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f the Greek extreme

Investigation of the effects of the Greek extreme wildfires of August 2021 on air quality and spectral solar irradiance

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Abstract. In August 2021, a historic heatwave was recorded in Greece which resulted in extreme wildfire events that strongly affected the air quality over the city of Athens. Saharan dust was also transferred over Greece on certain days of the same period due to the prevailing southern winds. The impact of these events on air quality and surface solar radiation is investigated in this study. Event characterization based on active and passive remote sensing instrumentation has been performed. The study shows that significantly increased levels of air pollution were recorded from the end of July to the first week of August. The smoke led to unusually high aerosol optical

depth (AOD) values (up to 3.6 at 500 nm), high Ångström exponent (AE) (up to 2.4 at 440–870 nm), and a strong and negative dependence of single-scattering albedo (SSA) on wavelength that was observed to decrease from 0.93 at 440 nm to 0.86 at 1020 nm, while the dust event led to high AOD (up to 0.7 at 500 nm), low AE (up to 0.9 at 440–870 nm), and a positive dependence of SSA on wavelength that was observed to increase from 0.89 at 440 nm to 0.95 at 1020. Furthermore, the smoke plume was also detected over the PANhellenic GEophysical observatory of Antikythera on 7 August, which is about 240 km away from Athens. Increased AOD values (up to ~ 0.90 at 500 nm) associated with a high fine-mode AOD (up to ~ 0.85 at 500 nm) and decrease in SSA with wavelength suggested the dominance of fine biomass burning aerosols. The impact of dust and smoke on solar irradiance revealed significant differences in the spectral dependence of the attenuation caused by the two different aerosol types. The attenuation of solar irradiance in the ultraviolet (UV-B) spectrum was found to be much lower in the case of dust compared to smoke for similar AOD₅₀₀ values. Differences were less pronounced in the near-infrared and visible spectral regions. The large AODs during the wildfires resulted in a decrease in the noon UV index by up to 53 %, as well as in the daily effective doses for the production of vitamin D (up to 50 %), in the daily photosynthetically active radiation (up to 21 %) and in the daily global horizontal irradiance (up to 17%), with serious implications for health, agriculture, and energy. This study highlights the wider impacts of wildfires that are part of the wider problem for Mediterranean countries, whose frequency is predicted to increase in view of the projected increasing occurrence of summer heatwaves.

1 Introduction

Climate change is becoming a harsh reality and leading to climate problems, one of which is the increased frequency of occurrence of large-scale wildfires around the globe which affect both the environment and human life (Weilnhammer et al., 2021). Wildfires lead to the loss of land vegetation; worsen air quality; and affect ecosystems, societies, economies, and climate (Jaffe et al., 2013; Jolly et al., 2015). There have been concerns about the frequency of occurrence of such events in the recent past (Ganor et al., 2010; Forzieri et al., 2017). Extreme weather events like severe heatwaves (Perkins-Kirkpatrick and Lewis, 2020; Fischer et al., 2021), which are more prominent in southern and southeastern Europe (Giorgi and Lionello, 2008; Fernandez et al., 2015; Forzieri et al., 2017; Weilnhammer et al., 2021), act as fuel for other extreme events like wildfires. The probable causes of ignition of wildfires can be categorized as lightning-induced and human-caused.

A wildfire event leads to a sudden rise in particulate matter (PM) and gaseous pollutants, such as nitrogen oxides, carbon monoxide, greenhouse gases, and volatile organic compounds (Andreae and Merlet, 2001; Knorr et al., 2017; Fernandes et al., 2022). Among these, greenhouse gases having longer lifetimes impact the global climate, while aerosols having short lifespans mainly have regional and local effects. The long-range transport of wildfire smoke can lead to a change in the chemical composition of the plume and also affect the local and regional air quality upon the planetary boundary layer entrainment (Colarco et al., 2004; Pani et al., 2018; Wu et al., 2021). Following a wildfire, the poor air quality due to the high gaseous and particulate pollutant levels imposes serious threats of asthma, respiratory diseases, cardiovascular effects, and lung cancer via human

inhalation exposure (Manisalidis et al., 2020; Rice et al., 2021; Andreadis et al., 2022). Biomass burning also reduces the amount of solar radiation, as presented in the study by Rosário et al. (2022) that showed that the mean drop in solar radiation was up to 200 W m⁻². In another study by Park et al. (2018), it was found that smoke significantly reduces the ultraviolet (UV) actinic flux, while the spectral light absorption by smoke aerosols depends upon the combustion conditions, smoke chemical properties, and atmospheric processing (Saleh et al., 2013; Srinivas et al., 2016). Moreover, Arola et al. (2007) found that biomass burning aerosols led to a decline of about 35 % in surface UV irradiance, while the reduction was comparatively smaller for total solar radiation.

The Mediterranean is considered a "climate change hotspot" (Founda et al., 2022; Zittis et al., 2022) due to its faster warming rates, as compared to the global average, as well as an increase in the frequency of heatwaves followed by forest fires and prolonged droughts. The Mediterranean region is also susceptible to increased aridity as a result of climate change (IPCC, 2022; Turco et al., 2018; Guiot and Cramer, 2016). The occurrence of fires leads to water stress that in turn reduces the post-fire vegetation recovery (Puig-Gironès et al., 2017; Cruz and Moreno, 2001; Pratt et al., 2014; Vilagrosa et al., 2014; Pausas et al., 2016), leading to an expansion of shrublands as a combined effect of fire and drought (Batllori et al., 2017, 2019; Baudena et al., 2020). Previous publications in the Mediterranean have analysed aerosol optical properties and mixtures due to seasonal forest fires. Castagna et al. (2021) analysed the 2017 summer wildfires in the region of Calabria (southern Italy), which resulted in the largest burnt area of the last decade (2008-2019), estimated to be more than 1679 ha of forests and shrubland. The impact of the wildfires on air quality, ecosystems, and human health was analysed using carbon monoxide and black carbon (BC) measurements at the high-altitude station of Monte Curcio (39.32° N, 16.42° E). In Gómez-Amo et al. (2017), the authors studied two wildfires in Spain that occurred near Valencia during 29–30 June 2012 affecting 48 500 ha of land.

On the other hand, the summer season in the Mediterranean region witnesses frequent dust activities, and the dust accumulation is favoured by stable weather conditions due to the absence of precipitation and depressions (Nastos, 2012). The eastern Mediterranean region has a marked seasonal cycle of the occurrence of the Saharan dust events, which maximize in spring (mainly) and summer (Moulin et al., 1998; Rodríguez et al., 2001; Fotiadi et al., 2006; Meloni et al., 2007). In Papayannis et al. (2009), the authors presented a statistical analysis of Saharan dust for a 3-year period between 2004 and 2006 over Athens, Greece, and they found that the Saharan-dust-related aerosol layers were prevalent for 79 d. In Marinou et al. (2017), the authors showed that Saharan dust layers arrive above Greece in altitudes between 2-6 km in spring (mean dust extinction coefficient values of \sim 70 Mm⁻¹), between 3–6 km in summer (\sim 50 Mm⁻¹), and between 2–5 km in autumn (\sim 40 Mm⁻¹). Recently, Soupiona et al. (2018) presented a statistical analysis of Saharan dust events over Athens for a 16-year period between 2000 and 2016. They found that the dust layers arrive over Athens, between 1-6 km a.s.l., with the number of these events being the highest in spring, summer, and early-autumn periods, and that during spring the dust layers were moved at higher altitudes than in other seasons. Saharan dust effects in various sectors including health, aviation, and solar energy have been presented in Monteiro et al. (2022) and references therein. In particular, extreme dust events can attenuate the global horizontal irradiance (GHI) by as much as 40 %-50 %, while a much stronger attenuation was recorded in the direct normal irradiance (DNI) (by 80 %-90 %); spectrally, this attenuation is distributed as 37 % in the UV, 33 % in the VIS, and around 30 % in the infrared regions (Kosmopoulos et al., 2017). Also, Papachristopoulou et al. (2022) showed that for the eastern Mediterranean the average attenuation of dust in GHI and DNI using a 15-year climatology is $\sim 3\%$ and ~ 10 %, respectively.

The wildfires of summer 2021 in Greece were the most severe in the decade, signifying a conflagration period of about 20 d in August, and were triggered by severe and prolonged heatwaves, as discussed in a few recent studies. The study by Founda et al. (2022) showed that the heatwave of 2021 was intense and persistent with the highest observed nighttime temperatures and cumulative heat, which were also intensified due to the urban heat island effect in Athens. A study of the Varympompi wildfire of 2021, in the northern suburbs of Athens, by Giannaros et al. (2022), showed that it was characterized by an unusual spread of fire followed by massive spotting as well as pyroconvection influence. The development of pyroconvection and ignition was supported by dry and hot conditions that began emerging in late June that deteriorated further, reaching its peak in July and resulting in the wildfires. The meteorological conditions also supported the event including a lack of significant precipitation and higher-than-average temperatures. Another study by Papavasileiou and Giannaros (2022) analysed the pyroconvection using satellite data and found that there was a presence of pyrocumulus and pyrocumulonimbus for many hours during the severe fire events.

