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Multiannual assessment of the desert dust impact on air quality in Italy combining PM10 data with physics-based and geostatistical models

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ABSTRACT

Desert dust storms pose real threats to air quality and health of millions of people in source regions, with associated impacts extending to downwind areas. Europe (EU) is frequently affected by atmospheric transport of desert dust from the Northern Africa and Middle East drylands. This investigation aims at quantifying the role of desert dust transport events on air quality (AQ) over Italy, which is among the EU countries most impacted by this phenomenon. We focus on the particulate matter (PM) metrics regulated by the EU AQ Directive. In particular, we use multiannual (2006–2012) PM10 records collected in hundreds monitoring sites within the national AQ network to quantify daily and annual contributions of dust during transport episodes. The methodology followed was built on specific European Commission guidelines released to evaluate the natural contributions to the measured PM-levels, and was partially modified, tested and adapted to the Italian case in a previous study. Overall, we show that impact of dust on the yearly average PM10 has a clear latitudinal gradient (from less than 1 to greater than 10 μg/m^3 going from north to south Italy), this feature being mainly driven by an increased number of dust episodes per year with decreasing latitude. Conversely, the daily-average dust-PM10 (≅12 μg/m^3) is more homogenous over the country and shown to be mainly influenced by the site type, with enhanced values in more urbanized locations. This study also combines the PM10 measurements-approach with geostatistical modelling. In particular, exploiting the dust-PM10 dataset obtained at site- and daily-resolution over Italy, a geostatistical, random-forest model was set up to derive a daily, spatially-continuous field of desert-dust PM10 at high (1-km) resolution. This finely resolved information represent the basis for a follow up investigation of both acute and chronic health effects of desert dust over Italy, stemming from daily and annual exposures, respectively.

1. Introduction

Poor air quality (AQ) is generally believed to be associated with anthropogenic emissions, but gases and particles of natural and biogenic origin might largely contribute to worsen the quality of the air we breathe (e.g., Viana et al, 2014; Xie et al., 2021). In highly anthropized environments the intricate mixing of man-made and natural atmospheric emissions can even amplify the detrimental effects of the former (Querol et al., 2019a). Over Europe and the Mediterranean, a non-negligible contributor to the particulate matter (PM) load measured by AQ monitoring networks is represented by desert dust (Escudero et al., 2007; Querol et al., 2009; Aleksandropoulou and Lazaridis, 2013; Gobbi et al., 2013; Pey et al., 2013; Achilleos et al., 2014; Barnaba et al., 2017; Flores et al., 2017; Greilinger et al., 2019; Milford et al., 2019; Gama et al., 2020; which is frequently transported from the arid regions of North Africa and Middle East (Barnaba and Gobbi, 2004; Basart et al., 2012a). Quantification of this natural contribution to AQ relevant metrics as PM10 or PM2.5 (mass of atmospheric particles having

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aerodynamic diameter less than 10 μm and less than 2.5 μm, respectively) is important for air quality legislation. In Europe, the AQ Directive 2008/50 establishes for PM10 an annual limit of 40 μg/m^3 and a daily limit value (DLV) of 50 μg/m^3, not to be exceeded more than 35 days during a calendar year (EC, 2008). Still, if proved to be caused by particles of natural origin, exceedances of the DLV limit value can be considered not as such for the purpose of the Directive.

At the same time, when performed for long periods and over wide areas, such quantification also represents the necessary basis to investigate the impact of desert dust particles on human health. This connection remains currently unclear thus representing a forefront multidisciplinary area of scientific investigation (e.g., Hashizume et al., 2010; Karanasiou et al., 2012; Morman and Plumlee, 2013; Zhang et al., 2016; Tobias et al., 2019; Kotsyfakis et al., 2019; Hashizume et al., 2020; Arnold et al., 2020; Fussel and Kelly, 2021).

An observation-based, dust-PM10 information with fine spatial and temporal resolution would be in this respect crucial to assess the potential role of these particles on human morbidity and mortality. In fact, the scientific community is still debating about possible negative effect of this atmospheric component on human health. Several epidemiological studies found association between atmospheric mineral dust and both morbidity and mortality (e.g. Stafoggia et al., 2016, among others), while some others found no or unclear impact in countries affected by this phenomenon (e.g. Samoli et al., 2011). A recent systematic review commissioned by the World Health Organization (WHO) showed that, due to such contrasting results, no robust conclusions can be drawn yet on the detrimental effects of desert dust PM10 on human health (Tobias et al., 2019) and further investigation from both epidemiological and toxicological perspectives is needed (e.g., Fussell and Kelly, 2021).

Sometimes, simulations from global or regional dust prediction models rather than measurements have been employed to evaluate exposure to dust (e.g., Giannadaki et al, 2014). Desert dust prediction models indeed have the great advantage of producing continuous and homogeneous (in space and time) desert dust fields, being thus able to provide both short- and long-term exposure data. However, given the complexity of the physical, chemical and meteorological processes involved, accurate quantitative predictions of the desert-dust mass transported to Europe and there affecting the lowermost PM10 atmospheric levels is still challenging.

For exposure evaluation purposes, geostatistical models are also receiving attention in the air quality scientific community (Hoek, 2017). Within the broad spectrum of spatiotemporal land use regression methodology, a model-based approach recently emerging to fill spatiotemporal gaps in spatially inhomogeneous environmental measurements is represented by mixed effects regression modelling and machine learning. Specifically, these methods establish a statistical relationship between predictor variables (representing proxies for air pollutants concentrations, as dust or satellite-based aerosol optical depths) and measured PM concentrations, and apply careful cross validation procedures to justify predictions of finely resolved exposures on large spatiotemporal domains, which lack monitoring stations (e.g., Stafoggia et al., 2019).

As a recognition of the need to prove and quantify the natural contribution to PM10 loads, in 2011 the European Commission (EC) released specific guidelines to Member States to suggest reference methods for its assessment (EC, 2011). For desert dust, the reference method (also referred to as EC-Methodology in the following) was built on several studies mainly focused over the Iberian Peninsula (Escudero et al., 2007). The EC-Methodology provided for the first time local, regional and national administrations with a tool to discern the dust contribution in the overall PM10. It certainly still represents a useful tool to assess the desert dust impact on air quality in Europe and has been indeed widely employed by the scientific community in different EU countries (e.g., among others: Achilleos et al., 2014; Cuspilici et al., 2017; Milford et al., 2019; Greilinger et al., 2019; Gama et al., 2020). The procedure first requires the identification of dust-affected dates at the AQ monitoring site locations. This is performed by visual inspection of different dust forecast model outputs, satellite true colour images and meteorological maps. Then, the quantification of the dust-PM10 is performed using a statistical methodology applied to the measured, daily average PM10 data series registered at regional background (RB) sites. For each site of the AQ network, the dust-PM10 is then set equal to the value derived at the closest RB site.

As an effort to both extend its applicability and tailor it to user needs, some modifications to both the identification and quantification phase of the reference EC-Methodology have been recently proposed and tested over Italy using a nationwide annual (2012) dataset (Barnaba et al., 2017). The modifications were introduced to overcome some difficulties encountered in the application of the reference method over the Italian territory, and, as a major objective, to simplify its operability by the potential stakeholders (i.e., regional/national Environmental Protection Agencies in charge of the AQ monitoring) and relevant reporting to the EC. The modified procedure is referred to in the following as 'Diapason-Methodology' for brevity, as it was co-designed with users in the framework of the EC-Life+ Project DIAPASON ('Desert dust Impact on Air quality through model Predictions and Advanced Sensors ObservatioNs', e.g., Rizza et al., 2017; Gobbi et al., 2019). With respect to the EC-reference method, main differences of the Diapason-Methodology are that a) it is fully automatic (i.e., neither the site-resolved identification of dust-affected dates nor its site-resolved quantification require the user intervention) and b) the daily dust-PM10 values are derived in each monitoring site using the own-site PM10 series (i.e., these are not assumed to equal the dust-PM10 value derived in the closest RB site within the network). More details are provided in the Methods section.

A specific validation activity (two dedicated field campaigns performed in autumn 2013 and spring 2014) was also performed to evaluate the 'Diapason-Methodology' (e.g., Struckmeier et al., 2016; Barnaba et al., 2017; Gobbi et al., 2019). This validation showed good ability of the modified procedure to quantify the mass of desert dust contributing to the overall PM10 measured in situ, and to reproduce the temporal, daily-resolved evolution of the advected dust plumes (e.g., Struckmeier et al., 2016; Barnaba et al., 2017). A direct comparison of results obtained applying the EC-Method and the Diapason one in the same RB site in Central Italy (Fontechiari) over a three-year period (2012–2014), showed agreement within 2% on the derived dust-PM10 yearly average (Barnaba et al., 2017 and Appendix E therein). In this study, we use the Diapason-Methodology to derive daily- and site-resolved desert dust-PM10 in over 300 monitoring sites in Italy during the multi annual period 2006–2012, thus assessing the contribution of this important natural component to the EU legislated, PM-related air quality metrics. We then use this dataset to feed a machine learning algorithm, the random forest, to obtain a finely resolved (1-km^2) desert dust-PM10 dataset over the whole Italian domain to be used for exposure evaluation purposes (e.g. Querol et al., 2019b).

2. Materials and methods

2.1. PM10 and PM2.5 datasets

For the analysis presented here we used the PM10 dataset obtained from the Italian air quality database 'BRACE' (https://www.brace.sinanet.apat.it). This is an open-access database set up and maintained by the Italian National Institute for Environmental Protection and Research (ISPRA), with data availability limited to the period 2006–2012.

From such multiannual record, we considered daily average PM data collected in more than 600 monitoring stations in Italy, including rural background (RB), urban background (UB), urban traffic (UT), suburban, rural industrial and urban industrial site types. For each site and for each site-dependent day affected by desert dust, we obtained a dust-PM10 value following the procedure described in Section 2.2.

We then fixed two criteria for filtering the PM10 record at each single site to be included in the statistically significant, national and multiannual assessment. These are:

1) a data coverage per year greater than 70%;
2) a minimum of 5 years of data within the 7-year period considered (2006–2012).

The first filter was applied to avoid inhomogeneous and/or insufficient data coverage over each year included in the statistics (its application led to a reduction of useful monitoring sites per year ranging from 9 to 18% as shown in Appendix A1, Table A1.1). Filter at point 2) was applied to guarantee that each monitoring site included in the overall statistics was sufficiently representative of the multiannual period considered (2006–2012). Its application left a total of 304 PM10 monitoring sites, which therefore represents our ‘selected dataset’ for the multiannual statistics presented is work (Section 3.1).

