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Aerosol-cloud interactions and impact on regional climate

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Abstract Aerosols interact with clouds through radiative and microphysical mechanisms in addition to their direct radiative effects of scattering and absorbing of solar and thermal radiation. Aerosol indirect effects consist of the modification of cloud droplet number concentrations, cloud albedo, and ice nucleating particle concentrations with ensuing effects on precipitation through aerosol perturbations. Different aerosol types present over the basin, notably dust, sea-salt and anthropogenic contribute to the formation of cloud condensation and ice nucleating particles, thus modifying cloud parameters. These processes notably occur during the frequent dust outbreaks over the Mediterranean Sea. Besides, the semi-direct aerosol effect, namely changes in cloud cover and atmospheric dynamics due to aerosol absorption, is another impact on regional climate. Regional climate simulations including aerosol-cloud interactions highlight the importance of considering aerosols, even if uncertainties are still important notably with regards to effects on cloud microphysics. To date, the direct and semi-direct effects seem to have larger impacts on the average radiative budget over the Mediterranean than the cloud-albedo indirect effect, but the question remains open concerning other indirect effects. Therefore, more observations of these interactions coupled with numerical simulations considering all these processes are needed to reduce uncertainties.

Keywords Climate-aerosol interactions; Aerosol indirect effects; Aerosol-cloud interactions; Cloud-albedo indirect effect; Semi-direct aerosol effect; Aerosol impact on surface temperature; Cloud microphysics; Cloud fraction; Cloud droplet effective radius; Cloud droplet number concentration (CDNC); Cloud condensation nuclei (CCN); Ice nuclei (IN); Aerosol impact on precipitation; Regional climate simulations; Aerosol observations; Mineral dust; Sea-salt aerosols; Anthropogenic aerosols.

1 Introduction

1.1 Climate-aerosol interactions

Atmospheric aerosols play an essential role in the climate system through different mechanisms. First of all, they interact with radiation insofar as they are able to absorb and scatter solar radiation, and absorb and emit thermal radiation. This is known as the direct effect (Haywood and Boucher 2000), and has been documented in the previous chapter (Mallet et al. this volume). The second, main impact of aerosols on climate is due to their

interactions with clouds. Indeed, aerosols are known to act as cloud condensation nuclei (CCN) and ice nucleating particles (INP) and thus impact the formation and the life cycle of clouds (Pruppacher and Klett 1980). These effects are known as indirect aerosol effects (Lohmann and Feichter 2005), and belong to what is now called aerosol-cloud interactions (Boucher et al. 2013). In addition to these direct and indirect effects, atmospheric circulation and dynamics can also be affected by local warming due to aerosol absorption of solar light, with ensuing effects on cloud cover and precipitation. These processes are referred to as semi-direct effects (Hansen et al. 1997; Johnson et al. 2004). All these effects have significant consequences on global and regional climate. This chapter will specifically deal with aerosol-cloud interactions and their impact on the regional climate over the Mediterranean area.

1.2 Aerosol indirect effects

Aerosol indirect effects are a generic term to refer to all effects arising from processes involving aerosol particles and cloud microphysics. They include notably cloud droplet activation and ice nucleation which lead to the formation of cloud droplets and ice crystals, respectively. Aerosols can alter warm, ice and mixed-phase cloud formation processes by increasing droplet number concentrations and ice particle concentrations. The various identified aerosol indirect effects are complex and interlinked. The first one has been identified by Twomey (1959), consisting of an enhanced number of cloud droplets due to the presence of aerosols. These cloud droplets have reduced sizes for a similar liquid water path (LWP), resulting in an increased cloud optical thickness, that is to say an increase in the cloud albedo, which could lead to surface cooling. Besides, the more numerous but smaller cloud droplets inhibit precipitation formation efficiency and therefore increase the cloud lifetime, LWP and cloud fraction (CF), which is referred to as the Albrecht effect or second indirect effect (Albrecht 1989). This effect could be as large as the Twomey effect. This change in droplet size can also induce changes in the cloud top height (Pincus and Baker 1994). Aerosols may thus lead to an increase in cloud optical thickness due to a combination of reduction in cloud droplet radius and increased water content. However, this simple chain of events has been challenged for warm stratocumulus clouds (Ackerman et al. 2004) and shallow cumulus (Jiang et al. 2006). The latter studies confirmed a reduction in precipitation with increasing aerosol but showed both positive and negative responses of parameters such as LWP and CF. Aerosols perturbing pristine convective clouds have also been proposed to result in the same amount of surface precipitation due to convective invigoration despite the slower conversion of cloud droplets to precipitation in polluted conditions (Rosenfeld et al. 2008). Furthermore, Koren et al. (2012) have shown that aerosols could intensify precipitation events in the lower and midlatitudes. As a matter of fact, these different studies show that it is difficult to establish climatically meaningful relationships between aerosols, clouds and precipitation (Stevens and Feingold 2009). More recently, Fan et al. (2018) have highlighted the role of ultrafine aerosol particles (less than 50 nm in diameter) in enhancing convection and precipitation although they are too small to serve as CCN at cloud base.

