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► **To cite this version:**

Marc Mallet, Pierre Nabat, Alcide Giorgio Di Sarra, Fabien Solmon, Claudia Gutiérrez, et al.. Aerosol and tropospheric ozone direct radiative impacts. François Dulac; Stéphane Sauvage; Eric Hamonou. Atmospheric Chemistry in the Mediterranean Region, Vol. 2 - From Air Pollutant Sources to Impacts, Springer International Publishing, pp.373-402, 2022, 10.1007/978-3-030-82385-6_19 . hal-03975238

HAL Id: hal-03975238

<https://hal.science/hal-03975238>

Submitted on 6 Feb 2023

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Aerosol and tropospheric ozone direct radiative impacts

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Chapter reviewed by **Olivier Boucher** (Laboratoire de Météorologie Dynamique, Paris, France), as part of the book Part IX also reviewed by **Lucia Mona** and **Simone Lolli** (CNR/IMAA, Tito Scalo (PZ), Italy)

Abstract This chapter describes the direct radiative effect (RE) over the Mediterranean region exerted by aerosols and tropospheric ozone. Recent results on the regional direct aerosol RE at the surface and at the top-of-the-atmosphere (TOA) are presented, together with trends in surface solar radiation. Absorption by aerosol particles within the troposphere affects heating rates and atmospheric stability, therefore playing a key role in the regional radiative impact of aerosols. In addition, the impact of aerosols on photochemistry and solar energy production over the Mediterranean region is also discussed with a focus on recent advances on the impact of the aerosol dimming on photovoltaic (PV) panels. Finally, the last section describes the regional RE of tropospheric ozone and drivers of its uncertainty.

Keywords Aerosol direct radiative impact; Tropospheric ozone direct radiative impact; Aerosol absorption; Aerosol scattering; Aerosol single scattering albedo; Surface radiative forcing; Top-of-Atmosphere radiative forcing; Heating rates; Atmospheric stability; Aerosol impact on photochemistry; Aerosol impact on photovoltaic production; Biomass burning aerosols; Anthropogenic Aerosols; Mineral dust; Marine aerosols; Volcanic aerosols; Surface solar radiation.

1. Introduction

Among the various impacts (air quality, biogeochemical cycle, snow deposition,...) of atmospheric aerosols, their radiative effect has been recognized of paramount importance on the climate (e.g., [Ramanathan et al. 2001](#); [Crutzen et al. 2003](#); [Boucher et al. 2013](#)). Indirect radiative effects through aerosol-cloud interactions are analyzed in the following chapter of this volume ([Nabat et al. this volume](#)). In this chapter, we focus on the direct radiative forcing exerted by different aerosols types observed in the Mediterranean region. Quantifying the radiative forcing of aerosols is very important because it can lead to climate change (surface temperature, surface-atmosphere exchange fluxes, precipitation) even if these feedback's are complex and not necessarily linear. Most of the studies are based on a 1D radiative transfer model using ground-based and aircraft in situ measurements as input data. Some estimates are provided by regional climate models and satellite data. We consider in this chapter the aerosol direct forcing exerted at the surface (section 2.1) and at the Top of the Atmosphere (TOA) (section 2.2). In the last part (section 3), we discuss the effect of changes in surface radiations due to aerosols on photolysis and photovoltaic production (PV).

2. Aerosol direct radiative forcing

In the literature, aerosol direct radiative forcing is classically defined at different levels, namely at the surface or bottom of atmosphere (BOA), at the tropopause or top-of-the-atmosphere (TOA), and within the atmosphere (ATM). The first forcing represents the effect of particles on the net radiation fluxes reaching the surface and the second one, on the radiation fluxes reflected back to space by aerosols. The last term indicates the part of solar energy which is absorbed by aerosols. Through absorption and scattering, aerosols typically induce a decrease in the solar energy reaching the surface, thereby possibly cooling it. At TOA, the aerosol particles generally produce an increase in the reflectivity of the Earth-Atmosphere system, inducing a cooling or parasol effect ([Crutzen et al. 2003](#); [IPCC 2013](#)). The atmospheric forcing results, for a part, from the possible absorption of solar radiation by particles. In case of purely scattering aerosols (e.g., pure sulphate or sea-salt), the single scattering albedo of particles (SSA) tends towards 1 and the loss of energy at the surface is mostly scattered upward to space. In case of particles absorbing light from biomass burning (BB), anthropogenic urban/industrial (UI) pollution or mineral dust (D), the SSA is lower than 1 and a part of this loss at the surface is due to absorption within the aerosol layer which is consequently heated ([Alpert et al. 1998](#); [Ramanathan et al. 2007](#)). Large particles, such as dust, produce a measurable impact on the longwave radiation budget, through radiation absorption, scattering, and emission. The longwave radiative forcing at BOA is generally positive (i.e., aerosols induce an increase in downwelling IR radiation due to IR emission by suspended particles), while it is positive at TOA. Although this effect is smaller than in the shortwave spectral range, it may offset a significant part of the shortwave effect when large particles are present.

2.1 Direct radiative effect at the surface

By scattering and absorbing solar and infrared radiation, natural and anthropogenic aerosols are able to strongly perturb the net shortwave and longwave (to a lesser extent) flux at the surface in the Mediterranean basin. As an example, observations performed at the island of Lampedusa in the western central Mediterranean (35.5°N, 12.6°E) have been used to

provide computations of the direct radiative forcing for three Saharan dust episodes, which occurred on May 18, 1999 (Meloni et al. 2003) and July 14–16, 2002 (Meloni et al. 2004). Radiative transfer computations denote a significant surface cooling, with respective instantaneous BOA forcings (Fig. 1; Table 1) of -13 , -24 and -70.8 W m^{-2} for 14, 16 July and 18 May. At the same location, di Sarra et al. (2008) report surface aerosol effects obtained measuring the shortwave fluxes and aerosol optical properties in the period May–November of 2003 and 2004 (the pristine flux is considered here as the one occurring in the absence of atmospheric aerosols). They indicate that the daily surface forcing of mineral dust is much larger with respect to other aerosol species, with a mean value at the summer solstice and equinox of -30 and -24 W m^{-2} , respectively. A significantly higher daily RE, compared to the previous seasonal averages, of about -85 W m^{-2} was measured at Lampedusa island in 2011 during a very intense dust event (aerosol optical depth (AOD) up to 2; di Sarra et al. 2011), while it was about -29 W m^{-2} during a dust event (AOD of 0.5 at 500 nm) occurred in spring 2008 (Meloni et al. 2015). At a different site (coastal French Mediterranean region), Saha et al. (2008) have also calculated a significant diurnal reduction of the downward shortwave radiation by about -62 W m^{-2} (daily mean) due to the presence of dust. As previously noted, regional climate models used to compute the direct radiative forcing of aerosols over the Mediterranean, put in evidence important values ranging between -5 and -20 W m^{-2} using the RegCM model (Nabat et al. 2012), with maximum values during the dusty season. In parallel, Nabat et al. (2015) have reported higher values of the direct forcing at the surface ranging from -10 to -30 W m^{-2} using the CNRM-RCSM regional climate model (period 2003–2009). Spyrou et al. (2013) have also shown the crucial effect of dust aerosols on the Mediterranean budget through the dust SKYRON model radiative transfer scheme for a 6-year simulation. Lastly, for specific case studies, Santese et al. (2010) indicated a daily-mean SW direct radiative effect of about $\sim -24 \text{ W m}^{-2}$ at the surface and at a large regional scale, during two dust outbreaks. Using the NMMB-MONARCH model, Gkikas et al. (2018) assessed the regional dust radiative effect during the most intense identified dust events in the Mediterranean in the period 2000–2013 (maximum dust AOD ranging from about 2.5 to 5.5). They found that dust outbreaks affect radiation budget at BOA, with values of the mean regional radiative effect from -22.2 to $+2.2 \text{ W m}^{-2}$.

