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## ***Interactive comment on “Mass and chemical composition of size-segregated aerosols (PM<sub>1</sub>, PM<sub>2.5</sub>, PM<sub>10</sub>) over Athens, Greece: local versus regional sources” by C. Theodosi et al.***

**C. Theodosi et al.**

mihalo@chemistry.uoc.gr

Received and published: 8 July 2011

Referee report on ACP-2010-936

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:

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- 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).

ANSWER: We thank the reviewer for his/her comment. Most probably he/she refers to Athens data and not to FKL (FKL data has been already published and referred in the manuscript). The data referred in the manuscript as Mihalopoulos et al., unpublished data are now under revision (Pateraki et al., Environmental research). Aerosol chemical composition was measured in 3 locations around Athens in summer and winter in 2008 in PM2.5 and PM1 samples. POM+EC account for 32 and 37% of the PM mass in agreement with our conclusions reported here. Page 19, lines 499-502.

- Despite FKL 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?

ANSWER: The idea of this article is to use data from Finokalia as characteristic of natural regional background. This is clearly demonstrated by the results of several campaigns performed in the area using aircrafts or ships. On the other hand it is well known that Athens in some cases is influenced by local recirculation patterns (sea-breeze) due to its topography. Thus using a station around Athens as regional background it would not be possible to avoid a direct influence from Athens sources which is not the case of Finokalia. To persuade the reviewer on the validity of our hypothesis we also compared PM10 levels recorded at Finokalia (FKL) with those measured at a

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remote suburb area of Athens Thrakomakedones (THR). Thrakomakedones is located at a foothill of Parnitha Mountain, about 550m above sea level and 23 Km from the city center. This site is considered as a background, because of the absence of primary sources affecting PM concentrations (Grivas et al., 2004a). The comparison between THR and FKL revealed significant covariations between the two sites in terms of PM10 during the sampling period. Correlation between monthly mean values gave a slope of 1 with  $r^2$  of 0.6 indicating that FKL can be considered as a background (reference) site for the GAA. Similar climatology between GAA and FKL can account for the observed similarity in PM10 (see below). Differences between THR and FKL exist only during strong dust events occurring few times during the year, with Finokalia values being higher compared to THR, which is expected given the vicinity of FKL to N. Africa. Apart of these extreme dust events coarse mode in FKL is lower compared to GAA, indicating significant contribution for local sources in GAA (wind and road dust). Page 6, Section 3.1. Trajectories: Detailed trajectory analysis performed during the 2005–2006 period revealed that Athens and Finokalia are under similar air masses influence most of the time of the year, especially during the dry period (May to September, see also answer to reviewer 2). Trajectory analysis revealed that influence of Finokalia from Athens although possible under North winds is not often occurring throughout the year and conformed also by the analysis done by Markou and Kassomenos (2010). Page 6, lines 140–170.

- 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?

ANSWER: Complementary conclusions were added as recommended to help decision makers and monitoring network to better tackling PM exceedances in Athens. Page 20, Conclusions.

More specific comments are listed below:

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- Page 7664, line 10: as already mentioned in one of the quick report, the European limit value of annual mean PM10 is not 50\_g/m3. ANSWER:The reviewer is correct the sentence has been changed. Page 8, lines 182.

-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". ANSWER:The sentence has been corrected as suggested by the reviewer. Page 8, lines 189.

- P. 7665, l. 3-5; P. 7667, l. 6-; : : :: please give values obtained within previous studies. ANSWER:As suggested by the reviewer more information have been provided by comparing with various works cited in the manuscript. Tables 2 and 3, Pages 32-33.

- 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? I ANSWER:n the area studied and in general the Mediterranean region Saharan dust is the major source of mineral dust. However resuspension from traffic or from constructions could be the source of elements such as Zn, Cr, Fe, Cu and Mn. The existence of vehicle/road dust has been well highlighted for traffic locations in Athens by the observed significant statistical associations between coarse particle and tailpipe emitted gaseous pollutants (Chaloulakou et al, 2003). The importance of wind induced resuspension of soil particles should also not be ignored (Harrison et al., 2001, Atmos. Environ. 35 3667-3679).