The increase in the frequency of occurrence of these extreme wildfire events enhances the necessity of a more indepth understanding of these phenomena and their impact on various domains. The analysis presented in this study focuses on such wildfire events that were prevalent in August 2021 around the city of Athens. The study aspires to better analyse the wildfire smoke and simultaneous dust activity from in situ, remote sensing, and modelling data and to analyse their respective and combined impact on spectral solar irradiance. The datasets used were collected from Athens, during the ASPIRE (Atmospheric parameters affecting SPectral solar IRradiance and solar Energy) (ASPIRE, 2022) campaign, and from the PANhellenic GEophysical observatory of Antikythera (PANGEA) of the National Observatory of Athens (NOA). The ASPIRE campaign was designed with the objective to investigate the effect of clouds, aerosols, water vapour, and absorbing trace gases on spectral solar irradiance and contributes to interdisciplinary aspects. Wildfire events during this campaign allowed for the in-depth investigation of atmospheric composition and its impact on the transfer of solar radiation using active and passive remote sensing instruments.

The aim of this work is to analyse the spatial and temporal spectral aerosol optical properties during the August 2021 wildfires in Athens and their effects on surface solar radiation. More specifically, the main objectives are (1) to discuss the effect of the smoke and dust events on air quality, (2) to show how observations from different sensors can be combined to identify and study such events, (3) to study aerosol optical and microphysical properties during the events, (4) to investigate the transport of the smoke plume and its characteristics to the PANGEA observatory in Antikythera, and (5) to analyse the contribution of dust and biomass burning aerosols to the attenuation of spectral surface solar radiation over Athens. This paper is organized into four sections. Section 2 deals with the observational data and the methodology and is followed by Sect. 3, which presents the results and discussions; finally, Sect. 4 summarizes the findings of this study.

2 Data and methodology

For a better understanding of the August 2021 wildfires in Greece, ground-based measurements, satellite images, and radiative transfer modelling are used synergistically. This section deals with the description of the datasets used in this work, as well as the methodology followed to study the wildfire event.

2.1 Ground-based measurements

Data that were collected during the intensive ASPIRE campaign have been used for the study. In addition to the instruments that are permanently installed and operating at the actinometric station (for observations and measurements of solar radiation) of NOA (ASNOA) (located at Thissio, in the centre of Athens; 37.97° N, 23.72° E; 107 m a.s.l.), new instruments were installed in the context of the ASPIRE campaign. To study the impact of wildfires in Athens, measurements that were performed at the Biomedical Research Foundation of the Academy of Athens (BRFAA), Greece (37.99° N, 23.78° E; at approximately 180 m a.s.l.; located in a green area 4 km from ASNOA), and at the National Technical University of Athens (NTUA) (37.96° N, 23.78° E; at approximately 212 m a.s.l.; about 3 km from ASNOA) were also used. In situ air quality measurements were also used, obtained from the stations of the Greek National Air Pollution Monitoring Network (GNAPMN) in Athens. From the PANGEA observatory (35.86° N, 23.29° E; 110 m a.s.l.), which is located on the remote island of Antikythera, measurements collected at the NOA aerosol remote sensing facility have been used. Details on the equipment and measuring sites are provided in Table 1.

2.1.1 Air quality

We have analysed air quality data for the greater Athens area (GAA) from the GNAPMN. More specifically, we analysed daily averages of particulate matter concentrations (PM₁₀, PM_{2.5}), as well as hourly concentrations of nitrogen oxides (NO, NO₂), for the period July–August 2021 at 11 sites. Since data of NO and NO₂ are provided on an hourly basis and not on a daily basis as the PM data, we calculated daily mean concentrations of NO and NO₂ when at least 12 hourly measurements were available. Analytical information about the stations contributing data to the GNAPMN is provided by Grivas et al. (2008). Columnar NO₂ from Pandora (operating at ASNOA) (Herman et al., 2009) and an MKIV, a single monochromator Brewer spectrophotometer (Brewer no. 001) (Kerr, 2010; Kerr et al., 1985) (operating at BRFAA), were used in this study. Spectral measurements from the Brewer instrument were also used to retrieve the total column SO2 and the total column ozone. Details about the instruments, calibration, and uncertainties are provided in Appendix A.

2.1.2 Aerosol properties

The Athens NOA AERONET (Aerosol Robotic Network) station was operating from 2008 to 2021, with a CE318 sun/sky photometer from Cimel Electronique (Cimel no. 440) in operation during the study period. The columnar aerosol optical depth (AOD), Ångström exponent (AE), fine/coarse AOD, single-scattering albedo (SSA), and volume size distribution (VSD) (Giles et al., 2019; O'Neill et al., 2003; Dubovik and King, 2000), retrieved from the AERONET Version 3 algorithm (Giles et al., 2019), are used here. For Athens, Level 2.0 AERONET direct sun products (AOD, AE, fine/coarse AOD) were used in this study except for the days with very high smoke and/or dust aerosol load (4, 5, 7, 11, 18, and 19 August), when Level 1.0 data were used. For these particular days, the AERONET automatic cloud-screening algorithm filtered out data related to the wildfire plumes, when going from Level 1.0 (unscreened) to Level 1.5 (cloud-screened) and Level 2.0 (cloud-screened and quality-assured) products, due to the very high temporal variations in AOD. The sun photometer measurements during high-aerosol events with extremely frequent changes in the radiation field are difficult to be captured due to a cloudflagging algorithm failure and are more likely to be rejected as cloudy, even in cloud-free situations (Evan et al., 2022). Manual control of sky-camera (SKYCAM) images from the cloud camera was used as additional evidence for non-cloud presence regarding the choice of the Level 1.0 products to be used (Appendix Fig. A1). Accordingly, Level 2.0 inversion products (SSA, VSD) were used except for the days mentioned above (where Level 1.5 data with a sky error limit up to 5 % and solar zenith angle (SZA) of $> 45^{\circ}$ were used with additional filtering of coincident AOD at 440 nm of > 0.4for SSA similar to the AERONET quality assurance criterion for Level 1.5 to rise to Level 2.0, except for SZA for which we have used 45° instead of 50° ; Holben et al., 2023) since the strict criteria for Level 2.0 filters out a lot of useful retrievals in summer months, as explained in Kazadzis et al. (2016). The approach of using lower-level data increases the uncertainty in the retrievals, but the evidence of the collocated data from other sources provides a relatively high degree of data quality assurance. Also, the climatological values of the aforementioned properties reported in previous studies (Raptis et al., 2020) are used as a reference. We note here that measurements from the NTUA AERONET station operating in Athens since January 2021 (CE318 sun/sky photometer) were used for 7 August when the data in morning hours were not available from the Athens NOA AERONET station. The same parameters (AOD, AE, fine/coarse AOD, and SSA) were also collected at the PANGEA observatory. For PANGEA, Level 2.0 products were used for both direct sun and inversion products, as there was not much difference in Level 1.0 and higher-level products, as was the case of Athens.

In addition to the columnar optical properties, in situ measurements of spectral scattering and absorption coefficients measured at the Thissio Air Monitoring Station by means of integrated nephelometer (TSI 3564) and aethalometer (AE-33) instruments were analysed. Nephelometer measures the spectral scattering coefficient ($b_{sca,\lambda}$) at three wavelengths (450, 550, and 700 nm). Aerosol absorption was computed via AE-33 measurements at seven wavelengths (370, 470, 520, 590, 660, 880, and 950 nm), while the instrument also provides the BC concentrations and through the "aethalometer model" the fractions of BC related to biomass (or wood)

Quantity	Instrument name/network	Location	Temporal/spectral resolution	Туре	Reference	
PM ₁₀ , PM _{2.5}	Eberline, Thermo Fisher/GNAPMN*	GAA*	Daily	Beta gauge partic- ulate monitor	Grivas et al. (2008)	
NO, NO ₂	Horiba Ltd./ GNAPMN	GAA	Hourly	APNA analyser	Grivas et al. (2008)	
Scattering coefficient	TSI/NOA network	ASNOA*	3 wavelengths (450, 550, 700 nm)	Nephelometer	Kaskaoutis et al. (2021)	
Absorption coefficient	Magee/ NOA network	ASNOA	1 min/7 wavelengths (370, 470, 520, 590, 660, 880, 950 nm)	Aethalometer	Drinovec et al. (2015), Liakakou et al. (2020)	
Columnar NO ₂	Pandora/Pandonia	ASNOA	10–20 min	Spectral radiome- ter	Herman et al. (2009)	
Columnar SO ₂	Brewer/EuBrewNet	BRFAA*	-	Spectrophotometer	Kerr (2010)	
AOD*, Ångström exponent	Cimel/AERONET	NOA*, PANGEA*	15 min	Sun photometer	Giles et al. (2019)	
Fine/coarse AOD	Cimel/AERONET	NOA, PANGEA	500 nm	Sun photometer	O'Neill et al. (2003)	
Single-scattering albedo	Cimel/AERONET	NOA, NTUA*, PANGEA	440, 675, 870, 1020 nm	Sun photometer	Dubovik and King (2000)	
Volume size distribution	Cimel/AERONET	NOA	-	Sun photometer	Dubovik and King (2000)	
Backscatter coefficient	Vaisala/E- PROFILE, Polly ^{XT} /EARLINET*	ASNOA PANGEA	910 nm, 1064 nm	Ceilometer, Lidar	Kotthaus et al. (2016), Engelmann et al. (2016), Baars et al. (2016)	
Spectral irradiance	PSR*/NOA network	ASNOA	300–1020 nm	SR	Gröbner and Kouremeti (2019)	
UV-B irradiance	Brewer/EuBrewNet	BRFAA	290–315 nm	Spectrophotometer	Garane et al. (2006)	
Global horizontal irradiance, diffuse horizontal irradiance	Kipp & Zonen/ NOA network	ASNOA	285–2800 nm	Pyranometer	WMO (2021)	
Near-infrared irradiance	PSR/NOA network, Kipp & Zonen/ NOA network	ASNOA	700–3000 nm	SR, Pyranometer	Gröbner and Kouremeti (2019), WMO (2021)	
Erythemal irradiance	Brewer/EUBrewet, PSR/NOA network	BRFAA, ASNOA	300–400 nm	Spectrophotometer, SR	Kerr (2010), Gröbner and Kouremeti (2019)	
Vitamin D dose	Brewer/EuBrewNet, PSR/NOA network	BRFAA, ASNOA	300–400 nm	Spectrophotometer, SR	Kerr (2010), Gröbner and Kouremeti (2019)	

Table 1. Description of ground-based measurements.

