Response to Referees for manuscript ACP-2016-781

Dear Editor,

Please accept our revised manuscript entitled: "AIRUSE-LIFE +: Estimation of natural source contributions to urban ambient air PM10 and PM2.5 concentrations in Southern Europe. Implications to compliance with limit values". We would like to thank the referees for their constructive comments and suggestions. Our answers to the referees' questions (*in bold italics*) as well as a detailed description of all changes made to the manuscript are included below. Please find attached also our revised manuscript with all changes marked.

We remain at your disposal.

Sincerely,

E. Diapouli

Anonymous Referee #2

Received and published: 12 October 2016

This manuscript describes a well performed analysis of the contribution of natural sources to PM concentrations in 5 cities in Southern Europe. One year of data collection of PM and chemical analysis was performed. The data analysis is based on various state-of-the-art techniques. The results are presented and discussed appropriately. The text is well written and the reasoning well documented. The manuscript is a nice piece of craftmanship. It lacks scientific inspiration or innovation. Considering the Aims and Scope of ACP, i.e. "...The journal scope is focused on studies with general implications for atmospheric science rather than investigations that are primarily of local or technical interest..." (http://www.atmospheric-chemistry-and-physics.net/about/aims_and_scope.html***), the manuscript is barely publishable there.***

We are obliged to the reviewer for the positive remarks. Although his suggestion on the "barely publishable" quality of the manuscript is vague and unclear, we make an effort below to respond to all the points raised. It is noted that the notion of net dust as described in the current literature has never been tested, as far as we know, against the physical components of dust as described by the stoichiometric calculated component, the PMF derived net dust component and the estimated dust component by transport models like Skiron. All this is done at the same harmonized dataset and for several cities. This kind of sensitivity analysis to our view is innovative, has never been tried before and is introducing new methodology which can be guidance to other studies, as well as having serious policy making impact in South Europe. The continent of Europe and the effect of the Sahara which is the largest dessert on earth can hardly be considered of local interest. We believe that these are the kind of elements that are very well within the Aims and Scope of ACP.

Specific comments:

The reasoning for the obtrusive mentioning of "AIRUSE-LIFE +", even in the title of the manuscript, becomes slightly clear after reading the acknowledgements, but not earlier than that. The acronym should be deleted from the entire manuscript except the acknowledgements.

It is not understood why the reviewer finds the title of the LIFE+ programme funding the major part of this research obtrusive and requests the removal from the title. This is against ethics of the scientific community where, when the scientific work is the result of an international programme like in this case AIRUSE, its title is often used throughout the text, e.g instead of mentioning the campaigns in each city we refer to them as AIRUSE cities, the results obtained as AIRUSE data and so on. In many cases in the literature large experiments or even smaller ones are mentioned even in the title (see ACE experiments, INDOEX, SUB-AERO, PARTEMIS, etc.). We therefore request to pass on this suggestion by the reviewer as it does not have any scientific nature.

page 2, line 10: it is not clear what is meant by "This natural background ..." because in

the line before, it is talked about "both to natural sources and anthropogenic long range transported particles".

This phrase has been revised in order to be more precise.

page 4, line 26: how is nss-Na computed?

nssNa is computed based on a typical earth's crust elemental ratio (with respect to Al), as:

$nssNa = 0.348 \times Al$

This formula is mentioned in the text on Page 4, Line 27 (of the initially submitted manuscript), just below the formula describing the calculation of ssNa.

Fig. 12: Figures should not have headings, just subscript captions

The headings have been replaced by relevant labels and the caption has been slightly modified in order to reflect the annotations on the two figures.

Anonymous Referee #1

Received and published: 18 October 2016

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 stoichiometrically 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 PM_{10} and $PM_{2.5}$ 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 PM_{10} 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:

$$[Mineral\ dust] = 1.15\times(1.89\times Al + 2.14\times Si + 1.67\times Ti + 1.4\times Ca + 1.2\times K + 1.36\times Fe)$$

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 PM_{10} and $PM_{2.5}$ 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, 79% of the mass concentration during exceedance days was related to African dust

(Page: 7, Lines: 5-6 of the initially submitted manuscript). The suburban character of the site is also commented on the text: "The Athens suburban site on the other hand is a characteristic example of the effect of natural sources in background urban environments." (Page: 7, Lines 3-4 of the initially submitted manuscript). The low levels of PM_{10} at this suburban site definitely govern this behaviour and the numerical results presented here; Sahara dust events are characterized by high PM_{10} concentration values definitely much higher thav the PM_{10} levels at the Athens suburban site and there is therefore nothing suspicious about the fact that almost all exceedances are occurring during these events.

As concentration values, Sahara dust on average provides 4 out of the 20 μg m^{-3} of PM_{10}, as shown in Table 2. However, during exceedance days, the average PM_{10} 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 PM_{10} 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 PM_{10} concentration data obtained from stationary monitoring stations and dust concentration data estimated by the SKIRON model. The results show that the monthly mean PM_{10} 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 PM_{10} limit value by 25 and 34% during the spring and autumn periods respectively. In addition, for the year 2003, 65.7% of the daily exceedances are attributed to "African origin".

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 10% of average estimated dust concentrations). Based on the reviewer's suggestions, we have re-analysed the data by applying the

Deming regression and we have included in the revised manuscript all new plots. The correlation coefficients remain the same. Some slopes have changed, while the intercepts are again very low (in some cases lower that the ones calculated through simple linear regression). In the new plots: 1) intercepts are reported along with slopes; 2) Squared-R is also reported; 3) the 95% confidence interval of the regression line is provided.

The 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 and possibly agree with us that the log graph provides a better representation of the relationship between the two parameters compared, especially in displaying the level uncertainty in the lower end of concentration values. In both cases, the Deming regression analysis has been applied, while intercepts are also included.

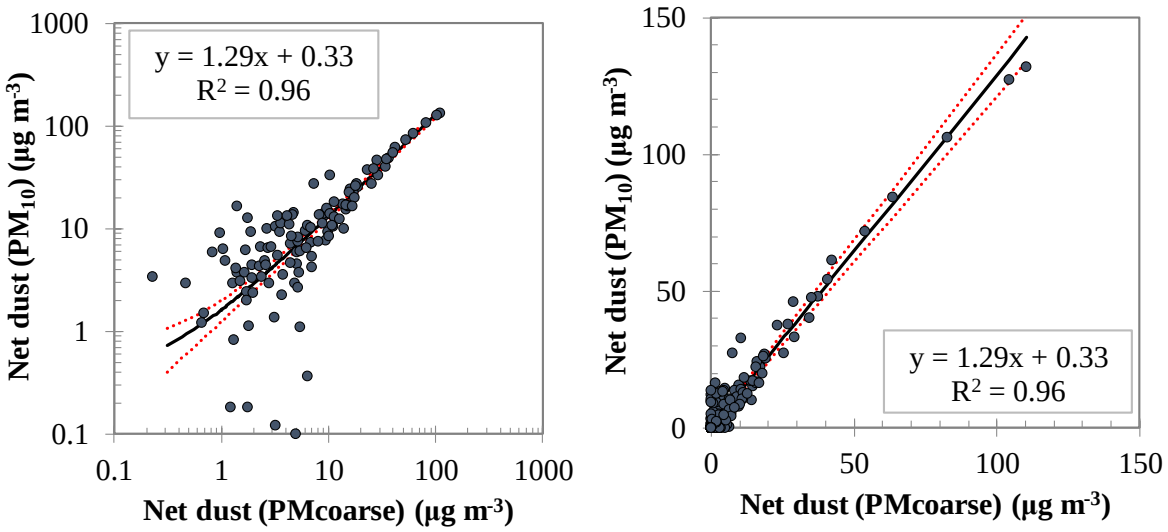

Figure: Regression analysis of Net dust concentrations calculated from regional background PM_{10} and PMcoarse ($PM_{2.5-10}$) concentrations for Athens, in log-log (left) and linear scale (right). The black line corresponds to the linear regression equation, while the red dotted lines are the upper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

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.

- Table 4: is there any explanation for the relatively higher intercept and slope given by BSC_DREAM model at surface level when compared to SKIRON model?

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 below. We believe that the log-log scale provides a better visual representation of the data.

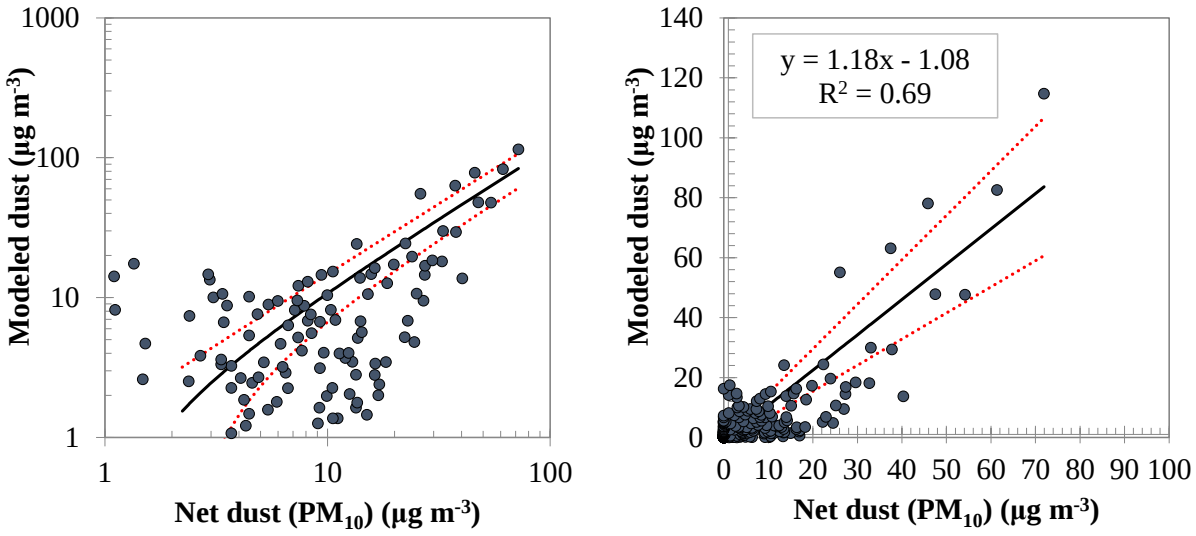

Figure: Regression analysis between net dust calculated through PM_{10} regional background data and dust concentrations modelled at surface level by SKIRON/Dust for the city of Athens, in log-log (left) and linear scale (right). The black line corresponds to the linear regression equation, while the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

Anonymous Referee #3

Received and published: 20 October 2016

General Comments

This paper deals with desert dust outbreaks in southern Europe, more specifically with the contribution of natural aerosols to mass concentrations measured in five urban environments in Southern Europe. This is an interesting work, well written and very well conducted, with results properly presented and examined (with the exception of the uncertainties on measured and calculated values). In this respect, I really appreciated the sensitivity analysis on the estimation of African dust contributions. However, if this study addresses some relevant scientific questions, many aspects of desert dust outbreaks in the Mediterranean environment have been broadly studied in recent years (e.g. Stafoggia et al., Environ. Health Perspect., 124 (4), 413-419, 2016 and references therein or Calastrini et al., Advances in Meteorology (2012), http://dx.doi.org/10.1155/2012/246874 and references therein). Therefore, the novelty of this work is limited anyway and it is difficult for me to assess the real contribution of this study to a better knowledge of the Mediterranean atmospheric environment. As the authors pointed out, in the studied urban areas, the natural contribution to the atmospheric particulate load during days in exceedance is very limited, except in Athens, which is not really new (see for example Grivas et al., STOTEN, 389 (2008) 165-177). From a general appraisal point of view, I suggest to the authors to strengthen their discussion about uncertainties in the quantification of the natural contributions, to reinforce their conclusions, before considering publication of this work in a high ranked journal as ACP.

