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Atmospheric Chemistry and Physics
Discussions

Interactive comment on "AIRUSE-LIFE+: Estimation of natural source contributions to urban ambient air PM_{10} and $PM_{2.5}$ concentrations in Southern Europe. Implications to compliance with limit values" *by* Evangelia Diapouli et al.

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The paper addresses the question of the natural contributions to PM levels which – although not dealing with novel concepts – has important implications for policy abatement strategies and measures. The paper novelty stands in the attempt of evidencing differences when comparing different approaches and assessing major causes of uncertainties. The paper is clear and well written. The datasets presented are suitable for such kind of analysis. As for the methods used, they are generally scientifically sound although a major concern is related to the algorithm reported for the stoichio-

metrically derived mineral dust which is not compliant to the mentioned reference and – in general– does not consider Ca, Fe, and K contributions. Maybe that it is simply a typo error but – if it is not the case – a large part of data analysis should be done again and the text modified accordingly. Another issue concerns the linear regression analyses which should be represented in more suitable way and the equations must be reported with all relevant parameters (e.g. with intercepts, uncertainties and confidence levels). The referee suggests to accept the paper with major revisions, which should take carefully into consideration the specific comments reported below.

The authors would like to thank the reviewer for the suggestions and positive remarks which assisted us in improving the manuscript. We address all general comments and suggestions within the answers given below to the specific comments.

Specific comments: - Please correct the misuse of the possessive case throughout the text (e.g. line 17 page 1 "sources' contribution", line 13 page 3 "pollutants' removal", etc.).

The possessive form has been corrected.

- Lines 16-17: Please specify if referring to aerodynamic diameter or other equivalent diameters.

At this point in the introduction, the term fine and coarse refers to atmospheric aerosol regardless of equivalent diameter. Equivalent diameters are necessary to consider when we refer to aerosol measured with a specific measurement technique. For example, optical particle sizers also separate fine and coarse particles in terms of their own equivalent optical size. Aerodynamic diameters are relevant to this work because, as can be seen further down, data were obtained by samplers using PM10 and PM2.5 heads which fractionate particles in terms of aerodynamic diameter, but at this point it is not appropriate to specify this yet. On the other hand, it is trivial to mention that PM10 is an aerosol metric by definition referring to aerodynamic diameter.

- It would be useful for the reader to add references for BSC-DREAM8b and FLEXTRA model.

References have been added for both models.

- Page 5 line 13: The algorithm reported in Marcazzan et al. (2001) is not the one written here. Please check it carefully in the original paper by Marcazzan et al. (2001) and change the data/comments accordingly if obtained with the wrong formula.

The formula proposed by Marcazzan et al. (2001) is: [Mineral dust]=1.15×(1.89×Al+2.14×Si+1.67×Ti+1.4×Ca+1.2×K+1.36×Fe) Marcazzan et al. (2001) also clarify that only the part of K and Fe of natural origin is included in this calculation. Taking this into account, and considering that Ca, K and Fe have shown to have in the study areas some anthropogenic sources (industrial, construction fugitive sources, traffic and biomass burning), these three elements were replaced in the calculation formula through their typical crustal ratios with respect to Al. For that reason, in the formula we used, Al is multiplied by 3.79 instead of 1.89 (as in the formula proposed by Marcazzan et al., 2001). This methodology has been initially proposed by Nava et al. (2012) and was also adopted in Amato et al. (2016). In the revised text this is better explained and two more references (Nava et al., 2012 and Mason, 1966) were added to Marcazzan et al. (2001), thus clarifying the calculation algorithm used in the present work.

- Line 18 page 5: Here mean contributions for African dust stands for the average obtained considering all the approaches reported in par. 2.2? Please specify.

The mean annual contributions of the studied natural sources to PM10 and PM2.5 concentrations are reported in Tables 2 and 3. This is now clearly stated in the text (in the beginning of section 3.1). In addition, the methodology applied for estimating these contributions is now described in this section.

- Line 2 page 6: Please give an explanation for the African dust events during winter in Porto while in Barcelona they were recorded mostly during summer and at the other two cities in springtime.

An explanation has been added, along with a new reference, where the annual cycle of African dust transport is discussed (Moulin et al., 1998).

- Figure 4: are you sure that the suburban character of the monitoring site in Athens does not affect the results? The large difference in the proportion between anthropogenic and natural sources is suspicious.