* AOD (aerosol optical depth), ASNOA (actinometric station of NOA), BRFAA (Biomedical Research Foundation of the Academy of Athens), EARLINET (European Aerosol Research Lidar Network), GAA (greater Athens area), GNAPMN (Greek National Air Pollution Monitoring Network), NOA (National Observatory of Athens), NTUA (National Technical University of Athens), PANGEA (PANhellenic GEophysical observatory of Antikythera), PSR (Precision Solar Spectroradiometer), SR (spectral radiometer).

burning (BC_{wb}) and fossil-fuel combustion (BC_{ff}) (Liakakou et al., 2020). Quality-controlled aerosol scattering, absorption, BC, and SSA values at Thissio are available on an hourly basis (Kaskaoutis et al., 2021), while daily averaged values are used in this study (1–20 August 2021). A Vaisala CL31 ceilometer installed at ASNOA provides information on the vertical distribution of the aerosol extinction coefficient (Kotthaus et al., 2016) and is part of the European Meteorological Network's programme "E-PROFILE" (ALCProfile, 2022).

2.1.3 Clouds

The Q24M Mobotix all-sky imager (ASI) was installed at ASNOA for observing the atmospheric conditions in Athens in the context of the ASPIRE campaign, which operated from December 2020 to September 2022, and provided images with a temporal resolution of 10s. Such ASIs can be employed for performing cloud detection and characterization (Kazantzidis et al., 2012) and/or retrieving aerosol properties (Cazorla et al., 2009; Román et al., 2022; Kazantzidis et al., 2017). Kazantzidis et al. (2017) proposed a methodology for retrieving AOD at 440, 500, and 675 nm using RGB channels of the ASI, the sun saturation area (a feature extracted from ASI images representative of AOD magnitude), and solar zenith position as inputs in a machine learning algorithm. In this study, the AOD from the ASI at 500 nm was analysed and compared with the AOD from the Cimel instrument at NOA. The ASI images have also been used to separate clouds from wildfire smoke, augmenting the AERONET datasets with cases erroneously characterized as clouds by the automated cloud-screening algorithm. At the PANGEA observatory, the Polly^{XT} lidar at NOA (part of the European Lidar Network, EARLINET, 2022; part of the Raman and polarization lidar network, POLLYNET, 2022) is installed. The Polly^{XT} lidar at NOA (Engelmann et al., 2016; Baars et al., 2016) is a multi-wavelength Raman-polarization system with 24/7 operational capabilities, which provides vertical distributions of the particle backscatter coefficient at 355, 532, and 1064 nm; the extinction coefficient at 355 and 532 nm; and the particle depolarization ratio at 355 and 532 nm, in altitudes from 0.2 up to 15 km above the surface. With these observations and using well-known methodologies, we can separate between aerosols and clouds, spherical and non-spherical particles in mixed aerosol layers (Tesche et al., 2009; Marinou et al., 2019), and absorbing and non-absorbing aerosols towards aerosol characterization and aerosol-cloud separation (Baars et al., 2017). Using the aforementioned parameters, we identified the times and altitudes where cloud-free smoke layers were observed above the PANGEA observatory, and we use these measurements as a complementary dataset in this study.

2.1.4 Solar irradiance

The Precision Solar Spectroradiometer (PSR) no. 007, operating at ASNOA since 2016, is a high-precision and highaccuracy state-of-the-art spectroradiometer (details are provided in Appendix A). It measures irradiance in the spectral range of 300-1020 nm with an average step of 0.7 nm and spectral resolution in the range of 1.5-6 nm (depending on the measured wavelength) (Raptis et al., 2018; Gröbner and Kouremeti, 2019). The total UV radiation has been calculated by integrating the PSR measurements in the range of 290–400 nm, while UV-A constitutes the integral in the range of 315-400 nm. The photosynthetically active radiation (PAR), which is equivalent to the visible (VIS) radiation, constitutes the integrated radiation in the spectral range of 400-700 nm (Poorter et al., 2019). The uncertainty budget of the instrument is less than 1 % in VIS, less than 1.7 % in UV-A, and higher than 2% in UV-B (Gröbner and Kouremeti, 2019).

Measurements of the global solar-spectral UV-B irradiance (integrated at 290–315 nm) from the Brewer (see Appendix A), which are available with a temporal resolution of about 30 min, were also analysed. In addition, measurements of the broadband GHI and diffuse horizontal irradiances have been also used. These measurements were performed by the two Eppley precision spectral pyranometers (S/N: 26069, 26070) that have been operating at ASNOA since 1986 (details are provided in Appendix A).

Near-infrared irradiance (700-3000 nm) was calculated from the difference between the GHI measurements from the pyranometer and the calculated integral of the PSR measurements in the 290-700 nm spectral range. The erythemal irradiance was calculated as the product of the UV spectra measured by the Brewer and the PSR with the action spectrum proposed by the International Commission of Illumination (ISO/CIE, 2023; McKinlay and Diffey, 1987; Webb et al., 2011). The effective dose for the production of pre-vitamin D3 in the human skin (hereafter referred as the vitamin D dose) was calculated similarly to the erythemal irradiance but using the respective effective spectrum (Bouillon et al., 2006). The spectral extension correction technique proposed by Fioletov et al. (2003) was used to calculate erythemal irradiance from the Brewer (which measures up to 325 nm, while the erythema effective spectrum extends up to 400 nm). A similar method was used for the calculation of the vitamin D doses from the Brewer. For vitamin D and PAR, we are interested in the cumulative daily dose (since their effects depend on the overall dose that a human or a plant, respectively, gets), while for erythema, we are interested in dose rates around local noon, when solar radiation is higher.

2.2 Satellite and reanalysis data

2.2.1 Copernicus Atmosphere Monitoring Service (CAMS)

The Copernicus Atmosphere Monitoring Service (CAMS) reanalysis product (Inness et al., 2019) was used to identify the dominant aerosol types over Athens during August 2021. Total AOD, dust AOD, and organic matter AOD at 550 nm were collected and analysed for a 2×2 pixel area centred over Athens for a month period in August 2021. The CAMS data are available at an interval of 3 h on a regular longitude–latitude grid ($0.75^{\circ} \times 0.75^{\circ}$) and is retrieved using the Copernicus Atmosphere Data Store (ADS, 2022).

2.2.2 Meteosat Second Generation (MSG)

The Spinning Enhanced Visible and InfraRed Imager (SE-VIRI) instrument on board geostationary Meteosat Second Generation (MSG) satellites of EUMETSAT provides full earth disc data at different channels every 15 min. In this analysis, the European high-resolution visible cloud RGB product was utilized, which is a product based on the highresolution visible and IR10.8 SEVIRI channels. These data are advantageous for cloud monitoring in high resolution. The images were analysed for August 2021 in order to identify the events, the initiation of the wildfires, and the smoke plume transport.

2.3 Modelling

2.3.1 Spectral surface solar radiation

The disort (DIScrete Ordinate Radiative Transfer) pseudospherical approximation (Buras et al., 2011) of the UVSPEC radiative transfer model that is included in the libRadtran v2.4 package (Emde et al., 2016) was used to simulate the spectral solar irradiance in the range of 290–3000 nm. Radiative transfer simulations were performed for August for the coordinates of the actinometric station of Thissio with a temporal resolution of 15 min. The libRadtran simulations were performed for three different groups of inputs.

- Case a. In the first case, the simulations were performed for the UV region with SSA = 0.85 and for the VIS and near-infrared (NIR) regions with SSA = 0.95. The inputs included AOD (at 340 nm for UV and at 500 nm for wavelengths above 400 nm), AE (440–675 nm), and the total column of water vapour (WV) that were obtained from the Cimel instrument at NOA. The Cimel measurements were interpolated to the time of the simulations (i.e. for all of August with a step of 15 min).
- Case b. In the second case, climatological values of AOD (at 340 nm for UV and at 500 nm for wavelengths above 400 nm), AE (440–675 nm), SSA (average of SSA at 440 and 675 nm), and the total column of WV

were used, which were derived by analysing Cimel measurements during 2008–2018 (Raptis et al., 2020).