Moreover, the complex interactions between particles and clouds depend on the characteristics of particles in question (concentration, size distribution, chemical composition, hygroscopic properties, mixing state) and the type of clouds involved (continental or maritime, convective or stratiform, cold or warm). Aerosols, on which cloud droplets form, determine the initial concentration and sizes of the droplets, influence the

effectiveness of precipitation production and affect cloud radiative properties. For instance, even insoluble particles like dust and biomass burning aerosols may affect convective clouds if coated with a small amount of soluble material (Levin et al. 1996; Kaufmann and Fraser 1997; Ackerman et al. 2000; Roberts et al. 2003; Levin et al. 2005). In return, the clouds affect the aerosols by modifying their size, concentration, and chemical composition due to heterogeneous reactions. Particles are modified by various chemical reactions that take place in the drops (or in ice crystals) and their concentration in the atmosphere is modified through various scavenging processes. Since most clouds evaporate and do not precipitate, aerosols of different compositions are released back into the atmosphere after droplet evaporation (Levin et al. 1996; Wurzler et al. 2000; Yin et al. 2002). For example, the study of Wurzler et al. (2000) shows that the interaction of insoluble mineral dust particles with clouds modifies the composition and size of the particles that are released into the atmosphere following cloud evaporation. Laboratory measurements indicate that dust particles may become better CCN after cloud processing (Kumar et al. 2011). These modified particles can serve as giant cloud condensation nuclei (GCCN) and thus can have a significant impact on the microphysical characteristics and the formation of precipitation in subsequent clouds (Rosenfeld et al. 2001).

It must be borne in mind that the treatment of aerosols and clouds in climate models encounters large uncertainties because of the complexity of these mechanisms and the large range of scales of these interactions (Seinfeld et al. 2016). Numerous observations of various types (remote sensing, in situ) are increasingly being used to constrain model simulations. The use of fine-scale models is therefore helpful to represent clouds, aerosols, and aerosol—cloud interactions with high fidelity, and to consider specificities of regions such as the Mediterranean area. The interactions with the larger scale are however absent from these regional models.

1.3 Other cloud-aerosol interactions

With regards to the semi-direct effect of aerosols, several studies have focused on the inhibition of cloud formation (Ackerman et al. 2000; Ramanathan et al. 2001; Koren et al. 2004; Johnson et al. 2004; Ramanathan et al. 2007). This aerosol effect has a strong impact on the radiative balance, but is also very sensitive to the vertical distribution of aerosols and clouds (Penner et al. 2003; Johnson et al. 2004). Studies have shown that absorbing aerosol in a cloud layer could increase evaporation of droplets, whereas an absorbing aerosol layer warming the air above a cloud deck (e.g., stratocumulus) could result in a thicker cloud via the reduction of above cloud subsidence and dry intrusions (Wilcox 2010). The cloud adjustments to absorbing aerosol such as black carbon can thus partially offset or reinforce the cooling due to the indirect aerosol effect (Penner et al. 2003; Johnson et al. 2004). Both the second indirect effect and the semi-direct effect involve feedback because of cloud lifetime and liquid water content changes.

1.4 Clouds and aerosols over the Mediterranean

Undoubtedly the aerosol types, seasons and regions of interest influence the impacts of aerosols on regional climate. As the Mediterranean is a crossroads of various natural and anthropogenic aerosol types (Lelieveld et al. 2002), their interactions with clouds and climate is all the more complex. For example, the presence of mineral dust and especially the high percentage of mixtures of mineral dust and sea-salt raise questions about the

potential effects of these particles in cloud formation processes and precipitation development. Previous works have shown that large dust particles coated with soluble material are efficient CCN (Levin et al. 1996) and that mineral dust particles are also efficient INPs (Grosch and Georgii 1976; DeMott et al. 2003). Anthropogenic particles coming from European sources (transport, industries) as well as ship transport are also responsible for the presence of CCN over the Mediterranean Sea. All these issues will be discussed in the following paragraphs, based on both observation (in-situ, satellite data) and modeling studies.

