basin, although on some occasions, minima in carbon dioxide are detected. The gradients were mainly attributed to the impact of long range transport of air masses originating either from Asia, North America, Europe, or Africa (Fig. 5). In the case of Asia, the Asian monsoon and its associated anticyclone are the main cause of the gradient by (1) trapping lower tropospheric pollutants and GHGs in the Asian monsoon; (2) updrafting pollutants and GHGs in the Asian monsoon up to the upper troposphere; (3) building up pollutants and GHGs within the Asian monsoon in the upper troposphere; (4) re-distribution of the pollutants and GHGs at a large scale by the Asian monsoon anticyclone to the Middle East and North Africa in the upper troposphere; and (5) subsiding pollutants and GHGs into the middle troposphere above the eastern basin (Ricaud et al. 2014). In the case of North America, biomass burning by long-lasting forest fires is the main source of aerosols, pollutants and GHGs. They are updrafted into the mid-to-upper troposphere by pyro-convection and/or through meteorological systems generally located in the Atlantic Ocean (warm conveyor pool, strong depression, etc.) following the jet stream towards the western Mediterranean basin (Ancellet et al. 2016; Ricaud et al. submitted). In the case of Africa, outbursts of Saharan desert dust are advected westward towards the Caribbean Sea, in the lower part of the troposphere, and, once in the central North Atlantic Ocean, are trapped within meteorological systems to be finally transported upward to the western Mediterranean basin like in the case of transport from North America.

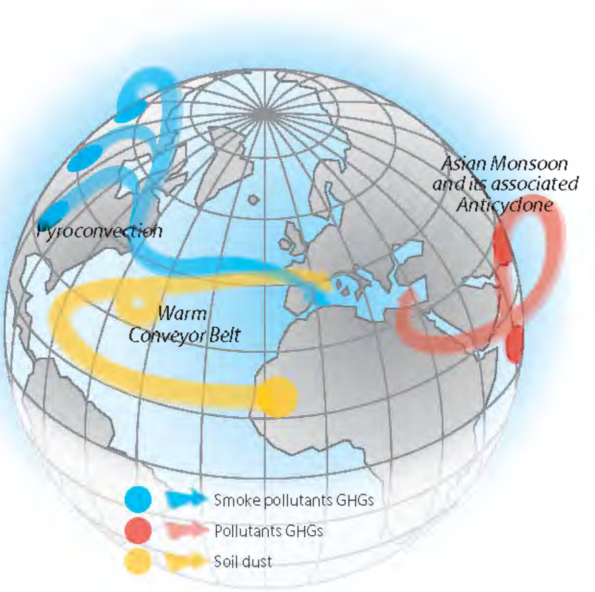

Figure 5
Very long-range intercontinental transport of air masses over the Mediterranean basin. Dots identify source regions and arrows identify transport pathways.

In addition to intercontinental transport of pollutants, GHGs and aerosols, the Mediterranean basin is a region that favors stratosphere-to-troposphere transport (Zbinden et al. in prep.). Stratospheric intrusions result in high ozone and low carbon monoxide or methane penetration into the troposphere. The depth, the irreversibility and the frequency of the stratospheric penetrations modify the tropospheric climatology and trends of these chemical species (Tyrlis et al. 2014).

Although measured and modeled data are still being analyzed in the ChArMEx program, their comparison underlines the difficulties models face representing 1) the significant fine structures observed both horizontally and vertically, 2) some processes that might be missing in the models (e.g. pyro-convection), 3) the mixing ratios of pollutants and GHGs with sufficient accuracy to estimate trends. Finally, chemical analyses that consist in assimilating satellite data with model data, widely used during the ChArMEx airborne campaigns, are efficient tools that benefit studies of the distribution and evolution of pollutants, GHGs, and aerosols over time.

The ChArMEx program allowed us to investigate the variabilities of pollutants, GHGs and aerosols in the Mediterranean basin in space and over time with particular emphasis on the summer period combining *in situ* airborne, spaceborne and model data, and to attribute these variabilities to several processes. Pyroconvection, convection, monsoon and the Asian monsoon anticyclone, the jet stream, etc., are dynamic processes that redistribute pollutants, GHGs and aerosols originating from desert dusts, biomass burning, etc., from Asia, Europe, Africa and North America over the Mediterranean basin. Some potentially key geographical areas that impact the eastern basin were revealed, e.g. the Arabian Sea, where future airborne campaigns may be deployed.

Box 1
The secondary organic aerosol (SOA)

The secondary organic aerosol (SOA) is made up of thousands of organic compounds originating from a wide range of natural and human sources (combustion of fossil fuel and biomass, gaseous emissions from the continental biosphere, marine emissions, etc.). SOA accounts for from 20% to 60% of very fine airborne particulate matter PM_1 (particles whose aerodynamic diameter is < 1 µm) (ZHANG et al. 2007). This aerosol fraction is the most relevant for health issues (POPE AND DOCKERY, 2006) but also climate effects. Organic aerosol particles scatter solar radiation, which cools the Earth's atmosphere, since part of the radiation is reflected back to space, but more recently their absorbing properties have also been put forward (ZHANG X. et al. 2011). This affects the radiative properties of aerosol observed over the Mediterranean basin (Di Biagio et al. 2016). Depending on its hygroscopicity and mixing state, organic aerosols affect cloud micro-physics (LOHMANN AND FEICHTER, 2005; ZHU et al. 2016). SOA forms from semi-volatile organic compounds (SVOCs) by nucleation or condensation. Oxidative processes are most efficient in lowering the volatility of initially volatile compounds of anthropogenic and biogenic origin (KROLL et al. 2008). Anthropogenic emissions play a role in the formation of oxidants and hence in the formation of SOA, so that SOA of biogenic origin may be strongly reduced by reducing anthropogenic emissions, especially above large cities around the Mediterranean Sea (SARTELET et al. 2012). Research to elucidate the formation of SOA is still very active. Once formed, SOA can become highly viscous, which prevents later evaporation of the organic material (VIRTANEN et al. 2010).

During ChArMEx, and for the first time in the western Mediterranean, intensive and long-term aerosol mass spectrometer measurements filled the gap of missing observations of SOA over this region. On a yearly average, organic aerosol made up about 50% of PM_1 at Cape Corsica (POM in Fig.B1, left), among which around 90% were of secondary origin (LV-OOA and SV-OOA in Fig.B1, right). Surprisingly, aged (highly oxidized) SOA (LV-OOA) concentrations were found throughout the year, dominating OA even in winter when photochemical conditions are low. This result provides further evidence for the highly oxidative atmosphere of the Mediterranean. Additional isotopic ^{14}C measurements during the intensive campaign in the summer of 2013 showed that the majority of the organic aerosol was of biogenic origin (from the continental biosphere), although still 20% at Cape Corsica and 35% at Mallorca were of anthropogenic origin (fossil fuel combustion).

SOA modelling is still very challenging as the many chemical reaction pathways are not explicitly known and need to be parameterized in 3D models. Including a volatility basis-set (VBS) scheme including multi-step functionalization and fragmentation of organic compounds and formation of non-volatile SOA in the CHIMERE regional chemistry-transport model (Shrivastava et al. 2015) made it possible to retrieve the SOA mass and its fossil vs. biogenic fractions observed at surface stations at Cape Corsica and Mallorca (Cholakian et al. 2016), and recently in the Paris area (Zhang et al. 2015). Simulations with the Polyphemus model including the new multiphase SOAP organic aerosol scheme were in good agreement with measurements of concentrations of organic aerosol at Cape Corsica. The formation of extremely low-volatility organic aerosols has been added to the model to better represent organic aerosol properties (Chrit et al., 2016). Well evaluated chemistry-transport models is a prerequisite for simulating the regional scale interaction of SOA with climate (radiation, micro-physics) over the Mediterranean region as the next step. Ongoing developments will be constrained and validated by the large number of field observations made recently in the western Mediterranean as well as new measurements in the eastern basin as part of the ChArMEx program.

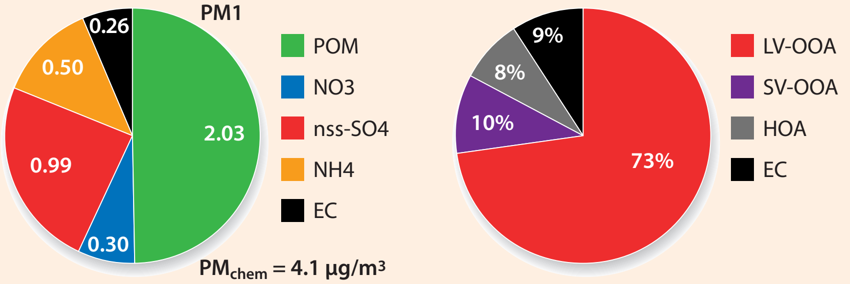

Figure B1

PM_1 composition in µg m^{-3} (left) and source apportionment (right) of carbonaceous aerosols at Ersa, Cape Corsica (42°58'N, 9°23'E, alt. 530 m), averaged over two years (from June 2012 to July 2014) from off-line filters and on-line aerosol mass spectrometer (Q-ACSM) measurements, respectively (after Nicolas, 2014). POM stands for particulate organic matter, NO3 for nitrate, nss-SO4 for non-sea salt sulfate, NH4 for ammonium end EC for elemental carbon. LV-OOA (low-volatility oxygen-like organic aerosol) is assimilated to aged secondary organic aerosol, SV-OOA (semi-volatile oxygen-like organic aerosol) to freshly formed secondary organic aerosol, HOA (hydrogen-like organic aerosol) to primary organic aerosol.