Fig. 1 summarizes the number of monitoring sites considered before (red) and after (blue) the data selection criteria, resolved by Italian administrative region. It clearly shows that, during the period addressed, there was no homogeneous coverage of monitoring sites within the country, with some southernmost regions suffering a quite low number of stations before and after the data filtering process.

To further investigate specific aspects emerged during the analysis, we also used PM2.5 daily average data to complement the analysis (Section 3.1). In this case, due to a limited availability of this information in the BRACE database, we restricted the analysis to two Italian regions (Lazio and Emilia Romagna) having a sufficiently long (≥3 years) PM2.5 record (2010–2012) and a suitable number of stations classified as RB, UB and UT in the addressed period.

2.2. Methodology to derive the desert dust contribution to PM10

The method we adopted to derive the desert-dust contribution to PM10 (and partially to PM2.5) values is a modified version of the reference EC-Methodology (EC, 2011). It was co-designed with potential stakeholders (namely regional and national air quality agencies) and tested within the EC-Life+ DIAPASON project (https://www.diapason-life.eu) to better adapt to the Italian case and to simplify the use of such kind of tools by expected final users. Here we summarize the ‘Diapason-Methodology’ scheme (Fig. 2), and briefly recall main modifications introduced, as well as pro and cons with respect to the EC approach, full details being available in Barnaba et al. (2017).

The Diapason approach introduces modifications to both steps of the EC-Methodology, i.e., the identification of desert-dust affected days (step 1), and the quantification of the desert dust contribution for those dates (step 2).

In the EC version, step 1 is made through a visual inspection of a series of resources (back trajectories, desert dust forecasts and true colour satellite images). This manual task gives optimal results (best identification of desert dust affected dates) when performed by an expert operator but requires a non-negligible time effort. With the intention to facilitate the use of this method by any potential user not necessarily trained in the field, in the Diapason-Methodology-step 1 we opted to only make use of the numerical outcome of desert dust forecasts to select desert dust days, thus making its outcome operator-independent and fast. In particular, the daily- and site-resolved identification of desert dust-affected days (binary yes/no dust-flag) is made using numerical data of the BSC-DREAM8b model (Basart et al., 2012b). With respect to the EC-Method, this alternative procedure loses the advantage of a supervised evaluation and the countercheck of desert dust presence by visual comparison to additional resources as satellite images and/or different dust models. This option was however preferred as it allows the procedure to be fully automatic and reproducible (operator-independent), which was a main requirement formulated by the involved stakeholders. On the other hand, even the satellite-based evaluation of the desert dust presence over the investigated area could suffer from some difficulties as the frequent co-presence of dust and clouds and,

Fig. 1. Region resolved number of AQ monitoring sites considered in the analysis before (red) and after (blue) the data selection criteria applied (see text) to filter the PM10 dataset to derive the multiannual dust-PM10 statistics. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

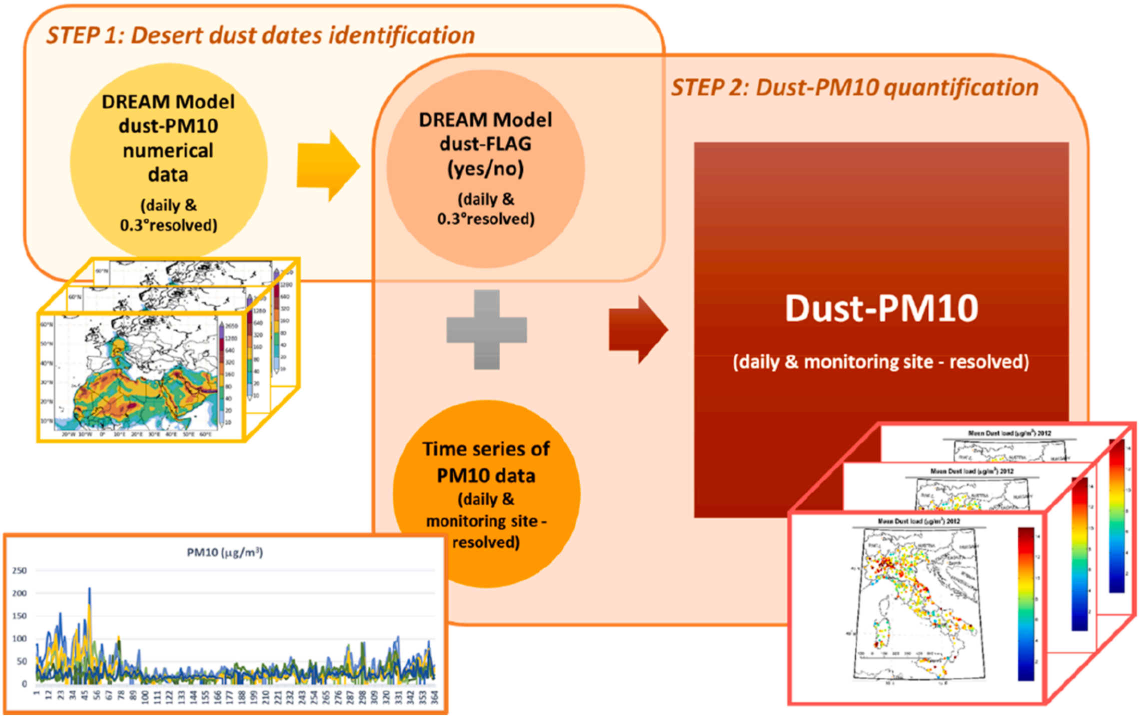

Fig. 2. Scheme of the DIAPASON-Methodology adopted in this study in which daily- and site-resolved model-based (BSC-DREAM8b) flags of desert dust presence (step 1) are combined to daily average PM10 values measured in the AQ network to derive the site-resolved daily average desert-dust PM10 value, if any (step 2).

primarily, from the satellite undetermined altitude of the dust plume over the observed scene. In fact, satellite-detectable desert dust plumes frequently impact the middle troposphere, but not the lowermost levels which are those of interest for air quality evaluations. A further drawback of trusting a single model rather than an ensemble of models (as in the reference method) could be to miss some of the desert dust events. This is however expected to mainly occur for the weakest dust events and on this aspect the Diapason-Methodology was already made ‘conservative’. In fact, in designing the dust-flag we opted to exclude from the statistics the weakest desert dust cases predicted by the BSC-DREAM8b model. In particular, the procedure was set to flag as dust-affected those days/sites where the modelled dust-PM10 daily average exceeds 5 $\mu g/m^3$. This lower limit threshold was fixed optimizing the matching between the BSC-DREAM8b model predictions and desert dust plumes observations performed year-round by ground based remote sensing in Central Italy within the DIAPASON Project (Gobbi et al., 2019). This test showed that, while BSC-DREAM8b predicted values of dust-PM10 at surface are generally higher than those observed (further quantification of this aspect is provided in Appendix A2), timing of the dust plume arrival and persistence at the test-observational sites (three sites in the Rome area) was well reproduced by the model. In fact, this was able to correctly identify over 85% of the dust events observed by in situ and ground based remote sensing instrumentation (e.g. Gobbi et al., 2019). Based on these numbers, it can be estimated that, when applied over Italy, step 1 of the Diapason-Methodology could lead to miss about 15% of the ‘real’ dust-affected dates, and likely within the weakest episodes. More in general, given the absolute values of the BSC-DREAM8b model dust-PM10 over Europe are mainly latitude-dependent (see Appendix A2), the minimum threshold of 5 $\mu g/m^3$ set in the procedure for Italy is expected to be suitable for other regions in Europe laying in the same latitude range. Overall, at the end of step 1, for each calendar date and each monitoring site considered the method provides a yes/no dust flag as indicated in Fig. 2.

Once the dust-affected dates are identified for each site, the quantification phase (step 2) is based on the relevant daily-resolved PM10 data record.

In the EC-Methodology, the quantification of the dust-PM10 is performed assuming that the presence of desert dust produces an increase (ΔPM10) in the observed PM10 values. This dust-induced increase is evaluated in all regional background (RB) sites of the AQ network following Eq. (1). More specifically, the estimated dust-PM10 is obtained as the difference between the daily PM10 value recorded at the RB site on dust-flagged days and a reference, average PM10 value ($<\mathrm{PM10_{RB}}$(out-of-dust)$>$) computed outside of the dust affected period (EC, 2011).

$$\text{dust-PM10}_{\mathrm{RB}}(\text{dust day}) = \Delta \mathrm{PM10} = \mathrm{PM10}_{\mathrm{RB}}(\text{dust day}) - <\mathrm{PM10}_{\mathrm{RB}}(\text{out-of-dust})> \quad (1)$$

In the EC-approach, the $\mathrm{PM10_{RB}}$(out-of-dust) value is computed considering a monthly moving (30-days-long) out-of-dust temporal window including the 15 days preceding and following the selected dust day (excluding any other dust-affected date within this window), and the $<\mathrm{PM10_{RB}}$(out-of-dust)$>$ value is generally taken as the PM10 40th-percentile within this temporal window. Note however that the choice of the 40th percentile is not strictly prescribed by the EC Guidelines (EC, 2011) which further specify that: ‘*the monthly moving 40th percentile is a site specific indicator which reproduces the background concentration existing in the Iberian Peninsula during days with prevailing atmospheric advective conditions. The use of this indicator in other countries has not been validated and no certainty exists on its accuracy. In absence of specific studies that identify the statistical indicator that better reproduce PM10 background* *concentration the use of a more conservative indicator, like the average of the PM10 concentrations registered during 15 days before and 15 days after the analysed dust outbreak episode excluding the days with the identified episode, or the moving 50th percentile of 30 days, should be preferred*'.

Once the dust-$PM10_{RB}$(dust day) values are computed in all RB sites of the AQ network, in any other monitoring site (MS) under evaluation it is assumed that dust-$PM10_{MS}$(dust day) equals that of the closest RB site.

The quantification phase (step 2) in the Diapason-Methodology keeps this ΔPM10 approach of the reference method, but introduces two modifications.

