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Characterization of carbonaceous aerosols during the MINOS campaign in Crete, July–August 2001: a multi-analytical approach

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Abstract. During the major part of the Mediterranean Intensive Oxidant Study (MINOS) campaign (summer 2001, Crete Isl.), the Marine Boundary Layer (MBL) air was influenced by long range transport of biomass burning from the northern and western part of the Black Sea. During this campaign, carbonaceous aerosols were collected on quartz filters at a Free Tropospheric (FT) site, and at a MBL site together with size-resolved distribution of aerosols. Three Evolution Gas Analysis (EGA) protocols have been tested in order to better characterize the collected aged biomass burning smoke: A 2-step thermal method (Cachier et al., 1989) and a thermo-optical technique using two different temperature programs. The later temperature programs are those used for IMPROVE (Interagency Monitoring of Protected Visual Environments) and NIOSH 5040 (National Institute of Occupational Safety and Health). Artifacts were observed using the NIOSH temperature program and identified as interactions between carbon and dust deposited on the filter matrix at high temperature (T>550ºC) under the pure helium step of the analysis.

During the MINOS campaign, Black Carbon (BC) and Organic Carbon (OC) mass concentrations were on average respectively 1.19±0.56 and 3.62±1.08 μgC/m^3 for the IMPROVE temperature program, and 1.09±0.36 and 3.75±1.24 μgC/m^3 for the thermal method. Though these values compare well on average and the agreement between the Total Carbon (TC) measurements sample to sample was excellent (slope=1.00, r^2=0.93, n=56), important discrepancies were observed in determining BC concentrations from these two methods (average error of 33±22%). BC from the IMPROVE temperature program compared well with non-sea-salt potassium (nss-K) pointing out an optical sensitivity to biomass burning. On the other hand, BC from the thermal method showed a better agreement with non-sea-salt sulfate (nss-SO_4), considered as a tracer for fossil fuel combustion during the MINOS campaign. The coupling between these two methods for determining BC brings here new insights on the origin of carbonaceous aerosols in a complex mixture of different sources.

It brings also to our attention that important deviations in BC levels are observed using three widely used EGA's techniques and most probably none of the EGA tested here are well adapted to fully characterize this aerosol mixture.

Spherical, smooth and silico-aluminated fly-ash observed by an Analytical Scanning Electron Microscope (ASEM) confirm the influence of coal combustion on the carbonaceous aerosol load throughout the campaign. A rough calculation based on a BC/nss-SO_4 mass ratio suggests that biomass burning could be responsible for half of the BC concentration recorded during the MINOS campaign.

From the plot of BC as a function of TC, two linear correlations were observed corresponding to 2 times series (before and after 12 August). Such good correlations suggest, from a first look, that both BC and OC have similar origin and atmospheric transport. On the other hand, the plot of BC as a function of TC obtained from the 2-step thermal method applied to DEKATI Low Pressure Cascade Impactor samples does not show a similar correlation and points out a non conservative distribution of this ratio with 2 super micron modes enriched in OC, correlated with sea salt aerosols and probably originating from gas-to-particle conversion.

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