Sub-chapter 1.4.3

Atmospheric deposition to nutrient depleted seawater

Atmospheric deposition is the removal from the atmosphere of particles and gas by sedimentation (dry deposition) and by rainfall (wet deposition). Atmospheric inputs of pollutants and nutrients are of the same order of magnitude or larger than the riverine inputs for the Mediterranean region (Koçak et al. 2010; Christodoulaki et al. 2013; Moon et al. 2016), and hence of primary importance for marine ecosystems in the particularly oligotrophic seawater (i.e. with very low nitrogen and phosphorus nutrient concentrations) of the Mediterranean basin (Dugdale and Wilkerson, 1988). Indeed, owing to the small size (2.51 10^6 km²) of the Mediterranean Sea and to many intense land-based sources of emissions, open waters receive significant loads of nutrients through atmospheric deposition (Fig. 6). Some deposition events qualified as 'extreme events', such as dust inputs as high as 22 g m^{-2} (Bonnet and Guieu, 2006), can occur at very short time scales (hours to days).

Atmospheric deposition is a significant source of major nutrients including inorganic (Kouvarakis et al. 2001) and organic nitrogen (Violaki et al. 2010), and phosphorous (Markaki et al. 2003). Even if water convection generally supplies most of the nitrogen (N) and phosphorus (P) available for biology in surface waters (where photosynthesis occurs) from deep waters, atmospheric inputs of inorganic N and P may be the most intense source of nutrients in summer when thermal stratification of the surface water column prevents vertical mixing (Pasqueron de Fommervault et al. 2015). Atmospheric deposition could also partly explain the increasing N:P ratio in the seawater column from the western

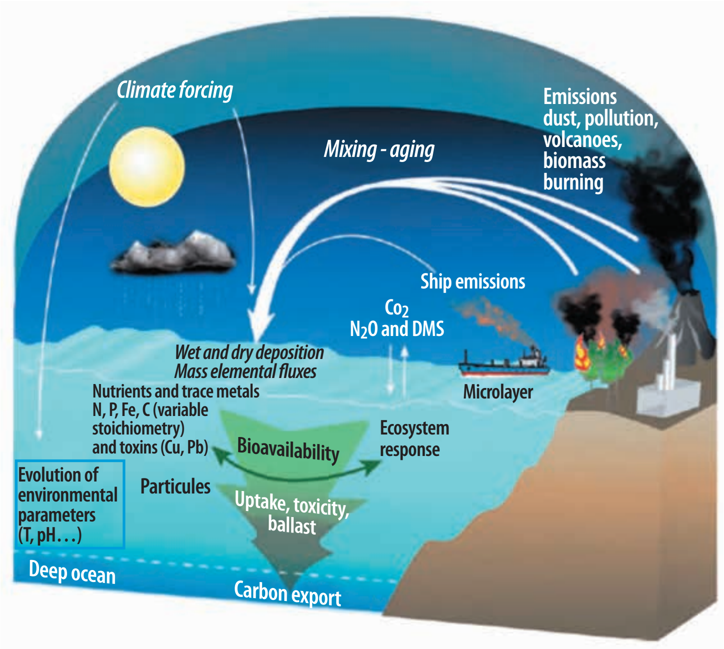

Figure 6
Main processes occurring at the air-sea interface driven by different atmospheric inputs (adapted from Law et al. 2013).

to the eastern Mediterranean basin, since a similar N:P trend is observed in atmospheric deposition in these areas (Markaki et al. 2010). It has been shown that atmospheric deposition of iron (Bonnet and Guieu, 2006) and trace metals (Theodosi et al. 2010) represents significant inputs to support the primary production in surface waters. Deposition also transfers different atmospheric organic contaminants from the lower atmosphere to the ocean surface, including organochlorine compounds or polycyclic aromatic hydrocarbons (PAH), but their low deposition fluxes and their degradation in the water column limit their impact on marine ecosystems (Castro-Jimenez et al. 2012; Berrojalbiz et al. 2014).

Deposited nutrients come from two main sources: anthropogenic sources and soil dust, the latter mainly from the Sahara but also from the Middle East in the easternmost Mediterranean basin. Anthropogenic inputs control the deposition flux of N, inorganic nitrogen being mainly supplied by dry deposition (Markaki et al. 2010), and organic nitrogen by wet deposition (Violaki et al. 2010). The deposition of desert dust plays an important role in the fluxes of P and trace metals due to sporadic but intense deposition events (Özsoy and Örnektekin, 2009; Guieu et al. 2010; Morales-Baquero and Perez-Martinez, 2016), even if the contribution of anthropogenic aerosol deposition is significant (between 10% for Fe to 90% for Zn; Guieu et al. 2010). Dust deposition releases dissolved inorganic phosphorous (DIP) and nitrate in N- and P-depleted surface waters (Ridame et al. 2014). The atmospheric deposition of mineral dust also determines enrichment of the sea-surface microlayer in dissolved trace metal micro-nutrients such as Cd, Co, Cu, Fe (Tovar Sanchez et al. 2014). However, it has been shown that dust deposition can result either in a net release or in scavenging of DIP and nitrate (Louis et al. 2015) and trace elements (Wagener et al. 2010; Wuttig et al. 2013) in seawater, depending on the quantity and quality of *in situ* dissolved organic matter at the time of the deposition. Indeed, the dissolved organic matter can control the dissolution of nutrients carried by dust particles.

Recent experiments in realistic conditions showed that, by providing P and N to the marine biosphere, wet Saharan dust deposition strongly stimulates primary production and phytoplankton biomass for several days after deposition (Ridame et al. 2014; Guieu et al. 2014b; Fig. 7). From such studies, the inputs of atmospheric trace metals into the Mediterranean Sea associated with dust deposition are also suspected of stimulating bacteria and phytoplankton species such as cyanobacteria that are able to assimilate atmospheric dinitrogen N_2 (Ridame et al. 2011). The extent of the fertilizing effect of dust deposition events in the Mediterranean was revealed by statistically positive correlations between dust deposition and surface chlorophyll concentrations in combined remote sensing and modeling approaches (Gallisai et al. 2014). However, a negative effect of atmospheric deposition on chlorophyll was observed in the regions under the influence of aerosols of European origin (Gallisai et al. 2014). Indeed, inputs of anthropogenic aerosols, such as Cu-rich aerosol, are suspected of inhibiting phytoplankton growth (Jordi et al. 2012). Dust deposition has also been shown to modify the structure of the bacterial community by selectively stimulating and inhibiting certain types (Pulido-Villena et al. 2014). By stimulating predominantly heterotrophic bacteria (i.e. that use organic carbon for their growth), atmospheric dust deposition can increase the recycling of carbon, thereby reducing net atmospheric CO_2 drawdown and the fraction of dissolved organic carbon that can be mixed and exported to deep waters (Pulido-Villena et al. 2008). In contrast, Saharan dust deposition in the Mediterranean can enhance the export of particulate organic carbon to the deep ocean by acting as ballast and facilitating aggregation processes (i.e. Bressac et al. 2014; Desboeufs et al. 2014).

To tackle these questions at the scale of the Mediterranean basin, numerical models need to be developed. The main challenges are to quantify the relative contributions of anthropogenic and natural deposition of nutrients and contaminants in this region in a context of Mediterranean climate change, and to improve our ability to simulate the chemical elements and their soluble fraction in 3-D atmospheric transport and chemistry models. Modeling the size distribution of desert dust particles and its evolution from emission to deposition contributes to the difficulty. *In situ* measurements by aircraft suggest a coarse mode of large soil dust particles over the Mediterranean (with a volume mean effective diameter in the range of 3.8-14.2 µm) as large as that observed close to the Saharan and Sahelian source regions (Denjean et al. 2016). Balloon-borne aerosol counters have also shown the frequent presence of large particles (> 20 µm) inside airborne desert dust plumes over the western Mediterranean (Renard et al. 2016). These particles, which appear to be transported for several days without significant gravitational sedimentation, in contradiction to their size, probably control the mass flux of deposition of dust.