The first is that the daily resolved PM10 record measured in each monitoring site is used to estimate the dust-PM10 at that site, i.e., Eq. (1) is kept valid at each site, so that:

$$\text{dust-PM10}_{MS}(\text{dust day}) = \Delta\text{PM10} = \text{PM10}_{MS}(\text{dust day}) - <\text{PM10}_{MS}(\text{out-of-dust})> \tag{2}$$

The second is that the out-of-dust reference value is computed using the 50th percentile over a shorter temporal window of ±3 days from the dust-affected dates.

Here we recall the main rationale behind these modifications, all the details being thoroughly explained in Barnaba et al. (2017), and Supplementary Material therein.

The first modification was introduced for two main reasons: 1) a specific user requirement was the method to be homogeneously applicable at the national level, but in Italy not all the administrative regions have monitoring sites of RB type; 2) although it is clear that being less affected by urban and industrial sources of PM10, advection of desert dust is more apparent in PM10 time series of RB sites, it is not necessarily true that monitoring sites apart from these receive the same dust input. This happens for three main causes: i) transported desert dust plumes show spatial inhomogeneity over distances of the order of tens of kilometers; ii) complex local orography may introduce differences in dust loads (Diapouli et al., 2017), and complex orography is typical of most Italian regions, iii) RB sites are sometimes located at elevated locations (hills, mountains), making them often unsuitable to represent dust conditions in stations located at low levels.

The second modification, i.e. the reduction of the length of the 'out-of-dust' background period used in Eq. (2), followed a detailed analysis of the variability of PM10 time series at Italian sites in- and out- of dust events. Among others, a major motivation for this reduction is that there are important meteorological variations occurring over a 30-day time window to consider this as an effective 'background period' for dust-affected dates.

Overall, taking benefit of, and building on the EC-reference approach, the described modifications introduced in the Diapason-Methodology produce a site-dependent selection of desert dust affected days and generate a site-resolved dust-PM10 field in a fully automatic and fast way.

Once daily- and site-resolved dust-PM10 values have been determined, dust-related metrics relevant for EC Air Quality legislation have been derived. These are:

1) The dust impact on the yearly average PM10, i.e., the yearly average dust-PM10 (μg/m^3). For each site, this is derived as the sum of the relevant daily average dust-PM10 in a calendar year, divided by 365 (or 366 in leap years);
2) The number of dust-driven PM10 DLV exceedances per year. For each site, this is the number of dust-affected dates in a calendar year in which the total (measured) PM10 exceeds the EC fixed DLV (50 μg/m^3), but this threshold would no longer be exceeded subtracting the estimated daily dust-PM10 from the total PM10. In these cases, it could thus be assumed that the exceedance is dust-driven, in the sense that it would not occur without the additional contribution of dust.

Although not directly linked to AQ legislated values, a third dust-related metric addressed in this study is the 'daily mean dust-PM10 per dust day (μg/m^3)'. For each site, this is derived as the sum of the daily dust-PM10 in a calendar year, divided by the total number of dust-affected dates. This metric is therefore independent from the frequency of dust events in a given site, and is useful to quantify the mean dust-PM10 associated to each dust-affected day at that site.

2.3. The desert dust chemical transport model

As mentioned, within the Diapason-Methodology, daily and site-resolved identification of dust-affected dates was performed based on numerical data from the BSC-DREAM8b daily dust forecast service maintained by the Barcelona Supercomputing Center (BSC).

The BSC-DREAM8b model (Nickovic et al., 2001; Pérez et al., 2006; Basart et al., 2012b) predicts the atmospheric dust cycle including emission, transport and deposition along with dust-radiation interactions. BSC-DREAM8b has been used and evaluated for long time periods over Europe, Northern Africa and the Middle East (Basart et al., 2012b) and it is being used for dust forecasting and as a dust research tool. It is also one of the models whose forecasts should be evaluated within the EC-Methodology step 1 described above.

The BSC-DREAM8b participated in the model inter-comparison of the Regional Center for Northern Africa, the Middle East and Europe of the World Meteorological Organisation Sand and Dust Storm Warning Advisory and Assessment System (WMO SDS-WAS, https://sds-was.aemet.es/). The full model description is available in the BSC daily dust forecast website (https://ess.bsc.es/bsc-dust-daily-forecast). For the present work, dust-PM10 forecasts (0.3° spatial- and 1-hour- temporal resolution) interpolated over the selected Italian AQ stations and averaged on a daily basis were used for the identification of the days affected by Saharan dust.

2.4. The spatio-temporal land use random forest model

To obtain a homogeneous, fine-scale (1-km spatial resolution) and daily-resolved field of the dust-PM10 over Italy in the period 2006–2012, we used the Diapason-Methodology daily estimates of monitor-based desert dust-PM10 to feed a 3-stage random forest model, the 3 stages being "imputation" (to fill gaps in satellite retrievals), "calibration" (to capture monitor-based PM10 variability by use of spatial and spatiotemporal predictions), and "prediction" (to predict daily PM10 concentrations for each km^2 of Italy and each day in 2006–2012). Details of the overall 3-stages approach are presented in Stafoggia et al. (2019). Briefly, for each 1-km^2 grid cell of Italy and each day in the study period we collected a number of spatial (changing from cell to cell but assumed constant over time) and spatiotemporal (varying also within cell by day) parameters, representing potential proxies of desert PM10 concentrations and variability. The first group includes among others: land use terms, elevation, imperviousness surfaces, administrative and geo-climatic regions. The second group includes meteorological parameters, satellite-based aerosol optical depth (AOD from MODIS sensor on board of NASA Terra and Aqua satellites in our case) and monthly vegetation indexes (Normalized Difference Vegetation Index – NDVI). In the first stage of the modelling strategy, the "imputation" one, we built a random forest model aimed at

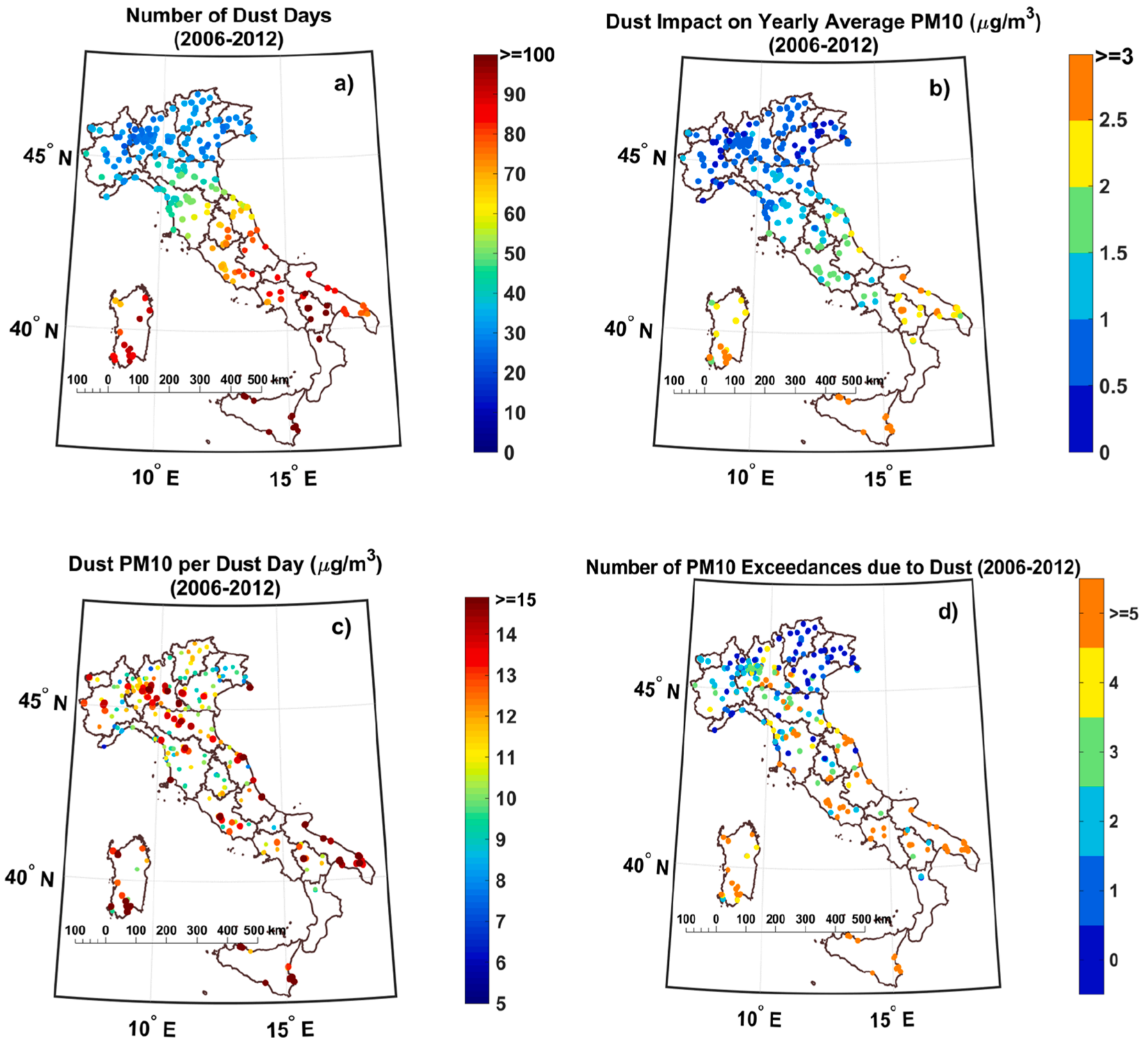

Fig. 3. Multiannual (2006–2012) assessment of the desert dust impact on PM10 air quality in Italy in terms of a) number of dust days per year, b) dust-PM10 yearly average, c) daily mean dust-PM10 per dust day, and d) number of dust-driven exceedances of the PM10 DLV (see text).

reconstructing missing satellite AOD retrievals using multi-band modelled AOD from the Copernicus Atmospheric Monitoring Service (CAMS, https://atmosphere.copernicus.eu/) as input variables. In the second stage, the “calibration” one, we trained another random forest model where the estimates of desert dust PM10 at the monitors were the target variable, and the spatial and spatiotemporal parameters mentioned above were the predictors. This model was carefully cross-validated to ensure generalizability of its output to external points. Finally, in the third stage, the “prediction” one, we applied the calibration model to the entire spatiotemporal domain and derived daily predictions of desert dust PM10 for each 1-km^2 of Italy.

