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Using Radiometric Measurements to Separate Dust and Smoke Radiative Effects during a Combined Smoke–Dust Event †

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Abstract: In August 2021, extreme wildfires burned wide areas in Greece. The most severe events took place in the region of Attica and significantly affected the air quality over the city of Athens. In the same period, southern winds transferred dust from the Sahara Desert to the city. We chose a day with high dust and smoke loads to test a new method for the determination of the individual components of the aerosol mixture, based on actinometric and radiometric measurements and radiative transfer modelling. Then, we estimated the radiative effect of each of the aerosol mixture components. Assuming that dust particles are coarse while all other particles are fine, coarse to fine mode ratios were estimated and were compared with the corresponding ratios from a CIMEL sun photometer, resulting in quite good agreement.

Keywords: aerosols; dust; smoke; radiative effects

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1. Introduction

Aerosols have a significant impact on Earth's climate. They affect the radiation budget in the Earth–atmosphere system through their interactions with solar and terrestrial radiation (direct radiative effects). They also affect the formation and the life of clouds, indirectly affecting the radiation budget (indirect and semi-direct aerosol effects). Aerosol radiative effects constitute a major factor of uncertainty in the projections of global climate models, since the interactions between different aerosol species and radiation are not completely understood, and in many cases, they cannot be modeled easily [1].

The Mediterranean Basin is a crossroads of many different aerosol types (marine, dust, anthropogenic, smoke, etc.) [2]. European countries lying on the shores of the Mediterranean Sea are frequently affected by the transport of Saharan dust in spring and summer [3,4]. Occasionally, dust events can result in extremely high concentrations with significant impacts on ecosystems, the economy, society and health [5]. It is estimated that on an annual basis, 3–10% of the global horizontal irradiance (GHI) is attenuated by dust aerosols over the Eastern Mediterranean [6]. The dry conditions during the summer months also result in frequent wildfires in mid-latitude Mediterranean countries, with implications for human health and climate (e.g., [7]). As discussed in recent studies [8,9], understanding the impact of dust and smoke mixing is essential to understand the Mediterranean climate.

In this study, we focused on an intense event of the combined presence of wildfire smoke and Saharan desert dust in Athens, Greece, and we tried to separate the direct radiative effects of each component of the aerosol mixture.

2. Data and Methodology

On 5 August 2021, wildfire smoke and Saharan dust aerosols were simultaneously present over Athens, Greece, [10] resulting in elevated levels of fine and coarse particles over the area (Figure 1). In the morning, the aerosol mixture was dominated by fine smoke particles that arrived from Varympompi (an area near the city of Athens). Gradually, the contribution of smoke became smaller throughout the day.

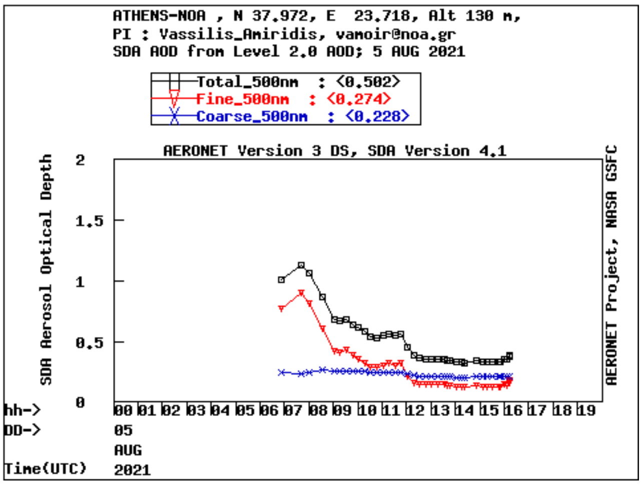

Figure 1. Fine- and coarse-mode aerosol optical depth (AOD) at 500 nm (Level 2.0 data) over the ASNOA station on 5 August 2021 (provided by AERONET).

On the same day, measurements of various atmospheric parameters (including solar radiation components) were performed at the Actinometric station of the National Observatory of Athens at the Thissio site (ASNOA, 38.0° N, 23.7° E, 110 m above mean sea level) in the context of the Atmospheric parameters affecting SPectral solar IRradiance and solar Energy (ASPIRE) campaign (https://aspire.geol.uoa.gr/; last access: 9 May 2023), which were used to certify the quality of the data used and to identify the event [10].

