Desert dust contribution to PM10 loads in Italy: methods and recommendations addressing the relevant European Commission Guidelines in support to the Air Quality Directive 2008/50

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Abstract

In 2011 the European Commission (EC) released specific 'Guidelines' describing the methods to quantify and subtract the contribution of natural sources from the PM10 values regulated by the European Air Quality Directive (2008/50/EC). This work investigates the applicability to Italy of the EC-Methodology suggested for desert-dust, describes main limitations encountered and proposes specific modifications embedded within a 'revised-Methodology' to extend/improve its use. The revised-Methodology capabilities are then evaluated using original, chemically-resolved mineral-dust mass concentration measurements, showing better performances in predicting timing and absolute values of the desert-dust contribution to the daily-PM10 with respect to the current EC-approach. The revised-Methodology is then translated into an automatic (user-independent) tool tailored to the expected final-users. This tool is applied over the Central Italy across a 3-year long period (2012-2014), and over the whole Italian country for a calendar year (2012). The derived results confirm and extend to Italian regions never addressed before some previously observed features of the desert-dust impact over the country, such as a clear latitudinal dependence of the desert-dust impact on the yearly average PM10 (from more than 5 µg/m^3 to less than 0.5 µg/m^3, going from south to north Italy). The modifications introduced within the revised-Methodology also suggest a non-negligible role of desert-dust resuspension in areas characterized by both high traffic levels and soil sealing (urban areas and along the major Italian routes). In the Rome area, such an effect is found to add a contribution of about 2 µg/m^3 (i.e., of 20-30%) to the mean desert-dust load per dust day (about 10 µg/m^3). At the national level, this effect contributes increasing the total number of desert-dust driven exceedances of the PM10 daily limit value even in the northern regions, where the desert-dust impact on the PM10 yearly average is otherwise limited. These results also indicate the direction for possible mitigation strategies to be applied over impacted areas. The successful application of the revised-Methodology over Italy suggests it could represent a valid option for a nationwide standard procedure to quantify the desert-dust contribution to PM10, promoting the homogenisation of the relevant values annually reported to the EC.

Keywords: desert-dust, PM10, air quality, Directive 2008/50/EC, resuspension, Italy

1. Introduction

A large share of the particulate matter (PM, or aerosols) load into the European atmosphere has an anthropogenic origin (Viana et al., 2012). Still, the contribution of natural sources needs to be quantified to support the EU Member State efforts to meet the PM10-related air quality standards set out by EU legislation (daily limit value, DLV, of 50 µg/m^3, not to be exceeded more than 35 times per year, and yearly average of 40 µg/m^3).

In the Euro-Mediterranean regions, the main natural particles contributing to PM10 (i.e., particles with aerodynamic diameter < 10 µm) are desert-dust, sea spray and aerosol from wildfires (e.g., Viana et al., 2014). In 2011, the European Commission (EC) released specific 'Guidelines' (EC, 2011) to provide the Member States with common methods to assess the contribution of particles of natural origin to the PM-related metrics regulated by the Ambient Air Quality Directive 2008/50/EC (EC, 2008). Standardized methods to quantify the contribution of natural particles to the measured

PM values are important as, once adopted to prove that limit values are exceeded for natural causes, the EC legislation allows not to consider PM exceedances as such for the purpose of the Directive.

In this work we focus on the contribution of desert-dust to PM10 levels, a natural component which has been proved to be significant, especially in southern and western European countries (e.g., Basart et al., 2012a; Pey et al., 2013), and such to cause exceedances of the EU air quality PM10 limit values. In fact, the understanding of the impact of desert-dust on air quality, and the subsequent application of mitigation measures, has been recognized as one of the three major challenges in the field of air pollution chemistry in the recent overview by Gimeno et al. (2013).

In this context, the methodology currently suggested by the EC-Guidelines to quantify desert-dust in PM10 (hereafter referred to as the 'EC-Methodology') is certainly a valuable tool to provide European environmental agencies with a common procedure to assess such impact on the regulated PM10 values. The EC-Methodology is however largely based on past, extensive work performed over the Iberian peninsula (Escudero et al., 2005; Escudero et al., 2007; Querol et al., 2009) and, although applied to other regions of Europe (e. g., Matassoni et al., 2009; Aleksandropoulou and Lazaridis, 2013; Pey et al., 2013) there has been little investigation on the actual applicability of this method to other areas, which may have desert-dust regimes different from the western Mediterranean ones in terms of seasonality, driving meteorology and atmospheric processing (e.g., Israelevich et al., 2012; Gaetani et al., 2012; Salvador et al., 2014; Varga et al., 2014). In some cases, non-negligible differences between the desert-dust contribution derived from the EC-Methodology and that derived by other methods or by chemically-derived desert-dust values have been reported (e.g., Viana et al., 2010; Marconi et al., 2014).