2 Aerosol-Cloud interactions by aerosol type

2.1 The case of dust particles

Mineral dust is the major source of coarse aerosol particles in the Mediterranean, together with sea-salt particles. They mainly come from the Saharan desert, from which they are frequently transported over the Mediterranean Sea (Moulin et al. 1998; Israelevich et al. 2012; Gkikas et al. 2013). Their optical properties make them able to both scatter and absorb solar radiation, which leads to a reduction in incoming solar radiation at the surface, with ensuing cooling as shown by several studies over the Mediterranean area (Spyrou et al. 2013; Flamant et al. 2015; Nabat et al. 2015a; Gkikas et al. 2018). Dust aerosols also have the ability to affect cloud microphysics through ice particle formation, and to act as CCN through their coating by other aerosol types such as sea-salt and sulfate (see Denjean, volume 1, for more details).

In particular, dust aerosols interact in the formation of giant cloud condensation nuclei (GCCN) as shown by Levin et al. (2005). These GCCN can notably enhance the development of the large drops, leading to enhanced growth by collection and rapid development of the rain (Johnson 1982; Cooper et al. 1997; Yin et al. 2000; Teller and Levin 2006). They have thus a positive effect on the amount of rain in continental clouds but have a negligible (even slightly negative) effect in maritime clouds. Levin et al. (2005) have used size distribution and chemical composition of the aerosols measured during a dust storm in the Mediterranean region as initial conditions for simulating cloud development. They concluded that the amount of rain falling from a maritime cloud is generally much higher than from a continental cloud. However, the development of the warm rain process in the latter is enhanced when allowing the soluble component of the mixed aerosols to act as an efficient GCCN and ignoring the ice-nucleating ability of the mineral dust. They also infer that the role of mineral dust particles acting as efficient INP reduces the amount of rain on the ground compared to the case when they are inactive. When the dust particles are active as both GCCN and effective INP, the continental clouds become wider, while the effects on the more maritime clouds are very small. It was also found that the rapid growth of large drops enhances ice formation in colder convective clouds through the more efficient freezing of drops. This efficient production of large drops and ice particles modifies the time, the amount, and the distribution of the precipitation. In contrast, when the concentration of small nuclei is low, as that in maritime clouds, the effect of these GCCN is insignificant and the total precipitation remains unaltered. On the other hand, for both marine and continental clouds, inclusion of the effects of INP by dust particles tends to delay the initiation of the rain and leads to a reduction in the total rainfall amount as compared with the cases having only GCCN.

Satellite observations and in-situ measurements have also been used to show that desert dust aerosols might also suppress precipitation in marine clouds (Rosenfeld et al. 2001; Mahowald and Kiehl 2003). The latter two studies found a strong relationship between thin cloud properties and mineral dust concentrations over North Africa. In fact, the introduction of modified dust into the clouds has two competing effects (Yin et al. 2002). The large particles acting as GCCN produce larger drops which tend to rapidly form precipitation by collecting small drops formed on the large number of small CCN particles. At the same time, the larger concentrations of drops compete for the available water vapor. Thus, the drops may be smaller, reducing the efficiency of precipitation.

More recent studies have relied on regional climate simulations to confirm the role of dust in CCN and INP formation. Bangert et al. (2012) reproduced a strong Saharan dust outbreak over Europe in May 2008 using the regional climate model COSMO-ART, including a two-moment cloud microphysics scheme coupled with comprehensive parameterizations of aerosol activation and ice nucleation. Even if dust aerosols do not significantly modify the cloud droplet number concentration, the simulation highlights the impact of dust on clouds under specific temperature conditions, namely between the freezing level and the level of homogeneous freezing. In this case, the number concentration of ice crystals is found to be strongly related to dust particles because of their efficient heterogeneous freezing. Dust aerosols thus modify the glaciation of mixed phase clouds, with ensuing effects on cloud optical properties that lead to optically thicker clouds, which scatter more efficiently shortwave radiation. As a result, Bangert et al. (2012) have found that the combined direct and indirect effects of dust aerosols caused a decrease in the incoming shortwave radiation at the surface of up to -75 W m^{-2} and a cooling of -0.2 to -0.5 K (locally -1.0 K) during this dust outbreak. However, at lower temperatures, the effects of dust particles are weaker because of a competition with the homogeneous freezing of liquid aerosol particles for water vapor during the ice nucleation. The same kind of dust outbreak was also studied by Bègue et al. (2015), who simulated the transport of dust over the Mediterranean and Europe with Meso-NH in May 2008 during the EUCAARI campaign. Their analysis puts forward heterogeneous reactions between dust and inorganic salts, making dust particles soluble enough to impact hygroscopic growth and cloud droplet activation. Another study by Abdelkader et al. (2015) focused on two dust events over the eastern Mediterranean in September 2011 has highlighted the role of synoptic disturbances and frontal systems in the transport of dust particles in this region. Using simulations with the atmospheric chemistry model ECHAM5/MESSy2 and back trajectory analysis, these authors have shown that frontal systems associated with precipitation control dust removal processes, thus impacting dust ageing, increasing particle size, dust deposition and scavenging efficiency during transport.