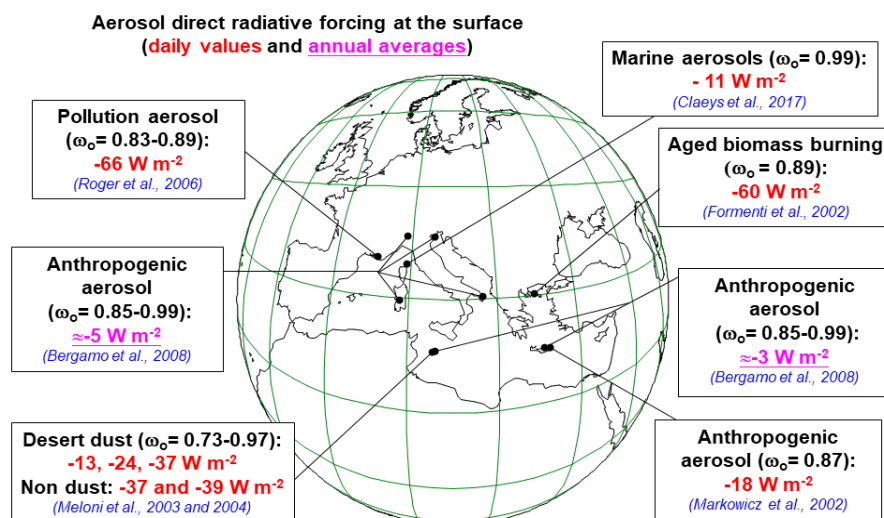


Fig. 1 Examples of literature-reported estimates of the direct radiative effect exerted by different aerosol species (mineral dust, smoke, anthropogenic and sea-spray) at the surface

in the Mediterranean region. Daily and yearly (underlined) estimates are indicated for different locations. ω_0 is the aerosol single scattering albedo. See **Table 1** for more results

Similar effects have been observed and/or calculated for urban/industrial aerosols over the Mediterranean region (**Table 1**). Horvath et al. (2002), Markowicz et al. (2002), Meloni et al. (2003), Roger et al. (2006), Saha et al. (2008), and Mallet et al. (2016) show a decrease in surface solar fluxes of 23, 18, 19, 34, 24 and 15 W m^{-2} (daily mean), at Almeria (South Mediterranean coast of Spain), Finokalia (Crete island in the Eastern Mediterranean; 35.3°N, 25.6°E), Lampedusa island, Marseilles (southeastern France), Toulon (South Mediterranean coast of France) and Barcelona, respectively. More recently, for continental pollution in the western basin observed in summer 2013, di Biagio et al. (2016) have suggested up to a 50% change of the forcing efficiency (FE), i.e., the DRE per unit of optical depth, at the surface (from -160 to $-235 \text{ W m}^{-2} \tau^{-1}$ at 60° solar zenith angle) for a SSA varying between its maximum and minimum value.

Table 1 Daily (unless specified) shortwave (SW) direct radiative forcing (DF, in W m^{-2}) and forcing efficiency (FE, in W m^{-2} per unit AOD) of aerosols in the Mediterranean region

Localisation	Period	Aerosol species	$DF_{BOA} (FE_{BOA})$	$DF_{TOA} (FE_{TOA})$	$DF_{ATM} (FE_{ATM})$	Reference
Eastern Med (STAAARTE-MED Exp.)	14/8/1998	Biomass burning	-64 (-164.1)	-22 (-56.4)	42 (107.7)	Formenti et al. (2002)
Ionian Sea	24–27/8/2007		-55	-20	35	Kaskaoutis et al. (2011)
Barcelona (NE Spain)	07/2009		--	-13(1)	--	Sicard et al. (2012)
Almeria (S Spain)	06/1999	Continental urban	-23.1 (-57.7)	-4.2 (-10.5)	18.9 (47.2)	Horvath et al. (2002)
Marseille (ESCOMPTE Exp.)	06/2001		-34.3 (-114.3)	-7.7 (-25.7)	26.6 (88.7)	Roger et al. (2006)
Toulon (SE France)	06/2006		-24.9 (-83.0)	-5.1 (-17)	19.8 (66)	Saha et al. (2008)
Realtor (ESCOMPTE)	24/6/2001	Continental industrial	-66* (-165.0*)	-9* (-22.5*)	57* (142.5*)	Mallet et al. (2006)
Lampedusa Isl.	25/5/1999	Anthropogenic	-18.8 (-117.5)	+1.1 (+6.9)	19.9 (124.4)	Meloni et al. (2003)
	27/5/1999		-20.0 (-90.9)	-0.6 (-2.7)	19.4 (88.2)	
Finokalia, Crete Isl. (MINOS Exp.)	07/2001		-17.9 (-85.2)	-6.6 (-31.4)	11.3 (53.8)	Markowicz et al. (2002)
Whole Mediterranean	1996–2007		--	-5	--	Zanis et al. (2012)
	1979–2016	Anthropogenic nitrate	-1.7	-1.3	0.4	Drugé et al. (2019)
Vinon-Sur-Verdon (ESCOMPTE Exp.)	24/6/2001	Continental rural	-61* (-164.9*)	-8* (-21.6*)	53* (143.2*)	Mallet et al. (2006)
Toulon (SE France)	19/6/2006	Continental dust	-61.8 (-77.2)	-7.7 (-9.6)	54.1 (67.6)	Saha et al. (2008)
Eastern Med. (24.5–34.5 °E and 32.5–35.5 °N)	June–Aug. 2010	Polluted continental	-17.41^ (-)	-10.27^ (-)	7.14^ (-)	Mishra et al. (2014)
		Polluted dust	-39.95^ (-)	-20.60^ (-)	19.35^ (-)	
Barcelona (NE Spain)	22–23/07/2009	Saharan dust	--	-8(1)	--	Sicard et al. (2012)
Lampedusa Isl.	18/5/1999		-36.7 (-72.0)	-1.6 (-3.1)	35.1 (68.8)	Meloni et al. (2003)
	May–Nov. 2003 and 2004		-(24–30) (-68.8–86.4)	--	--	di Sarra et al. (2008)
	14/7/2002		-13.1* (-56.9*)	-5.7* (-24.8*)	7.4* (32.2*)	Meloni et al. (2004)
	16/7/2002		North African dust	-24.1* (-92.7*)	-1.5* (-5.8*)	
Western Med.	Summer 2013	Marine	-11 (-)	-8 (-)	+3 (-)	Claeys et al. (2017)
7 km downwind Etna (Sicily)	Summer 2016–2017	Volcanic aerosol	-4.5 (-)	-7 (-)	2.5 (-)	Sellitto et al. (2020)
Lecce (South Italy)	Summer 2003–2004	Not specified	-15 (-)	-9.0 (-)	6 (-)	Tafuro et al. (2007)

(* instantaneous values (^) for a solar zenith angle of 60° (overestimates the daytime value at the surface and top of atmosphere, underestimates it within the atmosphere; see suppl. Fig. S8 by Mishra et al. 2014)

