

The paper presents 21 years (2003-2023) of aerosol optical size distribution in a WMO/GAW, ACTRIS-RI and ICOS-RI sampling place at Monte Cimone peak, which have been used to identify dust transport days on site and to analyze its interannual and seasonal patterns for both frequency of occurrence and observed particle concentration. Air masses pathways were also analyzed to confirm the selected days. The paper is an extension of the analysis shown in Duchi et al. (2016), which similar goals and methodology but with 11 years more of database.

We thank Reviewer 2 for the detailed and useful comments. Based on that we made the following main changes in the manuscript:

- Added a section of detailed description of the backward trajectories
- Added a section discussing the uncertainties
- Added a section comparing our work to Duchi et al. (2016)
- Added two tables to summarize the results
- Added supplementary material

The structure of our answer to the comments is: black – Comment of the reviewer, blue – Author's response, red – changes made in the manuscript

General comments

The topic of the paper is suitable for ACP, the methodology is clear, the results are logically interpreted and the manuscript is well written. But, it is well-known the doubts of the scientific community when using the aerosol optical size distribution to study desert dust episodes. Therefore, it is observed a lack of explaining the uncertainty in the experimental technique, and how this may affect the estimated parameters. While information of the inventory and duration of the events is strong, other parameters as total mass concentration, enhancement or the comparison with the WHO threshold are subject to some uncertainty. It is due to, as authors comment in Sections 2.1 and 2.3, OPS instruments measure the particle number size distribution, which is converted into a mass distribution using a particle density dependent by particle size, but it also depends on aerosol composition. The dependence on particles composition becomes much stronger, due to the variability in the aerosol typing studied in this work: desert dust and background aerosol.

Therefore, before publishing this work, the authors should resolve this issue, explaining the uncertainties in the estimations. Perhaps, the use of complementary information such as off-line mass concentration, multi-wavelength absorption coefficient, and/or aerosol optical depth could be useful, even if it is of shorter periods.

A similar point was raised by reviewer 1, so we included in the method section a detailed paragraph on the instrumental uncertainties. Moreover, their influence on the data is discussed in multiple parts throughout the manuscript. In the following we list the changes made in the manuscript:

- UNCERTAINTY DEFINITION: we added in the methods a section to describe the uncertainties of the individual terms in the calculation of the PMcoarse concentration. The new section in the methods reads as follows:

2.7 Uncertainties

The calculation of the PMcoarse concentration is subject to uncertainties. Given Equation 1, individual uncertainties of the particle diameter (d_i), the particle number concentration ($C_{n,i}$) and the particle density (ρ_i), are propagated into a final uncertainty of the PMcoarse concentration.

For $C_{n,i}$, the manufacturer provides for the same OPC model an uncertainty between 3% and 5%. In the literature, the characterization of the uncertainty is limited to one study by Burkart et al. (2010) who observed a 9 % higher total number concentration measured by the same OPC compared to a differential mobility analyzer. However, they did not convert the electrical mobility diameter to an optical equivalent diameter, which can lead to an increased uncertainty. We therefore apply in our calculation of the error propagation the uncertainty of 5 %.

An uncertainty for d_i is not provided by the manufacturer of the OPC; however, it should be accounted due to biases in the correct sizing introduced by non-spheric particles. Putaud et al. (2004) suggest in their study at Monte Cimone a particle sizing uncertainty of 10 % outside of DTDs and of 20 % during DTDs. The higher uncertainty during DTDs arises from the high degree of non-sphericity of dust particles.

The uncertainty for (ρ_i) is not given in the study by Wittmaack (2002), which we used to obtain the size dependent particle density. For our calculations we estimated an upper and lower uncertainty both for background conditions and during DTDs. For the upper limit we used the ratio between the mean PMcoarse concentration calculated as described in Sec. 2.4 and the mean PMcoarse concentration calculated with the highest density we used of 2.6 g cm^{-3} . On the other hand, for the lower limit, we used the lowest density of 2.1 g cm^{-3} for measurements during DTDs and 1.77 g cm^{-3} for background conditions. During background conditions the aerosol present at Monte Cimone is mainly organics, ammonium sulphate and unknown particles (Putaud et al., 2004) Based on this calculation, we obtained the following uncertainty ranges for the density: DTDs + 9.5 %/ - 9.8 % and background conditions + 11.4 %/ - 28 %. Applying the error propagation, we obtain the upper and lower uncertainty for the PMcoarse concentration during DTDs +/- 61 % and during background conditions + 32 %/ - 41 %.