A further element needed to promote a widespread exploitation of a shared methodology at the European level is the simplicity of its application by the end-users. This is still a weak aspect of the EC-Methodology, which requires a large number of datasets, images, and modelling resources to be considered in the evaluation process, also leaving large margins of subjectivity in its application.

Aim of the present study was testing the applicability of the current EC-Methodology to Italy, and contribute improving it in view of a more widespread use of this or similar methods by regional/national environmental services. The work was carried on in the framework of the DIAPASON Project ('Desert-dust Impact on Air quality through model-Predictions and Advanced Sensors ObservatioNs'), funded by the European Union LIFE+ program (www.diapason-life.eu; Gobbi et al., 2017). To reach the study objectives, we first applied the EC-Methodology in Central Italy (chosen as Pilot study area) over a 4-year-long period. Based on this 'pilot application', we then identified some drawbacks in the assumptions the EC-Methodology builds upon, and in its practical implementation in the Italian case (Sections 2.2, 2.3). We then propose specific solutions to overcome the identified drawbacks (Section 2.3), and embed such solutions into a modified, improved version of the EC-Methodology referred to as 'revised-Methodology'. The capabilities of this revised-Methodology in correctly predicting the desert-dust contribution to PM10 values were evaluated over the Pilot study area using original datasets of desert-dust mass concentration derived from chemical measurements on PM10 samples (Section 2.4). The revised-Methodology was then operationally implemented at both the regional and national scale deriving relevant statistics on desert-dust impact on PM10 levels in Italy (Section 3). Besides providing a basis for comparison with the corresponding EC-Methodology outcomes, this analysis allowed to unveil some features of the desert-dust impact over Italy undetectable following the EC-Methodology approach.

Due to the large amount of data and evidences contributing to this effort, the more specific/technical aspects of the study have been included in five relevant Appendices to the main text (Appendices A-E).

2. Material and Methods

2.1 The study area

Standing in the middle of the Mediterranean Sea, in-between continental Europe and North Africa, Italy is an ideal place where to investigate the transport of Saharan dust to Europe and related impacts (e.g., Gobbi et al., 2004; Gobbi et al., 2006; Matassoni et al., 2009; Mallone et al., 2011; Nava et al., 2012; Marconi et al., 2014; Mona et al., 2014; Stafoggia et al., 2016). Figure 1a shows the country coordinates and location of the official PM10 monitoring sites classified as 'rural background' (green bullets) within the Italian air quality network.