Case c. The third case uses AOD = 0 and the total column of WV from the Cimel instrument as inputs.

The total column of ozone from the Brewer was also interpolated to the time of the simulations and was used as input in all the three cases. A default concentration of 420 ppb was assumed for CO_2 . Surface albedos used were for 0.05 and 0.2 in UV and VIS, respectively. In all these cases, simulations were performed for cloudless conditions assuming climatological profiles of atmospheric molecules corresponding to mid-latitude summer (Anderson et al., 1986) and climatological profiles of the aerosol optical properties (Shettle, 1989). The extraterrestrial spectrum proposed by Kurucz (1994) was used for the simulations.

2.3.2 Aerosol source and transport

For analysing the source and transport of the wildfire plumes, the Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model (Stein et al., 2015) was employed that uses a hybrid of Lagrangian and Eulerian approaches. HYS-PLIT is used over the regional to global scale to account for the transport of pollutants, their dispersion, and deposition. In this analysis, 72 and 24 h backward trajectories ending at 12:00 UTC for Athens and PANGEA, respectively, were generated using Global Data Assimilation System meteorological data at seven levels varying from 0.5 to 3.5 km, with an interval of 0.5 km above ground level.

3 Results

3.1 Description of the event

A series of wildfires that severely affected Athens occurred at three locations, namely Varympompi, North Evia, and Vilia, as shown in Fig. 1. The first source was ~ 25 km north of Athens, near Varympompi, and affected the air quality from 4 to 9 August with about 8370 ha of area burnt. The second fire source was at a distance of ~ 190 km, in the northern part of the Evia Island that led to the worsening of air quality from 3 to 11 August and with a burnt area of about 51 000 ha. Another fire source that affected the air quality in Athens from 17 to 19 August was in the northwest at a distance of ~ 50 km from Vilia, with a resulting burnt area of 9400 ha. Giannaros et al. (2022) reported that a total area of 94 000 ha was burnt collectively by five large wildfires in 2021 in Greece.

According to NOA records, the summer (June–August) average temperature in Athens over the climatic period 1991–2020 was found to be around 28.5 °C, and the average daily maximum temperature was about 34 °C over the same period (Fig. A2a in Appendix A). However, the period from the end of July to the beginning of August 2021 was marked by a very high temperature surge, with positive air



Figure 1. (a) Map for the wildfire source sites (in red) and the study region (in white). Base map credits: © Google Earth 2022. Identification of the event on 5, 11, and 18 August using (b) Meteosat Second Generation (MSG) and (c) sky-camera images.

temperature anomalies of the order of 10 °C, compared to the long-term average (34 °C) (Founda et al., 2022). Moreover, the relative humidity in the same period was observed to be well below its climatic value (summer average humidity from 1991 to 2020, Fig. A2b). Apart from the temperature and relative humidity, the maximum wind speed from the end of July to early August was found to be around 5.4 m s^{-1} , well below 5 on the Beaufort scale $(8.0-10.7 \text{ m s}^{-1})$ (Fig. A2c and d in Appendix). Yet, total precipitation in Athens from March to July 2021 was found to be about 75 % lower than its climatic value (Founda et al., 2022). Such meteorological conditions characterized by warmer-than-average temperatures, extremely dry air, low wind speed, and a precipitation deficit served as the preconditions for the burning of the available fuel and then conversion into massive wildfires. In Giannaros et al. (2022), the authors found that warmer-thanaverage temperatures and a lack of precipitation catering to the two prolonged (greater than 10 d) heatwaves led to the efficient drying of the fuel until the ignition time, creating a highly flammable fuel. Also, the hot and dry atmospheric layer near the surface helps in maintaining intense burning, as well as the upthrust of the plume.

The spread of wildfire smoke was investigated using the MSG images (15 min frequency), while the presence of smoke over Athens was confirmed by visually inspecting SKYCAM images (available with a frequency of 10 s). Figure 1b presents satellite images from MSG on specific days/hours, which along with the respective sky-camera images (Fig. 1c) for Athens, confirm the presence of smoke in the region. In addition to the prevalent smoke due to wildfires, August 2021 also experienced episodes of dust, as can be seen in the maps of the dust extinction optical thickness from CAMS reanalysis (Fig. 2d and e). HYSPLIT back trajectories confirmed that the origin of air masses on particular days (5 and 11 August) was from the Sahara (Fig. 2a and b). According to CAMS reanalysis and HYSPLIT back trajectories, dusty air mass from northern Africa (Morocco, Tunisia, and northern Algeria) merged over the Mediterranean Sea. As a result, the air over Athens on 5 and 11 August included a mixture of smoke, marine, and dust particles at altitudes below 3 km. On the other hand, 18 August (Fig. 2f) was a dust-free day.

3.2 Impact on air quality

Both primary and secondary aerosols are produced during biomass burning whose chemical composition highly depends on the type of combustion (flaming or smoldering) and environmental conditions (Rickly et al., 2022). The time series of the air quality data are presented in Fig. 3, which shows the daily mean of PM2.5, PM10, NO, and NO2 concentrations ($\mu g m^{-3}$) at various sites within GAA. Note that not all stations measure the same air pollutants, which is why the number of stations is different in Fig. 3. It was observed that $PM_{2.5}$ values were generally below $20 \,\mu g \,m^{-3}$ during this period except for the beginning and middle of August (during wildfire events), when, for particular stations that were strongly affected by smoke, they exceeded 60 and $70 \,\mu\text{g}\,\text{m}^{-3}$, respectively (Fig. 3a). Elevated PM₁₀ levels were also found during the same period with values reaching up to $130 \,\mu\text{g}\,\text{m}^{-3}$ (Fig. 3b). PM₁₀ levels were at their maximum in the first week of August due to the presence of wildfire smoke and desert dust over all the stations.



Figure 2. Identification of dust transfer to Athens using (**a**, **b**, **c**) HYSPLIT back trajectories ending at 12:00 UTC and (**d**, **e f**) Copernicus Atmosphere Monitoring Service (CAMS) reanalysis time-averaged dust optical thickness over 00:00–21:00 UTC.

 NO_x is mainly generated during the flaming stage that occurs at high temperatures (Stefenelli et al., 2019). Very high NO and NO₂ concentrations were also recorded in the first week of August, as well as on 18 and 19 August, due to the wildfire events that obviously tend to increase NO_x emissions (Jin et al., 2021). Daily average NO reached $30 \,\mu g \, m^{-3}$ (it is usually below $10 \,\mu g \, m^{-3}$), while daily average NO₂ reached 75 μ g m⁻³ (it is usually below 30 μ g m⁻³). But it is interesting to note that high NO and NO2 values have also been recorded on days when the aerosol mixture is mainly constituted of dust (e.g. 11, 26, and 27 August). Elevated NO / NO₂ levels during dust events have been also reported in other studies (Milford et al., 2020). An increase in the total column of NO₂ (Fig. 3d) is generally in agreement with the increase in surface NO₂ concentration. According to our analyses, increased NO_x levels coincide with the presence of smoke and dust aerosols and/or low wind speeds (see Appendix Fig. A2). Low wind speed also favours increased NO_2 concentrations in urban environments, as NO_x concentrations are found to be in negative correlation with wind, precipitation, and relative humidity (Liu et al., 2020). Also, total NO₂ columns increased up to 6 times from the climatological mean during the forest-fire period (Fig. A3). High values of total column SO₂ were also observed during the first week of August (highest on 7 August) and, then, later on 19 August, with values reaching as high as 8 and 6 DU

(Dobson unit), respectively (Appendix Fig. A3), while the climatological average is ~ 1 DU. During wildfire events, unusually large amounts of SO₂ also have been observed in previous studies (Rickly et al., 2022; Weber et al., 2021; Ren et al., 2021).

The daily evolution of the near-surface aerosol scattering coefficient $(b_{sca,470})$ clearly detects the effect of Attica forest fires on the light scattering (Fig. 3e), with a daily mean b_{sca} value of $282 \,\mathrm{Mm^{-1}}$ on 4 August and enhanced $(> 100 \,\mathrm{Mm^{-1}}) b_{\mathrm{sca}}$ values on other days (like 6 and 10 August) significantly affected by transported smoke plumes over Athens. The mean SSA₆₆₀ during the measuring period was found to be 0.77 (0.02 higher than SSA₄₇₀), while under intense smoke conditions (4 August), this difference increased to 0.05 (SSA₆₆₀ = 0.70), suggesting the enhanced presence of brown carbon (BrC) aerosols. The peak values of the aerosol absorption coefficient (b_{abs}) on 4 and 6 August, associated with higher BCwb concentrations, are characteristic of the strong smoke effect on light absorption, while this effect was much more intense at 370 nm $(b_{abs,370} = 156.7 \text{ Mm}^{-1} \text{ on 4 August})$. The BC_{wb} concentrations in August 2021 $(0.43 \pm 1.21 \,\mu\text{g m}^{-3})$ were much higher - and variable as well - than the 4-year August mean value of $0.22 \pm 0.20 \,\mu \text{g m}^{-3}$ (Liakakou et al., 2020).