- UNCERTAINTY IN THE NUMBER OF DETECTED DUST TRANSPORT DAYS

To assess the uncertainty in the number of the detected dust days, we assumed that the $\pm 5\%$ uncertainty of the OPC number concentration accounts in the same way for the high frequency component and the threshold value for DTD identification. This leads +5 and -8 detected DTDs over a total of 1004 DTDs, which are negligible

numbers, that do not have an effect in our presented analysis. We changed the text in L.147 to the following:

The uncertainty in the quantification of DTDs was calculated assuming the +/- 5% uncertainty of the OPC counting for both the high frequency component and the threshold, which are the variables directly used to identify DTDs. Hence, the maximum overestimation of DTDs was calculated assuming a +5% on the high frequency component and a -5% on the threshold. The opposite was done to estimate the maximum underestimation of DTDs. Overall, we obtained +5 and -8 DTDs, which are negligible numbers given the total number of 1004 days. We can conclude that an effect of the measurement uncertainty on the analysis presented in the paper can be excluded.

- **UNCERTAINTY IN THE PMCOARSE CONCENTRATION**

In the original manuscript we showed in Fig. 3 and Fig. 4 the median and 25th and 75th percentiles. We agree with the reviewer that plotting the uncertainties provides more information. Therefore, we added the average values together with the error bars, defined as +/- 61% in Fig. 3 a) and 4 a). The average values are consistently higher than the median, which points out that the underlying dataset is not normally distributed and contains extreme values driving the average. This is underlined by the fact that the standard deviation is always larger than the average value. In such cases, it is recommended to rely on the median for the data analysis and interpretation. However, we think that we can add value to the discussion by showing the average, highlighting that a few dust transport events with very high PMcoarse concentrations may influence the statistics of the dataset. Concerning this point, we added a discussion in Section 3.3.1 L.204, which now reads as:

The average of the PMcoarse concentration during DTDs (Fig 4 (a), dark brown line) is consistently higher than the median. In most of the years the error bars of the average, given as +/- 61% include the median value. In exceptional years, such as 2014, the difference between the median and the average can be as high as a factor of 5, while the standard deviation is always higher than the average. These statistics point out that the PMcoarse concentration during DTDs is driven by one or two events per year transporting very high amounts of dust mass towards Monte Cimone and thus leading to a skewed distribution of the PMcoarse concentration. To reduce the weight of extreme events on the multi-decadal time series, it is recommended to rely on the median values for further analysis. All three variables, i.e., the median of the PMcoarse background, the median and the average PMcoarse during DTDs, showed a wave-like profile with a wavelength of about 12 years.