By considering annual mean values of the measured AOD at Lampedusa for the different aerosol types, and the corresponding estimates of the aerosol forcing efficiency forcing efficiency (FE), (i.e., the DRE per unit of optical depth), [Di Biagio et al. \(2010\)](#) estimated a surface shortwave daily average RE at the equinox of about -22 W m^{-2} for desert dust, -17 W m^{-2} for polluted/biomass burning particles, and -13 W m^{-2} for marine-mixed aerosol types. Volcanic aerosols originating from Etna have also been shown to produce a non negligible radiative effect during active phases. [Sellitto et al. \(2016\)](#) estimated daily shortwave radiative forcing efficiencies, i.e. radiative forcing per unit AOD, between -39 and $-48 \text{ W m}^{-2} \text{ AOD}^{-1}$ at the TOA and between -66 and $-49 \text{ W m}^{-2} \text{ AOD}^{-1}$ at the surface. Recently, [Sellitto et al. \(2020\)](#) have reported a daily average radiative forcing of -7 W m^{-2} at the surface for a volcanic plume with an ultraviolet AOD of 0.12–0.14.

At a regional scale, [Zanis et al. \(2009\)](#) have also studied the radiative forcing of anthropogenic aerosols for summer 2000 and indicated significant values from -30 to 70 W m^{-2} over the Central Europe (mainly due to sulfates particles) and lowest (between 0 and -10 W m^{-2}) over the Mediterranean basin. In addition, the COSMO-CLM version has been used to study the dimming–brightening phenomenon over Europe for the period 1958–2001 ([Zubler et al. 2011](#)). Recently, [Drugé et al. \(2019\)](#) have proposed a first estimate of the nitrate direct radiative forcing at the surface. Over the period 1979–2016, the DRF is found to be about -1.7 W m^{-2} over Europe. As mentioned in the subsection 3.3 and although only few studies have been dedicated to the radiative effect of biomass burning aerosols over the region, calculations performed by [Formenti et al. \(2002\)](#) during STAAARTE-MED reveal a significant surface dimming by smoke particles. Based on chemical, physical and optical measurements within a large aged BB plume on board a research aircraft, [Formenti et al. \(2002\)](#) indicated a shortwave radiative forcing of -64 W m^{-2} at the surface. Finally and for primary sea spray aerosols, [Claeys et al. \(2017\)](#) have reported values of $\sim -11 \text{ W m}^{-2}$ during summer 2013.

Numerous studies underline the fact that the LW radiative effect of large particles, in particular mineral dust, can offset for a part the one exerted in the SW ([Santese et al. 2010](#); [di Sarra et al. 2011](#); [Nabat et al. 2015](#); [Meloni et al. 2018](#)). At the surface, during desert dust events at Lampedusa, the longwave forcing has been computed to offset about 50% of the shortwave forcing ([di Sarra et al. 2011](#); [Meloni et al. 2015](#)). It has been estimated that longwave radiation scattering by desert dust contributes by 18% to the LW direct radiative forcing ([Sicard et al. 2014](#)). As discussed by [Gómez-Amo et al. \(2011\)](#) and [Gkikas et al. \(2018\)](#), while the shortwave forcing is more intense during day-time (SZA of 50 – 60°) and zero in nighttime, the longwave forcing acts continuously. The combination of the two may produce a modulation of the overall effect, producing time varying effects on the surface radiation budget and on the atmospheric temperature. Although estimations of the surface radiative effects over the Mediterranean Sea are scarce at this time, the results presented here clearly display a non-negligible surface dimming at different locations. In regards to such surface cooling, it appears now crucial to investigate its potential impact in terms of changes in regional Mediterranean climate. First, there is a need to better quantify how the change in solar irradiance at the sea/continental surface influences regional water cycles through modifications in evaporative, heat fluxes and SST. Secondly and since the percentage reduction in the UV and the visible region is as large as 10% to 20%, effects on terrestrial and marine biogeochemical cycles need to be examined. In particular, it should be

interesting to study how the change of solar irradiance at the sea surface influences carbon cycle and marine biological productivity through photosynthesis changes.

2.2 Aerosol direct forcing at TOA

Unless specified, results in this sub-section are for the shortwave domain. As reported in **Table 1** and **Fig. 1**, large differences are observed between the surface and top of the atmosphere forcing over the Mediterranean region. Concerning dust aerosols, the radiative transfer computations performed at Lampedusa show that the presence of dust increases the part of incident radiation that is scattered back to space but in magnitudes significantly lower (**Table 1**) than the surface energy losses. Similarly, [Saha et al. \(2008\)](#) indicate a diurnal direct TOA forcing of -7.7 W m^{-2} , which is significantly lower than the one estimated at the surface. At the regional scale and using the RegCM3 regional climate model, [Santese et al. \(2010\)](#) have indicated a daily-mean SW direct forcing for mineral dust outbreak of about $\sim -3 \text{ W m}^{-2}$ at top of the atmosphere. For some specific cases, model computation results put in evidence positive values of (SW) direct forcing of dust at TOA for extreme absorbing mixed dust particles ([Sicard et al. 2012](#)). [Di Biagio et al. \(2010\)](#) estimated a shortwave daily average RF at TOA at the equinox at Lampedusa. They found that the RF is about -14 W m^{-2} for desert dust, -5 W m^{-2} for polluted/biomass burning particles, and -4 W m^{-2} for marine-mixed aerosol types.

For polluted aerosols, results are quite similar and radiative transfer computations performed by [Roger et al. \(2006\)](#) indicate a mean TOA direct forcing four times lower compared to the surface one, consistently with computations performed by [Horvath et al. \(2002\)](#), [Markowicz et al. \(2002\)](#), [Meloni et al. \(2003\)](#) and [Saha et al. \(2008\)](#). Over the Crete island, [Vrekoussis et al. \(2005\)](#) indicated a radiative effect ranging from -12.6 to -2.3 W m^{-2} at TOA, for summer and winter (March 2001–June 2002), respectively. Over the Po Valley, where nitrate is an important source of aerosol emissions and over the Adriatic Sea, [Highwood et al. \(2007\)](#) report an instantaneous TOA aerosol forcing of $-4 \pm 2 \text{ W m}^{-2}$ over vegetation, and $-8 \pm 4 \text{ W m}^{-2}$ over ocean during the Aerosol Direct Radiative Impact Experiment (ADRIEX) project in late summer 2004.

[Bergamo et al. \(2008\)](#) indicate that anthropogenic particles produce a significant cooling effect over coastal and land sites of the central Mediterranean. At the TOA, the monthly average direct forcing is $-4 \pm 1 \text{ W m}^{-2}$ during spring-summer and $-2 \pm 1 \text{ W m}^{-2}$ during autumn-winter at the polluted sites. In parallel and based on MODIS Level 2 data in the eastern Mediterranean (Crete), [Benas et al. \(2011\)](#) report that anthropogenic aerosol forcing can reach values of -4 W m^{-2} at TOA (monthly mean instantaneous values).