Beyond these case studies, Hande et al. (2015) have studied the impact of dust on ice nucleation at a seasonal time scale. They used the COSMO-MUSCAT regional climate model to estimate dust concentrations and their role as ice nucleating particles during 2008. Dust concentrations are indeed highly variable over Europe and the Mediterranean between seasons, with the highest concentrations and variability in the lower to mid-troposphere, where the increase of potential INP concentrations is exponential with height. Spring has been found to be the season with the highest dust concentrations over Europe, and therefore the highest potential INP, contrary to summer about an order of magnitude weaker.

2.2 The case of sea-salt particles

Sea-salt aerosols constitute the other main natural aerosol type present over the Mediterranean region. Like other aerosol particles, they are able to scatter solar radiation (de Leeuw et al. 2011), and to a lesser extent thermal radiation (Li et al. 2008). In addition, they can act as CCN because of their size and hygroscopic properties, and thus impact marine cloud properties and lifetime (Pierce and Adams 2006; Korhonen et al. 2008). Sea salt aerosols are more CCN active than sulphate particles because of their larger size and lower supersaturation threshold required for their activation (O'Dowd et al. 1999). At the global scale, the direct radiative effect of sea-salt aerosols is well established, ranging from -0.38 to -0.65 W m⁻² (Reddy et al. 2005; Ma et al. 2008; Ayash et al. 2008; Partanen et al. 2014) with stronger values over the Southern Ocean. On the contrary, the estimation of the indirect effect is more uncertain. While Ma et al. (2008) have found a larger negative indirect forcing of sea salt aerosols (-2.9 W m⁻²), Ayash et al. (2008) have shown that indirect sea-salt aerosol effects were weaker than their direct radiative effects, and Partanen et al. (2014) have even identified a positive global indirect effect, putting forward competition effects. All these uncertainties argue for a better estimate of sea-salt climate effects at the regional scale.

Over the Mediterranean, the direct sea-salt aerosol effects have been estimated by Zakey et al. (2008) using the RegCM climate model, and by Lundgren et al. (2013), using the nonhydrostatic online-coupled regional-scale model system COSMO-ART for case studies under conditions of low to moderate wind speeds. The resulting clear-sky shortwave and longwave radiative budgets are found to be of the same order of magnitude, but with opposite signs. Therefore, the net radiative effect of sea-salt is close to zero, hence a weak impact of sea-salt on surface temperature. However, this conclusion is only relevant for clear-sky conditions without considering the indirect effects of sea-salt. The latter has been discussed by Kallos et al. (2016), focusing on low cloud formation over the southern Aegean and eastern coast of the Mediterranean. High sea-salt loads are indeed observed in this area. Using the modeling system RAMS/ICLAMS, they simulate the cloud formation of mixed-stratiform clouds and low-level cumulonimbus around Crete. The combination of sea salt and dust particles can lead in deep convection through orographic lifting and activation of ice nucleation processes. The authors conclude that in this case with slow lifting and thermal updrafts sea-salt particles influence mainly the cloud droplet number concentration.

2.3 The case of anthropogenic particles

Anthropogenic particles are also known to have both direct (Charlson et al. 1992) and indirect effects (Boucher 1995). As for sea-salt particles, their direct radiative forcing is better understood than their indirect forcing, probably because of the complexity of cloud system responses to aerosol perturbations that complicates the estimation of indirect effects (Boucher et al. 2013). Sulphate particles brought the highest contribution to the anthropogenic aerosol forcing which is negative at the global scale (Myhre et al. 2013), but nitrates (Bellouin et al. 2011) and secondary organic aerosols (Scott et al. 2014) also have significant impacts, which consist also in negative radiative forcing. The role of black carbon particles is however different, because of their absorbing properties that tend to warm the atmosphere (Chin et al. 2009; Myhre and Samset 2015). The Mediterranean region is also

affected by all these anthropogenic particles, coming essentially from European sources, Middle East and North African cities, and ship transport.

