

This manuscript addresses relevant scientific questions within the scope of the ACP special issue dedicated to megacities. Its overall presentation (including the title, the abstract and the figures) is appropriate, clear and globally well structured. It presents a novel dataset on the chemical composition and major sources of size-segregated aerosols sampled in the Athens area, Greece, using common scientific methods and, more generally, valid assumptions. Nevertheless, there are some major issues/lacks that might be considered before publication:

- Carbonaceous material: despite the probable high influence of such species on PM levels and health/climatic effects in the Athens area, no data directly related to these species is presented in the manuscript. Nevertheless, authors mentioned recent samplings/analyses of OC/EC, probably at the same sites (at least FKL). It might be worthy adding this information in the present manuscript, directly in the text or within supplementary info. This might help convincing readers that results presented here (EC + OM of about 20%, or less, within PM₁₀ on annual average) are actually in good agreement with previous studies in Athens (EC+OM over ~ 30% for PM₁₀ during short-time field campaigns).
- Despite FLK is more generally situated downwind of Athens emissions, this site is considered here as representative of background pollution. The validity of this assessment needs to be discussed. For instance: impact of the topography north of Athens, sea breeze? Any information on the aging of Athens emissions from the dataset?
- Further discussions on the major conclusions of the paper are still needed. Which information could help decision makers and monitoring network better understanding/tackling limit values exceedances? To what extent could some conclusions be generalized or contrasted to other megacities?

More specific comments are listed below:

- Page 7664, line 10: as already mentioned in one of the quick reports, the European limit value of annual mean PM₁₀ is not 50 µg/m³.
- P. 7664, l. 17 (also 7668, lines 17 and 23, ...): a satisfactory positive correlation doesn't demonstrate a direct link between both investigated parameters. Please replace "indicating" by e.g., "suggesting".
- P. 7665, l. 3-5; P. 7667, l. 6-; ...: please give values obtained within previous studies.
- P. 7665, l. 10-14: Are high mineral dust contents always due to Saharan dust events? At GAA sites: other mineral dusts related to resuspension from traffic; and wind gusts?
- P. 7666, l. 1: This sentence is a little bit confusing. I assume 25% of southern winds, and not of "dust events".
- P. 7667, l. 5: "In PM₁₀ and PM_{2.5}, levels ...".
- P. 7667, l. 15: "In Athens, the most likely formation pathway ... "
- P. 7669, l. 1-2: Possible implication of acidic conditions on SOA formation?

- P. 7669, section 3.2.3: Discussion on the scavenging of gaseous precursors by mineral dust maybe needed. Any impact on the concentration levels of these gaseous species?
- P. 7670, l. 4-5: How long could the long-range transport? What does mean “larger spatial scale”?
- P. 7671, l. 16: Resuspension only due to traffic? Wind? The distinction between soil dust and car/road abrasion could be clarified throughout the manuscript.
- P. 7672, l. 18-20: not accurate. Some North African countries banned leaded gasoline for several years.
- P. 7673, l. 10: “In total, crustal ...”
- P. 7675, l. 5: “... continuously monitored. “
- P. 7675, l. 16: “Crustal material was accounted ..”.