The suburban character of the site does influence the results, especially during exceedance days. The site is not close to direct anthropogenic emissions (as noted in Amato et al., cited in section 2.1), thus exceedances of EU limit values are rare and are almost entirely attributed to African dust events. During the studied year, 79As concentration values, Sahara dust on average provides 4 out of the 20 μg m-3 of PM10, as shown in Table 2. However, during exceedance days, the average PM10 concentration is 67 μg m-3 out of which 53 μg m-3 is African dust. It has to be noted that dust outbreaks lead to exceedances only in the case of the suburban Athens site. The significant impact of African dust on PM10 concentration levels observed in the city of Athens have been also documented elsewhere. Mitsakou et al. (2008) report on the effects of dust transport on air quality in several Greek urban areas during the period 2003-2006, based on PM10 concentration data obtained from stationary monitoring stations and dust concentration data estimated by the SKIRON model. The results show that the monthly mean PM10 concentrations measured at a suburban station in Athens have maximum during the month of April, when African dust concentrations are also high. Long-range transport of dust affect the exceedances of the 24 h PM10 limit value by 25 and 34

Mitsakou, C., Kallos, G., Papantoniou, N., Spyrou, C., Solomos, S., Astitha, M., and Housiadas, C.: Saharan dust levels in Greece and received inhalation doses, Atmos. Chem. Phys., 8, 7181-7192, doi:10.5194/acp-8-7181-2008, 2008.

- Fig. (not Fog.) 6-9: it is not clear to the referee why the authors represented all these regression lines in a log-log scale. Moreover, 1) the regression lines often show a clear intercept which has not been reported in the regression equation; 2) the values reported for squared-R seem not to correctly represent real data dispersion. How large is the associated uncertainty? How much is this linear regression compatible with a true-linear model? The referee suggests to represent the data in a linear scale, possibly making an orthogonal/Deming regression in order to take into account uncertainties in both x- and y-data as well as the compatibility with a linear model within a given confidence level. Last but not least, check if the MIN-STOICH data reported here have been calculated with the formula reported in the text or using the original Marcazzan et al. algorithm.

The typo has been corrected in Fig. 6. The MIN-STOICH data have been calculated according to Nava et al. (2012), as explained in details above. All intercepts in the regressions presented in Fig. 6-9 were very low (below 10The log-log scale has been selected for all regressions included in Fig. 6- 9, because of the wide range of values and the high number of zero values (due to the episodic character of African dust events). The reader can have a better visual understanding of the level of discrepancy in the lower values of calculated net dust metrics investigated here and the estimated dust calculated by transport models. This allows the reader to have an understanding of the dust mass concentration levels that this sensitivity analysis is meaningful (mostly > 5 μg m-3) We suggest to compare both graphs representations given here (Figure $1_{R}C1)andpossiblyagreewithusthattheloggraphprovidesabetterrepresentationoftherelations$

The advantage of log plots illuminating the concentration levels where the uncertainty on the dust component estimates by the different methods becomes significant, is also identified by reviewer 3.

- Line 28 page 8: also this “dirty” profile for African dust in Athens suggests that the suburban character of the monitoring site may affect the results. Please add a comment in the text.

The chemical profiles depicted in Fig 11 are: 1) the local dust profile for Florence, 2) the African dust profile for Florence and 3) the mixed (local and African) dust profile for Athens (denoted as “mineral dust”). There is no African dust profile for Athens, since we could not separate the local and African dust by PMF analysis in Athens (and similarly in Barcelona, Porto and Milan). This is clearly stated in Page 7, Lines 28-30 and Page 8, Lines 5-6 of the initially submitted manuscript. The Athens mineral dust profile is indeed “dirty”, as is the Florence local dust profile. This enrichment with anthropogenic components is already discussed and is found in the mineral dust profiles obtained by PMF for all 5 cities (Amato et al., 2016). So this is a common finding for all sites and although the urban character of the sites introduces a certain degree of contamination, it is not specific for Athens or the nature of the Athens site. The Saharan dust may be also enriched with anthropogenic components, as shown for the Florence Saharan dust chemical profile (depicted in Figure 11) and documented elsewhere as well (Levin et al., 1996; Sun et al., 2005).

Levin Z., Ganor E. and Gladstein V., (1996) “The effects of Desert Particles Coated with Sulfate on Rain Formation in the Eastern Mediterranean”, Journal of Applied Meteorology. 35, pp1511-1523.