Figure 3. Variation in (a) $PM_{2.5}$ (µg m⁻³), (b) PM_{10} (µg m⁻³), (c) NO (µg m⁻³), and (d) NO₂ (µg m⁻³) in the greater Athens area (GAA) during August 2021. Temporal variation in the daily mean values for the (e) scattering and (f) absorption coefficients in Athens during 1–20 August 2021. The data points in (e) and (f) are colour-coded as a function of SSA₄₇₀ and BC_{wb} concentration, respectively. Please note that the date format in this figure is day/month.

3.3 Aerosol columnar optical and microphysical properties

Figure 4a shows the variation in AOD at five wavelengths, namely 340, 440, 675, 870, and 1020 nm (from now on referred as C5), and AE at 440-870 nm, while Fig. 4b presents the variation in the fine-mode, coarse-mode, and total AOD at 500 nm and the fine-mode fraction during August 2021. For Fig. 4, AERONET Level 2.0 data were used except for the days with high dust and/or smoke events when Level 1.0 direct sun and Level 1.5 inversion products were used, as mentioned in Sect. 2.1.2. Extremely high levels of fine-mode AOD were recorded, up to 1.95 at 500 nm on 18 August, due to the presence of smoke, with the high fine-mode fraction reaching 0.99 (99%) during these days. When dust aerosols were dominant, the fine-mode fraction was much lower, e.g. 0.36 on 11 August. On other days with a strong effect from smoke and dust (i.e. 4 and 5 August), the coarse-mode AOD was relatively higher. The detailed values of aerosol properties on these days are presented in Appendix Table A1.

Figure 4c and d present the volume particle size distribution and variation in columnar SSA on specific days in August 2021 in Athens, which are produced using the daily averages of 22 logarithmically equidistant discrete points in the size range varying from 0.05 to 15 μ m and daily average SSA values (at 440, 675, 870, and 1020 nm), respectively. There were six interesting cases observed in Athens (with AOD values equal to 1 or above) on 4, 5, 7, 11, 18, and 19 August. For these days, Fig. 5 shows the time–height distribution of aerosol layers in the atmosphere using the attenuated backscatter collected by the ceilometer at Thissio, Athens. The most notable aerosol layers were observed from 4 to 7 August, on 11 August, and from 18 to 19 August. Henceforth, we consider three cases: case 1 with only smoke, case 2 with dust, and case 3 with both dust and smoke. The variations in the aerosol properties in these three cases are presented in Table 2.

3.3.1 Case 1: smoke

On 7, 18, and 19 August, only smoke was present; 7 August was characterized by very high AOD values varying from 3.19 to 0.34 at C5 with the mean and maximum AE being



Figure 4. Variation in (**a**) spectral aerosol optical depth, (**b**) fine- and coarse-mode AOD at 500 nm, (**c**) daily mean volume particle size distribution, and (**d**) single-scattering albedo during the wildfire event in Athens. The missing lines for 4 and 5 August (**c**, **d**) are due to the available data at Level 1.5 not meeting our filter criteria for the inversion products on these days.

Table 2. Average aerosol properties (maximum values in brackets) for smoke and/or dust events in August 2021.

	Event	AOD ₅₀₀	Fine AOD ₅₀₀	Coarse AOD ₅₀₀	FMF ₅₀₀	AE 440–870	SSA (440–1020)
Case 1: 7, 18, 19 August	Smoke	0.53 (1.42)	0.45 (1.38)	0.05 (0.10)	0.87 (0.98)	1.84 (2.19)	0.93–0.89 (0.98–0.94)
Case 2: 11 August	Dust	0.56 (0.73)	0.26 (0.33)	0.30 (0.39)	0.46 (0.54)	0.74 (0.90)	0.89–0.97 (0.90–0.99)
Case 3: 4, 5 August	Dust and smoke	0.63 (1.43)	0.39 (1.21)	0.23 (0.28)	0.57 (0.85)	1.46 (1.56)	-

FMF: fine mode fraction. AE: Ångström exponent. SSA: single scattering albedo.

1.97 and 2.44, respectively. The high values of AE denote the dominance of small smoke particles in the aerosol mixture; 18 August was characterized by maximum values of AODs between 3.57 and 0.42 at C5, and AE showed a mean and maximum value of 1.75 and 2.14, respectively. The particle size distribution on these days had the dominance of accumulation mode (Fig. 4c). A strong absorption characteristic and strong spectral dependence is observed on 7 August when the SSA is seen to monotonically decrease with wavelength from 0.93 at 440 nm to 0.86 at 1020 nm, indicating the presence of smoke (Reid and Hobbs, 1998; Dubovik et al., 2000), while the lower SSA values in this day indicate the presence of strong absorbing aerosols (Kaskaoutis et al., 2021; Wu et al.,

2021). A highly intensified aerosol layer is observed below 2 km altitude on 7 August as it appears from Fig. 5c, which persists for the entire day. On 18 August, a dense floating aerosol layer can be seen before about 03:00 UTC that descends down and gets mixed in the boundary layer (Fig. 5f). This aerosol layer remained there throughout the day.

3.3.2 Case 2: dust

On 11 August, dust was dominant in the aerosol mixture. The AOD at C5 reached maximum values varying from 1.03 to 0.45, respectively, and the AE displayed an average and maximum value of 0.74 and 0.90, respectively. The aver-



Figure 5. Variation in (a) spectral aerosol optical depth, (b) fine- and coarse-mode AOD at 500 nm, (c) daily mean volume particle size distribution, and (d) single-scattering albedo during the wildfire event in Athens. The missing lines for 4 and 5 August (c, d) are due to the available data at Level 1.5 not meeting our filter criteria for the inversion products on these days.

age fine-mode, coarse-mode, and total AODs at 500 nm and fine-mode fraction were observed as 0.26, 0.30, 0.56, and 0.46, respectively. Low AE on 11 August indicates the presence of larger particles (Pace et al., 2006) which, as can be seen in Fig. 2, are dust particles that have been transported to Athens, and the size distributions is bimodal with dominance by the coarse mode (Fig. 4c). From Fig. 4d (and in Appendix Table A1), the SSA on 11 August is seen to increase with wavelength from 0.89 at 440 nm to values above 0.97 at wavelengths between 675 nm and 1020 nm, which signifies large forward scattering due to the presence of dust particles (Gómez-Amo et al., 2017). This is a typical spectral behaviour of dust aerosols having more absorption in UV than in the near-infrared region (Dubovik et al., 2002; Derimian et al., 2008). From Fig. 5d, it is seen that on 11 August there was a floating dust aerosol layer around 1 km altitude until 10:00 UTC and after 16:00 UTC.

3.3.3 Case 3: smoke and dust

4 and 5 August were characterized by the presence of both dust and smoke. On 4 August, the maximum AOD values at C5 varied from 2.15 to 0.44, respectively, while the corresponding variation on 5 August was from 2.32 to 0.68, respectively. The maximum AE at 440–870 nm was found to be 1.57 and 1.83 for 4 and 5 August, respectively. An

enhanced aerosol layer was present throughout the day on 4 August below 2 km (Fig. 5a). Figure 2a indicated the transport of Saharan dust to Athens, thus signifying the presence of both smoke and dust on 5 August. On 5 August, there was a large difference in the average and maximum AE at 440–870 nm (Appendix Table A1). For that particular day, a constant dust layer and a smoke layer decreasing from morning to afternoon led to a decrease in AOD and in AE during the day. A dense floating layer of aerosol is observed on 5 August (Fig. 5b) at about 1 km altitude and mostly below 2 km around 07:00 UTC. During the nocturnal hours, the highlighted aerosol layers are observed below 1 km altitude. Also, in Gómez-Amo et al. (2017), the authors found that the wildfire-related smoke event and a dust episode were simultaneously detected, and the dust-smoke mixing was found to enhance the aerosol load and modify the aerosol properties. The bimodal size distribution of the mixture was found to be dominated by smoke and dust in fine and coarse modes, respectively.

3.4 Aerosol properties from CAMS and SKYCAM

Figure 6a shows the total, organic matter, and dust AODs from CAMS. It is observed that the organic matter AOD is high on 7 August and between 17 to 19 August, while the dust AOD on these days is low. The dust AOD is high on 11

and 27 August, while it also presented enhanced values from 1 to 5 August, when the organic matter AOD is also high (4 and 5 August) due to the smoke effect. Figure 6b and c compare the daily average AODs from the Cimel instrument with those from CAMS and SKYCAM retrievals. For this comparison, the AOD from CAMS at 550 nm has been extrapolated to 500 nm, using the daily average AE from the Cimel instrument. It must be noted that the SKYCAM retrievals used for the calculation of daily averages were simultaneous with the Cimel AOD retrievals. For most days, SKYCAM gives higher values than the Cimel instrument, with the difference between the AODs being less than 0.1 (Fig. 6c). However, during intense smoke events, the differences are much larger, up to 0.2. Similarly, the differences between the AODs from CAMS and the Cimel instrument are also below 0.1, with the exception of days with dust or smoke events. Dust AOD is generally overestimated by CAMS (e.g. by 0.2 on 11 August). During smoke events, differences between CAMS and the Cimel instrument are again larger.