The effective radiative forcing (ERF) of anthropogenic aerosols can be estimated with global simulations from the sixth Coupled Model Intercomparison Project (CMIP6), in which specific simulations using pre-industrial (1850) and 2014 aerosol concentrations have been carried out (Smith et al. 2020). The ERF is calculated according to the Ghan (2013) methodology, which enables us to have separately the aerosol-radiation and the aerosol-cloud interactions (ERFari and ERFaci, respectively). Fig. 1 presents the average over the Mediterranean from six CMIP6 models, for which this calculation of ERF is possible thanks to available radiation fluxes with and without aerosols (double-call radiation). The resulting ERFari at the top of the atmosphere is on average -0.51 W m⁻², in addition to -0.86 W m⁻² for ERFaci. The highest forcing is located over the eastern basin. Note that these ERF values are in absolute values lower than the one of anthropogenic greenhouse gases (2.85 W m⁻²). It can also be inferred that anthropogenic aerosol-cloud interactions could have a larger radiative forcing over the Mediterranean than aerosol-radiation interactions, even if we must bear in mind that uncertainties are important since the respective confidence intervals at 95% of ERFari and ERFaci calculated for this ensemble of six CMIP6 models are ±0.76 and ±0.96 W m⁻², respectively.

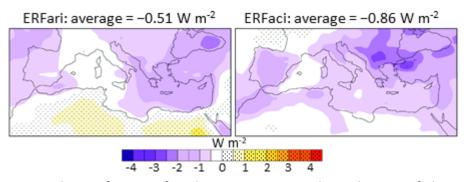


Fig. 1 Effective radiative forcing of anthropogenic aerosols at the top of the atmosphere estimated from multi-model CMIP6 simulations by difference between 2014 and 1850, separated between aerosol-radiation (ERFari) and aerosol-cloud (ERFaci) interactions. Adapted from Smith et al. (2020)

Because aerosols show much more variability at smaller time and space scales than greenhouse gases (e.g., Le Treut et al. 1998), it is of interest to study their radiative forcing at the regional scale with higher resolution models. The first modelling study using a regional model to estimate the anthropogenic aerosol effects over Europe and the Mediterranean area was carried out by Ekman and Rodhe (2003). They used the RCA2 regional climate model including tropospheric sulphate particles and a parameterization of their cloud-albedo effect to estimate the Twomey effect during 1993. The result was an anthropogenic sulfate indirect effect between –2.7 and –2.5 W m⁻², and a corresponding annual mean surface temperature response of –0.4 K over the model domain (western Europe and northern Mediterranean). With regards to direct and semi-direct effects, Zanis (2009) has found a negative radiative forcing over Europe and the Mediterranean during summer 2000, using RegCM3 regional simulations. This negative forcing leads to a decrease in surface temperature by sulfate particles of up to 1.2 K over the Balkan Peninsula and southern Europe. Semi-direct mechanisms have also been identified insofar as sulfate

aerosols induced changes in the circulation with a southward shift of the subtropical jet stream. These climate effects were better assessed in Zanis et al. (2012), using the same model but over a longer period (1996–2007). The annual average effect of anthropogenic aerosols on near-surface temperature was smaller (up to –0.2°C over the Balkan Peninsula) than in previous case studies, but it is associated with changes in atmospheric circulation. The maxima in aerosol loads are indeed uncorrelated with effects on surface temperature.

3 Implications for regional climate

3.1 Impact of aerosol-cloud interactions on regional radiative budget

After analyzing the aerosol effects on clouds and climate by aerosol species, this section aims at summarizing these effects at the scale of the Mediterranean area for the whole aerosol load. First of all, it is worth mentioning that the average direct effect of aerosols over the Mediterranean area has been estimated with both satellite and model data. Papadimas et al. (2012) have used MODIS data to find a direct radiative forcing of aerosols of –16.5 W m⁻² at the surface and –2.4 W m⁻² at the top of the atmosphere (TOA). These values are close to those given by regional climate models: Nabat et al. (2012) have respectively found at the surface and at TOA, –10.3 and –3.1 W m⁻², Nabat et al. (2015b) –19.9 and –7.8 W m⁻², and more recently Drugé et al. (2019) –8.7 and –2.4 W m⁻². Differences in model estimates may be attributed to the way aerosols are represented in the models (e.g., with monthly mean AOD values or interactive variables) and/or to the representation of radiation and clouds.

In addition to these direct effects, the semi-direct effect has also been estimated but in fewer studies. It consists of changes in cloud cover and atmospheric circulation due to aerosol absorption. Nabat et al. (2015b) have found a positive semi-direct effect over the Mediterranean of 5.7 W m⁻² on average, counterbalancing partly the negative direct forcing. A similar positive semi-direct forcing has been identified by Meier et al. (2012) in Europe in the case of absorbing aerosols in summer. In some cases, this semi-direct can even become stronger than the direct effect, as found by Forkel et al. (2012) in June–July 2006 in Europe.

