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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Source contributions to 2012 summertime aerosols in the Euro-Mediterranean region" *by* G. Rea et al.

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We thank the reviewer for the comments regarding the method used in the manuscript. Please find below our replies.

This study aims at quantifying the contribution of different aerosol sources over the Mediterranean area in summer 2012. A "reference" model simulation is compared to observations from surface network and remote sensing (AirBase, AERONET network, and the MODIS satellite instrument). Sensitivity simulations are performed: simulations are performed without mineral dust emission, without anthropogenic emission, without fire emission and without sea-salt emission. These simulations are compared to the reference simulation to quantify the impact of the sources on $PM_{2.5}$ and PM_{10}

concentrations.

In the model description, the authors mention that adsorption and desorption are taken into account in the model. I am however extremely surprised that they omitted absorption, one of the major process governing aerosol dynamic.

Although the kind of source apportionment done in this paper without considering the non-linearity of chemistry may be applied to inert chemical compounds such as dust, it does not apply to reactive chemical compounds. For example, most biogenic precursors may not condense onto particles without prior oxidation from anthropogenic-origin oxidants. The absorption of anthropogenic compounds onto particles is also influenced by biogenic compounds. The interaction between biogenic, anthropogenic and fire emissions also needs to be considered, when assessing the influence of fire emissions. For reactive species, different methodologies exist to determine source-receptor relationships: direct decoupled sensitivity analysis or emissions-labeled tracers analysis. Neglecting the non-linearity of chemistry when performing source apportionment, as done here, is very misleading and leads to misleading conclusions and messages. Therefore, I do not recommend this paper for publication, until the authors consider absorption in their model, and revise the methodology used for source apportionment or focus on the impact of inert compounds.

The reviewer mentioned the absence of absorption in the model description, resulting in a modelling of aerosol dynamic not correct. The CHIMERE model actually considers the condensation/adsorption/absorption in a same way, with the flux of a semi-volatile inorganic or organic species "fixed" themselves onto a monodisperse aerosol depending on a characteristic time, the gas phase and the equilibrium concentrations, the latter calculated with the ISORROPIA module for semi-volatile inorganic species and a partition coefficient for organic species. The missing term absorption in the paper is in fact forgotten, but is added on the new version of the manuscript because absorption is indeed taken into account in the CHIMERE simulations computed for this study: "Their life cycle is represented with a complete scheme of nucleation,

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absorption, adsorption, desorption, coagulation, as well as wet and dry deposition and scavenging. The size distribution is simulated using a sectional representation, i.e using five bins from a diameter of 40 nm to 40 μm" (Section 3.2, second paragraph).

The CHIMERE CTM has been used to perform a number of sensitivity simulations, each with one source eliminated. The aim is to provide the approximate impact from the main regional sources. The reviewer does not agree with the method used, as it does not account for the non-linearities inherent to reactive chemical compounds. Indeed, the authors are aware that this method can lead to imprecise contributions, for instance the sum of all individual sources may not amount to the reference concentrations of PM due to non-linarites in chemical processes (it is a disadvantage known with this method), and to missing sources and sink (transport from boundary conditions, wet and dry deposition). However, studies on particles sensitivities show that the main conclusions do not change with other methods such as source apportionment or direct decoupled sensitivity, if the indirect effect is not significant (Koo et al., 2007; Koo et al., 2009; Burr et al., 2011).

In our study, the amount of $PM_{2.5}$ and PM_{10} formed by non-linear processes can be evaluated in the reference simulation. Therefore, we estimate in the simulation the relative contribution from SOA and nitrates, which will be most impacted by non-linearities, to the total PM concentrations. On average over the domain and on the reference simulation during the summer 2012, 2.5% of total $PM_{2.5}$ is composed of SOA, and 1.3% of nitrates. Contributions to total $PM_{2.5}$ from the sensitivity simulations have been evaluated to be 1.3% and 1.1% of SOA and nitrates respectively from anthropogenic sources, and 1.5% and maximum 0.01% of SOA and nitrates from biogenic emissions. Contributions are 0.04% and 0.02% of SOA and nitrates respectively from fire sources. The complementarity between biogenic and fire emissions have not been taken into account, i.e. biogenic sources have not been removed where fires are present. This might lead to an overestimation of the contribution of biogenic sources during fires. The

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maximum of SOA contribution from biogenic emission on $PM_{2.5}$ surface concentration in a fire plume is 4.4%.

A paragraph with these results and bibliography is added in the new version of the manuscript (Section 3.4): "Note that the sum of all individual sources may not amount to the reference concentrations of PM due to non-linearities in chemical processes. However, studies on particles sensitivities show that the main conclusions do not change with other methods such as source apportionment or direct de- coupled sensitivity, if the indirect effect is not significant (Koo et al., 2007, 2009; Burr et al., 2011). In our study, the amount of PM2.5 and PM10 formed by non-linear processes can be evaluated in the reference simulation. Therefore, the relative contribution from SOA and nitrates, which will be most impacted by non-linearities, to the total PM concentrations is calculated. On average over the domain and for the reference simulation during the sum- mer of 2012, 2.5% of total PM2.5 is composed of SOA, and 1.3% of nitrates. Contributions to total PM2.5 from the sensitivity simulations have been evaluated to 1.3% and 1.1% of SOA and nitrates respectively from anthropogenic sources, and 1.5% and maximum 0.01% of SOA and nitrates from biogenic emissions. Contributions are 0.04% and 0.02% of SOA and nitrates respectively from fire sources."

Interactive comment on Atmos. Chem. Phys. Discuss., 15, 8191, 2015.