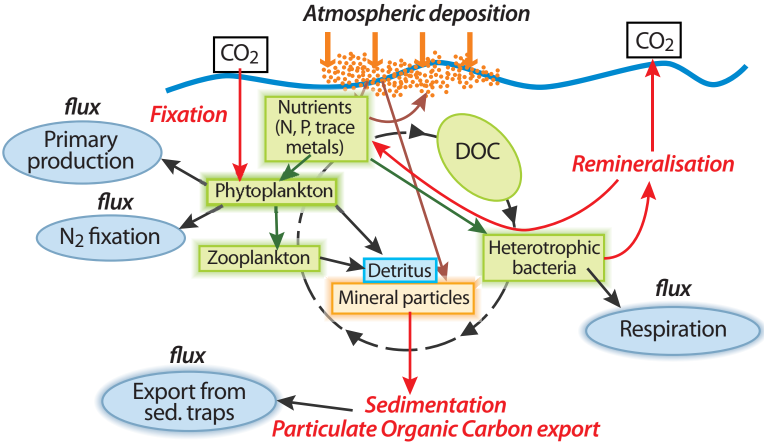

Figure 7
Stocks (green) and fluxes (blue) measured during mesocosm experiments after the simulation of a realistic atmospheric input in a water body large enough to be representative of natural processes (Guieu et al. 2014b; modified from de Leeuw et al. 2014).

Predicting inputs of atmospheric nutrients in the future is important to understand the vulnerability of the marine ecosystem and carbon fluxes. Climate change simulations predict a hotter dryer climate in the Mediterranean basin (e.g., Hertig & Jacobeit, 2008), a situation that could increase dust emissions and transport due to increased aridity (Moulin & Chiapello, 2006) but also intense deposition events (Beaulant et al. 2011). However, the most recent measurements of dust deposition in Corsica (2011-2013) show that deposition fluxes are much lower than in the three past decades shown by existing records in the area (11-14 g m^{-2} yr^{-1}) (Vincent et al. 2016). But it is not yet understood if the reduction in dust deposition is related to a decrease in the frequency of dust events and/or to a change in deposition processes and patterns in the Mediterranean region. The temporal dynamics of marine N and P concentrations since 1985 showed high sensitivity to anthropogenic atmospheric deposition and are expected to decline in the coming decades due to mitigation/control of pollutant emissions (Moon et al. 2016). In the same way, Christodoulaki et al. (2016) showed that even if the human-driven atmospheric deposition of N and P has led to a 16% increase in total phytoplankton biomass over the past one and a half centuries, small changes in carbon fluxes and planktonic biomasses are predicted for the near future with the projected inputs of N and P. The regulation of inputs of anthropogenic nutrients could be a key driver of seawater nutrient cycles and hence marine ecosystems in the future. However, findings concerning the sources of the atmospheric trace metals and effects need to be completed to enable high quality projections in a context of future changes.

Sub-chapter 1.4.4

Impact of atmospheric chemistry on the regional climate

In this section, we address the complex atmospheric-chemistry and climate interactions and feedback processes in the Mediterranean region.

Processes

Projecting climate changes at regional scale remains a challenge. This is especially true in regions where aerosols play a significant role in the radiative balance with effects on the climate and water cycle (Nabat et al. 2014 and 2015) like in the Mediterranean region. Regional projections of this type are required for the design of adaptation and mitigation strategies and a deeper understanding of regional processes is thus needed.

Atmospheric aerosol particles play a major role in the water cycle and also in the Earth's energy balance through their radiative effects, including direct effects (by extinction of radiation and attenuation of surface illumination), semi-direct effects (due to the heating of turbid air layers by particles absorbing solar radiation), and indirect effects (through their influence on cloud properties). The uncertainty of these climate effects exceeds that of any other forcing because the physical, chemical and optical properties of aerosols are highly variable in space and time since their atmospheric lifetime is short and their emissions are very heterogeneous (Forster et al. 2007).

The Mediterranean region is at the intersection of large scale circulation patterns with highly contrasted sources of particles. The region is subject to high particle loading as described in the sub-chapter 'High concentrations of aerosols and pollutant and greenhouse gases', high photochemical activity, with mixing of different origins: particles come from anthropogenic sources such as highly densified cities in Europe, Turkey, and the Middle East; desert dust from the Sahara and from the Middle East in the eastern part of the basin (Mamouri & Ansmann, 2015); maritime particles from the Mediterranean Sea; as well as organic particles from biomass burning (Lelieveld et al. 2002; Sciare et al. 2003, 2008). Expected impacts of climate change in the region include heat stress associated with poor air quality in the urban environment (Lelieveld et al. 2012, 2015). This is why the impact of particles in the Mediterranean region is high and is expected to change, and why each effect of different particles on the climate of the Mediterranean region, whether direct or indirect, needs to be understood and quantified.

Assessment of aerosol direct radiative forcing (DRF)

Aerosols observed over the Mediterranean basin are known to be able to interact with both shortwave (SW) and longwave (LW) radiation (Nabat et al. 2012; Papadimas et al. 2012; Zanis et al. 2012; Sicard et al. 2014). In the SW spectral range, due to their variability in size and in chemical properties, they can scatter (sea salt, sulfates, nitrates, ammonium and secondary organic particles) or scatter plus absorb (black and brown carbon from combustion [smoke], certain types of mineral dust) solar radiation. Only mineral dust and sea salt can interact with LW radiation due to their large particle size. Consequently, Mediterranean aerosols significantly affect the radiative budget of the Mediterranean by (1) decreasing surface incoming shortwave radiation, (2) increasing/decreasing outgoing shortwave fluxes depending on the surface albedo, and (3) possibly by heating turbid atmospheric layers when the particles absorb solar light.

Concerning pollution aerosols, SW direct shortwave radiative forcing (DRF) has been estimated at the local scale by many authors (Horvath et al. 2002; Markowicz et al. 2002; Meloni et al. 2003; Roger et al. 2006; di Sarra et al. 2008; Di Biagio et al. 2009, 2010). These authors showed a significant decrease in surface solar fluxes of 20–30 W m^{-2} (daily mean) at different locations including Almeria (Spain), Finokalia (Greece), Lampedusa (Italy), Marseilles and Toulon (France). In parallel, the combination of surface and satellite remote-sensing observations performed at Lampedusa has been used to calculate the radiative effects, in both the shortwave (Di Biagio et al. 2010) and longwave (di Sarra et al. 2011; Meloni et al. 2015) spectral ranges for different cases of Saharan dust intrusions. These studies emphasized that desert dust in the LW spectral range has a significant radiative effect, and offsets a large fraction of SW forcing (di Sarra et al. 2011; Meloni et al. 2015). More recently, based on remote-sensing observations in Barcelona and 1-D radiative transfer calculations, Sicard et al. (2014) also estimated the LW radiative effect of dust. Only a few studies are available concerning the radiative impact over the Mediterranean region of intense biomass burning events. One estimate was proposed by Formenti et al. (2002) for an aged Canadian biomass-burning plume and revealed a significant SW surface dimming of 60 W m^{-2}. In addition, the radiative effect induced by smoke aerosols at Lampedusa between August 3 and 23, 2003, during an exceptionally hot and dry season when the Mediterranean atmosphere was affected by massive forest fires, was estimated by Pace et al. (2005) to be between +22 and +26 W m^{-2}. For a complete review of local direct radiative forcing see Mallet et al. (2016).

At the regional scale, Papadimas et al. (2012) proposed an estimation of the aerosol DRF using MODIS data from 2000 to 2007 for both all-sky and clear-sky only conditions. These authors derived a multi-year regional mean surface DRF of -19 W m^{-2}, associated with a DRF of -4.5 W m^{-2} at the top of the atmosphere (TOA). It should be noted that such radiative forcing is regionally higher than that exerted by greenhouse gases. Regional modelling studies were also recently conducted by Nabat et al. (2012, 2015) using the coupled-chemistry RegCM and the CNRM-Regional Climate System Model (RCSM) models for multi-year simulations. These authors reported a mean SW regional surface (TOA) DRF of -13.6 W m^{-2} (-5.5 W m^{-2}) and -20.9 W m^{-2} (-10.5 W m^{-2}) for the RegCM and CNRM-RCSM (see Fig. 8) models, respectively. Zanis et al. (2012) also proposed a regional estimate of the DRF of anthropogenic particles for the 1996-2007 period using RegCM and showed a significant negative forcing of up to -23 W m^{-2} at TOA over Eastern Europe.

In parallel with the radiative forcing exerted by aerosols, Richards et al. (2013) investigated the effect of reducing certain sources of emissions on the radiative effect of ozone over the Mediterranean (ozone shows a marked but localized summertime maximum, especially over the eastern basin). These authors reported a mean radiative effect at the top of the atmosphere of between -1 and -40 W m^{-2}, depending on the types of emission tested. Hauglustaine & Brasseur (2001) reported mean radiative forcing associated with an increase in tropospheric ozone since the preindustrial era of around 0.70 W m^{-2}. Such studies demonstrate that the radiative effect of ozone is significantly lower than that exerted by natural/anthropogenic aerosols over the Mediterranean region.

The aerosol atmospheric load can also directly impact the production of secondary gaseous species (i.e. chemically produced from primary emitted species) by attenuating or scattering visible and UV radiation. Recently, MAILLER et al. (2016) showed a net reduction in the photolysis rates of ozone and nitrogen oxides due to the absorbing effect of a mineral dust plume observed in Lampedusa. This effect led to a change of several ppbs in ozone surface concentrations. Over the Mediterranean Sea and continental Europe, close to the sources of NO_x, the effect of dust leads to reduced ozone concentrations (but an increase occurs over remote areas such as the Sahara and the tropical Atlantic Ocean). By reducing photosynthetically active radiation (PAR), the aerosol may also reduce crop production, and hence biogenic emissions, leading to complex interactions in the formation of secondary organic aerosols that are not yet well understood.