Based on RegCM3 climatological computations, [Zanis et al. \(2012\)](#) have indicated a direct forcing of about -5 W m^{-2} over the Mediterranean for anthropogenic (sulfate + carbonaceous) aerosols from 1996 to 2007. Recently, [Drugé et al. \(2019\)](#) have proposed a first estimate of the nitrate direct forcing at TOA. On average over the period 1979–2016, it is found to be about -1.4 W m^{-2} over Europe. Lastly, [Formenti et al. \(2002\)](#) indicate that, over the sea, the SW radiative forcing is up to -64 W m^{-2} at the surface and up to -22 W m^{-2} at TOA for smoke aerosols. [Sicard et al. \(2012\)](#) have also documented smoke particles and showed significant instantaneous radiative forcing at TOA of about -13 W m^{-2} (at 17 UT) observed over Barcelona. Finally, [Di Biagio et al. \(2010\)](#) have reported a value of diurnal average forcing efficiency at the surface (and TOA) of -59.0 ± 4.3 (-19.2 ± 3.3) W m^{-2} for

urban-industrial plus biomass burning aerosols. During the EPL-RADIO campaigns (summer 2016–2017), [Sellitto et al. \(2020\)](#) reported a daily radiative forcing of about -4.5 W m^{-2} for volcanic aerosols 7 km downwind the Etna degassing craters.

[Di Biagio et al. \(2009\)](#) have highlighted a significant sensitivity of the direct radiative effects exerted by aerosols over the Mediterranean to the varying SSA of particles. Similarly to what happens at the surface, for desert dust the longwave effects compensates part of the shortwave radiative perturbation ([Santese et al. 2010](#); [Nabat et al. 2015](#); [di Sarra et al. 2011](#); [Meloni et al. 2018](#)). The dust longwave radiative forcing at TOA is generally positive. Measurements at Lampedusa show that on a daily basis over the Mediterranean the longwave effects offsets 26–35% of the shortwave radiation cooling ([di Sarra et al. 2011](#); [Meloni et al. 2015](#)). As shown by [Meloni et al. \(2015\)](#), the longwave radiation emission by dust may lead to a net cooling of the atmosphere at specific altitude ranges even in daytime.

Concerning primary sea-spray aerosols, [Nabat et al. \(2015\)](#) reported a mean direct radiative forcing (DRF) at the regional scale in the Mediterranean basin around -1 W m^{-2} at the surface and at the TOA using the CNRM-RCSM4 model (simulation covering the period from 2003 to 2009). In addition, the (daytime) DRF of marine aerosols has been estimated during summer 2010 over the eastern Mediterranean (clear-sky conditions) by [Mishra et al. \(2014\)](#) using satellite, in-situ observations and radiative transfer modeling. The direct radiative forcing exerted at the surface and at TOA are found to be between -5 and -10 W m^{-2} (wavelength range $0.25\text{--}20 \mu\text{m}$). In parallel, [Lundgren et al. \(2013\)](#) carried out a three-day computation (from July 24 to 26, 2006) over the whole Mediterranean basin with the COSMO-ART model. For clear-sky conditions and over the whole area (continental and oceanic surfaces), the solar DRF is about $-0.62 \pm 3.71 \text{ W m}^{-2}$ at the surface and $-0.47 \pm 3.21 \text{ W m}^{-2}$ at TOA. More recently, [Claeys et al. \(2017\)](#) reported higher direct forcings (at local scale) of $-11 \pm 4 \text{ W m}^{-2}$ at the surface and $-8 \pm 3 \text{ W m}^{-2}$ at TOA, during summer 2013. Marine aerosols also have the ability to interact with longwave radiation but few estimates are available over the Mediterranean basin. [Lundgren et al. \(2013\)](#) have reported an estimate of the LW forcing of $+0.19 \pm 1.08 \text{ W m}^{-2}$ at the surface and $+0.05 \pm 0.54 \text{ W m}^{-2}$ at TOA.

Figure 2 shows the seasonal-averaged SW DRE exerted by primary sea-spray particles at TOA as derived from the MACC reanalysis data ([Inness et al. 2013](#)) for 2006. Results indicate a significant seasonal variability with maximum occurring during spring and summer. During the summer (JJA) season, DRF can reach $\sim 6 \text{ W m}^{-2}$ over the eastern Mediterranean basin, notably due to the strong dry north winds over the Aegean Sea. During spring (MAM), important DRF is also observed over the western basin with values around -2 to -3 W m^{-2} . Compared to other aerosol regimes observed over the Mediterranean, the DRF due to marine particles is found to be similar to DRF exerted by anthropogenic aerosols. **Fig. 3** indicates yearly-mean (2006) DRF around -1 to -2 W m^{-2} at TOA for primary sea-spray consistently with estimates provided by [Nabat et al. \(2015\)](#). Compared to other natural aerosols, the DRF at TOA over the sea by marine aerosols is lower than the mineral dust solar forcing (~ -5 to -8 W m^{-2} ; **Fig. 3**).

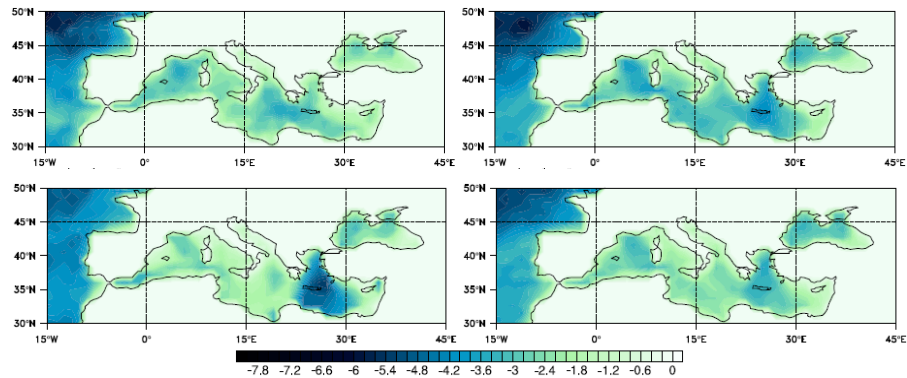


Fig. 2 Shortwave seasonal mean direct radiative forcing (W m^{-2}) exerted by primary sea-spray aerosols at TOA: **up left** December–February; **up right** March–May; **bottom left** June–August; **bottom right** September–November). MACC reanalysis data for the year 2006 from [Inness et al. \(2013\)](#)

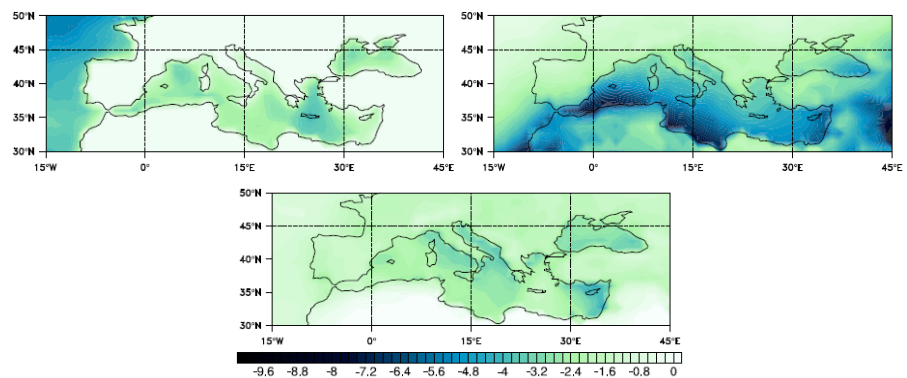


Fig. 3 Shortwave yearly-mean direct radiative forcing at TOA (W m^{-2}) due to various aerosols in the Mediterranean region: **up left** primary sea-spray; **up right** mineral dust; **bottom** anthropogenic aerosols. MACC reanalysis data for the year 2006 from [Inness et al. \(2013\)](#)