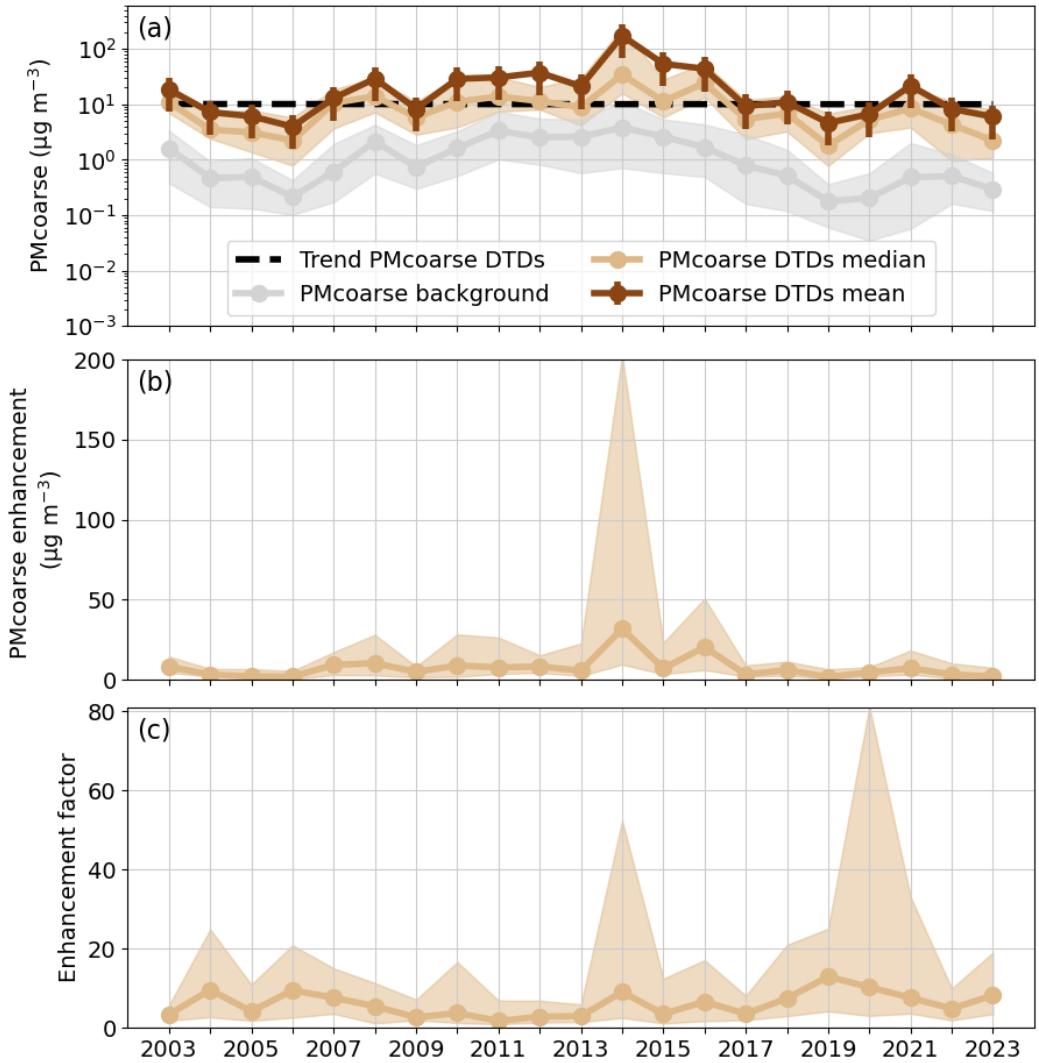


Figure 3. (a) Annual median PMcoarse concentration during (brown) and outside (grey) DTDs. The dark brown line shows the average values during DTDs including error bars, defined as $\pm 61\%$. The dashed line shows the trend in the PMcoarse concentration during DTDs. (b) Enhancement in the PMcoarse concentration during DTDs. (c) Enhancement factor (EF) of the PMcoarse concentration during DTDs. In all panels, the solid line shows the median values; the shaded area around is the 25th and 75th percentile.

and 3.3.2 L.223, which now reads as:

The average of the PMcoarse concentration during DTDs does not follow the seasonal cycle. While the average PMcoarse concentration aligns with the 75th percentile until October, from October to December it grows up to a factor of 5 higher than the 75th percentile. This increase suggests an increasing influence of intense dust transport events on the PMcoarse concentration and rises an issue on how the PMcoarse concentration during DTDs should be assessed statistically. While the median values help identifying recurring conditions or cycles and drawing a climatology over a long time period, averages may underline months containing strong dust transport events and may be used to isolate specific and intense anomalies. As the focus of this paper is the analysis of the climatology, the following results will be discussed based only on the median values.