Implication of aerosol direct radiative forcing (DRF) in the Mediterranean water cycle

Concerning surface and TOA atmospheric forcings, ZANIS et al. 2012; SPYROU et al. 2013; NABAT et al. 2014, 2015a,b recently investigated how changes in the radiative budget due to natural/anthropogenic aerosols influence the surface temperature (over both land and sea), relative humidity profiles, exchanges (latent heat fluxes) between ocean and atmosphere, cloud cover (semi-direct effect of absorbing particles), precipitation, and finally the whole Mediterranean hydrological cycle. Indeed, notable perturbations in sea surface-atmosphere fluxes are expected despite the relatively small size of the Mediterranean Sea, since the latter plays an important role at a much larger scale by providing moisture for precipitation to its surrounding land areas, which extend to northern Europe and northern Africa (GIMENO et al. 2010 and SCHICKER et al. 2010). In parallel, the absorbing particles over the Mediterranean Sea (MALLET et al. 2013) could have a semi-direct effect that could modify the vertical profiles of relative humidity and cloud cover.

In that context, using simulation ensembles of the direct radiative effect of aerosols (Fig. 8a) carried out with the regional climate system model CNRM-RCSM, NABAT et al. (2015) showed annual average cooling of the Mediterranean sea surface caused by aerosols of -0.5 °C, up to -1 °C locally, and higher in spring and summer when aerosol loads (mainly sulfate and dust particles) are maximal (see summer average in Fig. 8b). This cooling also affects the surrounding land, not only due to negative aerosol DRF over land, but also because of the advection of maritime cooler air over land regions. For example, in summer, northern winds over the eastern basin favor additional cooling over northern Libya and Egypt.

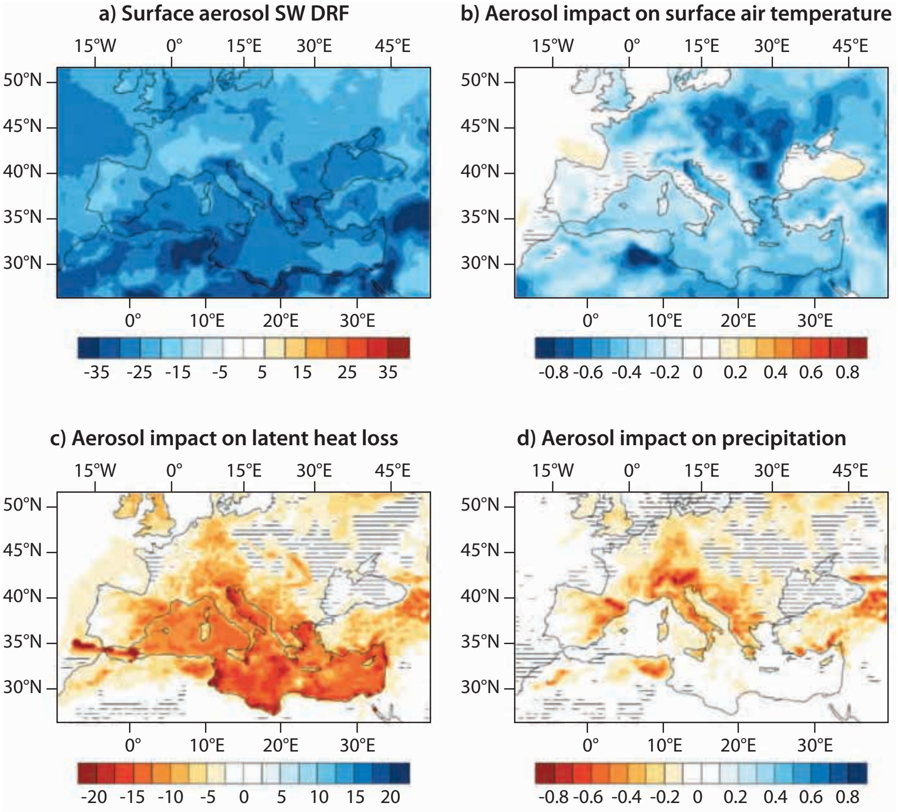

Figure 8

Average (JJA 2003-2009) surface aerosol direct radiative forcing (DRF, W m^{-2}) (a) and the resulting impact of the aerosol on surface air temperature (°C) (b), latent heat loss (W m^{-2}) (c) and precipitation (mm day^{-1}) (d), simulated by the CNRM-RCSM regional model. Hatched areas are not significant at the 0.05 probability level.

In addition, aerosols are responsible for a decrease in the latent heat loss by evaporation over the Mediterranean Sea (Fig. 8c) due to these lower sea surface temperatures (SST). Consequently a reduction in the whole hydrological cycle has been attributed to aerosols, resulting in an average 10% decrease in specific humidity in the lower troposphere, in cloud cover and in precipitation (Fig. 8d). In addition, dust aerosols also reduce atmospheric convection in the region by warming the lower troposphere by absorbing solar radiation, and hence stabilizing the atmosphere. The comparison of these results with the model response in atmosphere-only simulations shows that feedback is reduced if SST cannot be modified by aerosols, highlighting the essential role of the Mediterranean SST and the need to use atmosphere-ocean coupled regional models in regional aerosol-climate studies. In addition, the decrease in SST causes an increase in the density of surface Mediterranean waters that favors ocean convection in sub-basins where deep water masses are formed (Gulf of Lion, Adriatic Sea and Aegean Sea), and reinforces the Mediterranean thermohaline circulation.

It is also worth mentioning that since the 1980s, aerosol loads have been dramatically reduced over the Mediterranean basin (Nabat et al., 2013), because of the reduction in anthropogenic emissions of sulfate precursors in Europe (improved air quality standards plus economic crises). As a consequence, aerosol DRF decreased over the region between 1980 and 2012, leading to an increase in surface solar radiation (brightening period) and additional warming of land and ocean surface temperature: aerosol changes explain 23% of the warming in the region over this period (Nabat et al., 2014).

Aerosol-cloud interactions and impact on precipitation

The Mediterranean region is particularly vulnerable to climate change due to its location at the interface of the hot dry North African climate and the cooler wetter European climate. The water cycle in this region is very sensitive to the position of the descending branch of the Hadley cell, which is expected to move poleward following global warming, leading to increasingly scarce fresh water (Giorgi & Lionello, 2008). Understanding the formation of clouds and precipitation and its link with atmospheric composition is particularly important in this region. Clouds and precipitation strongly influence tropospheric composition and radiative properties, water transport and energy redistribution. At the cloud scale, clouds and the precipitation life cycle are controlled by the prevailing meteorology and aerosol particles, particularly the presence of cloud condensation and ice nuclei (CCN/IN) (Flossmann Wobrock, 2010). Indeed, hydrometeors cannot form spontaneously in the thermodynamic conditions encountered in the atmosphere. They need a substrate (a nucleus) during the early stage of formation. An aerosol particle that serves as a nucleus for a cloud droplet is called a cloud condensation nucleus (CCN), and for an ice crystal, an ice nucleus (IN).

An additional consequence of the enlargement of the Hadley cell (ICCP, 2014b) is the increasing frequency of dust particles transported over the eastern Mediterranean. Rosenfeld et al. (2001) reported that an excess concentration of atmospheric dust can prevent cloud precipitation. This argument is often referred to as the second indirect effect in the Twomey classification (1980): an excess of CCN causes an increase in the number of droplets and consequently the cloud droplet size distribution shifts to smaller sizes, thereby narrowing the droplet size distribution. This hampers precipitation. Indeed, precipitation is triggered by a mechanism that forms hydrometeors of millimetric sizes in a matter of minutes. Only the presence of a few large hydrometeors in the population that will fall and collect the population of small hydrometeors can lead to precipitation. The narrowing of the size distribution eliminates the largest hydrometeors and reduces precipitation ability. This will inherently amplify dryness due to extension of the Hadley cell, and is referred as the "desertification positive feedback loop".

Our understanding of the processes that take place inside clouds has considerably increased in recent decades (Flossmann and Wobrock, 2010, Lohmann et al. 2010, Levin and Cotton, 2008). Microphysical features of clouds in the Mediterranean region have already been characterized together with their link with the loading of aerosol particles (Levin et al. 1996, 2005; Rosenfeld et al. 2001; Teller et al. 2012). The impact of aerosols is uncertain but potentially maximal when aerosol particles can cause the formation of ice crystals in clouds. This is referred to as the ice indirect effect by Lohmann (2002). Indeed, global precipitation is predominantly produced by clouds containing ice crystals (DeMott et al. 2011). Aerosols that can act as IN are mostly insoluble particles that often mimic ice lattice structure. Only a few types of aerosols have this property, including mineral dust, volcanic ash, and bioaerosols such as bacteria, fungal spores, and pollen (Vali, 1985; Hoose & Möhler, 2012). Their IN ability, their likelihood of precipitating the ice phase, and to what extent anthropogenic aerosols can play a role in cloud ice formation are still not fully understood although the number of laboratory studies on the IN properties of different kinds of aerosol particles has been continuously increasing (e.g., Möhler et al. 2007, for bacteria; Connolly et al. 2009, and Ardon-Dryer & Levin, 2014, for dust).