2.3 Trends in surface solar radiation over the Mediterranean Basin

In parallel to the direct radiative forcing, the surface solar radiation (SSR) plays a vital role for the life on Earth as it controls the surface energy balance, the meteorological and climatic conditions, the water cycle, the plant photosynthesis and carbon cycle and the diurnal and seasonal temperature variations ([Mercado et al. 2009](#); [Sanchez-Lorenzo et al. 2009](#); [Wild et al. 2013](#)). In addition to the greenhouse gas increase effect on the longwave radiative budget, the multi-decadal trends in solar radiation (global dimming/brightening phenomena) partly controls (in addition to the global climate) the regional climate change, but the Mediterranean Basin, as a whole area, has not been studied extensively. A recent work ([Kambezidis et al. 2016](#)) examined the evolution and trends of the surface net solar radiation. The solar dimming/brightening phenomenon is temporally and spatially analyzed over the Mediterranean basin based on MERRA (Modern Era Retrospective-Analysis for Research and Applications) datasets and showed an increase in the spatially-averaged SSR over the whole Mediterranean Basin of $+0.36 \text{ W m}^{-2}$ per decade during the period 1979–2012. The SSR for all-sky conditions exhibited an overall increase of $+2.1\%$ from 1979 to 2012 over the Mediterranean Basin in spring, while the respective variations in the other seasons were below 1%. However, statistically significant trends in SSR either for all-sky or clear-sky conditions were observed only in May ([Kambezidis et al. 2016](#)). The higher

increase in SSR in spring was common for all Mediterranean regions (west: 6.67°W–7°E, central: 7°E–21°E, east: 21–35°E), whereas the central part exhibited slight negative trends during autumn and winter. However, these tendencies were not statistically significant except for June (west part), May (central and east parts) and spring (east part). Annual-mean variations of SSR for all-sky conditions in the west, central and east Mediterranean regions, exhibited an increasing tendency, with a higher rate in the western region, which is statistically significant at 95% CL (+0.82 W m⁻² per decade). The other two regions reveal slight and not statistically-significant increasing trends in SSR. However, these trends presented less increasing rates in the 2000s, in accordance with the results obtained in south Europe (Sanchez-Lorenzo et al. 2015). In general, the trends were higher (more positive or less negative) over the western Mediterranean, except in spring when the eastern Mediterranean exhibited higher increasing rates in SSR. An overall negative trend in SSR over the Mediterranean Basin (2000–2007) was found by Hatzianastassiou et al. (2012) based on satellite data, mostly detected over the sea and the north African coasts, while positive trends were observed over the Iberian Peninsula and Balkan countries during 2001–2006. In addition, a solar brightening was observed after the late 1990s over Europe (Ohmura 2009; Wild et al. 2009) including Spain (Sanchez-Lorenzo et al. 2009), Greece (Zerefos et al. 2009) and Israel (Stanhill and Cohen 2009) and Italy (Manara et al. 2015 and 2016). The comparison between the results from the various studies indicates a large variation or even reversible signs in the SSR trends attributed to the different periods used in the analysis, the different stations or spatial domains that are involved and the various techniques, making the study of solar dimming/brightening phenomenon over the Mediterranean a really difficult task (Kambezidis et al. 2016).

At local/regional scales, long-term actinometric measurements verified the increasing tendencies in solar radiation computed via satellite observations and reanalysis. Recent analysis in Athens (Kambezidis et al. 2018) showed that the global solar radiation exhibited a trend of +0.40% per decade during 1992–2017. For the case of clear skies, the trends were found to be +2.38% per decade, implying also a declining trend in cloudiness. Furthermore, it was found that the solar radiation in Athens had a winter trend of –2.46% per decade and a summer trend of +1.91% per decade during 1992–2017 (Kambezidis et al. 2018), while Kazadzis et al. (2018) revealed a +1.5% per decade in SSR in Athens during the brightening period (1980–2012). It was also found that the diffuse irradiance exhibited a trend of –5.19% per decade during 1992–2017 (Kambezidis et al. 2018), while under clear skies, the trend became –6.77% per decade. The declining trend in the diffuse radiation implies decrease in aerosol amount, or in cloudiness or/even both over the study location.

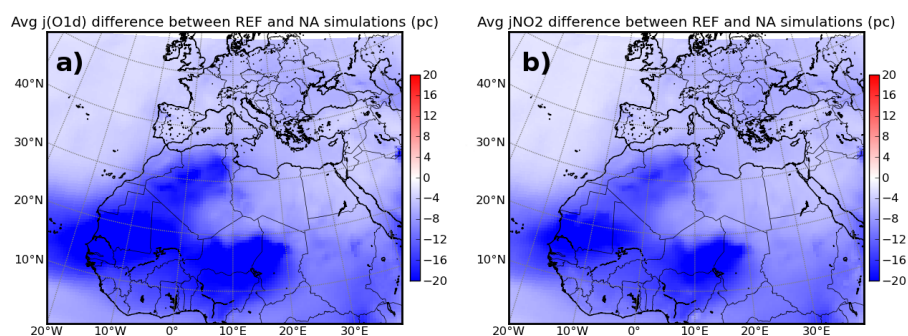
3. Implication for photochemical processes and solar energy production

3.1 Impact on photolysis

Impact of aerosols on photochemistry in the Mediterranean is a relevant question due to the presence of persistent, occasionally strong loads of aerosols over the area together with strong concentrations of tropospheric ozone over the Mediterranean Sea (Richards et al. 2013; Dulac et al. 2021). Balis et al. (2004) using lidar and total ozone observations at Thessaloniki, Greece found that aerosols can change by 10%–25% the UV irradiance thus being able to mask its changes due to total O₃ perturbations. The observational studies of Casasanta et al. (2011) and Gerasopoulos et al. (2012) have shown that the effect of aerosols on photolysis rates in this region is substantial. Both studies show a strong impact

of aerosol screening on two key photolysis rates, namely JO^1D and JNO_2 . From sunphotometer measurements at Lampedusa, [Casasanta et al. \(2011\)](#) have found a relatively linear effect of AOD at 416 nm on ground-level JO^1D , with a reduction of 62% in measured JO^1D for a unit AOD when the solar zenithal angle is 60° . From long-term measurements at the site of Finokalia, [Gerasopoulos et al. \(2012\)](#) found a substantial climatological effect of aerosols, from -6% in both JNO_2 and JO^1D when the aerosol load is low, down to -30% to -40% when the AOD exceeds 0.5, which is consistent with the findings of [Casasanta et al. \(2011\)](#). At the same location in the eastern Mediterranean [Benas et al. \(2013\)](#) identified maxima in the aerosol effect on JO^1D in springtime and autumn, corresponding to the season of maximal occurrence of mineral dust plumes in this basin. They computed daily surface JO^1D based on Terra MODIS aerosol optical depth data and total ozone MODIS and found a 13% decrease over a period of 11-years (2000–2010) at Finokalia station in the East Mediterranean, while daily effects of dust aerosol on JO^1D as high as 10% reduction were found. Note that a stronger impact of dust aerosols on the actinic flux and the photolysis rates has been recorded in Beijing by [Wang et al. \(2019\)](#).