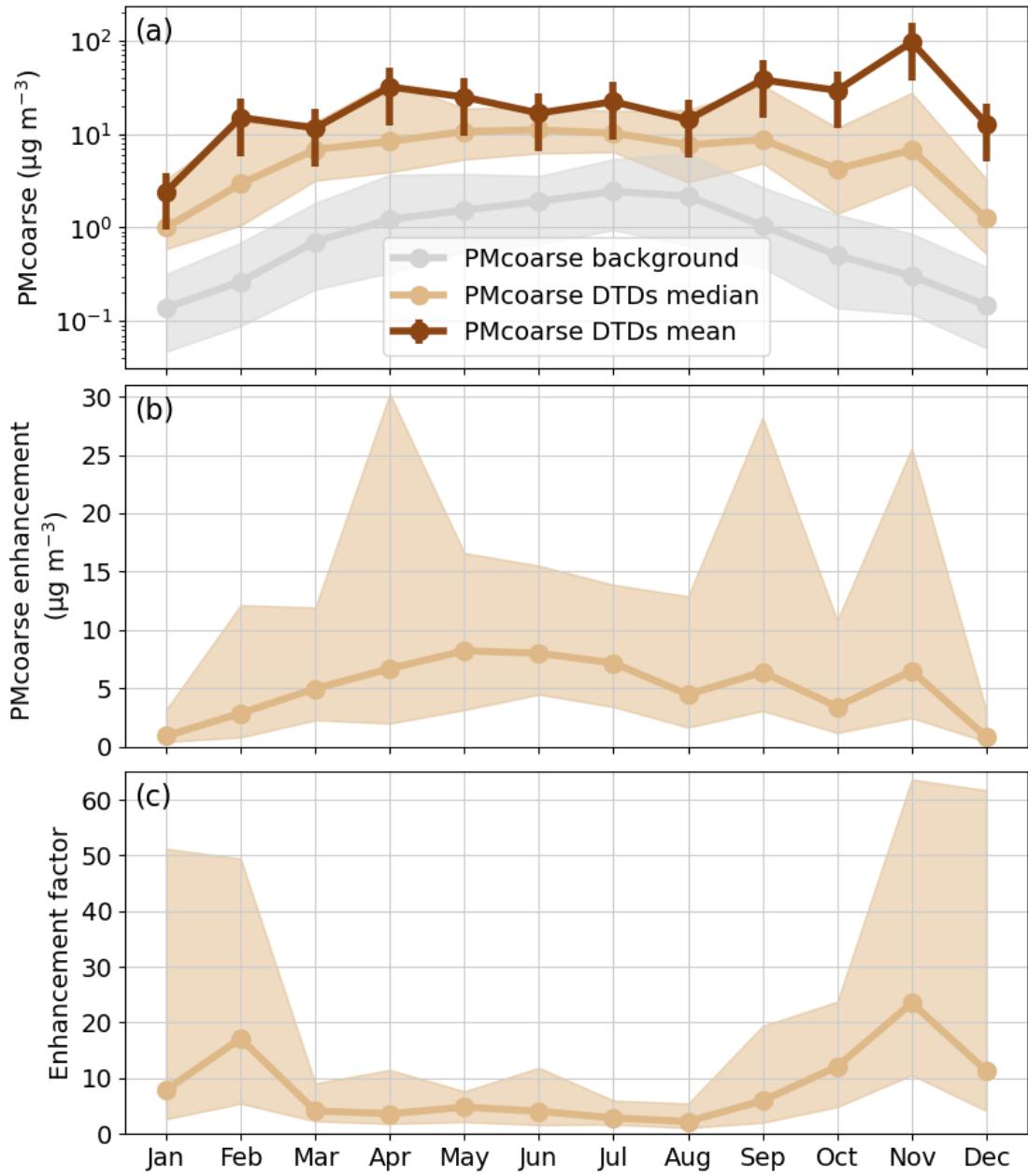


Figure 4. (a) Monthly median PMcoarse concentration during (brown) and outside (grey) of DTDs. The dark brown line shows the average values during DTDs including error bars, defined as $\pm 61\%$. (b) Enhancement in the PMcoarse concentration during DTDs. (c) Enhancement factor (EF) of the PMcoarse concentration during DTDs. In all panels, the solid line shows the median values; the shaded area around is the 25th and 75th percentile.

- **UNCERTAINTY IN THE PMCOARSE ENHANCEMENT AND INFLUENCE ON WORLD HEALTH ORGANIZATION THRESHOLD EXCEEDANCE**

The original manuscript showed the maximum daily PMcoarse enhancement as function of the duration as a bar graph. Based on the discussion around the uncertainties, we chose to modify Fig. 6 and 7 to show each dust transport event, the overall median and the WHO threshold, as function of the DTE duration. The results are summarized in a table. We accounted for the uncertainty by applying the lower and upper uncertainty on the data and counting the respective days that exceed the WHO threshold. The text is changed to the following:

To assess the intensity of DTEs as function of their duration and peak PMcoarse enhancement, we investigated the change of the maximum daily PMcoarse enhancement of the different DTE durations (Fig. 7). The median of the maximum daily PMcoarse enhancement increased steadily with the duration of DTEs from 2.28 $\mu\text{g m}^{-3}$ for the 1-day events to 19.47 $\mu\text{g m}^{-3}$ for DTEs lasting at least 4 days (Table 2). This means that the longer the DTE, the more likely it is to reach higher enhancements in the PMcoarse concentrations. The same pattern was observed when dividing the data into the different seasons (Figure 8 and Table 2). The median values of the maximum PMcoarse enhancement stayed consistently below the WHO threshold value of 45 $\mu\text{g m}^{-3}$. One reason for the observed behavior could be that for longer DTEs a more extensive dust plume reached CMN, which might be connected to a dust storm over the Saharan desert induced by strong winds. Short DTEs might originate from dust transport of the generally dust loaded air over the Sahara without a prior dust storm. Another important point to mark here is that longer lasting DTEs seem to be more likely to occasionally exceed the threshold value given by the WHO, as the fraction of DTEs above the threshold increases from 5.8% for 1-day events to 24.1% for events that last at least four days. To account for the uncertainty of the PMcoarse enhancement and its effect on the threshold exceedance, the +/- 61% uncertainty, as given in Sec. 2.7, is applied on the data and the respective days exceeding the threshold are counted (see Table 2). While the analysis of the full data set (Fig. 6) is based on enough data for each duration class, some of the seasonal data (Fig. 7) must be taken carefully as only very few events are available. Independent of the uncertainty, the numbers presented here give only a lower limit as we consider the PMcoarse and not the PM10 concentration and by that exclude some part of the aerosol mass concentration.