Even though air quality and water resources in the Mediterranean region have been the center of scientific interest, and have been studied intensively, many questions remain unanswered, in particular in the eastern part i.e., how and to what extent natural and anthropogenic aerosols can increase or prevent rainfall (e.g. Levin et al. 1996, Teller & Levin, 2006).

Ongoing research and recommendations

Recent studies clearly demonstrated the added value of using regional climate models including ocean-atmosphere coupling to study the impact of the aerosol direct radiative effect on the Mediterranean climate. One priority should now be to conduct an intercomparison of such models to check the robustness of individual models, as scheduled in the next phase of the MedCORDEX program (https://www.medcordex.eu/). In addition, the impact of the anthropogenic and natural aerosol DRF on future climate needs to be investigated using different RCP scenarios, with the 8.5 scenario as a priority. The second important point concerns the role of aerosols in cloud microphysical and macro-physical properties at climatic scale. To this end, regional climate simulations should be built including the first (aerosol-cloud albedo) and second (aerosol-cloud precipitation) indirect effects of anthropogenic and natural aerosols on warm clouds. A better representation of dust-IN interactions is also required over this region especially for the eastern Mediterranean. Specific experimental campaigns focused on this aspect associated with improvements in its representation in climate models should be encouraged.

Impacts of air quality on health

There is growing concern about the detrimental effects of air pollution on human health (WHO, 2013). However, data have only been collected in some countries in the Mediterranean region (WHO, 2014). A recent review systemically and qualitatively screened relevant papers and reports published between 2000 and 2014 on health impact of air pollution in the eastern Mediterranean region. The authors found only 36 published studies. A variety of indoor and outdoor exposures associated with various acute and chronic respiratory health outcomes were included. However, data were limited to a few studies in a few eastern Mediterranean countries and concerned both indoor and outdoor air pollution (Abdo et al. 2016). Several adverse respiratory health outcomes were positively associated with various indoor/outdoor air pollutants throughout the region. Respiratory health outcomes ranged in severity, from allergies and general respiratory complaints to lung cancer and mortality. In addition, although Mediterranean countries are highly exposed to dust storms and wildfires, their effects have rarely been studied. In this section, we present recent data on the consequences of exposure to particulate air pollution and related health impacts in the case of both anthropogenic and natural air pollution collected in the Mediterranean region. These include air pollution reduction scenarios.

Impact of air pollution on health in Bejaia (Algeria)

In Algeria, monitoring of air pollution is limited to three big cities (Algiers, Annaba and Oran), and little is known about the impact of air pollution on health in most of the country. *Ad hoc* measurements of ambient concentrations of particulate matter taken in the Bejaia region in July 2015 indicated that the annual average PM_{10} (particulate mass concentration of particles smaller than 10 µm in diameter) and $PM_{2.5}$ (*idem* for particles smaller than 2.5 µm in diameter) levels in this urban zone exceed the World Health Organization (WHO) air quality guideline (AQG) values, the EU AQG and Algerian AQG. As expected, the highest 24-hr average concentrations (PM_{10}= 103.7 ±15.1 µg m^{-3} and $PM_{2.5}$= 35.7 ±9.5 µg m^{-3}) were measured during peak traffic flow hours, pointing to a significant contribution of emissions from vehicles, which are generally old. These assessments of air pollution put forward that an estimated 55 deaths per year could be avoided by reducing the annual PM_{10} levels to the WHO AQG of 20 µg m^{-3}. Furthermore, not exceeding the PM_{10} WHO AQG would reduce respiratory and cardiac hospital admissions by 36 per 100,000 and 23 per 100,000, respectively (Benaissa et al. 2016). The same author previously showed that people who live in areas with high traffic density and high air pollution suffer from higher rates of asthma and COPD morbidity and mortality (Benaissa et al. 2014).

Impact of air pollution on health in Beirut (Lebanon)

Another study was conducted in Beirut in 2012 where the main sources of pollution are vehicles and dust storms as there is no industrial activity in the vicinity. Results (Farah et al. 2104; Farah et al. 2016) showed that the annual average concentrations of PM_{10} and $PM_{2.5}$ exceeded the annual average of WHO AQG (20 and 10 µg m^{-3}, respectively) by 150% and 200%, respectively. The mean $PM_{2.5}$:PM_{10} ratio for the entire study period was 0.61 ±0.12, indicating that in Beirut about 61% of PM_{10} is made up of $PM_{2.5}$, i.e. that particulate air pollution is dominated by fine particles. The highest daily averages of PM_{10} and $PM_{2.5}$ were measured in spring and summer (March to July) (Fig. 9), echoing the higher frequency of dust storms in this part of the Mediterranean at that period of the year. The correlation between particulate matter and nitrogen dioxide (NO_2) indicated that vehicle exhaust emissions contribute an average of 93% of $PM_{2.5}$ and 43% of PM_{10}.

Using data collected daily in 2012, the BAPHE (Beirut Air Pollution and Health Effects) study showed that total respiratory admissions were significantly associated with the same day (lag=0) level of PM_{10} (1.2% increase per 10 µg m^{-3} rise in daily mean pollutant concentration) and $PM_{2.5}$ (1.6 % per 10 µg m^{-3} rise in daily mean pollutant concentration) and that children and the elderly were at higher risk (Mrad Naklé et al. 2015). The results obtained in Beirut are similar to, and consistent with, those obtained in other international studies. Air pollution control is expected to reduce the number of disease admissions in Lebanon.

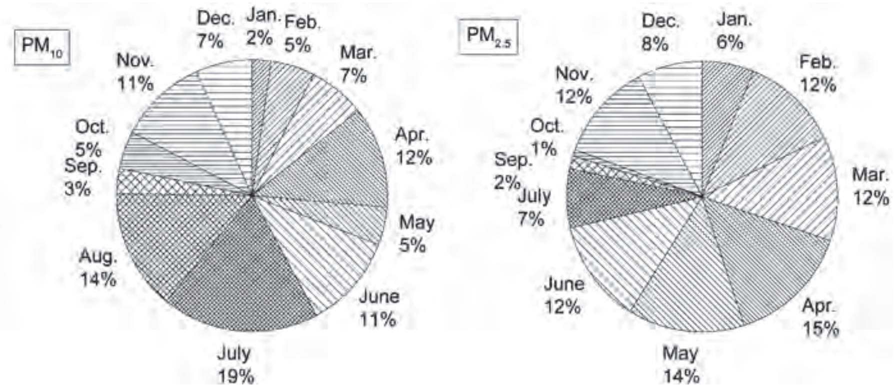

Figure 9
Relative distribution of WHO AQG exceedance days for PM_{10} and $PM_{2.5}$ during the different months of the study in Beirut (from FARAH *et al. 2016).*

Impacts of natural particles in the Mediterranean on health

Since climate change will accelerate desertification processes in arid and semi-arid regions, desert dust outbreaks and wildfires will increase substantially in both frequency and intensity in the near future in various regions of the world including the Mediterranean. The recent MED-PARTICLES ("Particles size and composition in Mediterranean countries: geographical variability and short-term health effects"; http://95.110.213.190/medparticles/) project studied the impact of dust storms and wildfires on human health in 13 cities of Euro-Mediterranean countries, including several on the Mediterranean coasts (Fig. 10): Barcelona and Madrid (Spain), Marseille (France), Bologna, Milan, Modena, Palermo, Parma, Reggio Emilia, Rome, and Turin (Italy), Athens and Thessaloniki (Greece). African dust outbreaks were highly frequent in southern sites during the period 2001-2011, i.e. occurred on 30% to 37% of the days, whereas they occurred on less than 20% of the days at northern sites (STAFOGGIA et al. 2016). The study also identified Saharan dust outbreaks as the largest source of PM_{10} in regional background southern sites of the Mediterranean (35% to 50% of PM_{10}), with seasonal peak contributions to PM_{10} of up to 80% of the total mass. Significant increases by 10-µg m^{-3} in non-desert and desert PM_{10} were associated with a lag of 0-1 day with increases in natural (non-accidental) total and cardio-respiratory mortality and hospital admissions (STAFOGGIA et al. 2016). The occurrence of wildfires assessed by satellite observations was also linked with health by the MED-PARTICLES project. A significant increase in natural and cause-specific mortality was observed on smoky days, with the biggest increase in mortality from cardiovascular diseases (FAUSTINI *et al.* 2015). PM_{10} had more marked effects on cardiovascular and respiratory mortality on smoky days than on other days, suggesting particulate matter is an effective component of fire smoke. This new evidence for adverse health effects of natural sources reinforces the need for control of anthropogenic sources, especially on days when natural dust levels are high, to avoid individuals being subject to excessive exposure resulting from the accumulation of anthropogenic and natural air pollution.

Figure 10
Urban cities involved in the MED-PARTICLES ("Particles size and composition in Mediterranean countries: geographical variability and short-term health effects") project.