Finally, the estimation of the aerosol indirect forcing is the most difficult to quantify. Using CERES and MODIS satellite data, Quaas et al. (2008) have estimated the average direct and indirect aerosol effects. Their maps show that the direct aerosol effect is an order of magnitude higher than the first indirect effect (cloud-albedo effect) over the Mediterranean area. From surface measurements over 1981–2005, Ruckstuhl et al. (2010) have underlined that the contribution of the first indirect aerosol effect to surface solar radiation changes over Europe was small compared to the direct effect. The same conclusion was drawn by Nabat et al. (2014). However, it should be noted that this comparison concerns only the first indirect aerosol effect, while the other aerosol-cloud interactions are more complex to quantify. Their impact on the radiative budget may not be negligible, and needs to be studied in future.

3.2 Implications for surface temperatures

All the effects of aerosols on the radiative budget including cloud-aerosol interactions have an impact on surface temperature, which generally leads to a cooling of the surface. Climate model simulations with and without aerosols are useful to quantify this impact on

temperature. Ekman and Rodhe (2003) have first shown an average cooling of 0.4°C over Europe due to the first indirect effect of sulphate particles. This order of magnitude was confirmed by the studies of Zanis (2009) and Zanis et al. (2012) as far as the direct and semi-direct effects of sulphate are concerned. Dust aerosols also contribute to this cooling over the Mediterranean, as noted by Santese et al. (2010), Bangert et al. (2012) and Nabat et al. (2015a). However, most of these studies concern only specific aerosol types and/or seasons.

A more complete study dealing with the impact of aerosols on surface temperature was carried out by Nabat et al. (2015b). They have used the fully coupled regional climate system model CNRM-RCSM including the main natural and anthropogenic aerosol types, as well as ocean-atmosphere coupling for the Mediterranean region. They concluded that the negative radiative forcing of aerosols at the surface was responsible for an annual average cooling of 0.5°C in near-surface temperature over the Mediterranean, and the same value for the Mediterranean Sea surface temperature (SST), an effect that atmospheric models forced with external SST cannot consider (Fig. 2). This cooling is stronger in spring and summer when aerosol loads are higher. Indeed, the temperature decrease can be much more important during episodes of strong aerosol concentrations, however, as shown by Gkikas et al. (2019) during dust outbreaks over the central Mediterranean.

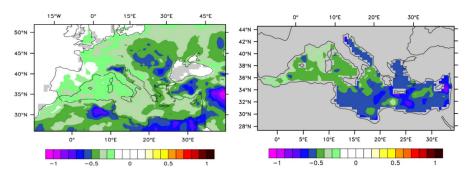


Fig. 2 Annual average aerosol impact on near-surface (2m) temperature **(left)** and on sea surface temperature **(right)** in CNRM-RCSM simulations (2003–2009 average, in °C). Only significant values at level 0.05 are shown. All resulting values are negative. Adapted from Fig. 11 in Nabat et al. (2015b)

3.3 Aerosol impact on cloud parameters

Both satellite data and model simulations have been used to show the impact of aerosols on cloud parameters. Myhre et al. (2007) have used MODIS data to study possible relationships between aerosol optical depth (AOD) and different atmospheric parameters. Their results over the Mediterranean area show a tendency for a weak increase in liquid water path (LWP) with AOD (Fig. 3a), a sharper increase in cloud fraction with AOD (Fig. 3b), and a more surprising increase in cloud droplet radius with AOD (Fig. 3c). With regards to LWP, this relationship is poorly reproduced by global models, making it difficult to draw conclusions from this observation to know whether anthropogenic or mineral aerosols impact the hydrological cycle. Tests performed with a non-hydrostatic large eddy simulation (LES) model (Sandu et al. 2005) demonstrate that the diurnal evolution of the cloud LWP is very sensitive to the type and amount of aerosol particles in the boundary layer. The increase of cloud fraction with AOD could be related to the role of aerosols as CCN, but also to their effects on cloud lifetime which are very complex. Jiang et al. (2006) have explored this

impact for shallow cumulus clouds by using models including size-resolved treatment of drop size distributions and warm microphysical processes, and a variety of sounding representing marine trade cumulus and continental convective clouds. Contrary to expectation, it was shown that an increase in aerosol concentration from very clean to very polluted conditions does not increase cloud lifetime even though precipitation is suppressed. This could be due to competing effects between precipitation suppression and enhanced evaporation. For a Mediterranean sounding adapted from measurements in the central Mediterranean Sea, individual cloud lifetimes may even show a decrease in lifetime of 10%-40% for polluted conditions. It is proposed that an enhanced evaporation tends to dominate in these shallow clouds. Absorption of incoming solar radiation by aerosols (like smoke and dust) can reduce cloud cover (Ramanathan et al. 2001; Kaufman et al. 2005). This effect can even be amplified if absorption of solar radiation by these aerosol particles occurs within cloud droplets (Chýlek et al. 1996). In fact, absorption of solar radiation, which is dependent on the chemical composition of the aerosol particles incorporated in cloud droplets, has a significant impact on the diurnal cycle of boundary layer clouds (Sandu et al. 2005).