We would like to thank the reviewer for highlighting the interesting aspects of this work and we will try to respond to the comment on uncertainty estimation in the specific comments below.

We have to point out that despite the numerous studies addressing African dust outbreaks, this study is one of the few that is based on an organized annual campaign simultaneously in 5 urban areas and also performs an innovative sensitivity analysis of the calculated African dust loads.

Specific Comments

Page 4, lines 9 to 13: Please add references about the BSC-DREAM8b and FLEXTRA models.

A reference has been added for each model.

Page 5, line 13: Equation (2) is not the correct formula reported in the Marcazzan's study! In Marcazzan et al. (2001), the mineral dust concentration is reconstructed from: Mineral Dust = 1.15(1.89Al + 2.14Si + 1.67Ti + 1.4Ca + 1.2K + 1.36Fe). Please check your "Min-Stoch" data to verify if they have been obtained with the equation (2) or with the original Marcazzan et al. (2001) formula.

We agree with the reviewer. A modified formula has been used for the calculation of the mineral dust concentration in the current work. Marcazzan et al. (2001) noted that only the part of K and Fe of natural origin is included in the calculation of mineral dust concentration. 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.

Table 2 (page 16) and Table 3 (page 17):

Please report uncertainties regarding mass contributions (g.m-3) and relative contributions (%) of natural sources to PM10 and PM2.5 concentrations for the five studied cities.

The uncertainties of the contributions of the different natural sources have been calculated and are reported in Tables 2 and 3. The text has been also modified in order to include information on the methodology used for calculating uncertainty and to comment on the estimated relative uncertainties.

Figures 6 to 9 (pages 22 to 24): They are clearly intercepts different from 0 in some reported regression lines, which are not considered in the regression equations: Could the authors examine and discuss the impact of these simplifications on their conclusions?

We agree with the reviewer. The intercepts were very low (below 10% of average concentrations); nevertheless, the regression equations should include both slopes and intercepts. All regressions have been now corrected and the new figures include the slope, intercept and R^2.

Page 9, lines 4 to 6 and Figure 12 (page 25): They are undoubtedly no correlation between measured and calculated dust concentrations for concentrations below 10 g.m-3. I suggest to the authors to clearly indicate that in their discussions on the use of the SKIRON and BSC DREAM8b v2.0 models.

The phrase has been modified according to the reviewer's suggestion and it is now stated that no correlation was observed between net dust loads and modelled dust concentrations for values below 10 $\mu g\ m^{-3}$, as shown in Figures 12a and b.

Technical corrections

- Page 4, line 27: please change Al for Al in brackets for the non-sea salt Na calculation.

Corrections have been made to equations (1) and (2).

- Page 22: Fig.6 not Fog.6

The correction has been made.

- Page 25, Fig.11: please, use a log-scaling for the y axis (Mass Fractions), as in Fig.12, for example.

A log-scale is already used for the y axis.

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.

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Abstract

~~Natural sources' c~~The contribution of natural sources to ambient air particulate matter (PM) concentrations is often not considered; however, it may be significant for certain areas and during specific periods of the year. In the framework of the AIRUSE-LIFE+ project, state-of-the-art methods have been employed for assessing the contribution of major natural sources (African dust, sea salt and forest fires) to PM concentrations, in Southern European urban areas. 24 h measurements of PM_{10} and $PM_{2.5}$ mass and chemical composition were performed over the course of a year in five cities: Porto, Barcelona, Milan, Florence and Athens. Net African dust and sea salt concentrations were calculated based on the methodologies proposed by EC (SEC 2011/208). The contribution of uncontrolled forest fires was calculated through receptor modelling. Sensitivity analysis with respect to the calculation of African dust was also performed, in order to identify major parameters affecting the estimated net dust concentrations. African dust contribution to PM concentrations was more pronounced in Eastern Mediterranean, with the mean annual relative contribution to PM_{10} decreasing from 21 % in Athens, to 5 % in Florence, and around 2 % in Milan, Barcelona and Porto. The respective contribution to $PM_{2.5}$ was calculated equal to 14 % in Athens and from 1.3 to 2.4 % in all other cities. High seasonal variability of contributions was observed, with dust transport events occurring at different periods in the Western and Eastern Mediterranean basin. Sea salt was mostly related to the coarse mode and also exhibited significant seasonal variability. Sea salt concentrations were highest in Porto, with average relative contributions equal to 12.3 % for PM_{10}. Contributions from uncontrolled forest fires were quantified only for Porto and were low on an annual basis (1.4 % and 1.9 % to PM_{10} and $PM_{2.5}$, respectively); nevertheless, contributions were greatly increased during events, reaching 20 and 22 % of 24 h PM_{10} and $PM_{2.5}$ concentrations, respectively.

1 Introduction

Atmospheric aerosols may be emitted by both natural and anthropogenic sources. Given that exposure to particulate matter (PM) is mainly related to urban environments where anthropogenic activities lead to increased concentration levels, natural sources are often not considered. Nevertheless, their contribution may be significant, especially for certain areas and during specific periods of the year. It has been estimated that the natural contribution to PM may range from 5 % to 50 % in different European countries (Marelli, 2007). Background annual average PM_{10} mass concentration for continental Europe is 7.0 ± 4.1 μg m^{-3} (Van Dingenen et al., 2004) and is attributed both to natural sources and anthropogenic long range transported particles. This ~~natural~~ background level shows regional variations, and in some cases (in particular for the Southern European countries) naturally emitted PM may contribute significantly, causing even ~~cause~~ exceedances of air quality standards (Pey et al., 2013; Querol et al., 2009; Rodriguez et al., 2001; Querol et al., 1998). The main natural sources affecting ambient PM levels are wind-blown soil dust, sea salt, wildfires, volcanic ash and biogenic aerosol (Viana et al., 2014).

Wind-blown soil dust relates to the transport of mineral dust particles from agricultural and arid or semiarid regions (Ginoux et al., 2012). North Africa is the main source of desert dust for European countries (Stuut et al., 2009). Most of these particles are very coarse (diameter ≥10 μm) and are thus deposited close to the source region, while a significant amount of coarse particles (diameter around 1-10 μm) can be transported over long distances. An estimation of the emission flux of desert aerosols that is subject to long-range transport is of the order of 1500 Tg/yr (Papayannis et al., 2005).

Sea salt aerosol is emitted from the sea surface, through bubble-bursting processes resulting in sea-spray particles with sizes ranging from sub micrometre to a few micrometres (O'Dowd and de Leeuw, 2007). Sea salt aerosols play an important role in atmospheric chemistry, providing the surface for heterogeneous reactions and acting as a sink for anthropogenic and natural gaseous pollutants (Tsyro et al., 2011). The presence of sea salt aerosols in the atmosphere was shown to significantly alter the regional distribution of other inorganic aerosols, namely sulphate, nitrate and ammonium (van den Berg et al., 2000). It may also appear in both the coarse and fine fraction (Eleftheriadis et al., 2014). Furthermore, sea salt helps to reduce the acidity of the air by providing base cations (Tsyro et al., 2011).

Wildfires relate to the burning of forests and other vegetation, mostly through natural processes. Large-scale forest fires are a major PM source, while smoke plumes may be transported over thousands of kilometres, affecting air quality at local, regional and global scale (Faustini et al., 2015; Diapouli et al., 2014). Volcanic ash can also have a global impact due to the fact that emissions may be injected into the stratosphere but have more infrequent occurrence (von Glasow et al., 2009). Biogenic aerosol is emitted by vegetation and may be of primary or secondary origin (Caseiro et al., 2007).

Taking into account that natural sources cannot be controlled, while their contribution varies between the European countries, EU legislation has allowed for the subtraction of PM concentrations of natural origin when Member-States assess and report

attainment of air quality standards. Apart from environmental reporting, quantification of natural contributions to PM levels is important in terms of exposure assessment as well. Many epidemiological studies have demonstrated the detrimental effects of particulate matter pollutants to human health (Ostro et al., 2015; Samoli et al., 2013). The distinct physico-chemical and toxic properties of anthropogenic and naturally emitted aerosol call for a differentiation of peak concentration days due to anthropogenic pollution or natural events, when assessing population exposure and dose-effect relationships. On the other hand, extreme natural events that lead to very high exposures may still adversely affect human health, especially in the case of exposures on markedly different aerosol size fractions (Zwozdziak et al., 2016) or sensitive population subgroups (Perez et al., 2008).

High background concentration levels are frequently reported in Southern European countries, often due to the enhanced contribution by natural sources. The Mediterranean climate, characterized by increased solar radiation and low rainfall rates, promotes aerosol production and reduces the potential for dispersion ~~pollutants' removal~~ and ~~-~~removal of pollutants~~dispersion~~ (Lazaridis et al., 2005). The vicinity of Southern Europe to North Africa on the other hand results in frequent and intense dust outbreaks, with high loads of dust from desert regions transported across the Mediterranean, which often leads to exceedances of air quality limit values (Pey et al., 2013; Nava et al., 2012; Athanasopoulou et al., 2010; Querol et al., 2009; Gerasopoulos et al., 2006; Kallos et al., 2006; Rodriguez et al., 2001; Querol et al., 1998-).

In the framework of the AIRUSE-LIFE+ project, the contribution of major natural sources to PM_{10} and $PM_{2.5}$ concentration levels was quantified for five Southern European cities (in Portugal, Spain, Italy and Greece). The project focused on two sources: the long-range transport of African dust and sea salt. The contribution from wildfires has been also detected and quantified in one city (Porto). In addition, a sensitivity analysis on the calculation of African dust contributions was performed, providing useful insight into the key factors affecting the quantified dust concentrations.

2 Experimental methods

2.1 Sampling and analysis

Year-long measurement campaigns were performed from January 2013 to February 2014 in five Southern European cities: Porto, Barcelona, Milan, Florence and Athens (Fig. 1). The cities were selected in order to cover Southern Europe from West to East, as well as sites by the sea and inland. 24 h sampling of PM_{10} and $PM_{2.5}$ (00:00 – 23:59) was performed once every 3 days for a full year in all cities. Additional sampling was conducted during days when African dust episodes were forecasted, in order to better characterize the contribution of this source to PM levels. Comprehensive chemical characterization of PM_{10} and $PM_{2.5}$ samples was performed for the determination of organic and elemental carbon, carbonate carbon, levoglucosan, ion species and major and trace elements. The measurement periods, monitoring sites and number of valid chemical speciation

samples for each city are presented in Table 1. Details about sites, sampling and analytical procedures are provided in detail in Amato et al. (2016).