Reducing air pollution in the Mediterranean region

Recent data from the Mediterranean region showed that reducing air pollution is beneficial (Benaissa et al. 2016). According to the VIIAS (Integrated Assessment of the Impact of Air Pollution on the Environment and Health) project (www.viias.it), 34,600 and 23,400 are the mean numbers of annual premature deaths in Italy that can be attributed to $PM_{2.5}$ and NO_2, respectively (http://www.viias.it/sites/default/files/ancona.pdf). Applying the 2020 Italian National Energy Strategy (NES) would prevent 17% of the $PM_{2.5}$-, and 57% of the NO_2-attributable deaths. However, compliance with the EU Directive for $PM_{2.5}$ would have an even higher impact with a 22% annual reduction in attributable mortality, with the highest reduction (-30%) in urban areas. For

NO_2, compliance with the EU Directive would result in a 25% annual reduction in attributable mortality, especially in urban areas (-31%). Like for ozone, VIIAS estimated 1,710 annual premature deaths from respiratory diseases due to long-term exposure, and 2,230 annual premature deaths from non-accidental causes due to short-term exposure. Applying the 2020 Italian NES would prevent 23% of the long term and 14% of the short term O_3 attributable deaths, especially in the south (-26% and -20%, respectively) and in rural areas (-27% and -21%, respectively).

Recommendations

Studies on the impact of air quality on health conducted in the Mediterranean region underline the need to improve assessment of exposure and estimations of anthropogenic and natural (especially dust storms and wildfires) related health outcomes in countries where they have been neglected. A better understanding of the role played by meteorology in the direction and the extension of dust events in space and over time is also important. Prevention needs to be promoted, since it has been shown to be effective in reducing effects on health.

Sub-chapter 1.4.6

The (uncertain) future of air quality

Assessing the future evolution of air quality requires taking climate projections into account and designing environmental policies. In the context of adaptation to climate change, the geophysical changes expected in coming decades will have an impact on chronic and extreme air pollution events (Jacob and Winner, 2009). But air quality is also sensitive to climate mitigation strategies: the social and technological changes required to reduce greenhouse gas emissions will be accompanied by changes in the emission of air pollutants and of their precursors. There are potentially large co-benefits between air quality and climate change mitigation that could help efforts to identify win-win strategies. Nevertheless, mitigating climate change can also cause collateral damage to air quality. It is thus very important to identify the co-benefits and possible collateral damage to maximize the former while minimizing the latter. Here we briefly review recent results on the impacts of climate change on air quality in the Mediterranean region in terms of ozone and particles, and describe the positive and negative feedback of climate change on air quality.

Adaptation: the impact of climate change on air quality

Surface ozone

Ozone concentrations in the troposphere are driven by many chemical and dynamic processes including emissions of ozone precursors and meteorological variables. Climate change has an impact on the tropospheric ozone through its effects on biogenic emissions of ozone precursors (mainly volatile organic compounds - VOCs), meteorological parameters (temperature, precipitation, humidity) and atmospheric chemistry (chemical budget, photochemical regimes). Climate change will be accompanied by a reduction in rainfall over southern Europe, creating wintertime deficits that reduce soil water content, thereby further increasing average temperatures and the frequency and severity of heat waves (Fiore et al. 2012; Vautard et al. 2013) with major consequences for summertime ozone pollution in Europe and the Mediterranean, which have already been pointed out (Langner et al. 2005; Meleux et al. 2007). A meta-analysis of the 25 projections of ozone pollution in Europe in the context of climate change published between 2007 and 2015 was conducted by Colette et al. (2015a) to explore the robustness of the projected impact of climate change on surface ozone (Fig. 11). The corresponding climate ozone penalty is defined as the incremental change in ozone that can be attributed to climate change alone, in the absence of changes in anthropogenic emissions of ozone or other drivers. The penalty was confirmed over most of continental Europe, especially in European countries located on the Mediterranean rim where such a penalty is robust, i.e. consistent in over two-thirds of the models in the ensemble (diamond symbols in Fig. 11).

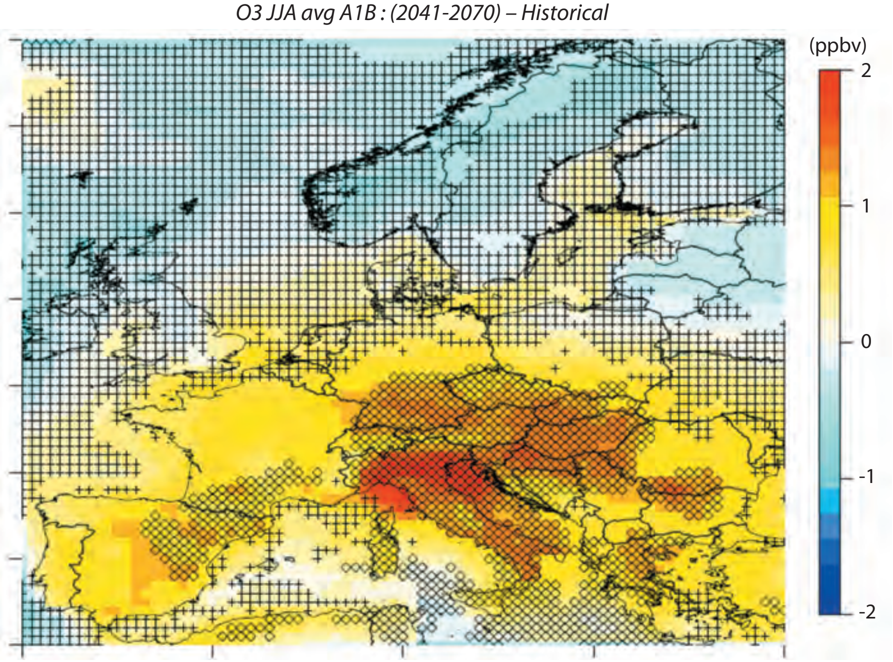

Figure 11
Increase in surface summertime ozone concentrations (ppbv) by the middle of the 21st century in the moderate climate change scenario A1B in an ensemble of all the published European model projections (adapted from Colette et al. 2015a).

The main effect of climate responsible for this increase in surface ozone pollution in Europe is the increase in temperature and solar radiation leading to an increase in biogenic isoprene emissions even if a possible inhibition of these emissions with increasing CO_2 concentrations occurs in the long run, thereby yielding major uncertainties (LATHIÈRE et al. 2010; LANGNER et al. 2012). The other impacts of climate on surface ozone are the direct impact of an increase in temperature on the kinetics of atmospheric chemistry, and the direct impact of solar radiation on photochemistry resulting from changes in cloud cover. Both increase photolysis rates, particularly that of nitrogen dioxide, which favors the formation of ozone. In both cases, the increase in temperature and solar radiation can result from gradual changes in the average climate, but they are exacerbated in the case of extreme heat wave events. In addition to meteorological factors, heat waves favor the accumulation of pollution in the absence of atmospheric dispersion.

In the context of the MISTRALS/ChArMEx project, the global model outputs from the Atmospheric Chemistry and Climate Model Intercomparison Project (ACCMIP; YOUNG et al. 2013) are being analyzed to assess future changes in surface ozone over the Euro-Mediterranean region (JAIDAN et al. in prep.). Under the pessimistic Representative Concentration Pathway (RCP8.5) scenario, mean temperature will increase by about 5.4K by 2100 compared to 2000 accompanied by a small increase (about 2%) in surface ozone.

Over European land surfaces, the 95% confidence interval of summertime mean ozone change is estimated to be [0.44; 0.64] and [0.99; 1.50] ppbv for the 2041–2070 and 2071–2100 periods, respectively. This change may seem small, but it is of the same order of magnitude as the ozone trends reported over Europe in the past two decades despite the implementation of ambitious policies (MONKS et al. 2015; COLETTE et al. 2016). This raises serious doubts about our ability to compensate for the climate change penalty by controlling the emissions of ozone precursors.

Particulate matter

The largest detrimental sanitary impacts of air pollution are currently attributed to atmospheric aerosols from various sources (WHO, 2013). Also called particulate matter (PM), they can originate from anthropogenic or biogenic gaseous precursors (this is the case for example of sulfate, nitrate, ammonium, and secondary organic aerosols), from primary emissions of particulate matter (e.g. elemental carbon (EC), but also heavy metals and persistent organic pollutants (POPs) such as polycyclic aromatic hydrocarbons (PAHs)), or from natural sources (desert dust, sea salt, volcanic ash).