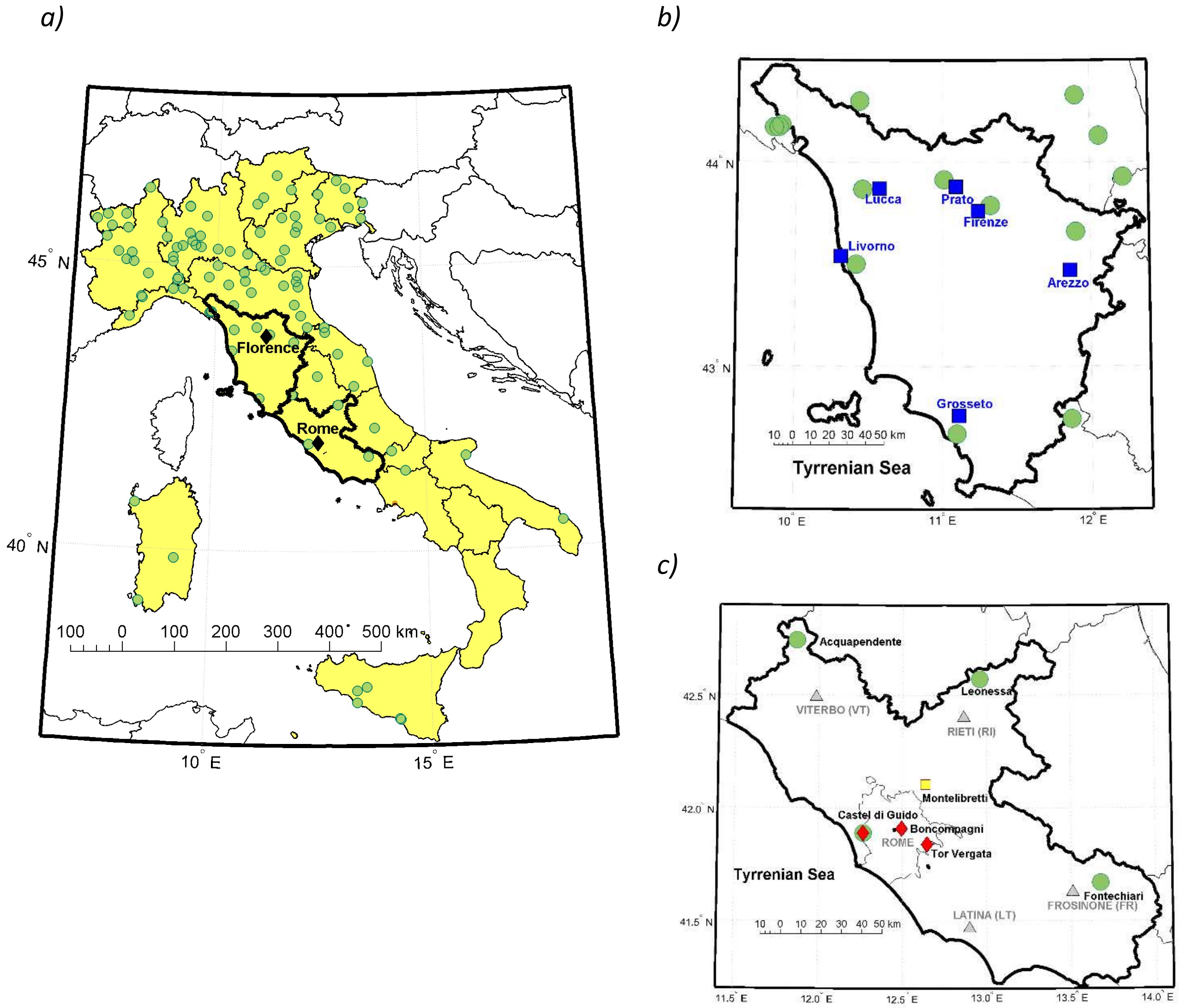

Figure 1: a) The Italian territory (yellow), its regional partitioning (thin black lines) and the PM10 monitoring sites classified as 'rural background' stations active in the last years (green bullets). The regions of Tuscany and Latium in Central Italy are highlighted (thick black lines), as most of the data presented in this work refer to these two regions. These are zoomed in panels b) and c), respectively. In addition to the relevant 'rural background' stations (green bullets), these panels also show location and naming of the sites (coloured symbols) where most of the datasets used in this work were collected. In panel c), boundaries of the Rome Metropolitan area (thin grey line) and location/names of the other 4 main cities (provincial capitals) of the Latium region (grey triangles) are also reported.

A special attention is given in this study to the central Italian regions, and namely to Tuscany (Figure 1b) and Latium (Figure1c), where most of the original PM10 datasets were collected in the framework of the PATOS ('Particolato Atmosferico in Toscana'; Nava et al., 2012; 2015) and

DIAPASON (Gobbi et al., 2017) projects, respectively. Figure 1b illustrates location and naming of the PATOS sites (blue squares) and their position with respect to the rural background stations (green bullets) in the area. Figure 1c shows the Latium region, chosen as DIAPASON Pilot Scale and 'test area' for both a long-term (4-year) application of the current EC-Methodology (see Sections 2.2 and 3.1) and for the test/validation of 'upgrades' to that methodology (Section 2.4). In particular, Figure 1c shows location and naming of the Latium region sites where most of the original observations used in this study were performed: the three DIAPASON monitoring sites in the Rome area (Rome-Tor Vergata, Rome-Boncompagni, and Rome-Castel di Guido, red diamonds) and the EMEP (European Monitoring and Evaluation Programme) station of Montelibretti (yellow square). As in the previous panels, rural background stations are represented by green bullets, among which the DIAPASON site of Rome-Castel di Guido. In the region, routine PM10 measurements are performed by the regional environmental agency (ARPA-Lazio) through a network of 37 air quality monitoring sites (not shown), 20 of which in the Rome metropolitan area and 17 distributed in the areas of the other four main cities of the region (grey triangles in Figure 1c).