3.5 Transport to PANGEA

On 7 August smoke was detected from the Polly^{XT} lidar at NOA above the PANGEA observatory at altitudes of 0.5–3.0 km above the surface. Figure 7 shows the attenuated backscatter coefficient at 1064 nm for the Polly^{XT} lidar at NOA and the 24 h air masses back trajectories above the station (ending at 12:00 UTC on 7 August 2021) from HYS-PLIT model simulations (Fig. 7b). Wildfire aerosol sources and transports, lidar measurements, and analyses with the HYSPLIT model confirm that the smoke plume was transferred to PANGEA from various fire events, as can be seen from the layers below 3 km in Fig. 7a.

On the morning of 7 August aerosol concentrations above PANGEA were increased at different heights, probably as a result of remaining smoke plumes of a fire that started and also ended on 6 August in the southern Peloponnese (Mani Peninsula), an area close to the island of Antikythera (PANGEA). Air mass trajectories also showed that smoke from the main Attica and Evia fires arrived at PANGEA (at altitude below $\sim 1500 \text{ m}$) in the midday of 7 August (Fig. 7a), possibly mixed with smoke from wildfires that were burning in the Peloponnese (at altitude above $\sim 2000 \text{ m}$) during the previous day and/or night. The fire on the Mani Peninsula on 6 August can be seen in Aqua MODIS satellite images (see for instance Aqua MODIS-corrected reflectance, NASA Worldview; https://go.nasa.gov/3SEK9XK, last access: 10 May 2023; MODIS, 2023).

Figure 8 presents the diurnal variation in total, fine-mode, and coarse-mode AODs at 500 nm and AEs on 7 August 2021. In Athens, high AOD values (up to ~ 0.75) were observed in the early morning hours that were further increased after 10:00 UTC (up to ~ 1.53), which were accompanied by a high fine-mode fraction. The AE for both stations was found to be above 1 for the entire day. The lower AE values at 340–440 nm compared to those at 500–870 nm indicate a negative curvature effect, signifying the dominance of fine particles (Schuster et al., 2006). At PANGEA, AOD was high in the morning (~0.90) and afternoon (~0.72) hours of 7 August (due to the Athens fire transport), with a significantly high fine-mode AOD (~0.85 and ~0.68, respectively) and high fine-mode fraction. The fires of the southern Peloponnese may have also affected the air composition on 7 August at PANGEA.

Figure 9 shows the daily averaged spectral SSA variations from 440 to 1020 nm on 7 August, when smoke was present over Athens and over PANGEA (only afternoon values). The median SSA value at PANGEA decreases monotonically from 0.95 at 440 nm to 0.90 at 1020 nm. In Athens, the median SSA value was found to have a more drastic decrease from 0.91 at 440 nm to 0.82 at 1020 nm. It should be noted that Level 1.5 inversions were used for SSA retrievals in Athens from the Athens NOA station and one morning measurement was taken from the Athens NTUA station to get the average SSA of the day. The SSA decreasing with wavelength indicates the presence of fine smoke, which is evident for both stations. Higher SSA values in the afternoon of 7 August at PANGEA, compared to Athens, could be an indication of changing optical properties of smoke through transport and ageing processes that reduced the absorbing capability (Dasari et al., 2019; Eck et al., 2023). However, the presence of smoke from the Peloponnese local fire makes this assumption quite uncertain.

3.6 Effect on solar radiation

3.6.1 Spectral and total solar radiation

Finally, we calculated the attenuation of solar radiation by dust and smoke in different spectral regions during specific high-AOD days of August 2021. For this purpose, we compared measured irradiances at different spectral bands with the corresponding modelled irradiances for aerosol-free skies (case c in Sect. 2.3.1). In order to ensure that the modelled and the measured irradiances are comparable, we also modelled the irradiances using Cimel measurements (case a in Sect. 2.3.1) and then compared measured and modelled irradiances for days with very low aerosol load. When AOD is low, uncertainties in the aerosol optical properties used for the simulations have a negligible impact on the simulated irradiances. The lowest-AOD days were 15 and 16 August as inferred from Sect. 3.3. The results for both days were nearly identical and yielded an agreement better than 2 % between the measured and modelled irradiances for SZAs below 80°. For 16 August, the ratio between the measured and modelled values (considering realistic aerosol conditions) is presented with dotted lines in Fig. 10o. The results of the comparison between measured and modelled (considering AOD = 0) irradiances for 4 different days are also presented in Fig. 10 for UV-B (Fig. 10a-d), VIS (Fig. 10e-h), and NIR (Fig. 10i-l).



Figure 6. (a) Total, organic matter, and dust AOD from CAMS at 550 nm. (b) Comparison of AOD from the Cimel instrument with AODs from CAMS and SKYCAM at 500 nm (see text). (c) AOD differences between CAMS and the Cimel instrument and between SKYCAM and the Cimel instrument at 500 nm.



Figure 7. (a) Lidar attenuated backscatter coefficient at 1064 nm and (b) HYSPLIT 24 h backward trajectories ending at 12:00 UTC on 7 August 2021 over PANGEA.

These days are chosen as representative of different events, as presented in Sect. 3.3, including 4 August with very high AOD due to the presence of both dust and smoke, 11 August with very high AOD due to the presence of dust, 16 August representing very low AOD (daily average below 0.05 at 500 nm), and 18 August with very high AOD due to the presence of smoke.

From Fig. 10a–d, it is observed that the attenuation of UV-B irradiance was the least on 11 August and was the highest on 18 August, followed by 4 August. It is to be noted that 4 and 18 August are the days corresponding to smoke aerosols with very high fine-mode AOD values (> 1), as presented in Sect. 3.3, while 11 August has low fine-mode AOD but high coarse-mode AOD. Also, the very high AE of smoke, combined with the low SSA, induced a steep gradient in the spectral dependence of the attenuation. Thus, on 4 and 18 August, the UV-B irradiance was attenuated by 60 %. Moreover, in the evening of 18 August, the smoke aerosols attenuated about 90 % or even more of the UV-B irradiance. The attenuation in NIR was comparatively less, as can be seen from Fig. 10i–l, which was mostly of the order of 20 % or less. However, the attenuation of NIR irradiance was higher in the evening of 18 August, as was the case of UV-B irradiance, reaching about 40 %.

Figure 11 shows the relative contribution of the different spectral regions (UV-B, UV-A, VIS, NIR) to the daily inte-



Figure 8. Variation in total, fine-mode, and coarse-mode AOD at 500 nm and Ångström exponents (**a**) in Athens (AERONET Level 1.0) and (**b**) at PANGEA (AERONET Level 2.0) during the wildfire event of 7 August 2021.



Figure 9. Single-scattering albedo for (a) Athens (AERONET Level 1.5 with filters as mentioned in Sect. 2.1.2) and (b) PANGEA (only afternoon measurements from AERONET Level 2.0 inversion) for 7 August 2021.

grals of the GHI irradiance. The contribution is calculated as the ratio between irradiance in a spectral region (NIR, VIS, and UV) and the GHI. Due to relatively large gaps in the Brewer measurements on 7 and 12 August, the UV-B integrals have not been calculated for these days. The theoretical integrals that have been calculated based on modelled irradiances are presented with dashed lines. It can be observed that the contribution of NIR to total irradiance is higher on smoke days than on dust days, while the opposite can be observed for the VIS range. Figure 11b and c show the contribution of UV-A and UV-B to total irradiance. As in the VIS range, the contribution from UV-A and UV-B is lower for smoke cases as expected, due to the higher spectral dependence (high AE) of AOD on smoke aerosols, being higher in the lower spectral ranges. The daily average AOD at 500 nm and at 340 nm is shown in Appendix Table A1. It is interesting that although the daily average AOD at 500 nm is almost same, ~ 0.58 , the average AE at 440-870 nm is 1.97 and 0.74 on 7 August (smoke) and 11 August (dust), respectively, signifying the large variation in AOD with wavelength on 7 August (1.18 to 0.15 for the 340-1020 nm) compared to that on 11 August (0.80 to 0.35 for the 340–1020 nm) (Appendix Table A1). Hence, the change in the composition of GHI is significantly more pronounced on 7 August, and the attenuation is more enhanced mainly because of stronger absorption of light by smoke aerosols relative to dust aerosols (Kaskaoutis et al., 2021).

The effect of smoke on the levels of daily and monthly GHI in August 2021 is presented in Fig. 11d. These ratios have been calculated as the ratio of the daily integrals from the pyranometer measurements divided by the daily integrals calculated from modelled irradiances for AOD = 0 and are represented by the light-blue rhombus. In order to exclude the effect of clouds there was a visual inspection of the measurements with respect to cloud camera images and the hours during which the sun disc was partially or fully covered by clouds were marked. Then, for these hours the modelled irradiances were assumed to be equal to the measured irradiances (assuming that the aerosol effects are negligible under cloudy conditions). Measurement-based integrals were then divided with the latter modified modelled integrals (grey and black bars). Intense smoke events were marked in black. The dashed light-blue line represents the ratio between the average of measurement-based daily integrals and the modelbased daily integrals, excluding the days corresponding to intense smoke events. The dotted grey line represents the ratio between the average of measurement-based daily integrals and the modified model-based daily integrals for clear-



Figure 10. Effect of aerosols on (**a**–**d**) UV-B, (**e**–**h**) VIS, and (**i**–**l**) NIR irradiance on 4, 11, 16, and 18 August, respectively, and the ratio between measured and modelled irradiances (**m**–**p**).