2.2 Quantification of natural sources contribution

Contributions of sea salt and African dust to PM_{10} and $PM_{2.5}$ concentrations were quantified based on EU guidelines (SEC 2011/208) for all five AIRUSE cities. Specifically for African dust, ~~P~~potential African dust transport events at each city were identified through: (i) 5-day backward air mass trajectories obtained every 3 hours and at 3 heights (500, 1000 and 1500 m a.s.l.) by Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT) Model (Draxler and Rolph, 2003); (ii) dust load surface concentrations provided by the Barcelona Supercomputing Centre (BSC)-DREAM8b v2.0 Atmospheric Dust Forecast System (Basart et al., 2012); (iii) dust concentrations at surface levels and at three additional heights (reaching up to ~950 m a.s.l.) provided by SKIRON/Dust forecast model (Spyrou et al., 2010); (iv) 7-day backward air mass trajectories obtained every 6 hours and at 3 heights (500, 1000 and 1500 m a.s.l.) by Flextra model (Stohl and Seibert, 1998). Following the identification of days potentially affected by long-range transport of African dust, net African dust concentrations were calculated based on continuous 24 h PM data from background sites representative of the regional background concentrations at the studied cities. PM_{10} and $PM_{2.5}$ data available from the National Monitoring Networks operating at the five cities were used for this analysis. 30-days moving averages of the previous and next 15 days of the regional background concentrations were calculated, excluding days with potential African dust transport. Averages corresponded to 40th percentiles in the case of Porto, Barcelona and Florence (Escudero et al., 2007). In Milan and Athens, a more conservative indicator, the 50th percentile, was selected since it was found to reproduce better PM background concentrations (SEC 2011/208). Net African dust load was quantified for each day forecasted as potential dust event by at least one of the above mentioned models, as the observed increase in concentration with respect to the calculated moving average for that day (representative of background concentration not affected by dust transport).

Sea salt (ss) was calculated based on major sea salt components (Cl and Na) and typical elemental ratios for sea water (Mg/Na, K/Na, Ca/Na and SO_4^{2-}/Na) and earth's crust (Na/Al) (Calzolai et al., 2015):

$$Sea\ salt = [ssNa] + [Cl] + [ssMg] + [ssK] + [ssCa] + [ssSO_4{}^{2-}], \quad (1)$$

where:

$$[ssNa] = [Na] - [nssNa]$$

$$[nssNa] = 0.348 \times [Al]$$

$$[ssMg] = 0.119 \times [ssNa]$$

$$[ssK] = 0.037 \times [ssNa]$$

$$[ssCa] = 0.038 \times [ssNa]$$

$[ssSO_4^{2-}] = 0.253 \times [ssNa]$.

The contribution of wildfires was estimated only for Porto, where several wildfires were registered during late August and September of 2013. A biomass burning factor was obtained by receptor modelling (Positive Matrix Factorization, PMF), with several peak concentrations during the wildfires' period; thus, these concentrations were attributed to wildfires and classified as natural source contributions. Details on PMF analysis and results are presented in Amato et al. (2016).

2.3 Sensitivity analysis on the estimation of African dust contribution

A sensitivity analysis was performed in order to assess the main parameters affecting the quantification of net African dust concentrations. Specifically, the following parameters were examined with respect to the calculation of net African dust: (i) the identification of dust transport episodes by different modelling tools; (ii) the use of PMF analysis for the identification of a mixed mineral dust source or a separate African dust source; (iii) the use of alternative input concentration data, such as the coarse PM fraction ($PM_{2.5-10}$) and the mineral component of PM_{10}, calculated either by PMF analysis or reconstructed from elemental concentrations based on stoichiometry (Nava et al., 2012; Marcazzan et al., 2001):

$$[Mineral\ dust] = 1.15 \times (3.791.89 \times [Al] + 2.14 \times [Si] + 1.67 \times [Ti] + 1.4 \times [soilCa] + 1.2 \times [soilK] + 1.4 \times [soilFe]). \quad (2)$$

where the soil fractions of Ca, K and Fe have been calculated using their typical crustal ratios with respect to Al (Mason, 1966): [soilCa]=0.45×[Al], [soilK]=0.32×[Al], [soilFe]=0.62×[Al].

Net dust concentrations calculated by PM_{10} regional background data were also compared to the dust concentrations provided by SKIRON/Dust and BSC - DREAM8b v2.0 transport models.

3 Results and discussion

3.1 Contribution of natural sources to PM concentrations and exceedances

Mean annual contributions of long-range transported African dust, sea salt and wildfires (estimated only for Porto) to PM_{10} and $PM_{2.5}$ concentrations at each city, are presented in Tables 2 and 3, along with their respective uncertainties. African dust and sea salt concentrations were calculated based on SEC 2011/208. Only in the case of Florence, where PMF analysis produced a separate source attributed to the long-range transport of Saharan dust, the concentrations of African dust for PM_{10} and $PM_{2.5}$ reported in Tables 2 and 3 correspond to the contributions of the Saharan dust PMF factor. Wildfires contribution in Porto was also estimated by PMF analysis. The uncertainties of African dust and sea salt concentrations were calculated based on the uncertainties of the parameters included in the respective calculation formulas (PM regional concentrations in the

case of African dust and Na, Cl and Al concentrations for sea salt). The uncertainties associated with PMF analysis (contribution of African dust in Florence and of wildfires in Porto) were calculated based on the standard error of the coefficients of a multiple regression between the measured PM concentration (independent variable) and the source contributions estimated by PMF analysis (dependent variables).

African dust contribution to PM concentrations was found to be more pronounced in Eastern Mediterranean (Athens), with peak concentrations during spring time reaching up to 127 μg m^{-3} (maximum 24 h mean dust concentration during a 15-day dust transport event on May 2013). Previous studies have also reported the high impact of dust transport events in Athens and Greece in general (Manousakas et al., 2015; Grigoropoulos et al., 2009; Mitsakou et al., 2008). The mean annual relative contributions of African dust to PM_{10} concentrations decreased from East to West: 21 % in Athens, 5 % in Florence, and ~ 2 % in Milan, Barcelona and Porto. The respective contributions to $PM_{2.5}$ concentrations were 13.7 % in Athens, 1.3-1.4 % in Florence and Milan and 2.3-2.4 % in Barcelona and Porto. The large difference between net dust loads calculated for Athens and the other cities is due to the Southern location of Athens, and the severity of some Saharan dust episodes in the eastern part of the Basin (Athanasopoulou et al., 2016). High seasonal variability of contributions was observed, with dust transport events occurring at different periods in western and eastern sides of the Mediterranean (Pey et al., 2013; Querol et al., 2009). African dust inputs were highest during spring and lowest during summer in Athens and Florence. Milan presented high contributions during spring and summer, Porto during winter and Barcelona during summer season. These results are in good agreement with Moulin et al. (1998) who reported that the annual cycle of African dust transport over the Mediterranean region starts during springtime in the eastern part, while during summer there is maximum transport in the western part. Porto was the only city deviating from this behaviour, suggesting that the studied year may not be representative for this city for assessing seasonal trends, probably due to the low frequency and intensity of dust events. Querol et al. (2009) have also noted that when intense dust transport events are recorded in the Eastern Mediterranean (such as the case for 2013), unusually low African dust contributions are observed in the Western Mediterranean.

Sea salt was mostly related to the coarse mode and exhibited significant seasonal variability as well. Sea salt concentrations were highest in Porto, with average relative contributions equal to 12.3 % and 4.6 % for PM_{10} and $PM_{2.5}$. The respective contributions for Athens and Barcelona were 7–8 % to PM_{10} and 2.3-2.5 % to $PM_{2.5}$. The lowest contributions were observed in Florence and Milan (1.3-3.3 % to PM_{10}). The results reflect the geographical distribution of AIRUSE sites: lower levels of sea salt at the inland Italian cities (Florence and Milan) and higher at the Mediterranean coastal sites, with the highest contribution observed at the Atlantic site (Porto). Similar observations were reported by Manders et al. (2010) who showed that the sea salt load in PM_{10} at the Atlantic side of Europe is much higher than at the Mediterranean region, especially the western Mediterranean. They also showed that the sea salt load in PM_{10} is reduced very fast as the air masses progress inland.

Large scale uncontrolled forest fires were observed only in Porto during the period of the study. The average contribution to PM levels was low (1.4 % and 1.9 % to PM_{10} and $PM_{2.5}$, respectively) due to the few event days during the year (after the 20th

of August and during September). Nevertheless, during event days, contribution to PM was greatly increased, reaching 20 and 22 % to PM_{10} and $PM_{2.5}$, respectively.

The uncertainties for the calculated contributions of the different natural sources were estimated on average around or below 10%. The relative uncertainties exhibited low variability during the studied period, except for the case of African dust, where a significant increase was observed for net dust concentrations below 5 μg m^{-3}. The relative uncertainties calculated for each city and PM size fraction were on average at 6-15%, 10-41% and above 100% for African dust loads above 5 μg m^{-3}, between 1 and 5 μg m^{-3} and below 1 μg m^{-3}, respectively.

The subtraction of natural sources' contribution from PM_{10} concentrations measured at the AIRUSE sites, according to EC regulation, led to a decrease in the mean annual PM_{10} concentrations in the range of 3.5 (Milan) to 29.5 % (Athens) (Fig. 2). Attainment of the annual limit value set by the EU through Directive 2008/50/EC was achieved at all sites during 2013, although the urban background site in Milan and the urban traffic site in Porto exhibited concentrations close to the air quality standard. Similar decrease (1.5–21 %) was observed in the 90.4th percentiles of PM_{10} concentrations. The 90.4th percentile corresponds to the maximum permissible number of exceedance days (35 during the year). The subtraction of natural sources' contributions led to marginal compliance with the 24 h limit value for Porto, while Milan continued to present more exceedances than the permitted 35 days (84 days for PM_{10} and 82 days for the adjusted PM_{10} after subtraction of natural sources' contribution). Regarding $PM_{2.5}$ concentrations, the subtraction of natural sources' contribution led to decreases in mean annual concentrations in the range of 1.3 (Florence) to 16 % (Athens) for AIRUSE sites. Despite the subtraction of natural sources' contribution, Milan did not attain the EU annual limit value, while in the urban traffic site in Porto, marginal attainment was achieved (Fig. 3).

Average contributions of natural sources to PM_{10} concentrations at each city, during all measurement days and only when exceedance days were considered, are presented in Fig. 4. Wildfires contributed to exceedances in Porto. Average concentration during exceedance days was low (below 4 μg m^{-3}), nevertheless it was much higher than the corresponding mean value during the yearly measurement campaign. Sea salt, on the other hand, is related to clean air conditions, while no African dust event was recorded during exceedance days. In Barcelona urban background site, 24 h concentrations did not exceed the respective EU limit. The highest concentrations were almost entirely attributed to anthropogenic sources. Again no dusts events were recorded during high concentration days. Similar results were obtained for Florence and Milan as well. The Athens suburban site on the other hand is a characteristic example of the effect of natural sources in background urban environments. Exceedances of the PM_{10} 24 h EU limit value were attributed to African dust by 79% in terms of mass concentration (53 out of 67 μg m^{-3}), with a total contribution from natural sources reaching 88%. Mean annual contribution of African dust was also significant (21%).

3.2 Sensitivity analysis on the estimation of net African dust

Based on the available tools for dust transport modelling, different potential dust event days may be identified. Analysis of AIRUSE data showed that models' results are not always in perfect agreement. A sensitivity analysis was performed in order to assess the effect of model selection, based on the Athens dataset, which included the largest number of dust events. Net African dust loads were calculated using SEC 2011/208. In this analysis, days were marked as dust events for the following scenarios: (N1) when at least 1 out of 4 models gave an event signal; (N2) when at least 2 models gave an event signal; (N3) when at least 3 models gave an event signal; (N4) when all models gave an event signal. The results of the calculated dust concentrations for each of the scenarios (N2) - (N4) (shown in blue) and the respective increments (shown in red) when a less strict criterion is selected, (N1) – (N3) respectively, are presented in Fig. 5.