- P. 7666, l.1: This sentence is a little bit confusing. I assume 25% of southern winds, and not of "dust events". ANSWER:As suggested by the reviewer the sentence has been rephrased. "On the other hand, southern winds carrying dust from arid areas of Sahara are very frequent in the area during spring and autumn contributing up to 25% of the prevailing air masses". Page 7, lines 159-161.

- P. 7667, l. 5: "In PM10 and PM2.5, levels : : :". - P. 7667, l. 15: "In Athens, the most likely formation pathway : : : " ANSWER:The sentences have been corrected as

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suggested by the reviewer. Page 11, line 253 and 271.

- P. 7669, l. 1-2: Possible implication of acidic conditions on SOA formation?

ANSWER: We agree with the reviewer remark and a sentence was added “ Laboratory studies suggest that heterogeneous acid-catalyzed reactions in the particle phase are important mechanisms for SOA formation and that particle acidity has an impact on SOA yield”. Page 13, lines 311-315.

- P. 7669, section 3.2.3: Discussion on the scavenging of gaseous precursors by mineral dust may be needed. Any impact on the concentration levels of these gaseous species? A sentence on the role of dust as scavenger of gaseous precursors was added in the manuscript. ANSWER: Field and laboratory studies indicated that mineral dust particles can serve as reaction surfaces for different species, including those of man-made origin (Mamane and Gottlieb, 1992; Kocak et al., 2007). Gaseous species such as SO<sub>2</sub>, N<sub>2</sub>O<sub>5</sub>, HNO<sub>3</sub> and O<sub>3</sub> can react with mineral dust particles (Mamane and Gottlieb, 1992; Dentener et al., 1996) and result in the modification of optical properties, size distributions and chemical composition of the aerosols (Kouvarakis et al., 2002, Vrekoussis et al., 2005). Page 16-17, lines 420-429.

- P. 7670, l. 4-5: How long could the long-range transport? What does mean “larger spatial scale”?

ANSWER: Markou and Kassomenos (2010) in their five years trajectory analysis investigated possible source areas contributing to Athens pollution and found fast moving air masses crossing the hotspots of the Po Valley and the Ruhr area in Central Europe, enriched with emissions from major industrial combustion operations in Northwestern Greece, on the their pathway to Athens and Crete. Transport of air masses originating from the Black sea and the Donbas area in Ukraine, and traverse the East of the Balkan Peninsula, is also frequent and could account for a possible pathway. In order to address this issue a sentence was added in the manuscript. Page 10, lines 244-246.

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- 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.

ANSWER: The reviewer is right. Soil dust and car/road abrasion co-exist in our locations in the GAA however the distinction between them is difficult in a qualitative way and is out of scope of this manuscript. Given the area characteristics we can assume that road dust is expected to be more important at Goudi, a down town Athens heavy traffic area with almost no unpaved zones. On the other hand Lykovrissi is an agricultural area with uncovered land and wind resuspension of soil particles, hence it is expected to be a main source as indicated by the higher Ca level.

- P. 7672, l. 18-20: not accurate. Some North African countries banned leaded gasoline for several years. ANSWER: As suggested by the reviewer the sentence has been rephrased. However based on UNEP, until January 2007 in several N. African countries such as Algeria, Tunisia and Morocco leaded fuel was still in use (<http://www.unep.org/transport/pcfv/PDF/LeadReport.pdf>).

- P. 7673, l. 10: "In total, crustal : : :" - P. 7675, l. 5: " : : continuously monitored. " - P. 7675, l. 16: "Crustal material was accounted ...". The sentences have been corrected as suggested by the reviewer. Page 19, line 450 and P. 21, line 504, 514.

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Interactive comment on *Atmos. Chem. Phys. Discuss.*, 11, 7659, 2011.

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