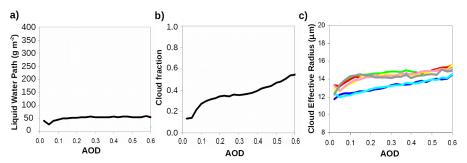


Fig. 3 a Liquid water path, **b** cloud fraction, and **c** cloud effective radius, as a function of the aerosol optical depth for the Mediterranean region. In (a) and (b), data are from the standard MODIS Terra product for the year 2001. Every line in (c) represents a different year from MODIS Aqua (2003 and 2004, in blue) or MODIS Terra (2000–2004). Adapted from Figs. 5, 7 and 13 in Myhre et al. (2007)

With regards to the relationship between cloud droplet radius and AOD, the classical theory (Twomey 1959) would imply a decrease in effective radius with the aerosol load, associated with an increase in cloud droplet number concentration. This is the basis of some parametrizations of the first indirect aerosol effect (e.g., Menon et al. 2002). At the global scale, this relationship between cloud droplet number concentration and AOD is observed in both satellite data (MODIS) and global models (Quaas et al. 2009). However, as mentioned previously Myhre et al. (2007) found the opposite result with MODIS data over the Mediterranean. This could be due to the observed decrease in cloud top pressure with AOD, given that cloud droplet radius increases with a decreasing cloud top pressure of convective clouds (Rosenfeld and Lensky 1998). This specificity of the Mediterranean area needs to be confirmed in the future by more observations and modelling studies. Besides, Calmer et al. (2019) have recently highlighted the role of entrainment on cloud optical properties and cloud-aerosol interactions, this variability in entrainment mixing also needs to be constrained in order to improve climate models.

3.4 Aerosol impact on precipitation

The impact on precipitation and hydrological cycle of the direct and semi-direct radiative effects has been studied by Nabat et al. (2015b). Using regional ocean-atmosphere coupled simulations, they concluded in a decrease of -0.2 mm yr⁻¹ in precipitation over the Mediterranean due to the increase in atmospheric stability caused by aerosols. It represents a reduction of about 10% of the activity of the hydrological cycle, which is also noted in evaporation.

In addition to the reduction of precipitation caused by the reduced radiative flux due to the direct aerosol effects, the sign and magnitude of the aerosol indirect forcing effect on precipitation is still controversial, because of various processes competing with each other. Solomos et al. (2011) have carried out RAMS/ICLAMS simulations to deal with the effects of pollution on precipitation in clean and polluted environments in the eastern Mediterranean. The onset of precipitation in hazy clouds is shown to be delayed compared to cleaner conditions. In particular, hygroscopic dust particles contribute to more vigorous convection and more intense up-drafts. In a case study, the simulated clouds extended about three kilometers higher, thus delaying the initiation of precipitation by one hour.

Using WRF simulations including an aerosol-aware microphysics scheme, Da Silva et al. (2018) have studied aerosol effects on summer precipitation in the Euro-Mediterranean area. They have found that increased aerosol loads lead to a reduction in precipitation, both with and without convection-permitting configurations. They propose a causal chain between aerosol effects on microphysics and their effects on precipitation (Fig. 4). These relationships involve the negative aerosol radiative forcing at the surface due to changes in cloud microphysics making clouds optically thicker (left part of the scheme), which increases atmospheric stability and thus reduces convection (right part of the scheme).

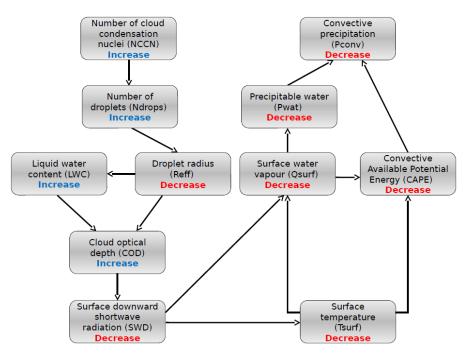


Fig. 4 Schematic summary of the proposed aerosol causal chain sequence for the indirect effect of aerosols on convective precipitation. Reprinted from Da Silva et al. (2018)