3. Results and discussion

3.1. Assessment of the desert dust impact on PM10 over Italy

The summary of the desert-dust contribution to PM10 values in Italy derived from the multiannual analysis is given in Fig. 3. This shows the site-resolved, multiannual (2006–2012) average values of four main metrics: a) the number of days per year classified as desert-dust affected, b) the desert-dust impact on PM10 yearly average (i.e., the yearly average dust-PM10), c) the daily average dust-PM10 per dust day, and d) the number of DLV exceedances driven by the additional desert dust contribution. Note that metrics b) and d) are those regulated by the EU Air Quality Directive 2008/50.

As expected, Fig. 3a reveals a clear latitudinal gradient of desert dust occurrence, from less than 40 to over 80 desert-dust affected days per year going from Northern to Southern regions. This also reflects into a similar, quite evident north-to-south gradient in the dust impact on the PM10 yearly average (Fig. 3b), with values in Southern Italy more than doubling those in the Northern regions. This is in agreement with the results by Pey et al. (2013) who, for this same dust metric analysed over the period 2001–2011, found a clear dependence of African dust contribution with latitude across the Mediterranean basin, including Italy. A quantification of this latitudinal gradient, and the direct comparison with relevant results obtained by Pey et al. (2013), is given in the Appendix A2 (Figure A2.1). It also includes the comparison of the dust-PM10 yearly averages as derived from the Diapason-Methodology with the corresponding values simulated by the DREAM8b model (Figures A2.2 and A2.3 and relevant discussion).

Results in Fig. 3 are also in agreement with, and extend in time, those obtained for the Italian peninsula by Barnaba et al. (2017), whose country-level analysis was limited to the year 2012. In that case, a slightly lower general impact of desert dust on PM10 values was found, and indeed, over the 7-year period considered in the present study, the year 2012 is associated to the lowest values of most metrics. This is evident in Fig. 4, showing the inter-annual variability of the four dust metrics, aggregated by administrative region.

The country-average summary of this year-to-year variability is also summarized in Table 1 (and in Appendix A1, Tables A1.2, A1.3 and A1.4

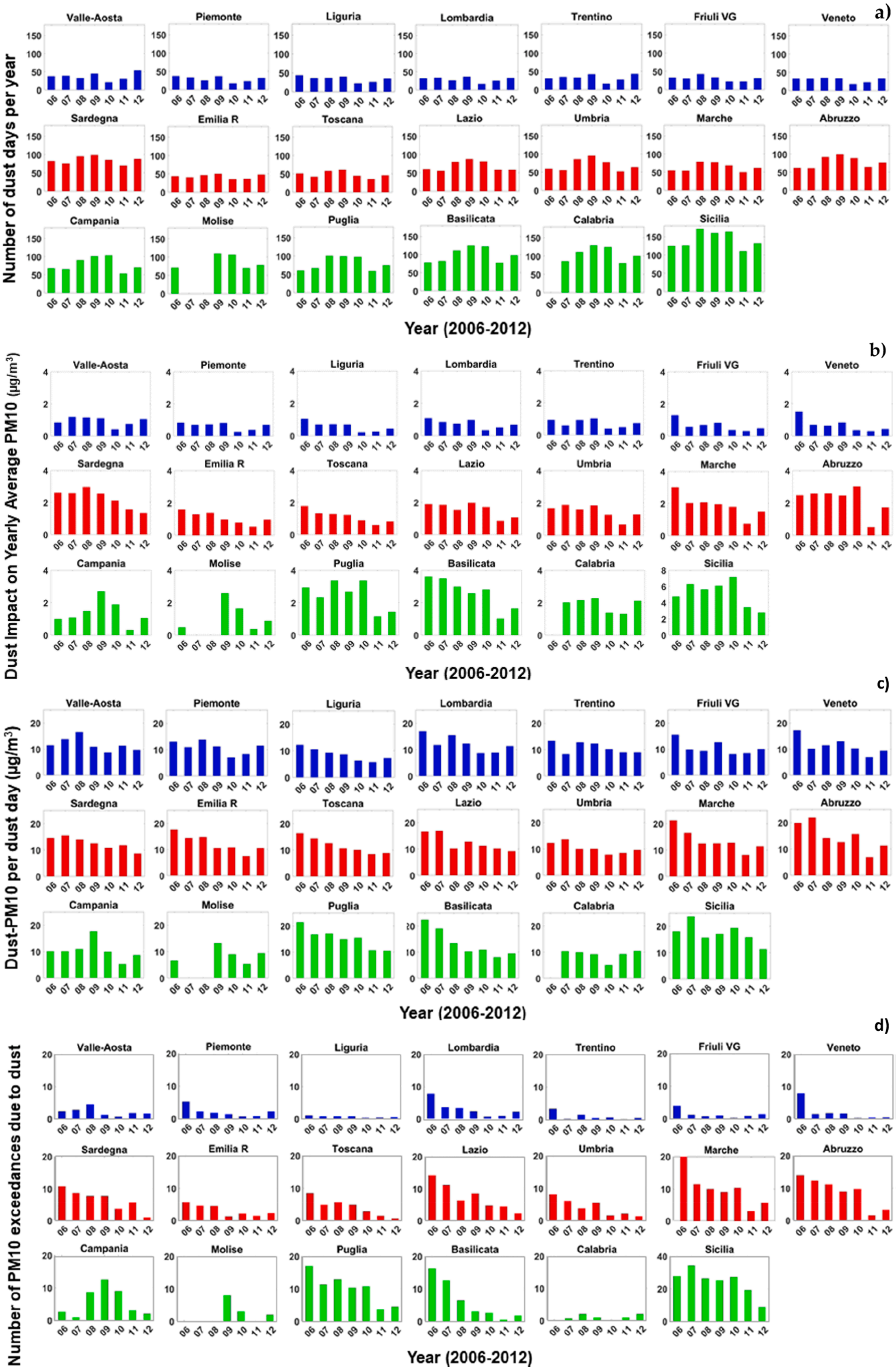

Fig. 4. Inter-annual variability of the four main desert-dust metrics investigated, with data aggregated by administrative region: a) number of desert-dust days per year, b) desert-dust impact on PM10 yearly average, c) average dust-PM10 per dust day, and d) number of desert-dust-driven exceedances of the PM10 DLV. Blue, red and green colours are used for Northern, Central and Southern Italian regions, respectively. Note the different Y axis scale for Sicily in panel b and d. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

Table 1

Inter annual variability of the four main desert-dust metrics investigated (first column) reported as country-average values (±standard deviation and min–max range in parenthesis) for each of the 7 years addressed.

Dust-related metric	2006	2007	2008	2009	2010	2011	2012
Number of desert dust days	53.8 ± 25.3 (28–131)	50.5 ± 22.9 (26–142)	61.7 ± 36.1 (15–184)	68.1 ± 35.2 (30–175)	51.7 ± 39.5 (13–175)	42.2 ± 21.5 (17–114)	51.8 ± 25.7 (22–138)
Yearly Average dust-PM10 ($\mu g/m^3$)	1.8 ± 1.2 (0.2–6.4)	1.6 ± 1.3 (0–10.4)	1.6 ± 1.3 (0.2–6.6)	1.7 ± 1.3 (0.4–10.5)	1.3 ± 1.6 (0.1–13.8)	0.7 ± 0.6 (0.1–4.8)	0.9 ± 0.6 (0.2–3.9)
Daily mean dust-PM10 per dust day ($\mu g/m^3$)	15.9 ± 4.8 (5.1–29.0)	13.7 ± 4.7 (4.8–34.6)	13.2 ± 3.7 (5.7–30.6)	12.1 ± 3.1 (5.8–25.9)	10.3 ± 3.9 (2.9–34.7)	8.9 ± 2.9 (3.8–22.2)	10.1 ± 2.4 (5.3–19.2)
Number of PM10 DLV exceedances due to desert dust	9.6 ± 9.0 (0–51)	6.5 ± 8.4 (0–69)	5.7 ± 6.8 (0–53)	5.3 ± 7.8 (0–83)	3.6 ± 7.4 (0–64)	2.3 ± 4.2 (0–37)	2.2 ± 3.4 (0–30)

by separately aggregating Northern, Central and Southern Italian regions respectively).

Fig. 4a shows that, in the period considered, the number of dust-affected dates was rather stable in the northern regions (blue), while some increase is observed in the central (red), and particularly in the southern (green) regions between 2008 and 2010. Corresponding data in Tables A1.2-A1.4 indicate a frequency of dust events per year ranging between 5% (2010) and 10% (2009) in the northern regions, 13% (2011) and 21% (2009) in the central ones, and between 21% (2011) and 34% (2009) in the southern regions. These numbers are also in line with results by Stafoggia et al. (2016), reporting an annual frequency of dust advection between 28.5% in Palermo (Sicily) and 8.8% in Emilia-Romagna.

On the other hand, it should be noted that variability in the number of dust affected days is somehow decoupled from that of dust-PM10 yearly average, being the latter also dependent on the absolute amount of dust each advection episode transports.

Overall, due to its proximity to the source region, Sicily is the Italian region receiving the largest impact of desert dust transport events, with the maximum number of desert dust days per year (over 150, i.e. over 40%, in 2008, 2009, 2010), and the largest impact of dust on the PM10 yearly average (≥5 $\mu g/m^3$ from 2006 to 2010, all other regions having values lower than 4 $\mu g/m^3$ during the whole period addressed). Interestingly, desert-dust values in Sicily are comparable to climatological mean values derived in the Canary Islands (Spain), close to the desert dust source area (31% of dust days and 7.9 $\mu g/m^3$ yearly average, Querol et al., 2019). In this region, our results for the period 2006–2012 also indicate a mean of 20 dust-driven PM10 DLV exceedances per year (Fig. 4d), out of a maximum of 35 allowed by EC legislation. For comparison, focussing on the period following our analysis (2013–2015) and addressing three monitoring sites in Sicily, Cuspilici et al. (2017) found that desert dust was responsible of 1-to-8 PM10 exceedances (depending on site and year), over a total number of exceedances ranging from 4 to 20. These numbers implied a reduction of the total number of PM10 exceedances after removal of the desert dust contribution ranging from 10% to 90%, depending on site and year. Also note that the lower number of dust-driven PM10 exceedances detected by Culpici et al. (2017) in the period 2013–2015 is compatible with our observed decrease over the years in Fig. 4d (from more than 25 in 2006 to less than 10 in 2012), possibly related to the decrease of the total PM10 in Sicily over that period (e.g. Abita et al., 2015) due to air quality improvement policies. This consideration also applies to the rest of the country, Fig. 4d showing a decreasing trend of dust-driven number of DLV exceedances in almost all regions, which is not associated to a similar trend in the number of dust events (Fig. 4a).

