Distribution, optical properties, and radiative effect of pollution aerosols in the western Mediterranean basin from TRAQA and SAFMED airborne observations

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Pollution aerosols strongly influence the composition of the western Mediterranean basin, but at present little is known on their distribution, optical properties and radiative effects. We report in this study in situ observations of pollution aerosol plumes obtained over the sea in the western Mediterranean during the TRAQA (TRansport and Air QuAlity) and the SAFMED (Secondary Aerosol Formation in the MEDiterranean) airborne campaigns in summers 2012 and 2013 (Di Biagio *et al*, 2015). The TRAQA and SAFMED flights explored an extended region of the western Mediterranean between 40°-45°N latitude and 2°W-12°E longitude including the Gulf of Genoa, Southern France, the Gulf of Lion, and the Spanish coasts. Measurements were performed over the sea at various distances from the coastline and up to 5000 m altitude. TRAQA and SAFMED successfully measured a wide range of meteorological conditions which favoured the pollution export from different sources around the basin.

Observations from the present study indicate that continental pollution largely affects the western Mediterranean both close to coastal regions and in the open sea as far as ~250 km from the coastline. Aerosol layers are distributed ubiquitously which indicates quite elevated levels of background pollution throughout the western basin. The measured aerosol scattering coefficient varies between ~20 and 120 Mm^{-1}, while carbon monoxide (CO) and ozone (O_3) mixing ratios are in the range of 60-170 ppbv and 30-85 ppbv, respectively. Pollution reaches 3000-4000 m in altitude and presents a very complex and highly stratified structure characterized by fresh and aged layers both in the boundary layer and in the free troposphere. Within pollution plumes the measured particle concentration in the Aitken (0.004-0.1 μm) and accumulation (0.1-1.0 μm) modes is between ~100 and 5000-6000 scm^{-3} (standard cm^{-3}), which is comparable to the aerosol concentration measured in continental urban areas. Additionally, our measurements indicate the presence of highly concentrated Aitken layers (10000-15000 scm^{-3}) observed both close to the surface and in the free troposphere, possibly linked to the influence of new particle formation (NPF) episodes over the basin.

Data from this study show a large variability of the aerosol shortwave single scattering albedo (ω) for pollution aerosols, with values between 0.84-0.98 at 370 nm and 0.70-0.99 at 950 nm. The single scattering albedo generally decreases with the wavelength, with some exceptions associated to the mixing of pollution with sea spray or dust. The lowest values of ω (0.84-0.70 between 370 and 950 nm) are measured in correspondence of a fresh plume possibly linked to ship emissions. The range of variability of ω seems to be independent of the source region around the basin, as well as of the altitude and aging time of the plumes. The observed variability of ω reflects in a large variability for the complex refractive index of pollution aerosols, which is estimated to span in the large range 1.41-1.75 and 0.002-0.068 for the real and the imaginary parts, respectively, between 370 and 950 nm.

Radiative calculations in clear-sky conditions were performed with the GAME radiative transfer model to test the sensitivity of the aerosol shortwave Direct Radiative Effect (DRE) to the variability of ω as observed in this study. Results from the calculations suggest up to a 50% and 30% change of the forcing efficiency (FE), i.e. the DRE per unit of optical depth, at the surface (-160÷-235 $Wm^{-2}\tau^{-1}$ at 60° solar zenith angle) and at the Top-Of-Atmosphere (-137÷-92 $Wm^{-2}\tau^{-1}$) for ω between its maximum and minimum value. This induces a change of up to an order of magnitude (+23÷+143 $Wm^{-2}\tau^{-1}$) for the radiative effect within the atmosphere.

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