Small increments in relation to peak dust concentrations were observed between (N1), (N2) and (N3) scenarios, with mean annual dust contribution calculated equal to 5.1, 4.3 and 3.9 μg m^{-3} for scenarios (N1)–(N3) respectively. Nevertheless, on a daily basis, these increments reached up to 16 μg m^{-3} for scenario (N1) in relation to (N2) and 25 μg m^{-3} for scenario (N2) in relation to (N3), which are of the same magnitude of typical PM_{10} concentration levels at this site (Triantafyllou et al., 2016). When full agreement between models was required, even very intense events were omitted, as is demonstrated by the comparison of (N3) and (N4) scenarios. The analysis highlights the need for employing as many available tools as possible for the identification of dust transport events, in order to ensure adequate coverage and reduce uncertainty.

Another parameter examined was the use of alternative input data in the net dust calculation algorithm. The net dust loads calculated by PM_{10} regional background concentrations according to the methodology adopted by EC, Net dust (PM_{10}), were used as reference. Net dust loads were also calculated by using the following input datasets: i) the coarse fraction of PM ($PM_{2.5-10}$) regional concentrations (instead of the PM_{10} fraction), Net dust (PMcoarse), and the mineral component of PM_{10}, ii) either reconstructed through stoichiometry, Net dust (MIN-STOICH), or iii) obtained by PMF, Net dust (PMF).

PMF analysis performed on the datasets of all five studied cities, reported in Amato et al. (2016), has shown that a distinct African dust factor is not easily obtained. Only in the case of Florence, a separate PMF factor profile for African dust was identified, providing a potential reference value for this city and insight into the chemical profile of transported African dust. The African dust concentrations estimated by PMF in Florence are in very good agreement with the Net dust (MIN-STOICH), while the method based on PM_{10} concentrations at the regional site seem to overestimate African dust loads (Fig. 6). This last observation may be related to the difficulty of finding a suitable regional background site representative for the city of Florence in connection to African dust transport, due to the orography of the region.

In all other AIRUSE cities, a mixed mineral dust factor was obtained, including both local soil and long-range transported dust. Comparisons of the net dust loads calculated based on the mineral component of PM_{10} (quantified stoichiometrically or by PMF) with the reference Net dust (PM_{10}), for the city of Athens, are shown in Fig. 7. For Porto, Barcelona and Milan no

regression between Net dust (MIN-STOICH) or Net dust (PMF) with Net dust (PM_{10}) was attempted, due to the much lower number of African dust event days and corresponding chemical speciation data. For the ATH-SUB dataset, the use of the mineral dust contributions estimated by PMF provided results in good agreement with Net dust (PM_{10}) concentrations, with the uncertainty increasing in dust concentrations below 10 μg m^{-3}. The net dust calculated from the PM_{10} stoichiometric mineral component (MIN-STOICH) exhibited very good correlation with Net dust (PM_{10}). Net dust (MIN-STOICH) displayed lower concentrations by a factor of 1.6 on average, while for net dust loads <10 μg m^{-3} this difference was higher (Fig. 7). Similar behaviour, with even higher correlation coefficient, was observed when PMcoarse concentrations were used in the calculation algorithm (Net dust (PMcoarse)) (Fig. 8). Barcelona exhibited comparable results with Athens (Fig. 8), while weaker correlations were observed for Porto and Milan (Fig. 9). Florence was not included in this analysis because no $PM_{2.5}$ or PMcoarse data were available from the regional background site. The results indicate that African dust is also found in sizes below 2.5 μm.

Regression analysis of the calculated Net dust (PM_{10}) and net dust (MIN-STOICH) versus $PM_{2.5}/PM_{10}$ concentration ratios was used in order to further examine the calculated dust loads with respect to particle size (Fig. 10). In the case of Net dust (MIN-STOICH), all intense dust events (with net dust loads greater than 10 μg m^{-3}) were related with the coarse fraction (low $PM_{2.5}/PM_{10}$ ratios). On the contrary, for Net dust (PM_{10}) several events with net dust loads from 10 to 20 μg m^{-3} or higher were related to fine particles ($PM_{2.5}/PM_{10}$ ratios greater than 0.6). This suggests that Net dust (PM_{10}) may include non-mineral fine particles.

The chemical profiles of mineral dust obtained by PMF (Amato et al., 2016) may provide further information on the discrepancies observed between the alternative methods. Comparison of the Athens mineral dust profile and the two mineral dust profiles obtained for Florence (for local and African dust), showed that the African dust profile differed with respect to the other two mineral dust profiles in the absence of organic carbon, Zn and Pb, while a much lower NO_3^- contribution was also observed. The presence of these species may reflect the enrichment of local dust with anthropogenic chemical components. On the other hand, the inclusion of non-mineral components in the African dust profile (Fig. 11) may explain the underestimation in Athens of net dust loads when the PM_{10} mineral component, Net dust (MIN-STOICH), is used (Rodriguez et al., 2001).

Net dust concentrations calculated by PM_{10} regional background data were also compared to the dust concentrations provided by SKIRON/Dust model (at surface level and at three different heights) and BSC-DREAM8b v2.0 model (at surface level). Very good correlation was obtained with the Athens dataset for both models. For the SKIRON/Dust model calculated and modelled dust loads at surface levels were comparable (Table 4). Nevertheless, no correlation~~s~~ was observed ~~became very weak~~ for dust concentrations below 10 μg m^{-3}, suggesting increased uncertainty at these dust levels (Fig. 12) possibly due to the applied dust cycle parametrization constrains and limitations. In the case of Porto, Barcelona and Milan, almost all modelled dust concentrations were below or equal to 10 μg m^{-3}, thus producing weak to no correlations with calculated dust loads.

Florence presented similar results with Athens, with somewhat lower Pearson's coefficients between modelled and calculated data, which may be attributed to fewer data with concentrations above 10 μg m^{-3}. In addition, the corresponding slopes of the regression lines were higher than 1.0 in all cases (Table 4). 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.

4 Conclusions

LIFE-AIRUSE project employed a large dataset of PM_{10} and $PM_{2.5}$ concentrations and chemical speciation from five Southern European cities (Porto, Barcelona, Milan, Florence and Athens), in order to examine the contribution of two major natural sources: long-range transport of African dust and sea salt. The results clearly show that natural source contribution may be significant during specific periods, leading to events of PM limit value exceedances. African dust contribution to PM concentrations was more pronounced in Eastern Mediterranean (Athens), with peak 24 h concentrations in spring time reaching up to 127 μg m^{-3} during a 15-day long African dust event in May 2013. The mean annual relative contributions of African dust to PM_{10} concentrations decreased from East to West: 21 % in Athens, 5 % in Florence, and ~ 2 % in Milan, Barcelona and Porto. High seasonal variability of contributions was observed, with dust transport events occurring at different periods in western and eastern sides of the Mediterranean. Sea salt was mostly related to the coarse mode and exhibited significant seasonal variability. Sea salt concentrations were highest in Porto, with average relative contributions equal to 12.3 % for PM_{10}. The respective contributions for Athens and Barcelona were 7–8 %, while the lowest contributions were observed in Florence and Milan (1.3-3.3 %). The results reflect the geographical distribution of AIRUSE sites: lower levels of sea salt at the inland Italian cities (Florence and Milan) and higher at the Mediterranean coastal sites, with the highest contribution observed at the Atlantic site (Porto). Uncontrolled forest fires were observed to affect PM concentrations only in Porto during the studied period. The mean annual contribution to PM levels was low (1.4 % and 1.9 % to PM_{10} and $PM_{2.5}$, respectively) due to the few event days during the year (after the 20th of August and during September). Nevertheless, during event days, contribution to PM was greatly increased, reaching 20 and 22 % of 24 h PM_{10} and $PM_{2.5}$, respectively.

A sensitivity analysis for the quantification of African dust contribution was performed, in order to assess the major factors affecting the calculated net dust concentrations. The analysis indicated that a key parameter to be considered is the selection of an appropriate regional background site. In addition, the use of as many available tools as possible for the identification of dust transport events is recommended, in order to ensure adequate coverage and reduce uncertainty. The results also indicated that the calculation of net African dust through the use of regional background data of PM_{10} (or $PM_{2.5}$) mass concentrations provides higher dust concentration estimates in comparison to the use of the same methodology with input data the mineral component of PM, derived stoichiometrically. Analysis of mineral dust source profiles obtained by PMF provides further

evidence that additional species to the crustal matter, usually secondary aerosol, are the source of this discrepancy, arriving together or associated with the crustal component during long range transport.

The present study has demonstrated that natural sources are often expressed with high intensity events, leading to very high daily contributions and exceedances of the EU air quality standards. Since these sources cannot be controlled, relevant mitigation measures can only be focused on minimizing the effects of this type of pollution. Namely, measures are recommended to target reducing the potential of particles deposited on the streets and other surfaces to resuspend, while emergency action plans, especially for sensitive population subgroups, may come into force during days when extreme dust events are forecasted.

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Table 1. Description of measurement campaigns: Measurement sites, periods and sampling days.

Monitoring site	Site acronym	Measurement period	Number of samples
Porto Urban traffic	POR-TR	01/2013-01/2014	122 (PM_{10}) / 125 ($PM_{2.5}$)
Barcelona Urban background	BCN-UB	01/2013-01/2014	125 (PM_{10}) / 109 ($PM_{2.5}$)
Milan Urban background	MLN-UB	01/2013-01/2014	276 (PM_{10}) / 357 ($PM_{2.5}$)
Florence Urban background	FI-UB	01/2013-01/2014	223 (PM_{10}) / 243 ($PM_{2.5}$)
Athens Suburban	ATH-SUB	02/2013-02/2014	192 (PM_{10}) / 212 ($PM_{2.5}$)

Table 2. Mean annual natural source contributions to PM_{10} concentrations and corresponding uncertainties, for~~at~~ the five AIRUSE cities.

	Contributions of natural sources (µg m^{-3})[1]				
	Porto	**Barcelona**	**Milan**	**Florence**	**Athens**
PM_{10} concentration	34.6	22.5	35.8	19.8	19.6
African dust	0.~~8~~75 ± 0.02	0.~~5~~49 ± 0.01	0.~~8~~76 ± 0.02	1.02± 0.14	4.~~2~~19 ± 0.55
Sea salt	4.~~3~~27 ± 0.41	1.~~5~~49 ± 0.18	0.~~5~~46 ± 0.03	0.64 ± 0.07~~7~~	1.64 ± 0.14
Wildfires	0.50 ± 0.02	NE[2]	NE[2]	NE[2]	NE[2]
Total natural	**5.52 ± 0.45**	**~~2.0~~1.98 ± 0.19**	**1.22 ± 0.05**	**1.~~7~~66 ± 0.21**	**5.83 ± 0.69**
	Relative contributions of natural sources (%)				
African dust	2.2 ± 0.1	2.2 ± 0.0	2.2 ± 0.0	5.2 ± 0.7	21.4 ± 2.8
Sea salt	12.3 ± 1,2	6.6 ± 0.8	1.3 ± 0.1	3.3 ± 0.3	8.1 ± 0.7
Wildfires	1.4 ± 0.1	NE[2]	NE[2]	NE[2]	NE[2]
Total natural	**15.9 ± 1.4**	**8.8 ± 0.8**	**3.5 ± 0.1**	**8.5 ± 1.0**	**29.5 ± 3.5**

[1]Contributions may slightly differ from the values reported in Amato et al. (2016) due to different statistics of the respective datasets.

[2]NE: Not estimated.

Table 3. Mean annual natural source contributions to $PM_{2.5}$ concentrations and corresponding uncertainties, for~~at~~ the five AIRUSE cities.