Despite the north-to-south variability of the metrics in Fig. 4a and 4b, it is worth noting that this latitudinal gradient of the dust impact is lost when quantifying indicators that are independent from the absolute number of dust days recorded, as the daily mean desert-dust PM10 associated to a desert-dust affected day (Fig. 3c and 4c). This finding suggests a sort of compensation effect between frequency and intensity of dust episodes. More specifically, the (expected) higher frequency of dust events in the southernmost Italian regions is associated to a wider range of their intensity while, even if sporadic, dust episodes reaching the northernmost regions are the most intense ones.

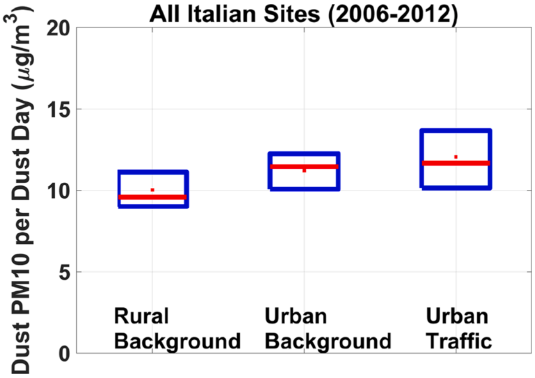

Fig. 5. Statistics of daily mean desert dust-PM10 per dust day as derived over the multiannual period (2006–2012) in the Italian AQ monitoring sites types classified as 'Rural Background', 'Urban Background' and 'Urban Traffic'. For each category the relevant box plot shows: the average (red square), the median (red line), and the 25th-75th inter-quartile range (blue box). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

If not much dependent on latitude, the daily mean desert-dust PM10 was rather found to be higher in correspondence to the high traffic routes and/or over the major urbanized areas of the country in the previous one-year investigation by Barnaba et al., (2017).

In this study we further checked existence of the same pattern over the whole Italian territory in the longer time series addressed (2006–2012). The results, summarized in Fig. 5, confirm that, grouping data by site-type, there is a statistically significant increase (about 2 $\mu g/m^3$, i.e. 20%) of dust-PM10 per dust day values in urban sites compared to those in rural background ones.

A possible explanation of this effect is that it could be related to resuspension of desert dust by traffic due to the vehicle-generated turbulence. In fact, Barnaba et al. (2017) highlighted a geographical correspondence between the highest desert-dust-PM10 per dust day and the path of main traffic routes along the Italian peninsula. To further check this hypothesis, we also used PM2.5 data for selected Italian regions. In fact, if from one side we know that Saharan dust advections over Italy transport particles down to sub-micrometer dimensions (e.g., Struckmeier et al., 2016; Gobbi et al., 2019), coarse particles still represent most of the dust-PM10 mass, and resuspension mostly acts on the coarse PM10 fraction (PM10-2.5) rather than over the fine one (PM2.5). For

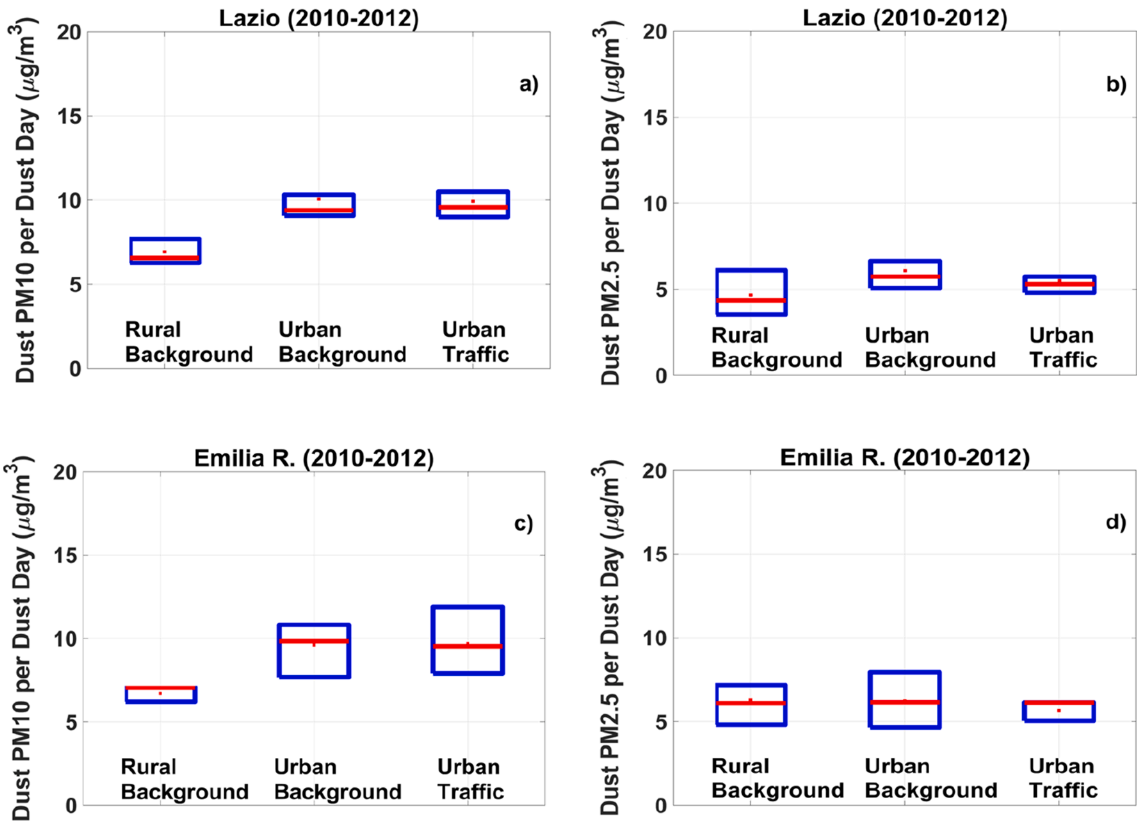

Fig. 6. Left column: As in Fig. 5 but limiting to AQ monitoring sites belonging to the two Italian regions of Lazio (Top) and Emilia Romagna (bottom) and to the period 2010–2012. Right column: As in the left column, but referring to estimated desert-dust PM2.5 data (see text). Note that the reference period for this figure is 2010–2012 as sufficient PM2.5 data were only available starting from 2010.

example, using size-fractionated aluminum and silicon measurements as a proxy for resuspension-related non exhaust emission, (Harrison et al., 2012) investigated the contribution to mass of resuspended particles (in the range 0.9–11.5 µm aerodynamic diameter). By comparing measurements at traffic and background sites in London (UK), they found the contribution of soil dust resuspension to mostly impact the mass size spectra for size ranges above 2 µm.

We performed such further analysis focusing over two Italian administrative regions having sufficient PM2.5 data coverage, and namely Lazio (main urban centre being Rome), and Emilia Romagna (main urban centre being Bologna). The results, summarized in Fig. 6, show that in these two regions the effect of desert dust on PM10 (left column) is similar to the one observed at national level (Fig. 5) while for PM2.5 (right column) this effect is markedly reduced (Lazio) or absent (Emilia Romagna).

This suggests that the dust-induced relative increase detectable on PM10 in the urban environment (Figure 5, and 6a, c) relates primarily to the coarser (i.e., PM10-2.5) fraction. Interestingly, similar conclusions were reached by Querol et al. (2019a) based on data in the Barcelona area (Spain). In fact, by analysing enrichment factor of specific chemical elements during desert dust advections, the authors concluded that mineral dust is not the only pollutant affecting air quality during African dust episodes but, among others, identified 'primary local species in the coarse fraction' as main co-pollutants.

A possible additional (or concurrent) phenomenon compatible with the derived enhanced ΔPM in urbanized environments during dust advections is represented by heterogeneous chemistry taking place on the advected dust particles, acting more effectively in polluted conditions, and possibly more efficiently on the coarse particle fraction (e.g., Dentener et al., 1996; Krueger et al., 2004; Bauer and Koch, 2005; Karagulian and Rossi, 2005; Laskin et al., 2005; Ndour et al., 2009; Wang et al., 2019; Wang et al., 2021). Indeed, some evidence of this effect was found by Conte et al. (2020) in southern Italy who found Saharan dust contributing to secondary organic aerosol in the coarse (PM10-2.5) fraction, with no increase of other species, as BC, that could indicate co-transport of other pollutants with dust.

Overall, this aspect would merit further, deeper investigation particularly for health impact evaluations, as this would imply a differential composition of desert dust particles transported to and thus inhaled in urbanized or background sites of downwind areas.