The effect of aerosols on photolysis rates is nowadays relatively well-known and included in many chemistry-transport models such as Polyphemus ([Real and Sartelet 2011](#)) or CHIMERE ([Mailler et al. 2016](#)). It has been shown by [Mailler et al. \(2016\)](#) that the dominant aerosol species affecting photolysis rates at the station of Lampedusa during the ChArMEx/ADRI-MED campaign was mineral dust, a conclusion similar to that for Crete from [Gerasopoulos et al. \(2012\)](#). Mailler et al. (2016) show that including the effect of aerosols on photolysis rates reduces CHIMERE model errors in the calculation of these rates and considerably improves the ability of the model to reproduce the observed day-to-day variations of the photolysis rate of ozone. Including the aerosol effects on photolysis rates has a double-edged impact on tropospheric ozone, reducing ozone production through NO_2 photolysis but also reducing ozone destruction through ozone photolysis. Using a chemical box model, [Gerasopoulos et al. \(2012\)](#) calculated that a 24% and 5% reduction in JO^1D and JNO_2 noon values, respectively, results in 12% reduction in the mean diurnal net chemical production of O_3 at Finokalia. The results of [Mailler et al. \(2016\)](#) with the CHIMERE model for their six-week study period in June–July 2013 suggest that, as a result of these competing effects, (i) a slight reduction of ozone concentrations over the Mediterranean Sea, stronger over the east-west shipping highway that crosses the Mediterranean between the Suez Canal and the Gibraltar Strait, (ii) a reduction as well over the surrounding land in Europe, and (iii) an ozone concentration increase further south over the Saharan desert. According to these authors, the net effect of aerosols on ozone concentrations through modulation of the photolysis rates would be relatively weak, hardly more than 1 ppb on average during their study period (**Fig. 4**).



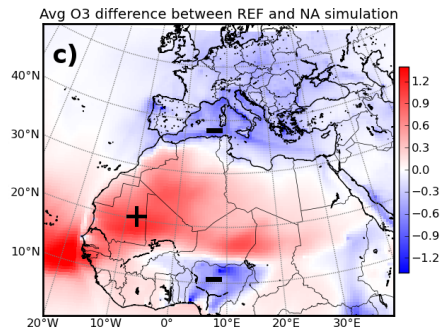


Fig. 4 CHIMERE simulations between June 6 and July 15, 2013, of: **a** Average effect of aerosols on JO^1D (in %); **b** Average modelled effect of aerosols on JNO_2 (in %); and **c** Average difference of low-level ozone concentration (in ppbv) due to the aerosol radiative effect on photolysis. Adapted from Mailler et al. (2016)

3.2 Impact of aerosol dimming on the solar energy production

The Mediterranean region is highly influenced by aerosols coming from different sources (e.g., Lelieveld et al. 2002), which affects the radiative budget (through the direct/indirect effect), climate, deposition and cloudiness in the region, so in the end, the amount of solar energy available at the surface. Despite of the fact of being a region with high solar resource due to its latitude, there are areas highly impacted by aerosols, like desert areas or some polluted areas in northern countries whose solar energy production potential could increase with a reduction of these anthropogenic aerosols emissions (Gutiérrez et al. 2018). Evaluation of the solar potential has been usually done using satellite-derived products (Sengupta et al. 2017), due to the lack of solar irradiance measurements at the surface. Satellite products have a high spatial resolution and have proven an accurate performance with low bias thresholds of errors (Posselt et al. 2012). However, aerosol retrieval from satellites perform worse in areas of high reflectivity, making the resource assessment more difficult in arid areas, which are abundant around the Mediterranean basin and are also sources of natural dust aerosols (Sengupta et al. 2017). The state-of-the-art approach for the evaluation of long-term solar resources consists of a solar irradiance retrieval algorithm from satellite, which is usually completed with a clear-sky model. The latter accounts mostly for the aerosols impact on solar irradiance. The AOD is the variable usually considered in these algorithms, and it is generally included, at a monthly resolution, which means that they cannot account for the interannual variability of aerosols (Ruiz-Arias et al. 2016; Sanchez-Lorenzo et al. 2017). Products with higher temporal resolution in aerosol input are becoming more frequent (Gschwind et al. 2019). A sensitivity of some satellite-based methods for solar irradiance to aerosols input was analyzed by Polo et al. (2015). An overestimation of 50% in AOD causes an error in the global horizontal irradiance of 3%–5% in southeastern Spain. Long-term trends in solar irradiance caused by an increase or reduction of aerosols (from anthropogenic sources) like the brightening period observed in Europe since the 1980's (Wild 2012; Nabat et al. 2014), has an impact on the potential PV power output. These trends can increase the resource assessment due to changes in solar irradiance of about ~3% by decade in areas of Central Europe. This change could be more for tilted panels and has an impact on the PV production estimates for the lifetime of a power plant (Müller et al. 2014; Gutierrez et al. 2018).

3.2.1 PV forecasting and aerosol impact over the Mediterranean area

In order to manage the photovoltaic production intermittency, an energy forecast needs to be made by power plant owners and operators of the systems. The forecast can be made at different time scales ranging from minutes to several weeks ahead and different methods are applied from statistical to physical models, usually depending on the forecasting horizon (Wild et al. 2015). Each model deals with aerosols in a different way (Inman et al. 2013). In the statistical methods, which are usually applied to a specific place, the availability of measurements of different atmospheric components at that location is important. Usually, some AOD satellite products, as well as aerosol type, are used but they are limited by their temporal resolution (Ruiz-Arias et al. 2016). On the other hand, numerical weather prediction models (NWP) that are accurate to forecast solar irradiance from intra-daily (beyond 4 h) to several days ahead (Perez et al. 2010), only include an AOD climatology, which has led to forecasting errors that generated big economic losses (Rieger et al. 2017). For instance, the dust outbreak of 14th April 2014 over continental Europe, ended in an overestimation of 5.3 GW of the PV electricity production in Germany (Rieger et al., 2017). The need to buy this energy at short notice in the intra-daily energy market caused a big economic damage. A combination of earth observation techniques and machine learning algorithms are applied to estimate the impact of particulate matter in Egypt (Kosmopoulos et al. 2018), finding a reduction in daily energy of more than 4 kWh m⁻² in a 10 MW plant, which can lead to important loss in daily revenues. Other studies have shown the impact on daily PV yield of different aerosol events around the Mediterranean area. In the Sahel zone, aerosols can potentially reduce the PV yield by 14%, and extreme events like dust storms can reach a -48% reduction (Neher et al. 2017). Other values are found in Romania, where the estimations show an impact of around 20% in PV energy produced due to different types of aerosols: volcanic ash, desert dust, biomass burning and urban aerosols (Calinoiu et al. 2013). In the western Mediterranean, extreme events of dust and smoke have caused a sporadic decrease in PV daily yield of 34% and 5%, respectively (Gómez-Amo et al. 2019). It can be noted that

3.2.2 Dust deposition on PV panels

Large scale PV projects are sometimes developed in arid and semi-arid areas, where a high concentration of atmospheric dust can be found. Apart from the reduction of solar irradiance from atmospheric aerosols, deposition (and accumulation) of dust over the optical surfaces also affects transmission and reflection of solar irradiance, causing a drop in the PV electricity production. Several review articles on these soiling effects have been carried out on this topic (Mani et al. 2010, Sarver et al. 2012, Costa et al. 2016). In general, the impact of soiling on a PV power plant depends on different factors, since the dust properties and the frequency of different dust episodes, to the rain events and the tilt of the panels. A wide range of reduction in cell performance have been reported in different studies for the Mediterranean (Mekhilef et al. 2012): 40% in a 6-month period in Saudi Arabia (Nimmo et al. 1979) or 32% in a 8-month period (Mani et al. 2010). In Egypt, 66% reductions in performance have been reported after six months exposure (Hassan et al. 2005) and in Algeria, 32-deg tilted panels have a 8% daily energy loss after several months (Semaoui et al. 2015). Other studies in Spain have presented average daily energy losses around 4% in a year and higher for long periods without rain (Zorrilla-Casanova et al. 2011). The accumulation of particles on the surface depends on the tilt of the panels, the rate of deposition and the removal due to enough wind speed. Strategies to prevent decrease in PV output and degradation due to soiling are needed.