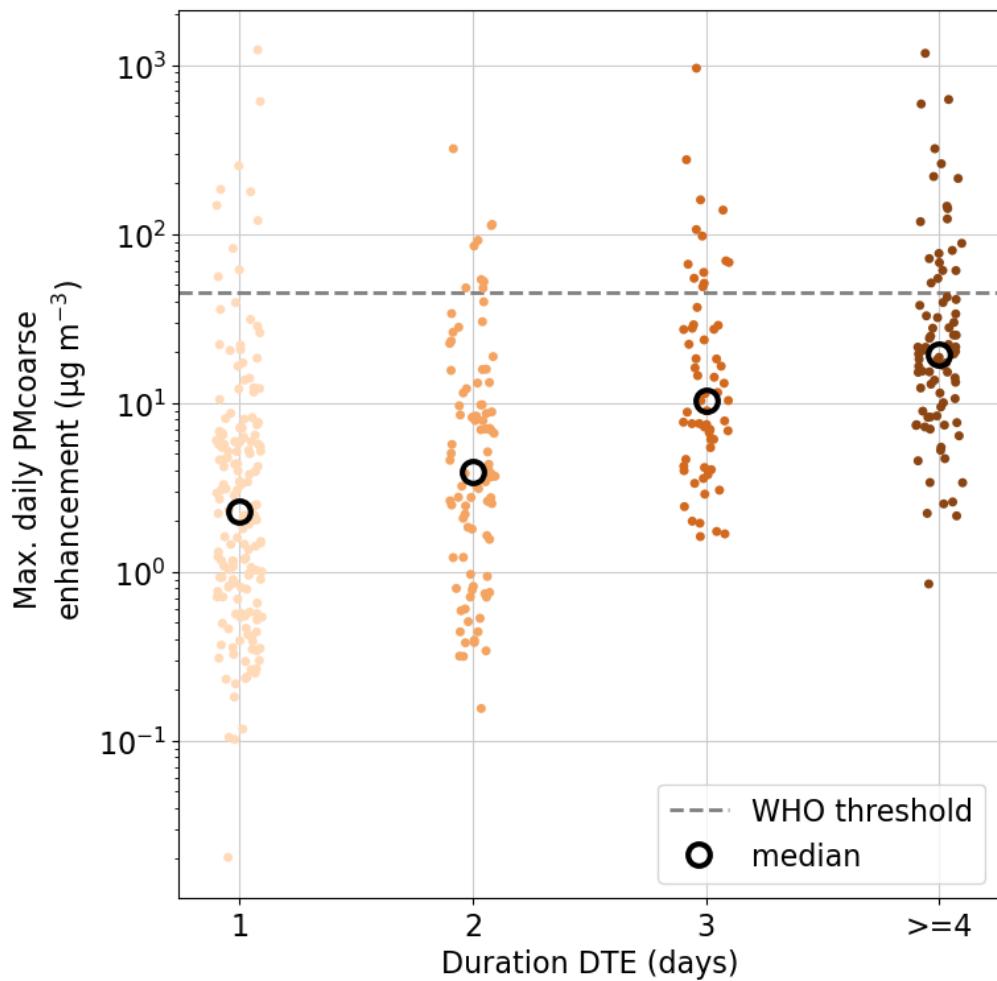


Figure 6. Maximum daily PMcoarse enhancement for the four different lengths of DTE. Each point represents one DTE, the black circle the median value and the dashed line the WHO threshold.