From an AQ legislation perspective, a question could also arise on whether this additional dust-PM10 contribution observed in urbanized environments could be considered of natural origin. In fact, although triggered by the presence of advected desert dust, the observed amplification effect due to traffic-related resuspension and/or secondary aerosol processes could be more properly considered as anthropogenically driven. By considering such an additional dust-PM10 as of natural origin rather than as anthropogenic, our approach could thus overestimate the number of dust-driven DLV exceedances. We quantified the error affecting our relevant statistics at urban traffic sites. In particular, being the observed urban-to-background dust-PM10 increase of the order of 20% (e.g., Fig. 5), we computed the decrease in the number of dust-driven DLV exceedances that would derive by considering as 'anthropogenic' 10-to-40% of each dust-PM10 value derived at urban traffic sites. The results, aggregated at regional level (see Appendix A3, Figure A3.1), show that, in most regions, total number of dust days with a dust-driven DLV exceedance reduce by less than 3% if attributing 20% of the estimated daily dust-PM10 to anthropogenic factors. This is because in those cases the actual total non-dust PM10 value is lower but close to the DLV, so that even a reduced amount of mineral dust is able to produce the PM10 exceedance. In the most 'unfavourable' cases in which our estimated dust-PM10 at urban sites contains up to 40% of a non-natural, dust–associated PM10 component, the reduction of the dates with DLV exceedances attributed to desert dust keeps lower than

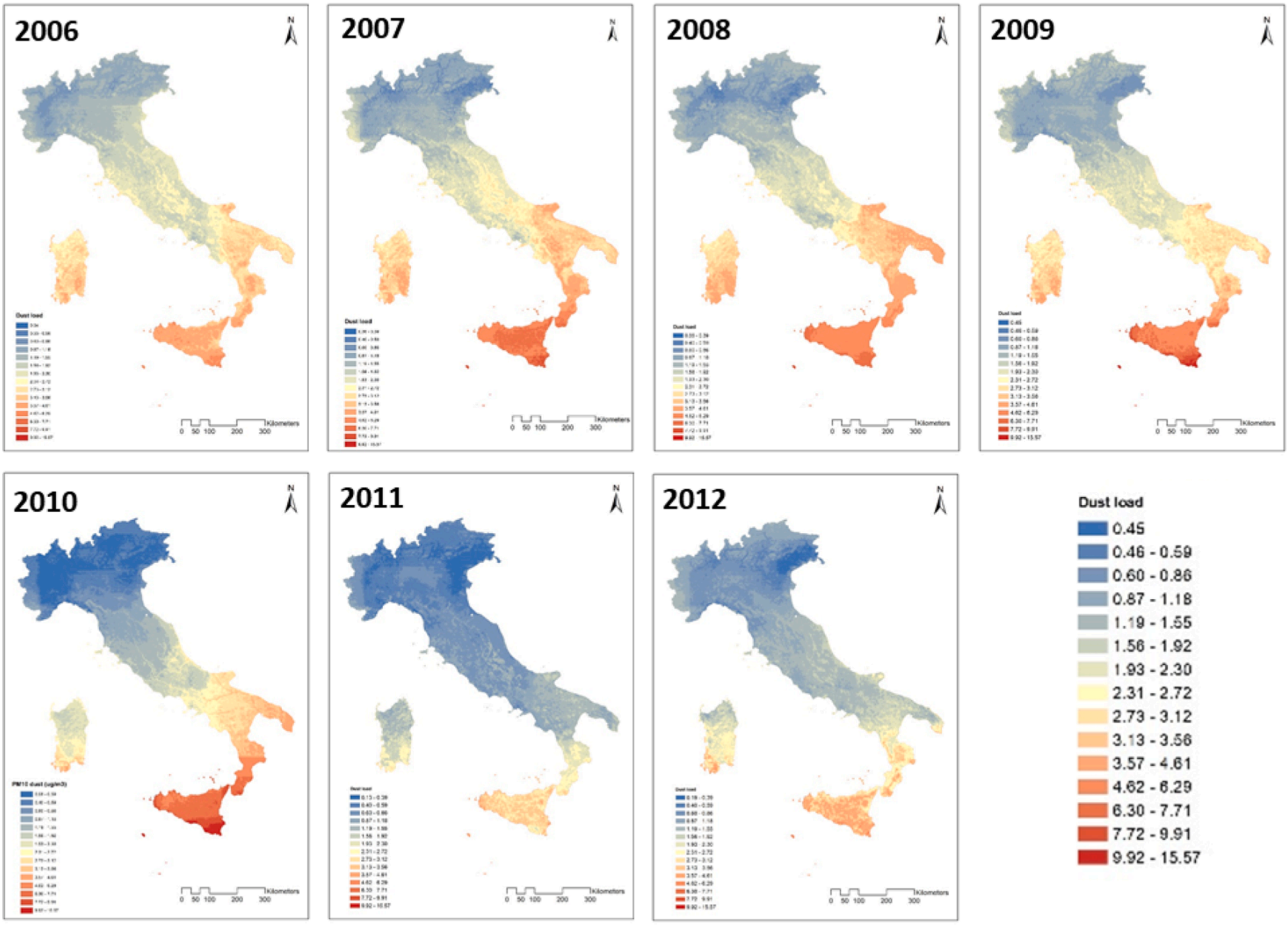

Fig. 7. Fine resolution (1 km) estimates of the dust-PM10 yearly average (μg/m^3) over Italy as obtained from the 3-stage random forest methodology fed with the measurements-based, monitoring site- and daily-resolved dust-PM10 data (2006–2012) derived in this study (see Section 3.1).

6%, with the exception of the Basilicata (-10%) and Sicily (-18%) regions. To be more conservative, as a reference to evaluate dust-driven DLV exceedances in urban areas for the purpose of the EC AQ Directive, dust-PM10 values derived at urban background sites, when available, should be preferred to those derived at urban traffic ones.

3.2. *Using the derived site- and daily dust-PM10 dataset to feed a statistical random forest model*

Numerical models are necessary to extend in time and space the inevitably limited information we can get from observational networks and, often, to predict future scenarios in the short and long time range. In fact, there is an increasing need of high-resolution maps on large domains in order to capture particle matter concentrations gradients both at the local and at the national scale (e.g., Cohen et al., 2017). A useful approach to generate a 'spatially continuous' PM field as a basis for health and epidemiological studies relies on random forest models. As summarized in Section 2.4, here we used such kind of approach (Stafoggia et al. 2019) to obtain daily maps (2006–2012) of desert PM10 concentrations for the entire Italy, at a very fine spatial resolution (1 km) starting from the derived site- and daily- resolved values of dust-PM10. Fig. 7 displays the relevant annual average concentration maps obtained for the investigated multiannual period 2006–2012. Besides showing the strong North-South increasing gradient of the estimated concentrations, these maps also allow to appreciate the fine scale variability of desert contributions to ground level PM10 concentrations. The calibration model set up was able to capture 74% (range: 66%, 83%) of the desert PM10 variability in held-out monitoring stations, without introducing meaningful bias in the final predictions (intercept and slope of the univariate regression between predictions and observations equal to −0.13 (range: −0.17, −0.07) and 1.03 (1.01, 1.04), respectively).

Fig. 7 also highlights a long-term variability, with similar loads between 2006 and 2009 followed by a marked decrease of desert dust PM10 in Northern Italy in 2010 extending to the whole country in the last two years of the period addressed (2011–2012). Such features of highly resolved maps (namely, strong spatial and temporal variability) are being used in a follow up study to investigate both acute and chronic health effects of desert dust stemming from daily and annual exposures respectively. In fact, availability of finely-resolved, nationwide and long-term datasets guarantees adequate statistical power to detect (even potentially small) associations with cause-specific mortality and hospital admissions.

4. Summary and conclusions

Air quality in Europe and the Mediterranean is affected by an intricate mixing of anthropogenic and natural sources. For particulate matter, a major natural source is represented by advected desert dust originating in the arid areas of Northern Africa, and partially Middle East.

Assessment of desert dust impacts on EU-regulated particulate matter loads is important both for complying with the EU Air Quality Directive and to provide a base for health impact studies. In this respect is worth highlighting that the new WHO Global AQ Guidelines (AQG) released in 2021 (WHO, 2021) fix target values of PM levels (annual and daily averages) stricter than before (WHO, 2005) and lower than the relevant EC ones. In particular, for PM10 the new WHO annual average limit is reduced from 20 to 15 μg/m^3 (it is 40 μg/m^3 in EU), while the

DLV is reduced from 50 to 45 μg/m^3 (it keeps 50 μg/m^3 in EU).

In this study we quantify the multiannual impact of desert dust over the whole Italian territory following a previously published method (Diapason-Methodology) which introduces some modifications to the reference, benchmark procedure suggested by the European Commission 2011 (EC, 2011) to better adapt it to the Italian case (Barnaba et al., 2017). In particular, we derive daily desert-dust-PM10 values for hundreds sites of the Italian air quality network, including site types ranging from rural background to urban traffic. These were derived exploiting the Italian PM database 'BRACE', covering the period 2006–2012. To keep the dataset self-consistent, we preferred not to merge it with more recent ones processed according to different QA/QC procedures. However, to further investigate long-term trends, a more extended analysis is currently in progress employing the European Environmental Agency dataset AirBase (https://www.eea.europa.eu/data-and-maps/data/aqereporting-9).

Overall, our results show that desert dust transport affects the lowermost atmospheric levels (i.e. those relevant for air-quality) in 10% (Northern regions)-to-30% (Southern regions) of the dates in a year. Correspondingly, the impact of desert dust on the EC regulated PM10-yearly average also shows a marked north to south gradient, ranging from less than 1 μg/m^3 in Northern Italy to over 3 μg/m^3 over the southern regions, with maxima reaching up to about 10 μg/m^3 in the southernmost parts of the peninsula. Overall, based on our data, we estimate that in the period considered (2006–2012) more than 30 Million people in Italy (i.e., half of the Italian population) have been exposed to an annual average desert-dust-PM10 greater than 1 μg/m^3. Of these, over 2.5 Million (mainly population of Sicily) have been exposed to yearly average desert-dust PM10 values greater than 5 μg/m^3, this representing alone 1/3 of the total-PM10 limit (i.e., including all chemical components) fixed by the WHO AQG (WHO, 2021).

Even if desert dust transport episodes are less frequent in the Northern part of the country, when advected to the lower atmosphere these are found to similarly impact the daily PM10 values over the peninsula. Mean daily contribution is of about 12 μg/m^3, with a site-to-site variability mostly related to the site type rather than to the site latitude. In some cases, the additional contribution of desert dust to local PM is found to produce exceedances of the EC and WHO AQG PM10 DLV. For the application of EU AQ legislation, this is particularly important considering that the Directive 2008/50 allows to discount DLV PM10 exceedances if it can be proved these are caused by a natural event. In fact, in May 2018 the Commission decided to refer Italy to the Court of Justice over persistently high levels of particulate matter (in particular for PM10). This referral concerned exceedances of air quality standards in 28 air quality zones in several regions including Lombardia, Piemonte, Lazio and Veneto, in which the DLV for PM10 have been persistently exceeded. Our results show that, despite the greater distance from source areas, in the northern Italian regions the mineral dust loads during dust episodes can still cause PM10 exceedances, due to the generally higher anthropogenic PM10 burden therein.