	Contributions of natural sources (μg m^{-3})[1]				
	Porto	**Barcelona**	**Milan**	**Florence**	**Athens**
$PM_{2.5}$ concentration	26.8	15.2	28.7	14.6	11.0
African dust	0.61 ± 0.01	0.~~4~~38 ± 0.01	0.41 ± 0.02	0.~~2~~19 ± 0.02	1.~~5~~49 ± 0.13
Sea salt	1.22 ± 0.15	0.~~4~~37 ± 0.10	0.42 ± 0.02	0.11 ± 0.01	0.25 ± 0.03
Wildfires	0.50 ± 0.02	NE[2]	NE[2]	NE[2]	NE[2]
Total natural	**2.33 ± 0.18**	**0.~~8~~75 ± 0.11**	**0.83 ± 0.04**	**0.30 ± 0.03**	**1.74 ± 0.16**
	Relative contributions of natural sources (%)				
African dust	2.3 ± 0.1	2.4 ± 0.1	1.4 ± 0.1	1.3 ± 0.2	13.7 ± 1.2
Sea salt	4.6 ± 0.6	2.5 ± 0.7	1.5 ± 0.1	0.7 ± 0.1	2.3 ± 0.3
Wildfires	1.9 ± 0.1	NE[2]	NE[2]	NE[2]	NE[2]
Total natural	**8.7 ± 0.8**	**4.9 ± 0.8**	**2.9 ± 0.2**	**2.0 ± 0.3**	**16.0 ± 1.5**

[1]Contributions may slightly differ from the values reported in Amato et al. (2016) due to different statistics of the respective datasets.

[2]NE: Not estimated.

Table 4. Deming ~~Regression~~ regression analysis of dust loads predicted by transport models versus the net dust concentration calculated: (i) through regional PM_{10} concentration data for Athens and (ii) by PMF analysis for Florence. The lower and upper bounds at 95% confidence interval for the calculated slopes and intercepts are presented in parenthesis.

		Height	Pearson's coefficient*	Slope	Intercept
ATHENS	SKIRON model	Surface	0.83	1.2 (0.9;1.5)~~0.95~~	-1.1 (-2.0;-0.1) ~~0.18~~
		450 m a.s.l.	0.86	1.9 (1.6;2.2)~~1.50~~	-1.8 (-2.9;-0.8) ~~0.28~~
		600 m a.s.l.	0.87	2.4 (2.1;2.7)~~1.93~~	-2.4 (-3.5;-1.2) ~~0.39~~
		750 m a.s.l.	0.87	3.0 (2.6;3.3)~~2.34~~	-2.5 (-3.9;-1.2) ~~0.01~~
	DREAM8b v2.0 model	Surface	0.77	2.4 (2.0;2.8)~~1.54~~	-1.3 (-2.6;0.0)~~2.18~~
FLORENCE	SKIRON model	Surface	0.64	1.9 (1.1;2.7)~~0.99~~	0.0 (-0.3;0.3)~~0.57~~
		590 m a.s.l.	0.65	2.1 (1.3;3.0)~~1.08~~	0.0 (-0.4;0.3)~~0.64~~
		760 m a.s.l.	0.66	2.5 (1.7;3.4)~~1.29~~	-0.1 (-0.5;0.3)~~0.72~~
		940 m a.s.l.	0.67	3.1 (2.1;4.0)~~1.51~~	-0.1 (-0.6;0.3)~~0.87~~
	DREAM8b v2.0 model	Surface	0.61	4.9 (3.0;6.8)~~1.95~~	-0.6 (-1.7;0.5)~~1.30~~

* All correlations were significant at p = 0.05.

List of figures

Figure 11: Source chemical profiles obtained by the application of PMF model (Amato et al., 2016): Two mineral dust sources were identified in Florence (African dust_FI and Local dust_FI) while a mixed mineral dust profile was found in Athens (Mineral dust_ATH).

Figure 12: Deming ~~R~~regression analysis between net dust calculated through PM_{10} regional background data and dust concentrations modelled at surface level by (a) SKIRON/Dust ~~(left)~~ and (b) BSC DREAM8b v2.0 model ~~(right)~~, for the city of Athens. The black line corresponds to the linear regression equation, while the red dotted lines are the upper and lower bounds, at 95% confidence interval.

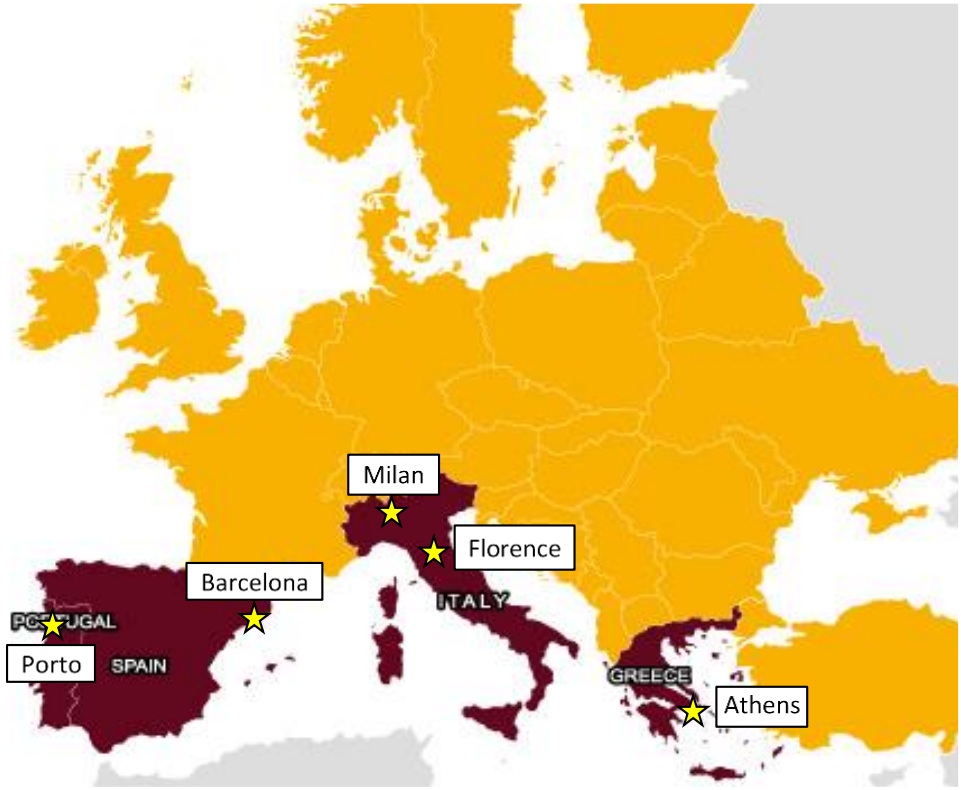

Milan
Florence
Barcelona
ITALY
PORTUGAL
Porto
SPAIN
GREECE
Athens

Fig. 1

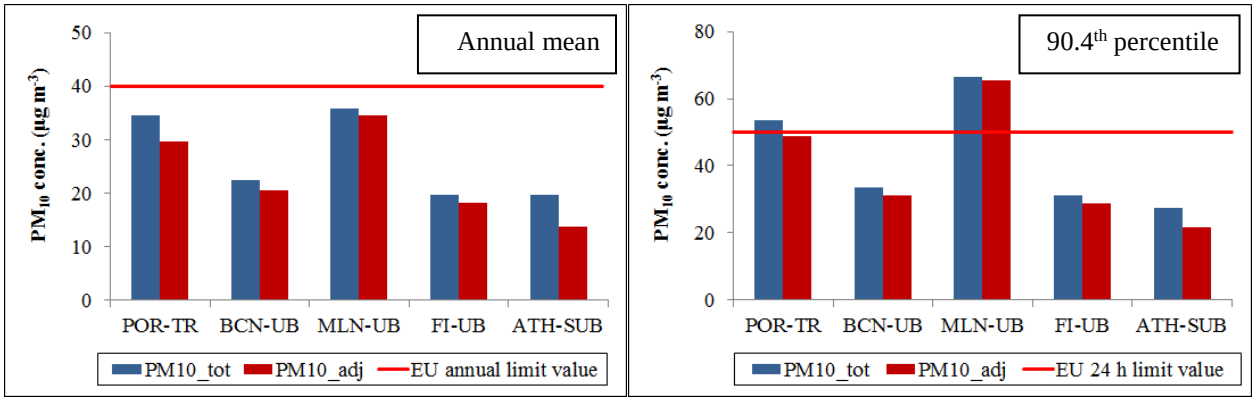

Annual mean
PM10 conc. (µg m-3)
50
40
30
20
10
0
POR-TR
BCN-UB
MLN-UB
FI-UB
ATH-SUB
PM10_tot
PM10_adj
EU annual limit value
90.4th percentile
PM10 conc. (µg m-3)
80
60
40
20
0
POR-TR
BCN-UB
MLN-UB
FI-UB
ATH-SUB
PM10_tot
PM10_adj
EU 24 h limit value

Fig. 2

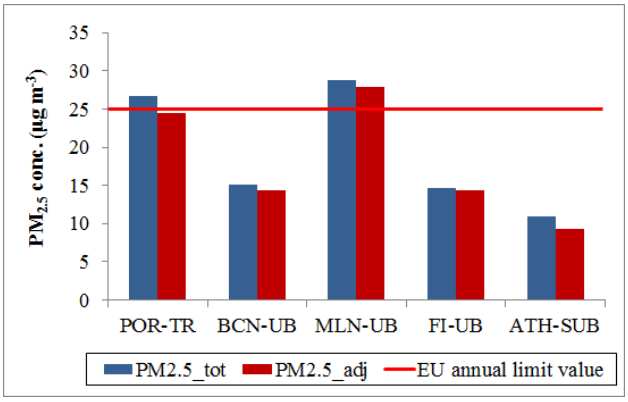

35
30
25
20
15
10
5
0
PM2.5 conc. (µg m-3)
POR-TR
BCN-UB
MLN-UB
FI-UB
ATH-SUB
PM2.5_tot
PM2.5_adj
EU annual limit value

Fig. 3

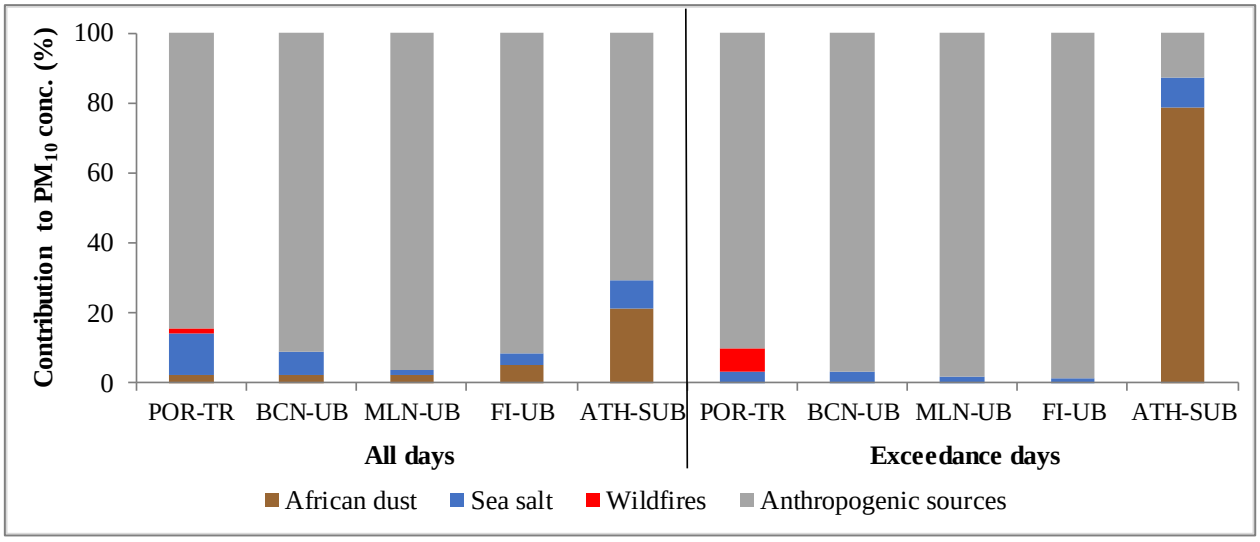

Contribution to PM10 conc. (%)
100
80
60
40
20
0
POR-TR
BCN-UB
MLN-UB
FI-UB
ATH-SUB
POR-TR
BCN-UB
MLN-UB
FI-UB
ATH-SUB
All days
Exceedance days
African dust
Sea salt
Wildfires
Anthropogenic sources