Aerosol optical properties were measured using a CE318 sun/sky photometer from Cimel Electronique (CIMEL). For this study, we used the Level 2 AOD from the AERONET Version 3 algorithm [11]. The GHI was measured using an Eppley PSPs (Precision Spectral Pyranometers, S/Ns: 26069, 26070) radiometer [12]. Aerosol backscatter coefficient profiles from a collocated Vaisala CL31 ceilometer were also used to approximate the aerosol profiles. Radiative transfer simulations were performed using the radiative transfer model UVSPEC, which is included in the libRadtran v2.4 package [13].

In order to estimate the composition of the aerosol mixture, we made a number of assumptions. The first assumption we made was that all coarse particles are mineral dust (miam), while fine particles are a mixture of water-soluble (organic carbon, sulfate, nitrate, etc.; waso) particles and soot (black carbon), and maybe a very small fraction of them can be dust. Although it is very possible that more species contribute to the aerosol mixture, the contribution of the above three species is expected to be dominant (Tables 3 and 4 in Hess et al. [14]). Additionally, we made the assumption that the mixture of aerosols is external, which is a common assumption in similar studies (e.g., [15]). Then, we proceeded as follows:

1. With a step of 0.05, we created all possible combinations of dust AOD, waso AOD, and soot AOD.

2. For the UVSPEC simulations, we used the following extinction coefficient profiles:
   a. waso: climatological urban from OPAC [14].
   b. soot: maximum invariant concentration between 0.5 and 1.5 km that drops to 20% of the maximum at the surface and at 2 km (estimated roughly from the eilometer).
   c. dust: homogeneous concentration from 0 to 2 km.

   Deviations from the exact profiles are not expected to induce large uncertainties in the results of the radiative transfer simulations (e.g., [16]).
3. Using the methodology developed by Siomos et al., [17] we calculated the mass concentrations that correspond to each AOD combination, assuming a relative humidity of 50%, and then scaled the profiles.
4. For each combination, we performed UVSPEC simulations of the GHI and then compared model outputs with the pyranometer measurements.
5. Finally, we considered that the combination for which the simulated GHI is closer to the measured represents the composition of the mixture.

Different analogies in waso, soot, and miam correspond to different effective aerosol optical properties such as single-scattering albedo (SSA), Angstrom exponent (AE), and phase function. Optical properties for each of the aerosol species from the OPAC package [14] were considered. Differences between the nearest modeled and measured GHI were within ±1% in all cases.

After defining the contribution of each component of the mixture, we performed radiative transfer simulations for aerosol-free atmosphere and for each of the components of the aerosol mixture in order to estimate the attenuation of the GHI due to each component. The AOD was interpolated to the time of the pyranometer measurements. The attenuation of GHI by aerosols was also estimated based on the aerosol optical properties (SSA, AE for 440–670 nm) provided by AERONET.

3. Results and Discussion

As a first step, the contribution of each aerosol type to the mixture was estimated (Figure 2). While from Figures 1 and 2 it is evident that the AOD in the morning (min < 700) is mainly due to small particles (waso, soot), the mass of dust is comparable or larger relative to the mass of small particles, obviously due to the much bigger size of the dust particles. The attenuation of GHI is shown in Figure 3. More specifically, the following is shown:

- Attenuation of the GHI assuming that only one of each of the assumed aerosol types (1. waso, 2. soot, 3. dust) is present.
- The summary of the attenuation that has been estimated individually for each aerosol type (1 + 2 + 3).
- Attenuation of the GHI assuming that all aerosol species are simultaneously present (all).
- Attenuation of the GHI that has been estimated using the optical properties provided by AERONET.

Soot (which, as defined in OPAC, is practically black carbon) is a very small fraction of the overall mass of the mixture but plays a very significant role in the attenuation of GHI, as shown in Figure 3, mainly because it strongly absorbs the GHI. In the first morning hours, although the AOD due to soot is 5–8 times smaller relative to the AOD due to waso particles, they result in similar attenuation (10–15%) of the GHI. In the evening, the GHI is attenuated mainly by dust.