The importance of quantifying desert-dust PM10 does not limit to relevance for the current legislation and associated limit values. In fact, if from one side EC allows not to consider exceedances as such for the purpose of the EU AQ Directive, on the health perspective airborne mineral dust represents a significant risk factor for human health (e.g., de Longueville et al., 2013; Goudie, 2014; Aghababaeian et al., 2021). Still, evidences based on both epidemiological (e.g., Tobias and Stafoggia, 2020) and toxicological (e.g., Fussel and Kelly, 2021) studies remain unclear and are thus a current object of scientific research. Among the factors that produce inconsistent results from epidemiological studies are the different metrics and methodologies used to assess the exposure of the population to desert dust, i.e. evaluate the presence and quantify the relative impact of the advected mineral component on the overall PM10 mass. The results presented here were obtained following a single, homogeneous methodology and are provided over each AQ monitoring site of the network, regardless of the site type classification. This is particularly important since, over highly anthropized regions as Europe and the Mediterranean, a complicating aspect in the understanding of the desert dust-health relationship is to disentangle the role of the desert-dust and non-desert-dust components during the mineral dust transport events. In a preliminary assessment over the Italian peninsula based on a single-year record (Barnaba et al., 2017), the site-resolved capacity of the Diapason-Methodology adopted unveiled a differential impact of desert-dust on highly and scarcely urbanized areas. The long-term analysis presented in this work confirms and extend this finding and provide further evidences that, during desert dust transport events, traffic sites show a higher enhancement of PM10 values with respect to background sites (this feature being weakly or not evident for PM2.5). This is compatible with possible resuspension of coarser particles during desert dust events in highly urbanized areas (related to both vehicle-generated turbulence and soil sealing), this contributing increasing PM10 values with respect to unsealed areas (as in regional background sites). This aspect deserves further attention, as current efforts in reducing tail-pipe traffic emissions may enhance the relative role of traffic-related resuspension of dust on PM10 levels in the near future. Interestingly, this finding is also compatible with results obtained by Querol et al. (2019a) over Spain who found a major role in PM10 increase during African advections to be played by 'primary local species in the coarse fraction'. In that work the authors also emphasized that particulate matter increase during desert dust advections is caused not only by the mineral dust load, but also by an increased accumulation of locally emitted or co-transported anthropogenic pollutants. In our long-term analysis, enhancement of PM10 due to co-transport of other pollutants with mineral dust is unlikely to play a major role in the urban-to-rural variability detected, as co-pollutants would have affected the results in downwind regional background and urban sites in the same way. However, an additional (and concurrent) explanation of the observed enhancement of dust-PM10 in urban sites could be the interaction of mineral dust with local pollution via heterogeneous chemical processes (and preferentially occurring over coarse particles), this playing a role in the more polluted urban environment rather than over suburban or background areas. This aspect, which is already documented in literature (e.g., Dentener et al., 1996; Krueger et al., 2004; Laskin et al., 2005; Karagulian and Rossi, 2005; Ndour et al., 2009; Wang et al., 2019; Wang et al., 2021), is particularly intriguing for the dust-health relationship as it could imply a different health impact of transported dust in different downwind environments. In this respect it would be key in the future to couple a spatially and temporally detailed desert dust PM information with health data. A preliminary attempt in this direction is provided in the present study in which the dust-PM10 data derived in hundreds Italian AQ monitoring sites has been used to feed a random forest model to obtain spatially finely-resolved (1 km), daily dust-PM10 data that will be combined to health data (e.g. cause-specific mortality and hospital admissions) in a forthcoming effort.

CRediT authorship contribution statement

Francesca Barnaba: Conceptualization, Methodology, Software, Formal analysis, Data curation, Writing – original draft, Writing – review & editing, Supervision, Funding acquisition. **Nancy Alvan Romero:** Software, Formal analysis, Data curation. **Andrea Bolignano:** Software, Formal analysis, Data curation. **Sara Basart:** Data curation, Writing – review & editing, Funding acquisition. **Matteo Renzi:** Software, Formal analysis, Data curation. **Massimo Stafoggia:** Software, Formal analysis, Data curation, Writing – review & editing.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary material

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envint.2022.107204.

References

Abita, A. R., Antero, G. Ballarino, M. Condò, I. Ferrara, G. Madonia, 2015. Relazione annuale sullo stato della qualità dell'aria nella regione Sicilia anno 2015 e revisione dell'Inventario delle Emissioni per gli anni 2005-2007-2012, https://www.arpa.sicilia.it/temi-ambientali/aria/#1548864447572-6f2b02e1-2783 (in italian).

Achilleos, S., Evans, J.S., Yiallouros, P.K., Kleanthous, S., Schwartz, J., Koutrakis, P., 2014. PM10 concentration levels at an urban and background site in Cyprus: the impact of urban sources and dust storms. J. Air Waste Manag. Assoc. 64 (12), 1352–1360.

Aghababaeian, H., Ostadtaghizadeh, A., Ardalan, A., Asgary, A., Akbary, M., Saeed Yekaninejad, M., Stephens, C., 2021. Global Health Impacts of Dust Storms: A Systematic Review. Environmental Health Insights 15, 1–28. https://doi.org/10.1177/11786302211018390.

Aleksandropoulou, V., Lazaridis, M., 2013. Identification of the influence of African dust on PM10 concentrations at the Athens air quality monitoring network during the period 2001–2010. Atm. Res 13 (5), 1492–1503.

Arnold, C., 2020. Dust Storms and Human Health: A Call for More Consistent, Higher-Quality Studies. Environ. Health Perspect. 128, 11. https://doi.org/10.1289/EHP7845.

Basart, S., Pay, M.T., Jorba, O., Pérez, C., Jiménez-Guerrero, P., Schulz, M., Baldasano, J. M., 2012a. Aerosols in the CALIOPE air quality modelling system: evaluation and analysis of PM levels, optical depths and chemical composition over Europe. Atmos. Chem. Phys. 12, 3363–3392. https://doi.org/10.5194/acp-12-3363-2012.

Basart, S., Pérez, C., Nickovic, S., Cuevas, E., Baldasano, J., 2012b. Development and evaluation of the BSC-DREAM8b dust regional model over Northern Africa, the Mediterranean and the Middle East. Tellus B: Chemical and Physical Meteorology 64 (1), 18539.

Barnaba, F., Gobbi, G.P., 2004. Aerosol seasonal variability over the Mediterranean region and relative impact of maritime, continental and Saharan dust particles over the basin from MODIS data in the year 2001. Atmos. Chem. Phys. 4, 2367–2391. https://doi.org/10.5194/acp-4-2367-2004.

Barnaba, F., Bolignano, A., Di Liberto, L., Morelli, M., Lucarelli, F., Nava, S., Perrino, C., Canepari, S., Basart, S., Costabile, F., Dionisi, D., Ciampichetti, S., Sozzi, R., Gobbi, G.P., 2017. 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. Atmos. Environ. 161, 288–305.

Bauer, S.E., Koch, D., 2005. Impact of heterogeneous sulfate formation at mineral dust surfaces on aerosol loads and radiative forcing in the Goddard Institute for Space Studies general circulation model. Journal of Geophysical Research: Atmospheres 110 (D17).

Cohen, A.J., Brauer, M., Burnett, R., Anderson, H.R., Frostad, J., Estep, K., Balakrishnan, K., Brunekreef, B., Dandona, L., Dandona, R., Feigin, V., Freedman, G., Hubbell, B., Jobling, A., Kan, H., Knibbs, L., Liu, Y., Martin, R., Morawska, L., Pope, C.A., Shin, H., Straif, K., Shaddick, G., Thomas, M., van Dingenen, R., van Donkelaar, A., Vos, T., Murray, C.J.L., Forouzanfar, M.H., 2017. Estimates and 25-year trends of the global burden of disease attributable to ambient air pollution: an analysis of data from the Global Burden of Diseases study 2015. The Lancet 389 (10082), 1907–1918.

Conte, M., Merico, E., Cesaria, D., Dinoi, A., Grasso, F.M., Donateo, A., Guascito, M.R., Contini, D., 2020. Long-term characterisation of African dust advection in south-eastern Italy: Influence on fine and coarse particle concentrations, size distributions, and carbon content. Atmos. Res. 233 (16) https://doi.org/10.1016/j.atmosres.2019.104690.

Cuspilici, A., Monforte, P., Ragusa, M.A., 2017. Study of Saharan dust influence on PM10 measures in Sicily from 2013 to 2015. Ecol. Ind. 76, 297–303. https://doi.org/10.1016/j.ecolind.2017.01.016.

de Longueville, F., Ozer, P., Doumbia, S., Henry, S., 2013. Desert dust impacts on human health: an alarming worldwide reality and a need for studies in West Africa. Int J Biometeorol. 57 (1), 1–19.

Dentener, F.J., Carmichael, G.R., Zhang, Y., Lelieveld, J., Crutzen, P.J., 1996. Role of mineral aerosol as a reactive surface in the global troposphere. J. Geophys. Res.-Atmos. 101 (D17), 22869–22889.

Diapouli, E., Manousakas, M.I., Vratolis, S., Vasilatou, V., Pateraki, S., Bairachtari, K.A., Querol, X., Amato, F., Alastuey, A., Karanasiou, A.A., Lucarelli, F., Nava, S., Calzolai, G., Gianelle, V.L., Colombi, C., Alves, C., Custódio, D., Pio, C., Spyrou, C., Kallos, G.B., Eleftheriadis, K., 2017. 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. Atmos. Chem. Phys. 17 https://doi.org/10.5194/acp-17-3673-2017.

EC, 2008, Directive 2008/50/EC of the European Parliament and of the Council of 21 May 2008 on ambient air quality and cleaner air for Europe (OJ L 152, 11.6.2008, p. 1–44), available at: http://eur-lex.europa.eu/LexUriServ/LexUriServ.do?uri=OJ:L:2008:152:0001:0044:EN:PDF.

EC, 2011, Establishing guidelines for demonstration and subtraction of exceedances attributable to natural sources under the Directive 2008/50/EC on ambient air quality and cleaner air for Europe. 38 pp., February, 2011, available at: http://register.consilium.europa.eu/doc/srv?l=EN&f=ST%206771%202011%20INIT.

Escudero, M., Querol, X., Pey, J., Alastuey, A., Pérez, N., Ferreira, F., Alonso, S., Rodríguez, S., Cuevas, E., 2007. A methodology for the quantification of the net African dust load in air quality monitoring networks. Atmos. Environ. 41 (26), 5516–5524.

Flores, R.M., Kaya, N., Eser, Ö., Saltan, S., 2017. The effect of mineral dust transport on PM10 concentrations and physical properties in Istanbul during 2007–2014. Atmos. Res. 197, 342–355.

Fussell, J.C., Kelly, F.J., 2021. Mechanisms underlying the health effects of desert sand dust. Environ. Int. 157, 106790.