Future changes in PM pollution in the context of adaptation to climate change is less clear than that of ozone because of the complexity of the often competitive processes involved (FUZZI et al. 2015). Recent evidence points to a climate change benefit (with a reduction in PM loads, in particular because of an increase in volatility with increasing temperature; Lecœur and Seigneur, 2013; Colette et al. 2013; Lacressonnière et al. 2016; Lemaire et al. 2016) but increases have been reported in the southern parts of Europe (Manders et al. 2012; Hedegaard et al. 2013). Changes in biogenic precursor emission of secondary aerosols (SOA), which are likely to increase substantially in a warmer climate, could lead to an increase in PM concentrations (Megaritis et al. 2013). Changes in scavenging by precipitation, transport patterns and persistence of anticyclonic conditions leading to PM accumulation could also play a role in shaping future aerosol concentrations (Pausata et al. 2013). The frequency of precipitation is more likely to affect PM scavenging than the intensity of precipitation. Simulating accurate precipitation frequencies is very challenging for climate models, and projections are still subject to large uncertainties. Extreme heat events associated with stagnation of air masses are projected to increase, but the relative contribution of changes in their frequency and duration versus changes in the intensity of heat waves is not yet clear (Clark and Brown, 2013). PM pollution is likely to be more sensitive to the extended duration of the events.

The potential change in PM loads in southern Europe will be largely determined by the mineral dust fraction. Both advection from the Sahara and North African deserts and local mobilization e.g. from agricultural land during dry conditions (Bessagnet et al. 2008) contribute to this fraction. Global and regional climate changes as well as changes in land use may have significant impacts on dust emission and transport. African dust activity has been shown to be correlated with different aspects of climate variability including the El Niño/Southern Oscillation, the North Atlantic Oscillation, the meridional position of the intertropical convergence zone, Sahelian rainfall and surface temperatures over the Sahara Desert, which can affect surface wind activity to varying degrees (Evan et al. 2016). The same authors conclude that the likely tendency for African dust activity is a decrease in a warmer climate. However, changes in PM_{10} exceedances due to dust over Europe are more likely to be sensitive to changes in the frequency and transport pathways of dust storms rather than to variations in mean emissions or in mean concentrations. Currently, there is no consensus on the sign and magnitude of future regional change in dust concentrations affecting Mediterranean regions and southern Europe.

As another source of natural aerosol, sea sprays, can account for a major fraction of PM in the coastal regions of Europe. Beside sea salts, a significant proportion of the submicron fraction of sea sprays is organic and comes from biogenic sources. Studies have revealed no significant trend in the activity of sea sprays in the North Atlantic in recent decades (Korhonen et al. 2011) and this is unlikely to change significantly with climate change (Jacobson and Streets, 2009).

Wildfires are another major source of aerosol and ozone precursors that can severely impact air quality (Hodzic et al., 2007; Miranda et al., 2008) and for which climate and land use change may be determining factors. A dryer climate would tend to increase wildfires but man-driven changes in land use also have a very strong impact, especially in Europe, where the population density is high. Landscape management and fragmentation and fire suppression tend to reduce wildfires (Knorr et al. 2014). For these reasons, an increase in fire frequency with climate change will not necessarily lead to a net increase in PM emissions as these are not only determined by the number of fires but also by their duration, extent and intensity.

Mitigation: towards win-win solutions to limit global warming and improve air quality

The evolution towards a low carbon economy will be accompanied by reductions in the emission of air pollutants. A vast array of mitigation measures will have beneficial impacts on both air pollution and climate mitigation, of which several belong to the category of energy efficiency measures, which represent a very substantial pathway towards win-win solutions (Colette et al. 2015b), even if some strategies that favor climate mitigation may be detrimental to air quality (for instance the use of diesel fuel and the domestic burning of wood with outdated appliances).

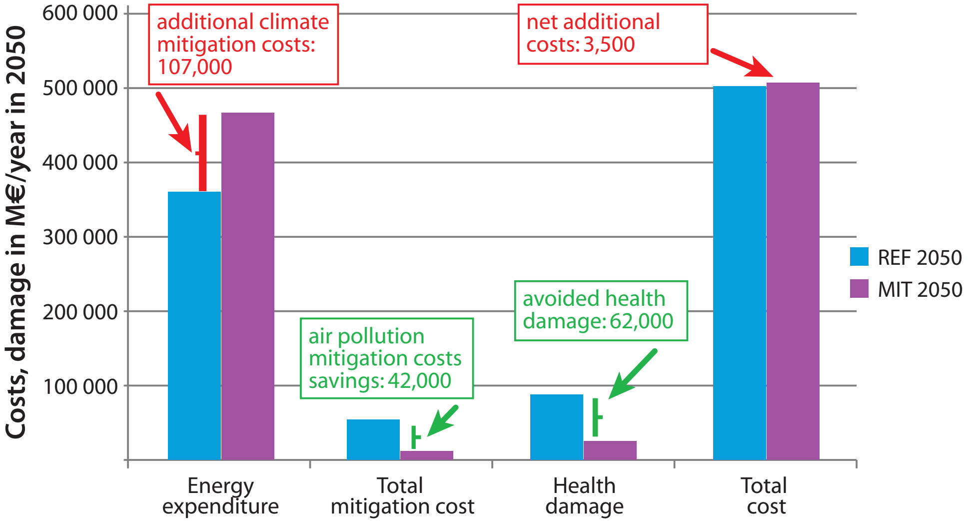

Figure 12
Win-win strategies: cost benefit analyses at European scale demonstrate that the additional costs related to the climate mitigation (MIT) scenario aiming at limiting warming to 2 °C at the end of the 21st century compared to the business-as-usual scenario (REF) could be largely offset by savings in end of pipe air pollution mitigation costs and avoided damage to health. Adapted from Schucht et al. *(2015).*

The sanitary benefits that can be expected from the future evolution of climate and air quality policies were quantified in IIASA (2013) and LIKHVAR et al. (2015), for example. These authors demonstrate that European countries located along the Mediterranean coasts will benefit the most from a reduction in ozone exposure.

A quantitative assessment of the costs and benefits associated with climate mitigation in Europe was proposed by SCHUCHT et al. (2015). These authors found that the very substantial costs of shifting to an energy mix that would comply with the 2 °C warming target would be offset by the positive externality represented by reduced air pollution. This is because the low-carbon scenario also yields (i) reduced cost of end of pipe technologies and (ii) direct sanitary benefits (Fig. 12).

Way forward

Recent evidence demonstrated the link between climate change and air pollution both regarding adaptation and mitigation strategies. It should be emphasized that, at present, most work has been performed at continental scale, through Europe-wide assessments, in addition to a few global studies (ANENBERG et al. 2010; WEST et al. 2013; LELIEVELD et al. 2015). There have been few dedicated assessments of such impacts on the Mediterranean region, thereby opening new research perspectives, in which the proposed contribution of the MISTRALS/ ChArMEx Program could be instrumental.

Overall, major uncertainties remain on the likely evolution of aerosols, especially over southern Europe and the Mediterranean basin. Beside process studies, ensembles of high resolution modeling approaches combining climate and aerosols, and including land use change/management scenarios are one possible way to characterize key mechanisms and to quantify and reduce these uncertainties.

As far as climate adaptation is concerned, the role of climate change in land use and, in turn, in dust resuspension and dispersion remain a key uncertainty. The role of biogenic emissions, as ozone precursors, but also of secondary organic aerosols, is also an important topic.

There are win-win strategies to be developed in the years to come to improve air quality and to engage in progress towards a low carbon economy. Such benefits have been pointed out in several European studies, but the specific situation of Mediterranean countries deserves a closer look to tailor the most efficient sustainable strategies.

Sub-chapter 1.4.7

Conclusion and recommendations

Ambient air is an important common resource. Its quality affects human and ecosystems health, and its composition impacts the regional climate. Climatological surveys show that atmospheric pollution in the form of both gaseous and particulate compounds is generally higher over the Mediterranean basin than over most European continental regions, especially during the long dry season, due to (i) the confluence of long range transported continental air masses that add to local sources of air pollution (e.g. heavy ship traffic), (ii) the scarce precipitation scavenging, (iii) intense photochemistry, and (iv) local circulations and poor ventilation rates that recycle polluted air layers in the western basin, or accumulate pollution in the eastern basin. Future levels of atmospheric trace compounds will be significantly impacted by the changes in climate conditions expected in the Mediterranean region, especially the significant increase in temperature and decrease in precipitation frequency. This will have, in turn, impacts on the Mediterranean climate and human health.

Atmospheric composition and air quality depend on natural and anthropogenic mechanisms, some of which are directly affected by climate change. This is the case of emissions of volatile organic compounds (VOCs) by vegetation, which depend on the temperature, photosynthetically active radiation and availability of water. The most recent observations made in the framework of ChArMEx show that climate change will likely lead to an increase in these emissions, which play a crucial role in the chemical formation of ozone and fine organic particles. But the impact of climate change on emissions is not always straightforward. This is the case of forest fire emissions that also play a role in ozone and particulate pollution. One could expect that a dryer climate would increase fire frequency and consequently particulate air pollution. Nevertheless, emissions by fires depend not only on their frequency, but also on their duration, intensity and extent. There is also no consensus on the impact of climate change on dust emissions. Although climate change will affect dust emissions, which depend on precipitation, land cover and surface wind, it is still uncertain whether these emissions will increase or decrease in the future, especially because dust emissions also depend on agriculture pressure and field preparation techniques. Neither is the effect of climate change on marine emissions of sea salt and VOCs (as marine VOCs depend both on the biological activity and environmental parameters) yet clear. Rather than the usually suspected sulfur compounds, iodine-containing compounds, whose emission is linked to seawater microorganisms, appear to be at the origin of new particle formation over the northwestern Mediterranean. Despite the fact anthropogenic emissions are also important contributors to the composition of the air, their quantification are still associated with considerable uncertainty. Finally, recent studies in large urban centers in the eastern Mediterranean basin indicate that large scale anthropogenic emission inventories are seriously underestimated. What is more, all these inventories agree on a marked increase in anthropogenic emissions of major pollutants in the Middle East area (MEA). Higher levels of primary emitted pollutants combined with higher temperature in the future will lead to more frequent intense pollution events that will have major health impacts in urban areas.