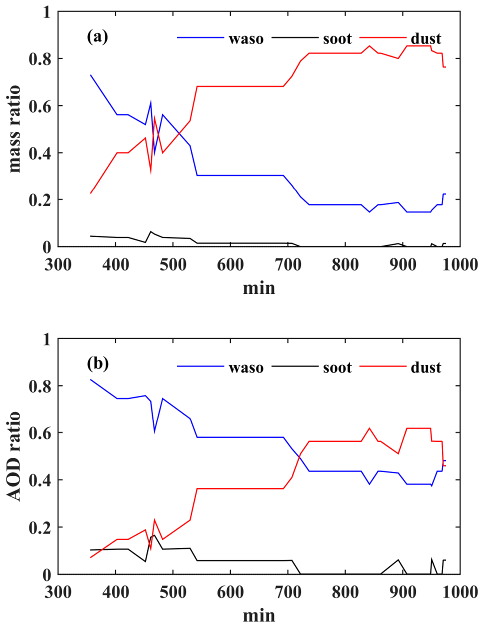

Figure 2. (**a**) Ratio between the mass of each aerosol type and the total mass of the mixture and (**b**) ratio between the AOD at 500 nm for each type and the total AOD.

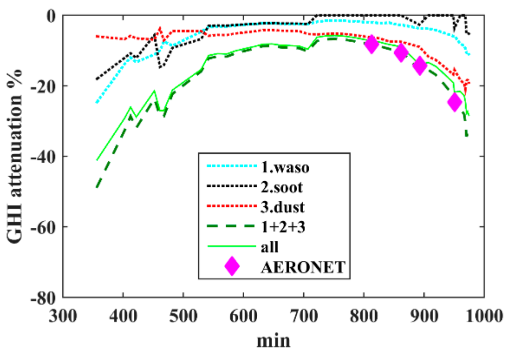

Figure 3. Attenuation of the GHI due to each of the different aerosol species (assuming that no other aerosols are present), cumulative attenuation that has been calculated for individual aerosol types, attenuation due to the simultaneous presence of all aerosol types, and attenuation due to all types as has been calculated from optical properties provided by AERONET.

The summary of the attenuation by different species (1 + 2 + 3) is practically equal (differences smaller than 3%) to the attenuation due to the combined mixture (all) during most of the day. In the morning, however, differences are larger, up to ~8%, possibly because the increased path of the radiation (larger solar zenith angles; SZAs) during its propagation in the atmosphere increases the relative significance of the interactions of scattered photons with different aerosol types. When SZAs are large, for the particular aerosol mixture, it is possible that multiple scattering increases the amount of GHI that reaches the surface. The attenuation that has been estimated based on the AERONET optical properties is in very good agreement with the attenuation that has been estimated based on our method. However, measurements of the SSA were available only in the evening when mainly dust—and no smoke—was present.

4. Summary and Conclusions

In this study, we present a novel method for the estimation of the radiative effects of the different aerosol species that constitute a mixture that includes more than one aerosol type. The method has been applied to a mixture of Saharan dust and wildfire smoke aerosols during an intense combined dust–smoke event.

The identification of the contribution of the different aerosol species to the mixture has been achieved using a modified version of the methodology proposed by Siomos et al., (2020) [18]. The method is simple and employs measurements of the AOD and the GHI that are available at many sites around the globe. Nevertheless, these results are preliminary and consider only one case, and further validation of the method is necessary, possibly against a more sophisticated model (e.g., GRASP; [19]). In the future, we intend to apply our method to more similar cases, validate the results to identify the uncertainties, and use spectral measurements of the surface solar irradiance instead of the GHI to improve the accuracy of the method.

Author Contributions: Conceptualization, I.F., N.S. and S.K.; methodology, I.F. and N.S.; software, I.F. and N.S.; validation, I.F.; formal analysis, I.F.; investigation, I.F. and N.S.; resources, B.E.P., I.-P.R., N.S., D.K. and V.A.; data curation, B.E.P., D.K. and I.-P.R.; writing—original draft preparation, I.F.; writing—review and editing, all authors; visualization, I.F.; supervision, S.K.; project administration, K.E. and S.K.; funding acquisition, K.E. and S.K. All authors have read and agreed to the published version of the manuscript.

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