Figure 11. Contribution of different spectral regions to total solar irradiance in August 2021. (a) Ratios of NIR and VIS with GHI, (b) ratio of UV-A with GHI, (c) ratio of UV-B with GHI, and (d) effect of smoke on the levels of GHI. Continuous lines represent ratios calculated using measured values, while dotted lines represent ratios calculated using modelled values for aerosol-free conditions. Black dots represent smoke events, while yellow dots represent dust events. The dashed lines (grey, light blue, and black) represent the ratio based on average GHI for August, while the ratio based on daily GHI is represented by grey shading, black bars, and a light-blue rhombus.

sky days, excluding again the days corresponding to intense smoke events. The ratio represented by the dashed black line has been calculated in the same way (as the ratio represented by the dashed grey line) but including the days with intense smoke events.

During intense smoke events, the daily GHI was attenuated by 10%–20% leading to a decrease of ~ 1.5 % in the monthly GHI. If days with smoke are not taken into account, the overall GHI decrease due to aerosols is ~ 5.5 % (dashed grey line). By taking the effect of clouds into account for the same days (dashed light-blue line), the decrease becomes 6.5 %. If only cloudless conditions are considered, including intense smoke events, the overall monthly GHI attenuation is 7 % (dashed black line). Also, during an intense wildfire event in Spain (Gómez-Amo et al., 2019), the radiative impacts of smoke and dust on photovoltaic plant performances were studied, revealing a loss of energy due to smoke with an average of 34 % on a daily basis, while due to dust it was around 6%, signifying the much higher efficiency of smoke in diminishing the solar-energy generation, as compared to dust.

3.6.2 Biologically effective doses

The effect of the intense smoke events on the levels of different biologically effective doses was also investigated. For this part of the study, the measured doses were compared with modelled ones that were calculated for climatological aerosol optical properties (case b of Sect. 2.3.1). In Fig. 12, the vitamin D and PAR daily doses, as well as the maximum UV index are presented, as they were calculated from the Brewer (maximum UV index and daily vitamin D), PSR measurements, and libRadtran simulations. The corresponding ratios between measured and modelled doses are also presented. The maximum UV index was calculated as the average UV index within ± 30 min around local noon.

Despite the distance of 6 km between the PSR and Brewer, the calculated doses from the two instruments agree quite well (within less than 5 % during the dust and smoke events), confirming that the effects of dust and smoke aerosols were quite homogeneous over the city centre during the events. The presence of smoke at noon on 4 and 18 August resulted in UV indices that are 4.5 and 2 units, respectively, below the climatological levels. A decrease of 30%-50% in the daily vitamin D doses was also estimated for the extreme-smoke events. Attenuation of the daily vitamin D dose on 7 August is 40%-50% and, thus, nearly double than the attenuation by dust on 11 August ($\sim 20\%$), although the daily average AOD₅₀₀ was larger on 11 August (see Sect. 3.6.1).

4 Summary and conclusions

A significant impact of severe forest fires on air quality and solar irradiance was observed in Greece in August of 2021. The AOD values increased up to 12 times, and total columnar NO_2 was up to 6 times above their climatological means. Total columnar SO_2 reached as high as 8 DU, while the climatological average is ~ 1 DU. Significantly elevated levels were also recorded in the surface $PM_{2.5}$, PM_{10} , and NO / NO_2 concentrations. In situ measurements showed that the transported smoke plumes over the Athens urban environment also exhibited a large effect on near-surface aerosol properties by significantly increasing the scattering and absorption coefficients, along with the BC concentrations, and especially the component related to biomass burning (BC_{wb}), which in August 2021 was double the long-term climatological August value.

Wildfire smoke was also observed to be accompanied by the Saharan dust on a few days in August. Based on AOD, AE, volume size distribution, spectral variation in SSA, and the synergistic use of the ceilometer vertical distribution, it was inferred that 4 and 5 August were characterized by the presence of both dust and smoke, while 7, 18, and 19 August were characterized by the presence of smoke and 11 August was characterized by the presence of dust. Only dust days were found to have high AOD, low AE, and positive spectral dependence of SSA. While the days with the presence of smoke exhibited high AOD, high AE, and a negative spectral dependence of SSA with increasing wavelength. Separate analyses of total, organic matter, and dust AODs from CAMS showed similarly the presence of large amounts of organic matter on only smoke days (7, 18, and 19 August) and peak dust AOD on 11 August.

On 7 August a smoke plume was also detected over PANGEA, which is about 240 km away from Athens. The transport of the plumes was detected below 3 km using lidar and HYSPLIT backward trajectories ending at PANGEA. AOD was found to be high in the morning and afternoon hours (smoke transported from Athens) with significant finemode AOD and AE values above 1 for the entire day. The negative curvature effect of AE further indicated the dominance of fine particles. Also, the SSA value was observed to decrease with wavelength from 440 to 1020 nm and to be considerably higher than the SSA measured in Athens.

Furthermore, the attenuation of solar irradiance in different spectral regions due to the presence of dust and smoke was analysed. It was found that the attenuation of UV-B irradiance was least in the presence of dust and highest due to smoke (up to 60 % or more) and intermediate when there was a mixture of smoke and dust. The attenuation in NIR was less compared to UV and VIS and mostly of the order of 20 % or less, although it reached up to 40 % in the presence of smoke. In the VIS region, the attenuation was greater than in the NIR region but less than in the UV-B region. The relative contribution of the different spectral regions, as compared to the daily integrals of the GHI irradiance, was also analysed, and it was found that the higher spectral dependence of AOD on smoke particles leads to lower relative contributions of the irradiance at lower wavelengths (UV, VIS) and higher relative



Figure 12. (a, c, e, g) Variability in biologically effective doses from the Brewer, PSR, pyranometer, and libRadtran simulations for climatological aerosol optical properties and (b, d, f, h) the corresponding ratios between the Brewer, PSR, pyranometer, and modelled doses. Please note that the date format in this figure is day/month.

contributions of the irradiance in NIR, compared to the ones for the dust cases.

The effect of smoke on the levels of daily and monthly GHI was also considered, and it was observed that during intense smoke events, the daily GHI got attenuated by 10 %–20 %. In August, the energy demand is high due to high temperatures, especially under extreme heat events (such as the one in August 2021). In the future, where a significant fraction of consumed energy will emerge from photovoltaics, decreases in GHI of 10 %–20 % could have a significant impact on many human activities that are strongly related to solar-energy production. Also, the smoke effect during the wild-fires period, as compared to climatology, led to decrease in the UV index of up to 53 \%, in vitamin D of up to 50 %, in PAR of up to 21 \%, and in GHI of up to 17 \%, with implications for health, agriculture, and energy.

Our results showed that extreme wildland fires such as those in August 2021 in Greece have considerable effects on

air quality (e.g. aerosol concentrations, aerosol properties, air pollutants) and solar radiation effective doses related to human health, ecosystems, and energy (e.g. UV index, vitamin D, PAR, GHI). Wildfires are part of the wider problem for Mediterranean countries, and the frequency of summer wildfires is predicted to increase in view of the projected increasing occurrence of summer heatwaves (Zittis et al., 2022). According to recent projections by Ruffault et al. (2020) the frequency of heat-induced fire weather is expected to increase in the Mediterranean basin until 2071-2100 under the RCP4.5 and RCP8.5 (Representative Concentration Pathway) scenarios, by 14 % and 30 %, respectively. In combination with extreme drought, extreme wind, and prolonged heatwave conditions in the future, it may well be speculated that the adverse effects of the projected increased frequency and extent of summer wildfires on vitamin D and PAR, as well as solarenergy production, will worsen across Mediterranean countries.

Appendix A: Measuring instrument description

A1 Brewer spectrophotometer

Brewer no. 001 has been automatically measuring the direct, diffuse, and global spectral irradiances in the UV and visible regions since 2003, and every 2-3 years it is calibrated on site by International Ozone Services (https://www. io3.ca/, last access: 7 May 2023). Since 2020, the Brewer has been calibrated using a set of three 200W lamps that are traceable to the scale of spectral irradiance established by the Physikalisch-Technische Bundesanstalt (PTB). More detailed information about the Brewer including measurements, quality control/assurance procedures, and calibration can be found in Eleftheratos et al. (2021) and Diémoz et al. (2016). The uncertainty in the Brewer measurements is estimated to 5% for wavelengths above 305 nm and SZAs lower than 70° (Garane et al., 2006). There is about 1 DU uncertainty in Brewer direct sun SO₂ measurements (Fioletov et al., 1998). During the wildfires, the SO₂ levels rose high enough in Athens, well above the mean $\pm 2\sigma$ (with the mean being 0.9 DU and σ being 0.6 DU), and, hence, the uncertainty was not of as much importance as it is in the case of low SO₂ values.

A2 Pandora spectral radiometer

Pandora uses the BlickP algorithm to calculate the total optical depth by estimating a synthetic reference spectrum and cross sections of NO₂ at an effective temperature of 254.5 K (Vandaele et al., 1998) that are fitted to a fourth-order polynomial, which results in the derivation of the slant column density (SCD). Then, it calculates the vertical column density (VCD) by applying a direct sun air mass factor. In clear-sky conditions, the precision of the slant column is 0.01 DU (Herman et al., 2009). Measurement uncertainties related to noise, systematic errors, drift, and wavelength shift are quantified during the monitoring process, and quality flags are provided (Cede and Tiefengraber, 2013). In this study, only the highquality post-processed data are used in order to eliminate any artefacts.