Fig. 4

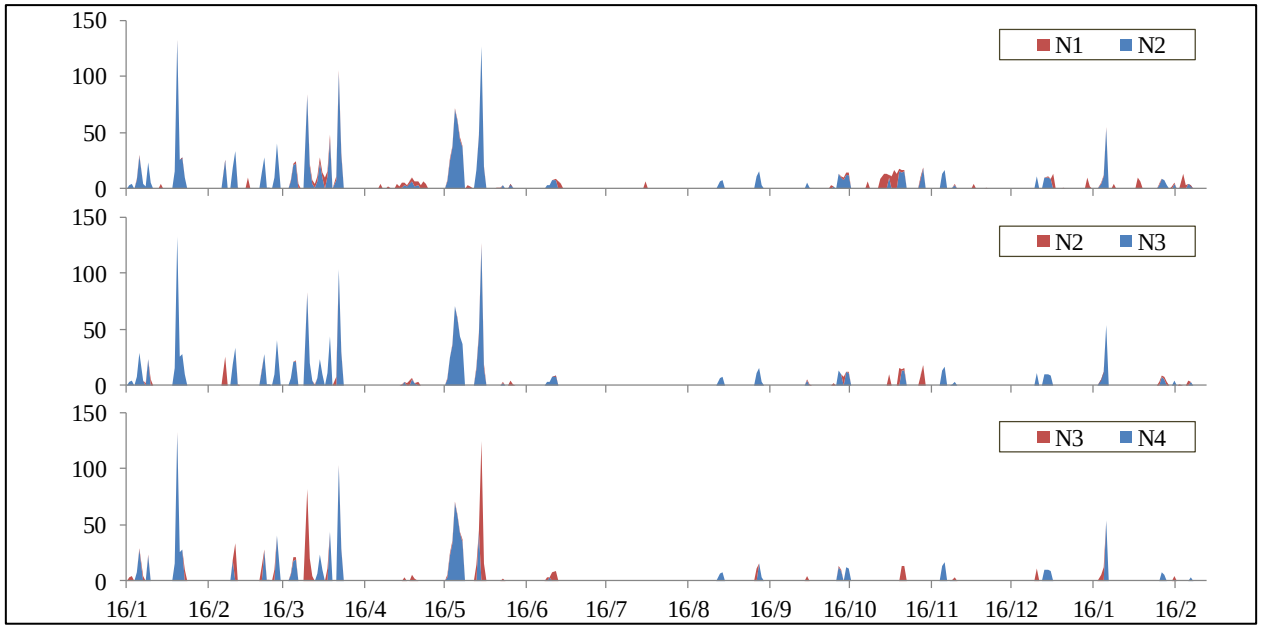
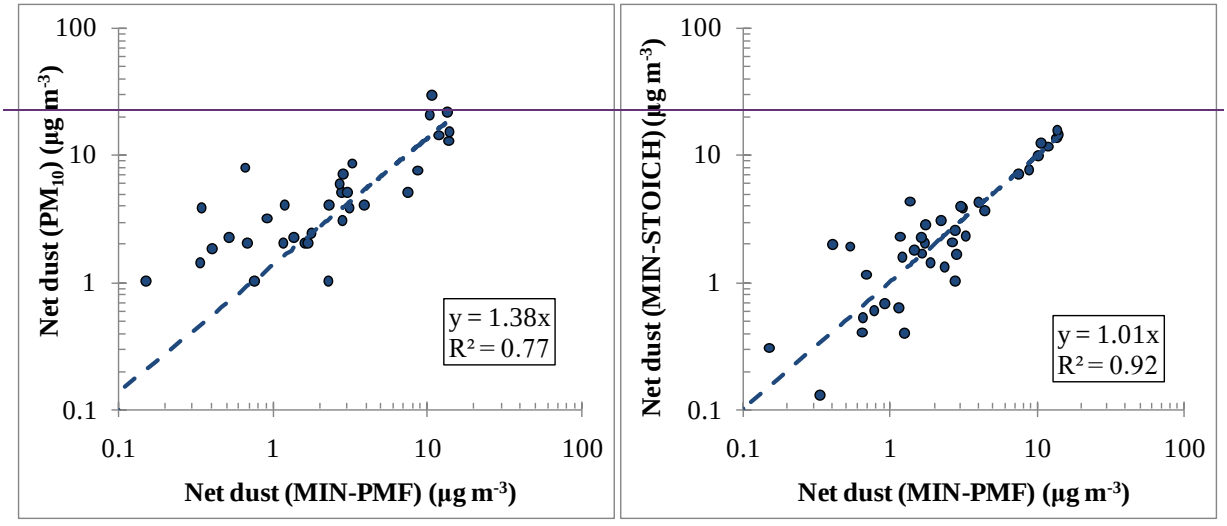

150
100
50
0
N1
N2
N2
N3
N3
N4
16/1
16/2
16/3
16/4
16/5
16/6
16/7
16/8
16/9
16/10
16/11
16/12
16/1
16/2

Fig. 5


Net dust (PM10) (µg m-3)
Net dust (MIN-PMF) (µg m-3)
y = 1.38x
R² = 0.77
Net dust (MIN-STOICH) (µg m-3)
Net dust (MIN-PMF) (µg m-3)
y = 1.01x
R² = 0.92
100
10
1
0.1

~~Fog~~

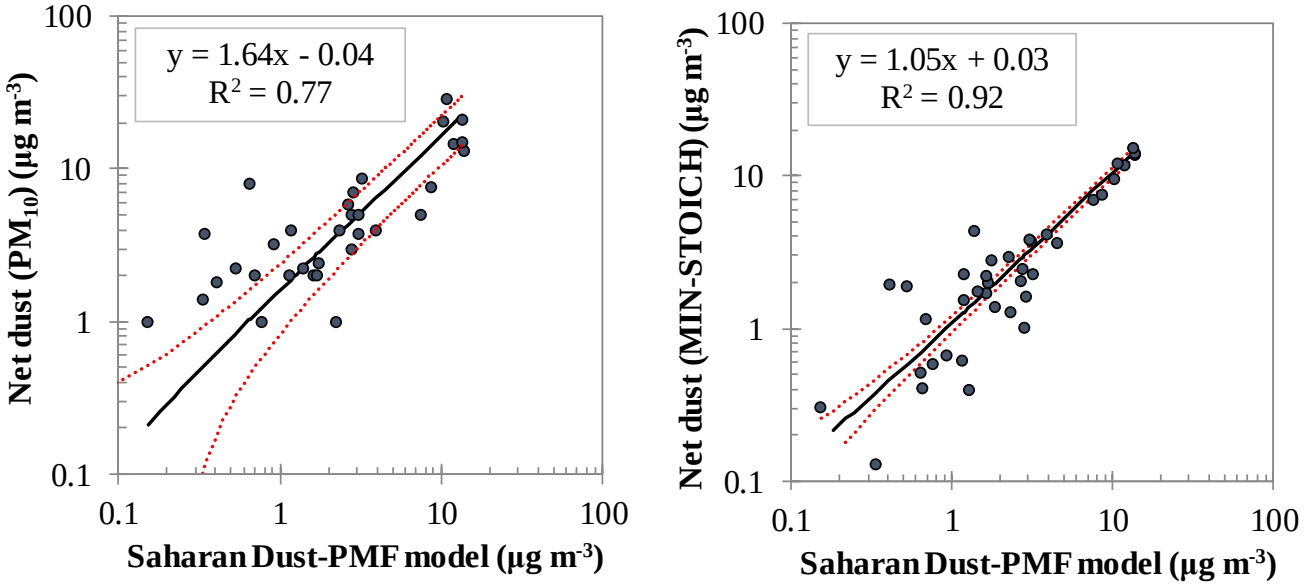
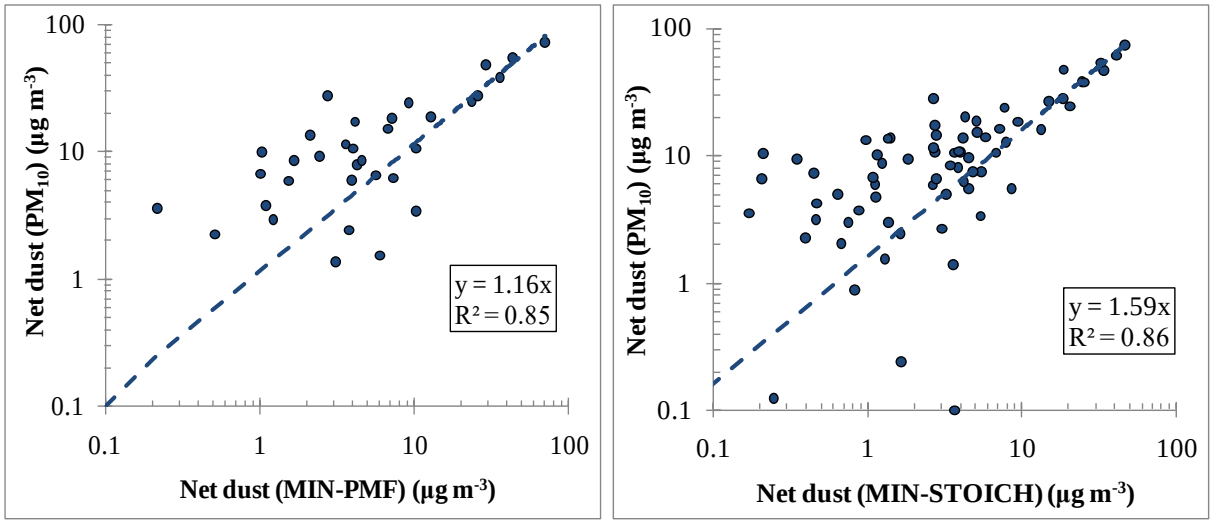

y = 1.64x - 0.04
R² = 0.77
Net dust (PM10) (µg m-3)
Saharan Dust-PMF model (µg m-3)
y = 1.05x + 0.03
R² = 0.92
Net dust (MIN-STOICH) (µg m-3)
Saharan Dust-PMF model (µg m-3)

Fig. 6


y = 1.16x
R² = 0.85
Net dust (PM10) (µg m-3)
Net dust (MIN-PMF) (µg m-3)
y = 1.59x
R² = 0.86
Net dust (PM10) (µg m-3)
Net dust (MIN-STOICH) (µg m-3)