3.2.3 Future PV projections and the role of aerosols

Planification of future photovoltaic power plants, made by different stakeholders of the industry, can take advantage of modeling tools like climate model projections instead of relying on historical data. In [Gaetani et al. \(2014\)](#), the impact of future trends in anthropogenic aerosol emissions on photovoltaic potential is shown using a global climate model (ECHAM5-HAM) with simulations between 2000 and 2030 for the scenario SRES B2. In general, the future reduction in aerosols results in an increase in the projected global warming. This indirectly results in an increase of PV productivity with more clear and less cloudy conditions in western Europe and eastern Mediterranean (10%). A reduction in PV productivity is projected in northern Africa due to augmented cloudiness ([Gaetani et al. 2014](#)). A positive trend has also been projected in Europe for the mid century, using CMIP5 models and the RCP8.5 scenario, partly due to a positive trend in clear-sky radiation that is likely related to a decrease in aerosol burdens ([Wild et al. 2015](#)). Different studies have also evaluated photovoltaic future potential in the Euro-Mediterranean area using higher resolution models (RCM) and they project an overall decrease in PV productivity over the Mediterranean area, more important in central and northern Europe. However, the limited representation of aerosols in the simulations makes it impossible to evaluate their impact on future PV potential and it adds uncertainty to the power projections presented. Nevertheless, regional climate models including evolving aerosols have simulated an increase in near-future PV potential, of up to +10% in summer in central and eastern Europe with uncertainty in magnitude related to the model ([Gutiérrez et al. 2020](#)).

4. Radiative forcing of tropospheric ozone

Tropospheric ozone is a secondary anthropogenic pollutant (see [Kalabokas et al. 2021](#), for a review of ozone studies in the Mediterranean region) and a short-lived greenhouse gas. The Intergovernmental Panel on Climate Change (IPCC) current best estimate for global mean tropospheric ozone radiative forcing (RF) over the industrial era is $0.4 \pm 0.2 \text{ W m}^{-2}$ with a 5%–95% confidence interval, making tropospheric ozone the third most important anthropogenic greenhouse gas after CO_2 and CH_4 ([Myhre et al. 2013](#)). This is also confirmed by the recent Copernicus Atmosphere Monitoring Service (CAMS) RF estimate for tropospheric ozone based on the CAMS Reanalysis ([Huijnen et al. 2020](#)), which is 0.32 W m^{-2} . The concentration and distribution of tropospheric ozone in the present-day are well constrained by satellite observations, resulting in robust estimates of the present-day radiative effect (RE) of tropospheric ozone ([Rap et al. 2015](#)). Estimates of present-day tropospheric ozone annual mean radiative effect using the TOMCAT-GLOMAP model are $1.17 \pm 0.03 \text{ W m}^{-2}$ globally and 1.63 W m^{-2} over the Mediterranean region (defined here as 28°N – 47°N , 9°W – 36°E) ([Rap et al. 2015](#); [Rowlinson et al. 2019](#)). The large uncertainty range in global mean RF (0.2 – 0.6 W m^{-2}) is primarily caused by the poor understanding of pre-industrial ozone concentrations, due to a lack of reliable PI measurements ([Myhre et al. 2013](#); [Stevenson et al. 2013](#)). Ozone is not stable in ice or snow, meaning proxy records are not available and although measurements of tropospheric ozone exist as far back as the late 19th, the accuracy and coverage of these early measurements is limited ([Volz and Kley 1988](#); [Cooper et al. 2014](#)). As well as anthropogenic sources, ozone precursors such as CH_4 , CO and NO_x have natural emission sources such as wildfires, wetlands, lightning and biogenic emissions. The role of these natural sources in the pre-industrial era is highly uncertain, introducing a large uncertainty when attempting to simulate tropospheric ozone in the PI atmosphere. [Checa-Garcia et al. \(2018\)](#) found that differences in pre-industrial estimates

between Coupled Model Intercomparison Project phase 5 (CMIP5) and CMIP6 cause an 8%–12% variation in ozone RF estimates but did not explicitly assess uncertainty in natural pre-industrial emissions. A recent analysis of oxygen isotopes in polar ice cores by [Yeung et al. \(2019\)](#) indicates that tropospheric ozone in the northern hemisphere increased by less than 40% between 1850 and 2005, suggesting that global mean tropospheric ozone RF is likely lower than the 0.4 W m^{-2} IPCC estimate.

Annual mean tropospheric ozone in the Mediterranean basin is characterised by high concentrations in the East Mediterranean, with lower ozone in the North Mediterranean and Adriatic Sea (**Fig. 5a**). Background tropospheric ozone concentrations in the eastern Mediterranean are amongst the highest in the world due to enhanced photochemical production ([Zanis et al. 2014](#)), stratospheric injection of ozone ([Akritidis et al. 2016](#)) and being at the “crossroad” of emissions from Europe, Asia and Africa ([Lelieveld et al. 2002](#); [Zerefos et al. 2002](#)). High solar intensity and cloud conditions mean the Mediterranean region exhibits a pronounced summertime maximum in tropospheric ozone concentration (**Fig. 5b**) ([Kanakidou et al. 2011](#); [Richards et al. 2013](#); [Zanis et al. 2014](#); [Kopanakis et al. 2016](#)). In-situ observations show that summer concentrations regularly exceed European Union (EU) target levels, up to 88% of the time over a 7-year period at the Akrotiri monitoring station in Greece ([Kopanakis et al. 2016](#)). [Richards et al. \(2013\)](#) found that tropospheric ozone in the Mediterranean region is sensitive to natural volatile organic compounds emissions, whereas surface ozone is most sensitive to local anthropogenic NO_x emissions.