Sun Y., Zhuang G., Wang Y., Zhao X., Li J., Wang Z., An Z. (2005) “Chemical composition of dust storms in Beijing and implications for the mixing of mineral aerosol with pollution aerosol on the pathway”, Journal of Geophysical Research. 110, D24209, doi:10.1029/2005JD006054. Sun Y., Zhuang G., Wang Y., Zhao X., Li J., Wang Z., An Z. (2005) “Chemical composition of dust storms in Beijing and implications for the mixing of mineral aerosol with pollution aerosol on the pathway”, Journal of Geophysical Research. 110, D24209, doi:10.1029/2005JD006054.

- Table 4: is there any explanation for the relatively higher intercept and slope given by BSC$_{D}REAMmodelatsurfacelevelwhencomparedtoSKIRONmodel?$

The differences observed in the slopes and intercepts calculated for SKIRON/Dust and

BSC-DREAM8b v2.0 models are related to the parametrizations used by each model for simulating the desert dust cycle, and more specifically with respect to the dust uptake scheme and the soil characterization. This explanation has been also added in the revised manuscript.

- Figure 12: same comment reported above for Figs. 6-9

Based on the reviewer's suggestions, we have re-analysed the data by applying the Deming regression and we have included the new plots, in log-log scale. The Deming regression has been also applied for the comparison between the calculated net dust loads and the modelled dust concentrations presented in Table 4. All new results are now included in the revised manuscript. A comparison between the linear and log-log scale figures is given in Figure $2_RC1.Webelievethatthelog-logscaleprovidesabettervisualrepresentationofthedata.$

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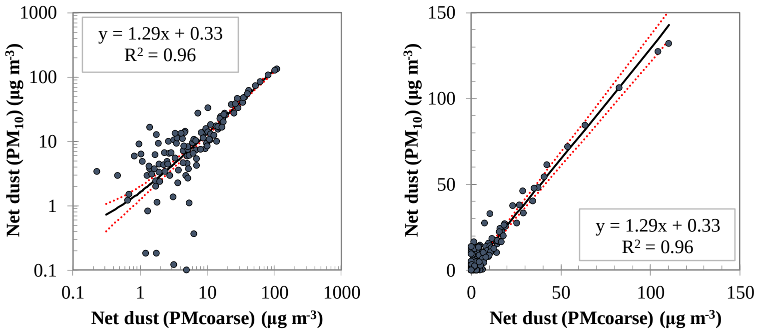

Figure 1_RC1: Regression analysis of Net dust concentrations calculated from regional background PM_{10} and PMcoarse ($PM_{2.5-10}$) concentrations for Athens, in log-log (left) and linear scale (right). The black line corresponds to the linear regression equation, while the red dotted lines are the upper and lower bounds, at 95% confidence interval.

Fig. 1. Figure 1_RC1: Regression analysis of Net dust concentrations calculated from regional background PM10 and PMcoarse (PM2.5-10) concentrations for Athens, in log-log (left) and linear scale (right).

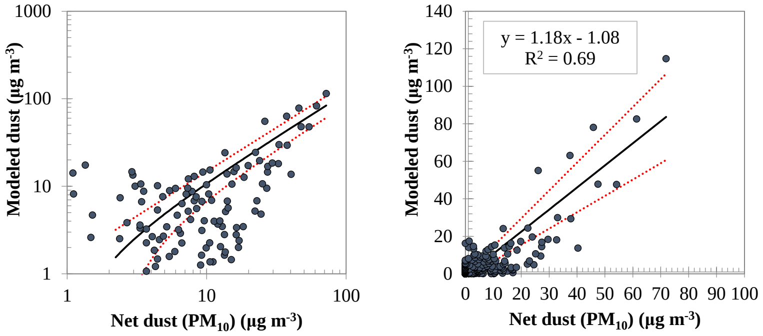

Figure 2_RC1: Regression analysis between net dust calculated through PM_{10} regional background data and dust concentrations modelled at surface level by SKIRON/Dust for the city of Athens, in log-log (left) and linear scale (right). The black line corresponds to the linear regression equation, while the red dotted lines are the upper and lower bounds, at 95% confidence interval.

Fig. 2. Figure 2_RC1: Regression analysis between net dust calculated through PM10 regional background data and dust concentrations modelled by SKIRON/Dust, in log-log (left) and linear scale (right).