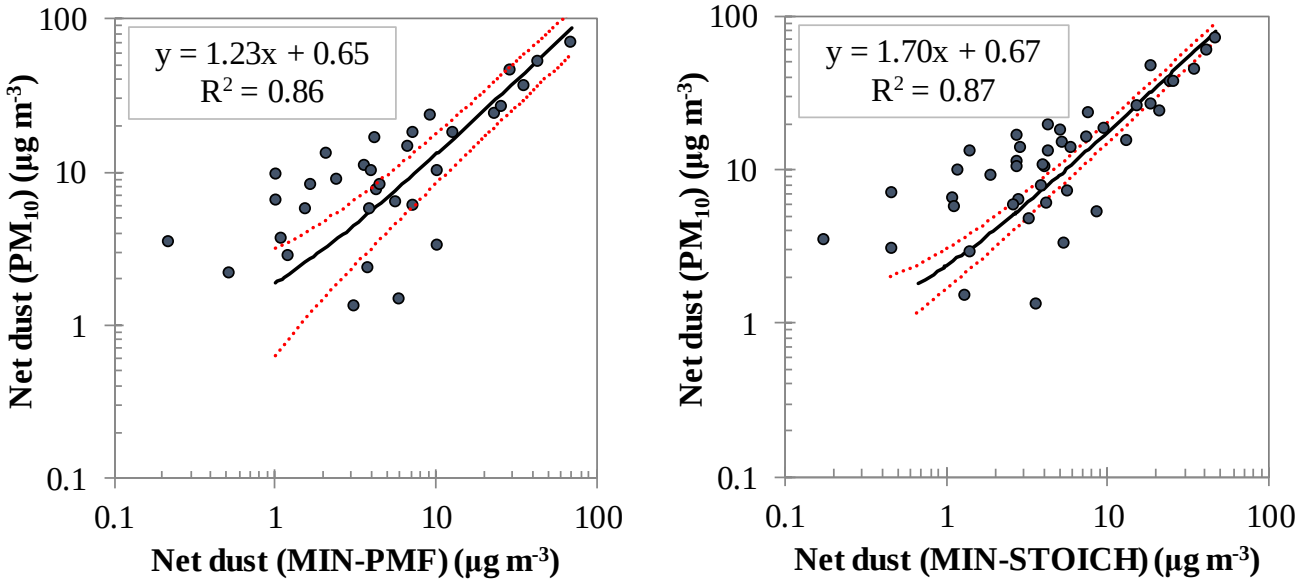
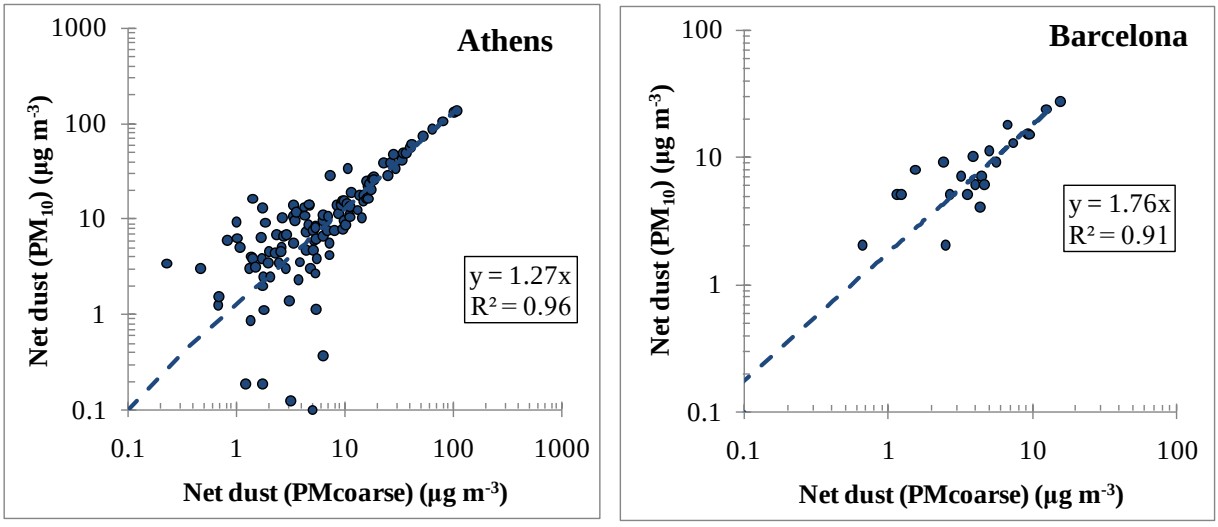

y = 1.23x + 0.65
R² = 0.86
Net dust (PM10) (µg m-3)
Net dust (MIN-PMF) (µg m-3)
y = 1.70x + 0.67
R² = 0.87
Net dust (MIN-STOICH) (µg m-3)

Fig. 7


Athens
y = 1.27x
R² = 0.96
Net dust (PM10) (µg m-3)
Net dust (PMcoarse) (µg m-3)
Barcelona
y = 1.76x
R² = 0.91

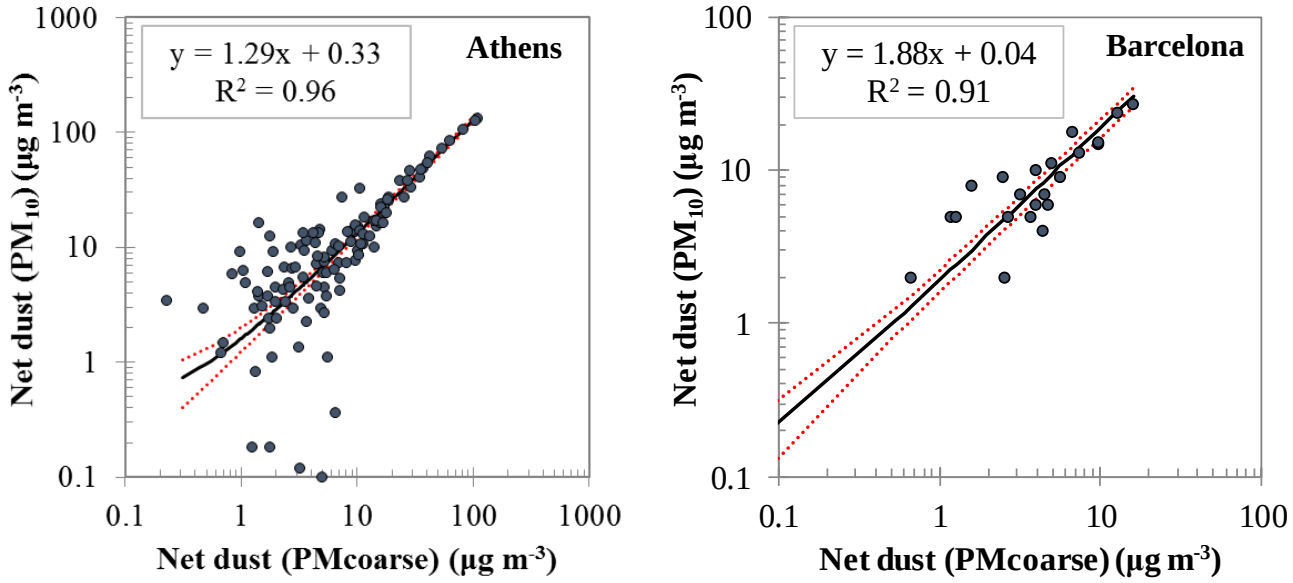
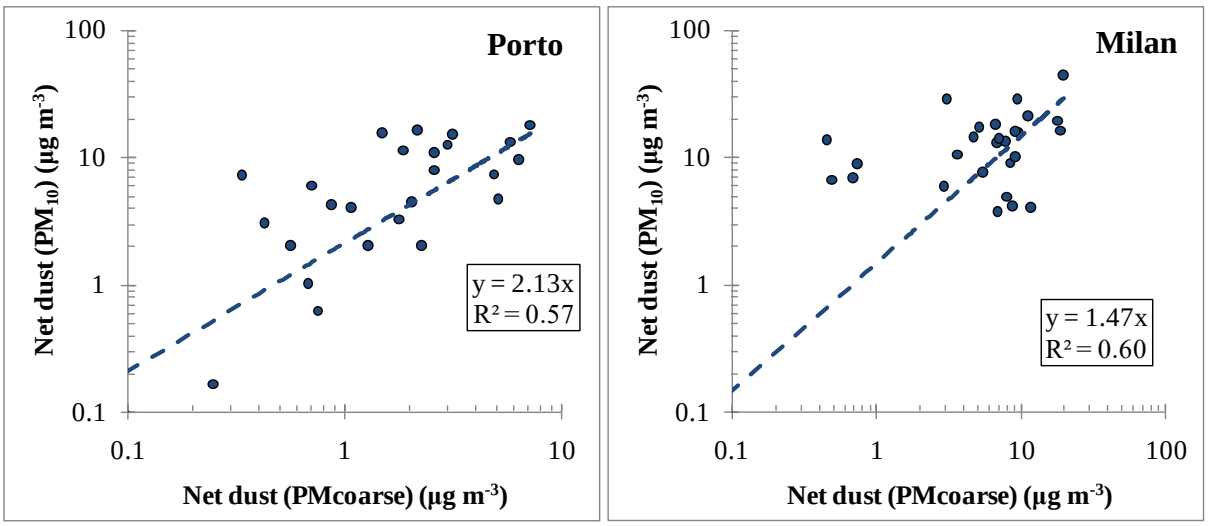

Athens
y = 1.29x + 0.33
R² = 0.96
Net dust (PM10) (µg m-3)
Net dust (PMcoarse) (µg m-3)
Barcelona
y = 1.88x + 0.04
R² = 0.91
Net dust (PM10) (µg m-3)
Net dust (PMcoarse) (µg m-3)

Fig. 8


Porto
y = 2.13x
R² = 0.57
Net dust (PM10) (µg m-3)
Net dust (PMcoarse) (µg m-3)
Milan
y = 1.47x
R² = 0.60
Net dust (PM10) (µg m-3)
Net dust (PMcoarse) (µg m-3)