2.2 The EC-Methodology and its operational application

We summarize in this section those elements of the EC-Methodology useful for the discussion, its thorough description being available in the specific report released by the European Commission in 2011 (EC, 2011). Some more technical details are also provided in Appendix A, in which we describe the specific tool developed within DIAPASON to facilitate its operational implementation over the Pilot Scale (i.e., the Latium region, Figure 1c).

The EC-Methodology is based on two steps:

1) Identification of the dates affected by desert-dust transport;
2) Quantification of the net desert-dust load on the daily PM10 record.

1) The identification of the days affected by desert-dust transport requires: a) the daily collection of a series of support information (e.g., model forecasts, back-trajectories, satellite images), mostly available via web, and b) the visual evaluation of these datasets/maps by an operator in order to classify each single date as dust-affected or dust-free. Overall, main weak points of this first step are: the non-negligible efforts required to collect all this material; the high degree of subjectivity in evaluating the presence/absence of desert dust based on it; and the occurrence of cases in which such information provides contradictory indications (e. g., a back-trajectory suggesting a Saharan origin, but no desert-dust predicted by the models). In our operational implementation of the EC-Methodology, this last aspect led to a number of cases classified as 'doubtful-dust' (Appendix A).

2) The quantification phase of the EC-Methodology requires a continuous record of daily PM10 data at a single, selected Regional Background (RB) site within the region under investigation, this acting as 'reference' for the entire region. For those days identified as 'dust-affected' at step 1), the net desert-dust contribution ($PM10_{net\_dust}$) to the daily PM10 value recorded at the RB site ($PM10_{RB}$) is estimated as a difference ($\Delta PM10$) with respect to the PM10 values measured at that site within an 'out-of-dust' reference period, i.e.,:

$$PM10_{net\_dust} = \Delta PM10 = PM10_{RB} - PM10_{outofdust\_RB} \tag{1}$$

In the work by Escudero et al. (2007), on which the EC-Methodology is mostly based, the out-of-dust PM10 background ($PM10_{outofdust\_RB}$) is computed as the 30th-percentile PM10 value over a 30-days-moving period, ranging from the 15 days before to the 15 days following any identified 'dust day' (and once any other dust-affected date within this time-window is removed). In the same work, the effect of changing this percentile between the 10th and the 50th is investigated showing, as expected from Eq. 1, that the estimated dust load ($PM10_{net\_dust}$) decreases as this percentile increases. In the EC-Methodology, the use of the 40th percentile is suggested as more conservative. This has been used for example by Pey et al. (2013) to estimate the dust load over Mediterranean countries in the period 2001-2011.

In order to follow a conservative approach, we chose to use the 50th percentile in applying the EC-Methodology, this leading to the lowest estimated desert-dust impact among the tested options (see further details in Appendix A).

Overall, the EC-Methodology has the great advantage of being simple and quantitative and, although with the limitations described in this work, both these aspects are worth to be maintained in any revision/upgrade/advancement of this (or similar) tools.

2.3 EC-Methodology drawbacks and proposed solutions within a 'revised-Methodology'

In addition to the limitations mentioned above, in this section we focus on two further, important drawbacks identified when applying the EC-Methodology to the Pilot area, and mainly related to the 'scientific' assumptions behind it. These are:

1) The choice of a 30-day-long period over which to compute the out-of-dust 'reference' PM10 ($PM10_{outofdust\_RB}$ in Eq. 1);
2) The need to have (or choose) a Regional Background site for this computation.

1) In our application to Italy, the time-window length (±15 days preceding and following the dust date) indicated in the EC-Methodology to evaluate the out-of-dust background PM10 was found to be problematic for three main reasons:

a) There are important meteorological variations occurring over a 30-days period to consider this as an effective 'background period'.

b) The auto-correlation function of PM10 series at 'background sites' becomes low (<0.5) after 1-to-3 days of time lag.

c) By estimating the desert-dust load as a difference (ΔPM10) between 'in-dust' and 'out-of-dust' PM10 conditions, Eq. 1 implicitly assumes this ΔPM10 is (or should be) zero in desert-dust-free periods. We tested this 'null hypothesis' and found that it is not. By testing different time-window lengths, we then found that the shorter the time-window, the minimum this difference keeps, with the closest-to-zero values obtained for a time window of ± 3 days.