Gama, C., Pio, C., Monteiro, A., Russo, M., Patrícia Fernandes, A., Borrego, C., Baldasano, J.M., Tchepel, O., 2020. Comparison of Methodologies for Assessing Desert Dust Contribution to Regional PM10 and PM2.5 Levels: A One-Year Study Over Portugal. Atmosphere 11 (2), 134. https://doi.org/10.3390/atmos11020134.

Giannadaki, D., Pozzer, A., Lelieveld, J., 2014. Modeled global effects of airborne desert dust on air quality and premature mortality. Atmos. Chem. Phys. 14, 954–968. https://doi.org/10.5194/acp-14-957-2014.

Gobbi, G.P., Angelini, F., Barnaba, F., Costabile, F., Baldasano, J.M., Basart, S., Sozzi, R., Bolignano, A., 2013. Changes in particulate matter physical properties during Saharan advections over Rome (Italy): a four-year study, 2001–2004. Atmos. Chem. Phys. 13 (15), 7395–7404.

Gobbi, G.P., Barnaba, F., Di Liberto, L., Bolignano, A., Lucarelli, F., Nava, S., Perrino, C., Pietrodangelo, A., Basart, S., Costabile, F., Dionisi, D., Rizza, U., Canepari, S., Sozzi, R., Morelli, M., Manigrasso, M., Drewnick, F., Struckmeier, C., Poenitz, K., Wille, H., 2019. An inclusive view of Saharan dust advections to Italy and the Central Mediterranean. Atmos. Environ. 201, 242–256.

Goudie, A.S., 2014. Desert dust and human health disorders. Environ. Int. 63, 101–113.

Greilinger, M., Zbiral, J., Kasper-Giebl, A., 2019. Desert Dust Contribution to PM10 Loads in Styria (Southern Austria) and Impact on Exceedance of Limit Values from 2013–2018. Applied Sciences 9, 11.

Harrison, R.M., Jones, A.M., Gietl, J., Yin, J., Green, D.C., 2012. Estimation of the contributions of brake dust, tire wear, and resuspension to non-exhaust traffic particles derived from atmospheric measurements. Environ. Sci. Technol. 46 (12), 6523–6529.

Hashizume, M., Ueda, K., Nishiwaki, Y., Michikawa, T., Onozuka, D., 2010. Health effects of Asian dust events: a review of the literature. Nihon Eiseigaku Zasshi 65 (3), 413–421.

Hashizume, M., Kim, Y., Ng, C.F.S., Chung, Y., Madaniyazi, L., Bell, M.L., Guo, Y.L., Kan, H., Honda, Y., Yi, S.-M., Kim, H.o., Nishiwaki, Y., 2020. Health Effects of Asian Dust: A Systematic Review and Meta-Analysis. Environ. Health Perspect. 128, 6. https://doi.org/10.1289/EHP5312.

Hoek, G., 2017. Methods for assessing long-term exposures to outdoor air pollutants. Curr. Environ. health Rep. 4, 450462.

Karagulian, F., Rossi, M.J., 2005. The heterogeneous chemical kinetics of NO3 on atmospheric mineral dust surrogates. *Phys. Chem. Chem. Phys.* 7, 3150–3162. https://doi.org/10.1039/b506750m.

Karanasiou, A., Moreno, N., Moreno, T., Viana, M., de Leeuw, F., Querol, X., 2012. Health effects from Sahara dust episodes in Europe: literature review and research gaps. Environ. Int. 47, 107–114.

Kotsyfakis, M., Zarogiannis, S.G., Patelarou, E., 2019. The health impact of Saharan dust exposure. Int. J. Occup. Med. Environ. Health 32 (6), 749–760. https://doi.org/10.13075/ijomeh.1896.01466.

Krueger, B.J., Grassian, V.H., Cowin, J.P., Laskin, A., 2004. Heterogeneous chemistry of individual mineral dust particles from different dust source regions: The importance of particle mineralogy. Atmos. Environ. 38 (36), 6253–6261.

Laskin, A., Wietsma, T.W., Krueger, B.J., Grassian, V.H., 2005. Heterogeneous chemistry of individual mineral dust particles with nitric acid: A combined CCSEM/EDX, ESEM, and ICP-MS study. J. Geophys. Res. 110, D10208. https://doi.org/10.1029/2004JD005206.

Milford, C., Cuevas, E., Marrero, C.L., de Bustos, J.J., Gallo, V., Rodríguez, S., Romero-Campos, P.M., Torres, C., 2019. Impacts of Desert Dust Outbreaks on Air Quality in Urban Areas. Atmosphere 11, 1.

Morman, S.A., Plumlee, G.S., 2013. The role of airborne mineral dusts in human disease. Aeolian Res. 9, 203–212.

Ndour, M., Nicolas, M., D'Anna, B., Ka, O., George, C., 2009. Photoreactivity of NO2 on mineral dusts originating from different locations of the Sahara Desert. Phys. Chem. Chem. Phys. 11 (9), 1312.

Nickovic, S., Kallos, G., Papadopoulos, A., Kakaliagou, O., 2001. A model for prediction of desert dust cycle in the atmosphere. J. Geophys. Res. 106 (D16), 18113–18129.

Pérez, C., Nickovic, S., Pejanovic, G., Baldasano, J.M., Ozsoy, E., 2006. Interactive dust-radiation modeling: a step to improve weather forecasts. J. Geophys. Res. 11, D16206.

Pey, J., Querol, X., Alastuey, A., Forastiere, F., Stafoggia, M., 2013. African dust outbreaks over the Mediterranean Basin during 2001–2011: PM10 concentrations, phenomenology and trends, and its relation with synoptic and mesoscale meteorology. Atmos. Chem. Phys. 13, 3.

Querol, X., Pey, J., Pandolfi, M., Alastuey, A., Cusack, M., Pérez, N., Moreno, T., Viana, M., Mihalopoulos, N., Kallos, G., Kleanthous, S., 2009. African dust contributions to mean ambient PM10 mass-levels across the Mediterranean Basin. Atmos. Environ. 43 (28), 4266–4277.

Querol, X., Pérez, N., Reche, C., Ealo, M., Ripoll, A., Tur, J., Pandolfi, M., Pey, J., Salvador, P., Moreno, T., Alastuey, A., 2019a. African dust and air quality over Spain: Is it only dust that matters? Sci. Total Environ. 686, 737–752.

Querol, X., Tobías, A., Pérez, N., Karanasiou, A., Amato, F., Stafoggia, M., Pérez García-Pando, C., Ginoux, P., Forastiere, F., Gumy, S., Mudu, P., Alastuey, A., 2019b. Monitoring the impact of desert dust outbreaks for air quality for health studies. Environ. Int. 130, 104867.

Rizza, U., Barnaba, F., Miglietta, M.M., Mangia, C., Di Liberto, L., Dionisi, D., Costabile, F., Grasso, F., Gobbi, G.P., 2017. WRF-Chem model simulations of a dust outbreak over the central Mediterranean and comparison with multi-sensor desert dust observations. Atmos. Chem. Phys. 17, 93–115. https://doi.org/10.5194/acp-17-93-2017.

Samoli, E., Kougea, E., Kassomenos, P., Analitis, A., Katsouyanni, K., 2011. Does the presence of desert dust modify the effect of PM10 on mortality in Athens, Greece? Sci. Total Environ. 409 (11), 2049–2054. https://doi.org/10.1016/j.scitotenv.2011.02.031.

Stafoggia, M., Zauli-Sajani, S., Pey, J., Samoli, E., Alessandrini, E., Basagaña, X., Cernigliaro, A., et al., 2016. Desert Dust Outbreaks in Southern Europe: Contribution to Daily PM10 Concentrations and Short-Term Associations with Mortality and Hospital Admissions. Environ. Health Perspect. 124, 4.

Stafoggia, M., et al., 2019. Estimation of daily PM 10 and PM 2.5 concentrations in Italy, 2013–2015, using a spatiotemporal land-use random-forest model. Environ Int. 124, 170–179.

Struckmeier, C., Drewnick, F., Fachinger, F., Gobbi, G.P., Borrmann, S., 2016. Atmospheric aerosols in Rome, Italy: sources, dynamics and spatial variations during two seasons. Atmos. Chem. Phys. 16, 15277–15299. https://doi.org/10.5194/acp-16-15277-2016.

Tobias, A., Karanasiou, A., Amato, F., Roqué, M., Querol, X., 2019. Health effects of desert dust and sand storms: a systematic review and meta-analysis protocol. BMJ open 9, 7.

Tobias, A., Stafoggia, M., 2020. Modeling desert dust exposures in epidemiologic short-term health effects studies. Epidemiology 31 (6), 788–795.

Viana, M., Pey, J., Querol, X., Alastuey, A., de Leeuw, F., Lükewille, A., 2014. Natural sources of atmospheric aerosols influencing air quality across Europe. Sci Total Environ. 472, 825–833. https://doi.org/10.1016/j.scitotenv.2013.11.140.

Wang, W., Liu, M., Wang, T., et al., 2021. Sulfate formation is dominated by manganese-catalyzed oxidation of SO_2 on aerosol surfaces during haze events. Nat Commun. https://doi.org/10.1038/s41467-021-22091-6.

Wang, Z., Uno, I., Yumimoto, K., Pan, X., Chen, X., Li, J., Wang, Z., Shimizu, A., Sugimoto, N., 2019. Dust Heterogeneous Reactions during Long-Range Transport of a Severe Dust Storm in May 2017 over East Asia. Atmosphere. 10 (11), 680. https://doi.org/10.3390/atmos10110680.

WHO, World Health Organization, 2005. Air quality guidelines for particulate matter, ozone, nitrogen dioxide and sulfur dioxide, Global update 2005, Summary of risk assessment, WHO Regional Office for Europe, WHO/SDE/PHE/OEH/06.02.

WHO, World Health Organization, 2021. Air Quality Guidelines - Update 2021. WHO Regional Office for Europe, Copenhagen, Denmark.

Xie, W., Li, Y., Bai, W., et al., 2021. The source and transport of bioaerosols in the air: A review. Front. Environ. Sci. Eng. 15, 44. https://doi.org/10.1007/s11783-020-1336-8.

Zhang, X., Zhao, L., Tong, D., et al., 2016. A systematic review of global desert dust and associated human health effects. Atmosphere 7, 158.