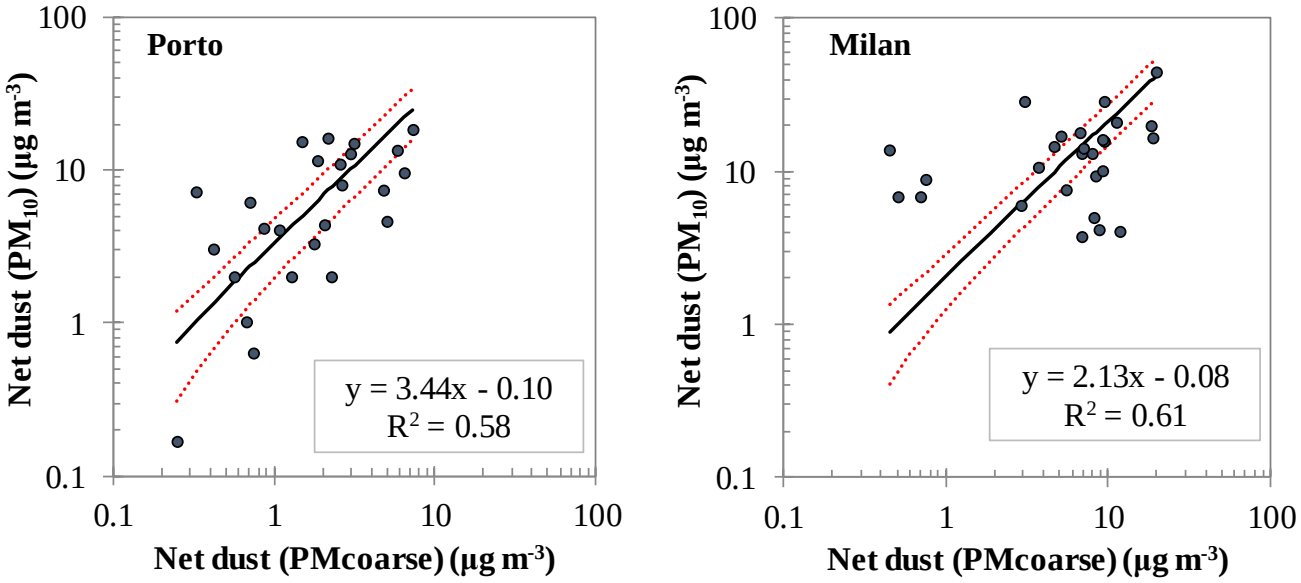
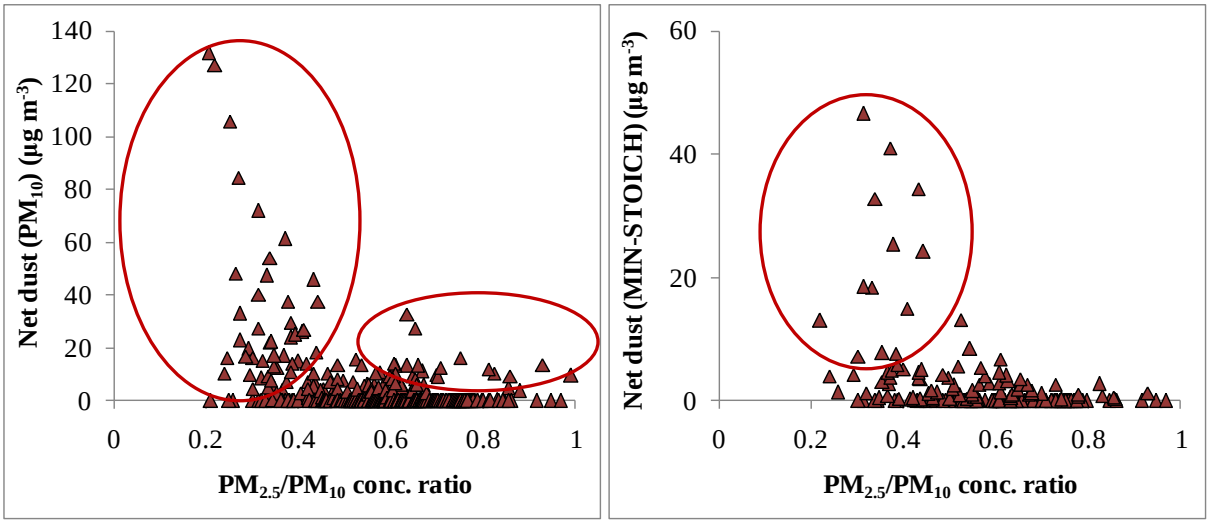

Porto
Net dust (PM_{10}) (µg m^{-3})
Net dust (PMcoarse) (µg m^{-3})
y = 3.44x - 0.10
R^2 = 0.58
Milan
y = 2.13x - 0.08
R^2 = 0.61

Fig. 9


Net dust (PM_{10}) (µg m^{-3})
$PM_{2.5}/PM_{10}$ conc. ratio
Net dust (MIN-STOICH) (µg m^{-3})
$PM_{2.5}/PM_{10}$ conc. ratio

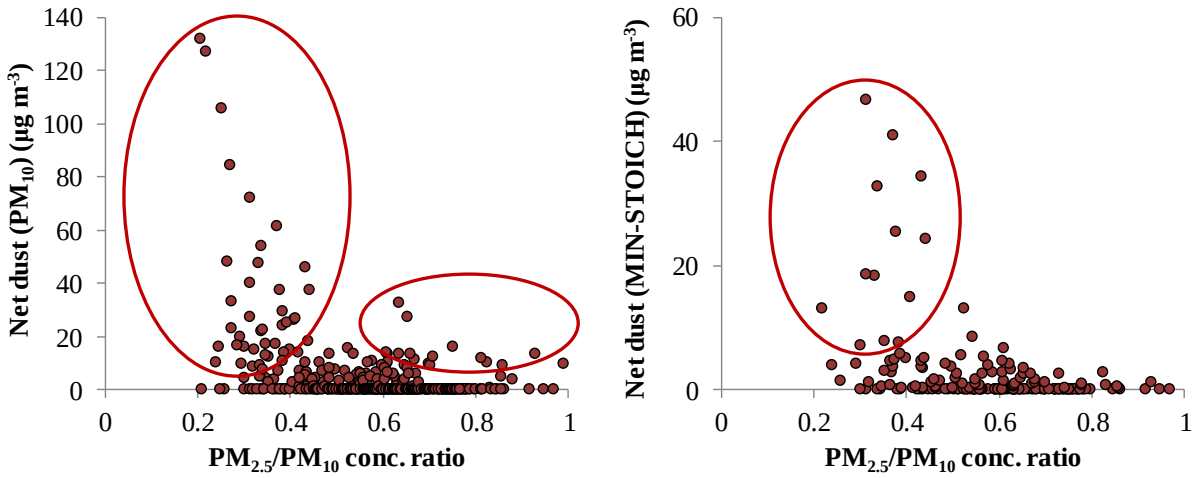

Net dust (PM10) (µg m-3)
140
120
100
80
60
40
20
0
0
0.2
0.4
0.6
0.8
1
PM2.5/PM10 conc. ratio
Net dust (MIN-STOICH) (µg m-3)
60
40
20
0
0
0.2
0.4
0.6
0.8
1
PM2.5/PM10 conc. ratio

Fig. 10

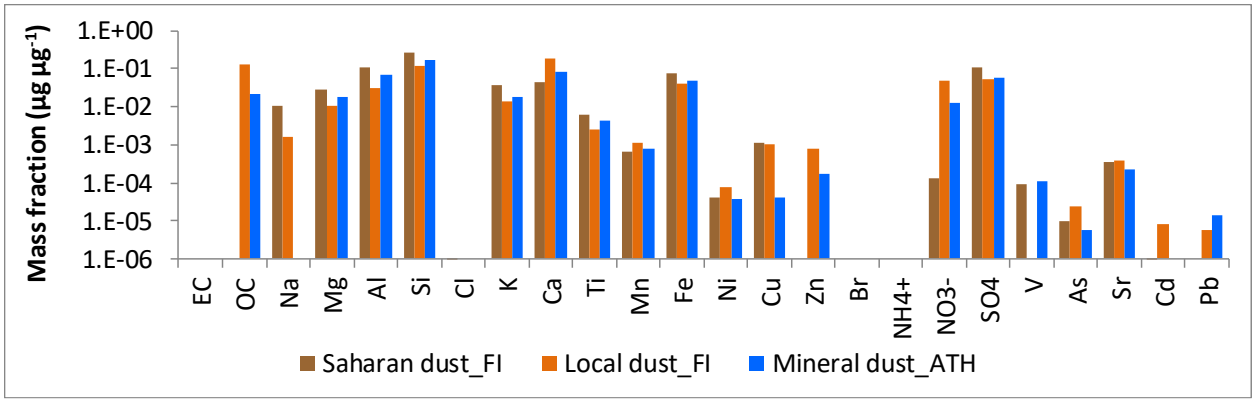

Mass fraction (µg µg-1)
1.E+00
1.E-01
1.E-02
1.E-03
1.E-04
1.E-05
1.E-06
EC
OC
Na
Mg
Al
Si
Cl
K
Ca
Ti
Mn
Fe
Ni
Cu
Zn
Br
NH4+
NO3-
SO4
V
As
Sr
Cd
Pb
Saharan dust_FI
Local dust_FI
Mineral dust_ATH

Fig. 11

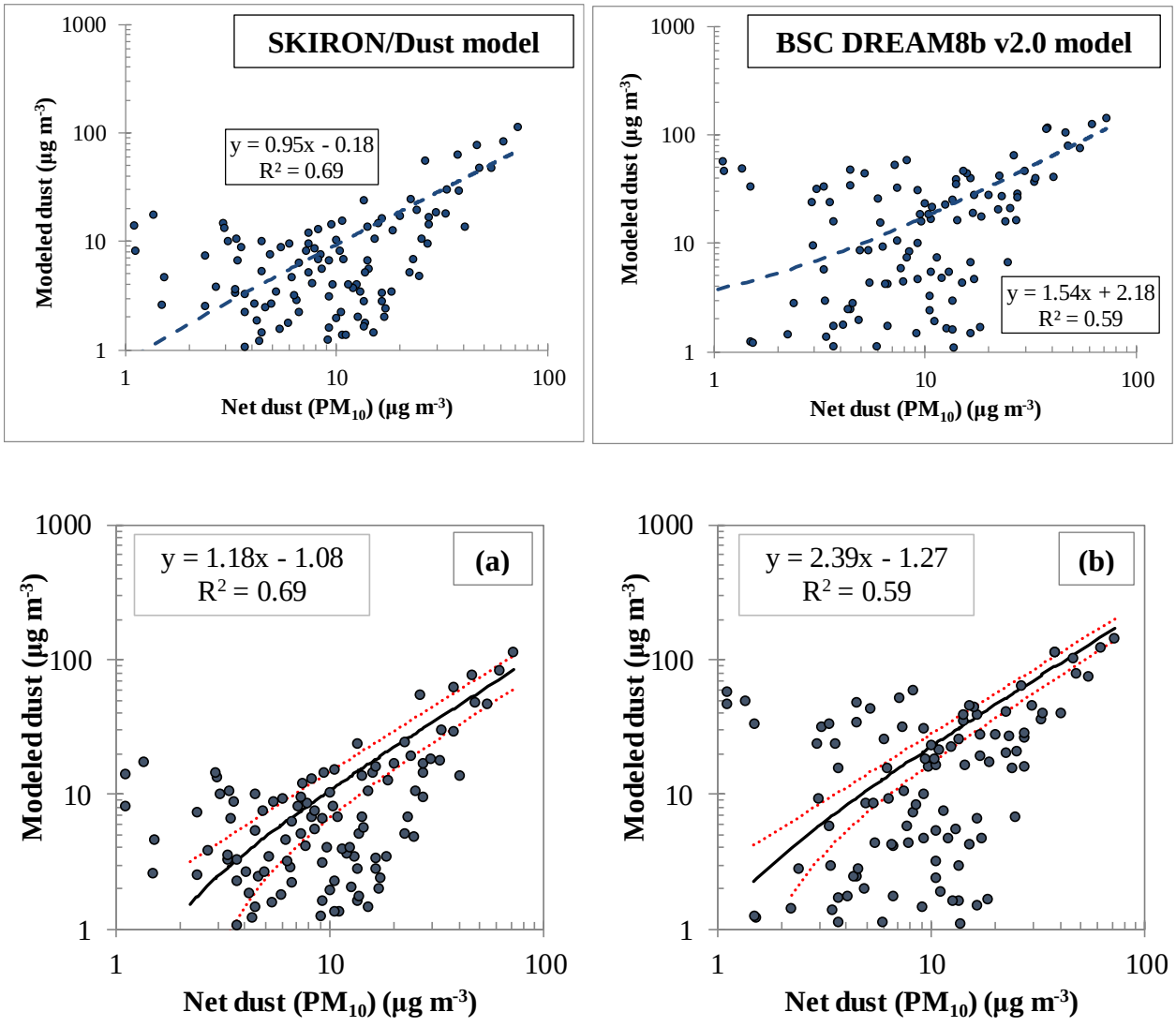

SKIRON/Dust model
y = 0.95x - 0.18
R² = 0.69
BSC DREAM8b v2.0 model
y = 1.54x + 2.18
R² = 0.59
y = 1.18x - 1.08
R² = 0.69
(a)
y = 2.39x - 1.27
R² = 0.59
(b)
Modeled dust (µg m-3)
Net dust (PM10) (µg m-3)
1000
100
10
1
1
10
100

Fig. 12