Global climate models (GCM) can also be used to identify the aerosol effects on precipitation, taking advantage from the higher number of available simulations despite coarser horizontal simulations. Tang et al. (2018) have compared the GCM responses of Mediterranean precipitation to greenhouse gases and aerosols. Both of them can indeed cause a drying of the Mediterranean, but they conclude that precipitation is more sensitive to black carbon forcing than to greenhouse gases and sulfate aerosol. Black carbon has the ability to strongly absorb solar radiation, thus heating the atmosphere locally. However, this is not the only mechanism that explains the impact of aerosols. An enhanced positive sea level pressure pattern similar to the North Atlantic Oscillation-Arctic Oscillation has been shown because of black carbon particles, characterized by higher pressures over the Mediterranean and lower pressures at northern latitudes. Greenhouse gases can lead to similar pressure changes. Due to both factors, the northward diversion of the jet stream and storm tracks tend to decrease precipitation in the Mediterranean while increasing precipitation in northern Europe. Finally, the authors warn of a possible underestimation of the aerosol-cloud interactions in these GCMs because many of them did not fully include such processes.

3.5 Trends in aerosol-cloud interactions

As described in the preceding chapter (Mallet et al. this volume), an increasing trend in surface solar radiation has been observed since 1980, which has been attributed to the decrease in anthropogenic aerosols over the same period by Nabat et al. (2014). This study also underlines that the role of clouds and of the indirect aerosol effect in this brightening seems to be negligible compared to the direct effect of aerosols at the scale of the Mediterranean area. However, weak trends in cloud cover and other cloud parameters have been noted in the same period. Using the MERRA reanalysis, Kambezidis et al. (2016) have shown that the brightening over the Mediterranean during the last decades was mostly associated with decreasing trends in cloud optical depth (COD), especially for the low (<700 hPa) clouds and during the spring season. On a monthly basis, those low clouds contributed 58%–87% to the total spatially-averaged COD in the Mediterranean. This decreasing trend in cloudiness and cloud thickness over southern Europe and the Mediterranean is also supported by other studies. In this respect, the slight brightening observed over Europe by Hatzianastassiou et al. (2012) was associated with a decrease of 5–10% in cloud cover.

As for precipitation, a slight decrease of -0.56 mm per month per decade (-1.3%) has been noted during 2002–2014 by Floutsi et al. (2016), in consistence with the decrease of convection and low-level cloudiness observed in Kambezidis et al. (2016).

4 Conclusion and recommendations

All these studies put forward the various and strong effects of natural and anthropogenic aerosols on the Mediterranean regional climate. In addition to the direct radiative effects, the interactions between clouds and aerosols also modify the radiative budget and regional climate over the Mediterranean. Absorbing aerosols such as dust and carbonaceous particles can modify cloud formation and atmospheric dynamics, which leads to semi-direct aerosol effects. Many aerosol types can also act as CCN and/or INP, with ensuing impacts on cloud cover, cloud lifetime and precipitation. However, there remains large uncertainties about the role of aerosols in modifying cloud microphysics, cloud radiative properties, and precipitation. More research in terms of both field experiments, laboratory studies and

modelling efforts is clearly needed in order to better understand and quantify the effects of aerosol on clouds and the hydrological cycle and predict future evolution. Most of the results presented above, which rely on numerical simulations, must be taken with caution because of the uncertainties associated with the different parameterizations used in models. In order to better understand and quantify the microphysical impact of aerosols on clouds, and thus better predict climate change, it is critical to particularly seek information on aerosol-cloud interactions in observations (Seinfeld et al. 2016). For that, the Mediterranean region appears as an excellent natural laboratory for studying the interactions between clouds and aerosols because this region is affected by air masses from different sources containing anthropogenic pollution from Europe, marine aerosols from the sea, mineral dust particles from the Sahara desert, biomass burning from summertime forest fires, and biogenic material from various land and marine sources.

Representation of cloud microphysical processes in climate models is challenging because fundamental microphysical details are poorly understood. Although there are several parameterizations that correlate the aerosol concentration and the CCN/INP concentrations as a function of aerosol properties, too many unknowns exist in order to accurately predict the cloud droplet and ice crystal number concentrations. To date, in-situ observations of the latter in the different cloud regimes over the Mediterranean are lacking. Calls for observations of cloud droplet and ice crystal number concentrations in the different cloud regimes have already been expressed (Kanji et al. 2017; Grosvenor et al. 2018), specifically to provide a more complete assessment of the relationship between CCN/INP concentration and cloud droplet/ice crystal number concentrations. To accomplish this, the next generation of field experiments should focus on extended characterization of aerosol and enhanced cloud observations (e.g., mixed-phase and ice clouds developing in different air masses) by combined in situ and remote sensing observations. These observations would be very helpful to improve climate models over the Mediterranean, and notably better understand the specificities of this region in aerosol-cloud interactions described in this chapter.

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