A3 Precision Solar Spectroradiometer

PSR no. 007 has a global sensor mounted on the auxiliary port, and by using the built-in shutter of the instrument, spectral GHI can be measured. Each cycle of measurements consists of 10 spectra of GHI and 5 dark measurements, which are eliminated, and the average spectra are stored before applying the calibration. Calibrations of the instrument were performed in the field on 7 July and 3 November in 2021 using a 200 W quartz halogen lamp that is traceable to the Physikalisch-Technische Bundesanstalt (PTB). The mean ratio between the calibrations was 1.0004 with a range between 0.9902 and 1.0276. Visual inspection of the data showed no possible jump/drift in the time series. Hence, a linear interpolation between the two calibrations provided the calibration for each day in the study period (August 2021). The uncertainty budget of the instrument is presented in Gröbner and Kouremeti (2019) and is less than 1 % in VIS, less than 1.7 % in UV-A, and higher than 2 % in UV-B.

A4 Pyranometer

The two pyranometers used in this study, manufactured by the Eppley Laboratory, have a black-coated thermopile acting as a sensor (or detector) which is protected against meteorological conditions by two concentric hemispherical domes. They both comply with the International Organization for Standardization (ISO) (ISO 9060, 2022) criteria for an ISO secondary standard pyranometer, being classified as "high quality" according to the World Meteorological Organization (WMO) nomenclature (WMO, 2021). Additionally, the corresponding pyranometer measuring the diffuse component was mounted on a shading device (Eppley shadow band) to block the direct irradiance and prevent it from reaching the sensor. Measurements from both pyranometers (for global and diffuse) have also been corrected for the "dark-signal" offset, also known as the "nighttime" offset, which is mainly due to thermal gradients between the dome and the sensor. As in any optical system that does not use cryogenic cooling or balanced operation, the transfer of infrared radiation between components affects the performance of pyranometers by generating an internal infrared signal that is superimposed on the output signal. The temperatures of the detector and of the outer dome are the main drivers of the temperature gradients that generate the internal, spurious signal. The inner dome acts as a "heat shield"; it reduces the amount of infrared radiation being transferred between the detector and the outer dome (Taylor, 1985). Both pyranometers were calibrated by the Laboratory of Meteorological Device Calibration of NOA (LMDC, 2022; Basil E. Psiloglou, personal communication, 2021) on 28 and 30 June 2021. In order to ensure high-quality measurements, LMDC follows the standard calibration procedure for thermopile pyranometers (ISO 9847, 2022), with exposure to real sunlight conditions and comparison with a working standard thermopile pyranometer (secondary standard), under constant clear-sky conditions and for solar altitude greater than 20°. This method is simple and provides sufficient accuracy because errors related to the dependence on solar incident angle and the instrument's spectral response are avoided. Traceability is ensured as LMDC's reference pyranometer, a Kipp & Zonen CMP21 (S/N: 150561), is regularly calibrated at the Physikalisch-Meteorologisches Observatorium Davos/World Radiation Center (PMOD/WRC), Davos, Switzerland. Also, utilizing the measurements during the nighttime period, from 21:00 to 03:00 UTC of the following day, it was possible to calculate the dark-signal error and correct the measurements of both pyranometers. The maximum daily error (daily integral) expected from these thermopile pyranometers is about 1 %-2 % (Hulstrom, 2003). These instruments also have imperfect angular response (Gueymard and Vignola, 1998), and, hence, a model-based correction for this effect was applied using a methodology similar to Bais et al. (1998).



Figure A1. Sky-camera images for Athens for (a1-a5) 4 August, (b1-b5) 5 August, (c1-c5) 7 August, (d1-d5) 11 August, (e1-e5) 18 August, and (f1-f5) 19 August, respectively.



Figure A2. Variation in (a) air temperature, (b) relative humidity, and (c, d) maximum wind speed during August 2021. Air temperature and humidity data are from the historical climatic data record of NOA. Wind speed data are from the ASNOA station. Please note that the date format in this figure is day/month.



Figure A3. Variation in (a) total SO₂ (DU) from Brewer measurements and (b) total NO₂ (DU) from Pandora measurements at Athens during August 2021. Please note that the date format in this figure is day/month.

Properties	4 August	5 August	7 August	11 August	18 August	19 August	26 August	27 August
Event	Dust and smoke	Dust and smoke	Smoke	Dust	Smoke	Smoke	Dust	Dust
AOD ₃₄₀	1.13 (2.15)	0.92 (2.32)	1.18 (3.19)	0.80 (1.03)	1.02 (3.57)	0.73 (1.30)	0.39 (0.50)	0.37 (0.49)
AOD ₄₄₀	0.81 (1.56)	0.74 (2.36)	0.75 (2.05)	0.63 (0.82)	0.73 (2.55)	0.51 (0.93)	0.34 (0.43)	0.31 (0.43)
AOD ₆₇₅	0.47 (0.77)	0.53 (1.40)	0.32 (0.75)	0.45 (0.58)	0.33 (1.08)	0.24 (0.41)	0.26 (0.34)	0.22 (0.32)
AOD ₈₇₀	0.36 (0.53)	0.39 (0.88)	0.19 (0.44)	0.38 (0.49)	0.21 (0.60)	0.15 (0.24)	0.23 (0.31)	0.19 (0.30)
AOD ₁₀₂₀	0.32 (0.44)	0.34 (0.68)	0.15 (0.34)	0.35 (0.45)	0.16 (0.42)	0.11 (0.17)	0.22 (0.30)	0.17 (0.28)
AE 440-870	1.12 (1.57)	1.04 (1.83)	1.97 (2.44)	0.74 (0.90)	1.75 (2.14)	1.79 (2.00)	0.57 (0.98)	0.76 (1.16)
AE 340-440	1.10 (1.35)	0.96 (1.26)	1.68 (2.15)	0.85 (1.13)	1.43 (1.80)	1.40 (1.70)	0.67 (0.87)	0.78 (1.12)
AE 500-870	1.07 (1.54)	1.00 (1.83)	1.96 (2.50)	0.69 (0.85)	1.75 (2.21)	1.79 (2.04)	0.52 (0.95)	0.71 (1.14)
Total AOD ₅₀₀	0.68 (1.24)	0.59 (1.43)	0.58 (1.53)	0.56 (0.73)	0.56 (2.00)	0.40 (0.73)	0.30 (0.39)	0.27 (0.38)
Fine AOD ₅₀₀	0.43 (0.99)	0.36 (1.21)	0.51 (1.49)	0.26 (0.33)	0.49 (1.95)	0.36 (0.70)	0.11 (0.13)	0.12 (1.15)
Coarse AOD ₅₀₀	0.24 (0.28)	0.23 (0.27)	0.06 (0.17)	0.30 (0.39)	0.06 (0.08)	0.04 (0.06)	0.19 (0.26)	0.15 (0.25)
FMF ₅₀₀	0.61 (0.80)	0.53 (0.85)	0.87 (0.99)	0.46 (0.54)	0.84 (0.98)	0.89 (0.96)	0.38 (0.57)	0.47 (0.65)
SSA ₄₄₀	_	_	0.93 (0.99)	0.89 (0.90)	0.94 (0.97)	0.93 (0.99)	_	0.89 (0.89)
SSA ₆₇₅	-	-	0.90 (0.99)	0.95 (0.97)	0.94 (0.96)	0.91 (0.98)	_	0.95 (0.95)
SSA ₈₇₀	_	_	0.88 (0.98)	0.96 (0.98)	0.94 (0.95)	0.89 (0.98)	_	0.97 (0.97)
SSA ₁₀₂₀	-	-	0.86 (0.98)	0.97 (0.99)	0.94 (0.95)	0.88 (0.98)	-	0.97 (0.97)

Table A1. Daily average values (maximum values in brackets) of aerosol properties for smoke and dust events of August 2021.

Code and data availability. The code and data used in this paper are available upon request from the authors. The AERONET aerosol optical depth data are freely available from the NASA Goddard Space Flight Center at https://aeronet.gsfc.nasa.gov/new_web/aerosols.html (Giles et al., 2019).

Author contributions. AM prepared the first draft. AM, IF, SK, and KE were the main concept organizers and main contributors to the writing of this paper. KE and SK organized the ASPIRE campaign, from which most of the data were collected. IPR, DK, IF, and NK contributed to the spectral solar and Pandora measurement analysis. AKam, SS, and AM contributed to air mass trajectory modelling. KP and IF contributed to radiative transfer modelling and the solar radiation analysis. EM, AG, and VA provided the Antikythera aerosol data. BEP provided solar radiation measurements and contributed to the data quality control and analysis. DF provided meteorological data and contributed to the analysis. VS and AKaz contributed to the sky-camera data analysis. DK and NM contributed to the in situ aerosol data provision and analysis. AP provided Athens NTUA data. CSZ, KE, and SK contributed to the paper overview and section organization. All authors contributed by critically reviewing the writing and gave final approval for publication.

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