The detailed supporting evidences of each of these points are provided in Appendix B. Overall, analysis of each of the three points listed above indicated that the closer the background period to the desert-dust event, the better it represents the 'most likely' PM10 background level over which the desert-dust event builds on. A symmetrical, out-of-dust background period of ±3 days is found to be particularly suitable for the purpose, having the further advantage of avoiding any possible weekly cycle in the PM10 record. This ±3 days- time-window option is therefore implemented within the proposed revised-Methodology.

2) An important disadvantage of the current EC-Methodology application to Italy is the need to define/select a Regional Background (RB) site as a 'clean' reference location to quantify the desert-dust contribution. This choice is challenging for a number of reasons:

a) A site with the required characteristics does not always exist in many Italian regions (Figure 1).

b) In some regions more than one site could be classified as RB (Figure 1): how to choose the most suitable one among the possible options?

c) Saharan dust plumes may exhibit a not-negligible variability over horizontal scales of the order of 20-30 km (e.g., Schepanski et al, 2015; Yuval et al., 2015). To which extent can a value of Saharan dust load derived in the selected RB site be representative of the Saharan dust load affecting larger areas? RB sites are usually far from urban areas where PM10 exceedances are more critical, how much is the desert-dust quantification at the RB site representative of the dust loads in a city area?

d) Saharan dust plumes typically exhibit a not-negligible variability over the vertical scale (e.g., Gobbi et al., 2004; Kishcha et al., 2005, Mona et al., 2014; Gkikas et al., 2016). As Regional Background sites are often elevated sites, this may introduce a further element of potential difference between what is observed at the RB site and what is observed at lower altitudes.

Examples to substantiate these points based on the DIAPASON and PATOS projects datasets are provided in APPENDIX C.

Overall, our results indicate that the need for, selection of, and use of a Regional Background site as required by the EC-Methodology is too constraining, particularly considering that evaluation of the dust load is mostly important at urban sites. We then propose to overcome this limitation by using each single site as a ‘background reference’ for its own PM10 record (‘self-calibration’ approach). This means that Eq. 1 is applied at each site on the basis of its own PM10 record, thus deriving site-dependent $PM10_{net\_dust}$ values. Besides solving the problems/ambiguities as of point 2 above, this ‘self calibration’ approach has the major advantage of producing a regional dust load field (map) rather than a single dust load value to be considered valid over a wide region, as in the EC-Methodology.

These suggested modifications (use of a background period length of ±3 days and of the ‘self-calibration approach’) represent a main revision of the EC-Methodology embedded within the proposed revised-Methodology.

An example of the type of outcome derived applying the revised-Methodology approach over the Latium region is shown in Figure 2. In particular, Figure 2a and 2b show the estimated desert-dust contribution to the daily-average PM10 as obtained for two days within a Saharan advection in May 2014. To insert the results into a more complete picture, in Figure 2 we also include the corresponding view obtained in the same dates from a modelling approach (mean desert-dust-PM10 predicted over Italy by the Chimere model (Menut et al., 2013), Figure 2c, 2d) and from a satellite perspective (AQUA satellite true-colour image for 22 May 2014, Figure 2e). This example depicts the effectiveness of the ‘self-calibration’ approach at capturing the spatial variability of the dust plume contribution, which is typically observed over our regions. Figure 2a highlights a marked northwest-to-southeast gradient, both predicted by the model and captured by the satellite image, with estimated desert-dust loads above 20-25 $\mu g/m^3$ on the north-western part of the Latium region and values lower than 10-15 $\mu g/m^3$ on the South-eastern side (a magenta line has been superimposed to Figure 2a to better highlight this discontinuity). In a similar way, the outcome of the ‘self-calibration’ procedure on May 24 (Figure 2b) captures the southward retirement of the dust plume predicted by the model (Figure 2d). It is also worth noticing how, in this specific case, the position of the RB Fontechiari site (black symbol) makes it unsuitable to represent the desert-dust conditions over the Rome city (thin grey line). This aspect will be further addressed throughout the text.