The level of pollution of Mediterranean air also depends on long range and intercontinental transport. Recent ChArMEx field campaigns identified air masses from North America and tropical Africa in the western Mediterranean and from South East Asia in the eastern Mediterranean. It was shown that imported Asian pollution builds up in the eastern Mediterranean, leading to a sharp west-to-east increasing gradient in aerosols and trace gases such as ozone and methane. These intercontinental pathways come into play under specific meteorological configurations that will be impacted by climate change. However, it is not yet known if climate change will favor intercontinental transport, or not.

The Mediterranean surface water ecosystem largely depends on atmospheric inputs for most of its crucial nutrients (N and P), especially in summer when thermal stratification prevents the upwelling of nutrients from deeper waters. Anthropogenic nitrogen and airborne dust deposition to nutrient depleted surface seawater could favor phytoplankton development and this fertilization effect may stimulate the transfer of atmospheric CO_2 to sediments, reducing atmospheric CO_2 and climate change. Due to air quality mitigation measures, it is also predicted than anthropogenic N fluxes will decrease in the coming years, limiting the atmospheric input of nutrients and possibly related biological activity. In addition, long term series of deposition measurements suggest that the atmospheric input of dust has decreased by one order of magnitude in the last decade compared to previous decades. The reasons are not well understood, satellite observations show that Saharan dust transport events are still common. Recent *in situ* studies in the northwestern Mediterranean showed that Saharan dust deposition by rain stimulates heterotrophic bacteria growth, which reemits CO_2. Thus, dust deposition has two opposite effects on the atmospheric CO_2 that need to be further studied in order to estimate the net effect at large scales.

One very important impact of atmospheric chemistry on the regional climate is the impact of aerosols on the water cycle, which is often neglected in climate models. The Mediterranean region is a special place where most of the moisture that fuels precipitation comes from evaporation from the Mediterranean basin. One direct effect of aerosols is reducing the solar energy delivered to the surface by scattering it back to space or absorbing it within turbid layers containing desert dust or carbonaceous aerosols. This dimming effect decreases surface temperature and consequently evaporation from the sea surface. Atmospheric models with externally forced sea surface temperature do not properly account for this effect, which can be simulated by atmosphere-ocean coupled models. A recent study performed in the framework of ChArMEx and HyMEX calculated that this radiative effect reduces the regional precipitation by 10%, which is a major issue in a region where water is already scarce. But aerosols can further reduce precipitation by indirect effects through aerosol-cloud interactions. Since water vapor condenses around aerosol particles to form the cloud droplets, an excess of aerosols leads to smaller cloud droplets that do not get big enough to fall. Another important and even more complex issue is the formation of ice crystals that trigger the precipitation cycle, which critically depends on the icing properties of aerosol particles. These properties are highly contrasted in different types of particles, some bacteria and soil dust being the most efficient. Aerosol-cloud interactions remain a major source of uncertainty in climate models and climate change projections, and should thus be an important field of study in the near future in the Mediterranean due to its possible impact on the scarcity of water resources.

The most dramatic effect of bad air quality is on human health. People poison themselves by breathing polluted ambient air, the result being chronic diseases or even premature death. Most inhabitants of the Mediterranean region, especially on the southern and eastern sides of the basin, are more or less regularly exposed to high loads (well above WHO air quality recommendations) in the form of soil dust particles, smoke emitted by forest fires, ozone, and anthropogenic emissions from almost unregulated large urban centers. All around the basin, the rare available epidemiological studies all show an excess of premature deaths associated with an increase in particulate pollution. Reducing pollution levels would reduce the death toll and hospital admissions, and prevention policies should be established with a view to reducing effects on health.

Recent evidence demonstrated the link between climate change and air pollution both regarding adaptation and mitigation strategies. There are important possible win-win strategies to be developed in the years to come to improve air quality while engaging in a process aimed at a low carbon economy. Such benefits have been pointed out in several European studies, but the specific situation of Mediterranean countries deserves more specific investigations.

To conclude, it is clear that both-way interactions between atmospheric chemistry and climate are not yet fully understood and quantified in the Mediterranean region. Robust predictions of the future living conditions in the Mediterranean require that such interactions are included in regional models. Positive feedback is expected between climate change and air pollution, but quantification of natural and anthropogenic emissions, process studies, and the development of chemistry-transport models are still necessary for a good assessment of future regional atmospheric environmental and climate conditions. In particular, *air pollution health* risk *assessment* is still lacking at the regional scale and requires a major research effort on the southern and eastern side of the basin where the dose-response functions established in well-developed countries can be questioned.

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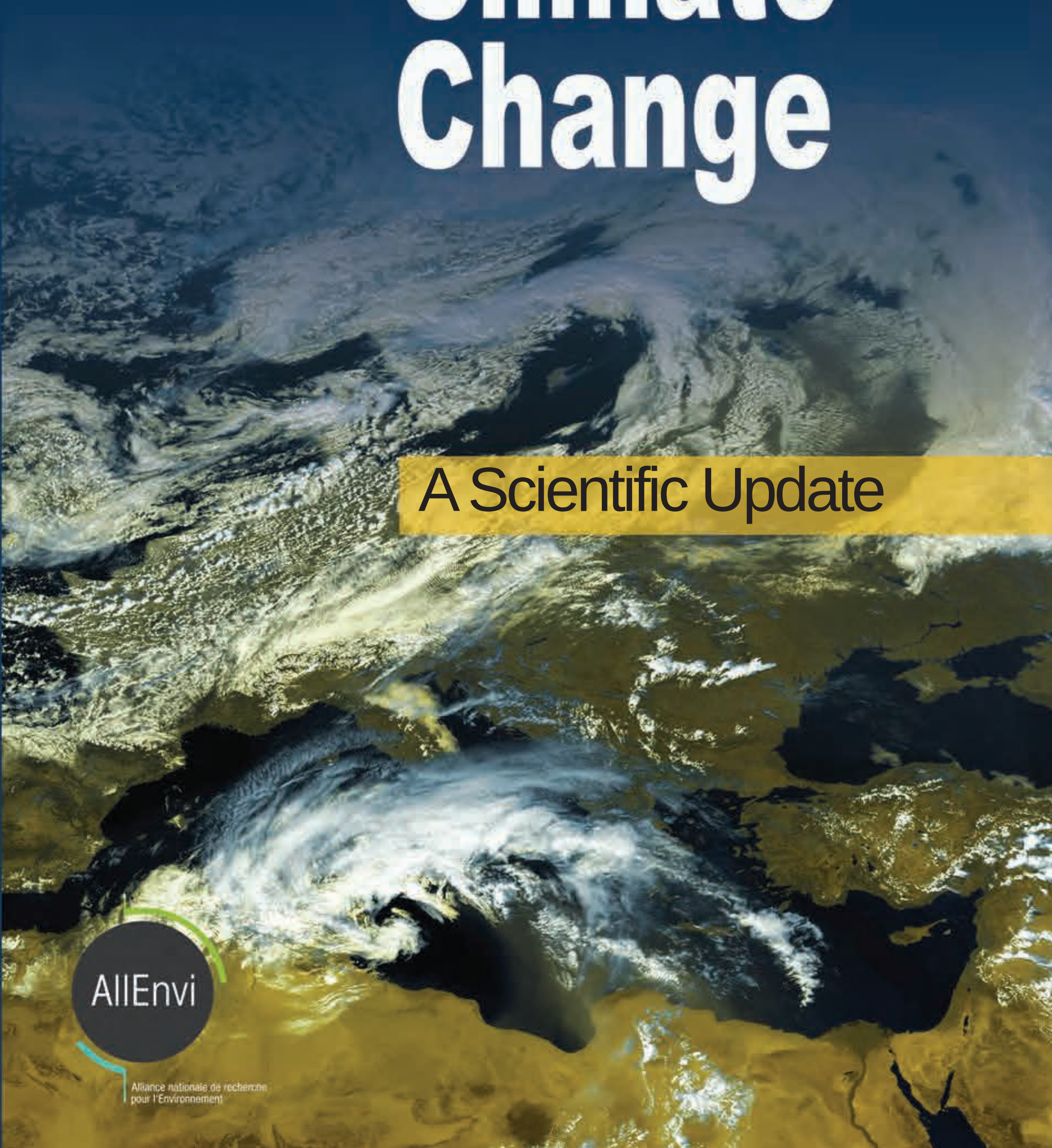
A Scientific Update



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The Mediterranean Region under Climate Change

A Scientific Update

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