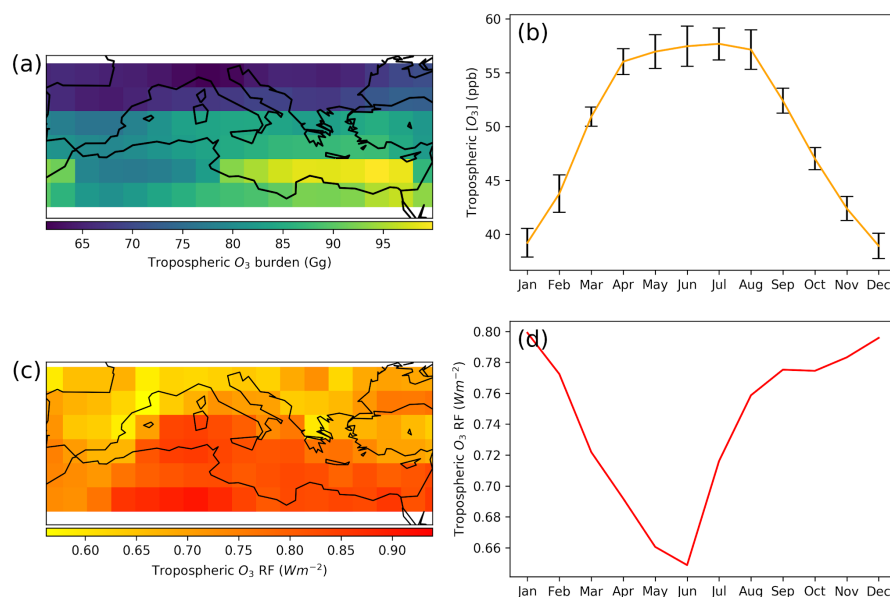


Fig. 5 TOMCAT-GLOMAP simulated tropospheric ozone in the Mediterranean region: **a** Annual mean tropospheric ozone burden (Mg m^{-2} averaged over 1997–2015); **b** Mean seasonal cycle of simulated mean tropospheric ozone concentrations (ppbv), with error bars showing one standard deviation of the mean; **c** Geographical distribution of the annual mean preindustrial to present-day stratospherically adjusted radiative forcing (W m^{-2}) of tropospheric ozone; **d** Seasonal cycle of the global mean pre-industrial to present-day tropospheric ozone radiative forcing (W m^{-2}). Adapted from [Rowlinson et al. \(2020\)](#)

Tropospheric ozone RF since the pre-industrial era is largest over the south and eastern Mediterranean (**Fig. 5c**), with a TOMCAT-GLOMAP model estimated Mediterranean

region mean of 0.74 W m^{-2} when using CMIP6 present-day and pre-industrial emissions (Rowlinson et al. 2020). Uncertainty in pre-industrial conditions (Hamilton et al. 2018; Rowlinson et al. 2020) means that this estimate might be in fact substantially lower. Using the TOMCAT-GLOMAP simulations from Rowlinson et al. (2020) we estimate this could be as low as 0.49 W m^{-2} when accounting for uncertainty in biogenic and biomass burning emissions in 1750. Strong localised forcing due to high tropospheric ozone production in the central and south-east Mediterranean may cause changes in atmospheric dynamics. Richards et al. (2013) showed that ozone in the upper troposphere, where ozone has the largest radiative effect, is most sensitive to global emissions rather than local sources, necessitating global action on emissions to mitigate the regional climate impact.

Interestingly, unlike the seasonal cycle of present-day regional tropospheric ozone concentrations (Fig. 5b), the tropospheric ozone RF over the Mediterranean region is larger in winter (Fig. 5d). Therefore, while the summer tropospheric ozone maximum is problematic for air quality, the climatic impact due to anthropogenic emissions is relatively larger during the winter months.

5. Conclusion and recommendations

The direct radiative effects exerted by aerosols and tropospheric ozone are now well documented at the local and regional scales by a large number of studies. Most of the direct aerosol effect estimates have been done in the solar spectral range. Based on the available literature, there is a clear consensus showing that Mediterranean aerosols exert a large and variable direct radiative effect at the surface and TOA. At the basin scale, this negative annual mean direct forcing is even larger in absolute value than the greenhouse gas positive forcing. However, the decrease in sulfate aerosol during the last decades has contributed by about 20%–25% to the surface warming in the Euro-Mediterranean region between 1980 and 2012 (Nabat et al. 2014).

The direct radiative forcing exerted by desert aerosols in the longwave spectral range has been demonstrated locally, but few studies have been carried out at regional and climatic scales. An effort could be made using updated optical properties (Di Biagio et al. 2020). In addition, the analysis of current decadal time series of dust data over the Mediterranean from research infrastructures and satellites (in particular CALIPSO) would be very informative.

A lot of studies report important differences between the surface and the TOA direct forcing, indicating that an important part of solar radiation is absorbed within absorbing aerosol layers (anthropogenic, mineral dust and smoke, typically), producing an important heating. Fig. 6 indicates that the yearly-mean atmospheric forcing due to anthropogenic particles can reach important values of about $5\text{--}10 \text{ W m}^{-2}$ over the Mediterranean basin. In that context, more meso-scale and RCM simulations are needed to quantify the semi-direct effect of natural/anthropogenic aerosols on the thermodynamic properties of the atmosphere and the circulation at regional and event/climatic scales. It appears also crucial to investigate more deeply the impact of the direct radiative effect of aerosols over both continental and marine surfaces on the Mediterranean climate, and more specifically the impact on humidity fluxes between the sea and the atmosphere and more largely the hydrological cycle. For this, coupled atmosphere-ocean models seem necessary because

they can account for changes in sea surface temperature and evaporation due to the aerosol dimming at the surface (Nabat et al. 2014 and 2015).

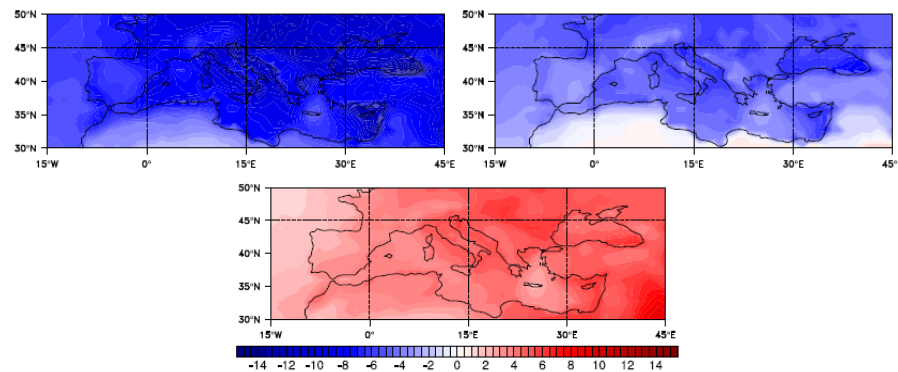


Fig. 6 Annual average shortwave direct radiative effect (in W m^{-2}) of anthropogenic tropospheric aerosols: **Top left** At the surface; **top right** At the TOA; and **bottom** Within the atmosphere. MACC reanalysis data for the year 2006 from Inness et al. (2013)

It would also be very interesting to study the effects of aerosol radiative forcing on air quality through the impact of absorbing polluted aerosols on the boundary layer development (Péré et al. 2011).

In parallel, investigating the possible link between the aerosol direct forcing and some characteristics of the Mediterranean heatwaves (intensity, duration), especially for mineral dust and anthropogenic particles, should also be a priority for future research given the expected regional climate change and positive aerosol feedback on heatwaves (Nabat et al. 2015). The aerosol radiative impact on PV production is an issue of recent concern, especially driven by the need of energy production and distribution networks to have accurate forecasts of the daily production yield. More studies dedicated to the impact of aerosols on solar energy production (by both reduction of surface solar radiations and wet/dry deposition) are needed in the context of the energy transition towards renewable energy sources, especially over the southern Mediterranean and islands.

The radiative effect of tropospheric ozone is strongest in the eastern Mediterranean during the summer maximum, but the influence of anthropogenic emissions is the greatest in winter, as the summer maximum is primarily driven by meteorological conditions. Further research is needed to understand the impact of strong regional forcing due to tropospheric ozone on atmospheric dynamics and chemistry, including how elevated ozone and OH production interacts with other pollutants. Studies should also focus on how effective future emissions reduction scenarios will be at decreasing tropospheric ozone in the Mediterranean basin, contrasting the projected changes in climate which may enhance ozone production and counteract mitigation measures.

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