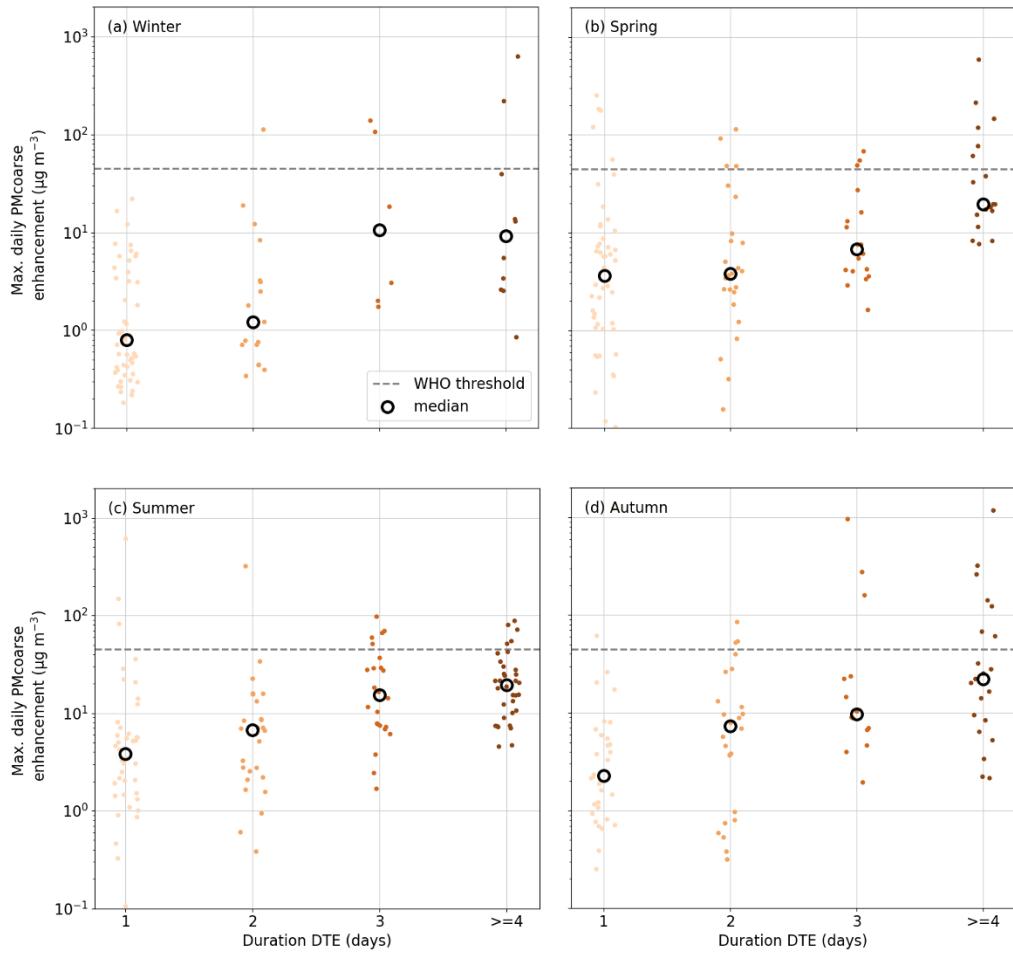


Figure 7. Maximum daily PM_{coarse} enhancement for the different durations of dust transport events. The data are divided into the four seasons: (a) winter, (b) spring, (c) summer and (d) autumn. Each point represents one DTE, the black circle the median value and the dashed line the WHO threshold.

Table 2. Summary of the results from Fig. 6 and 7 indicating the median of the maximum daily PMcoarse enhancement for the four dust transport event duration categories (1 day, 2 days, 3 days and ≥ 4 days) the median of the maximum daily PMcoarse enhancement. The percentage of how many DTEs exceeded the WHO threshold value is also provided. To account for the uncertainty, the lower and upper uncertainty was applied on the dataset and the respective number of days above the threshold were counted. The number in brackets give the total number of events

		1 day	2 days	3 days	≥ 4 days
all	Median	2.28	3.93	10.31	19.47
	Threshold exceedance	5.8 %	9.8 %	21.0 %	24.1 %
	Min. events - max. events (total events)	7 - 14 (174)	1 - 13 (92)	4 - 16 (62)	11 - 28 (83)
Winter	Median	0.80	1.22	10.66	9.19
	Threshold exceedance	0.0 %	5.9 %	33.3 %	20.0 %
	Min. events - max. events (total events)	0 - 0 (50)	0 - 1 (17)	1 - 2 (6)	2 - 3 (10)
Spring	Median	3.65	3.81	6.82	19.48
	Threshold exceedance	10.4 %	16.0 %	16.7 %	33.3 %
	Min. events - max. events (total events)	4 - 7 (48)	0 - 5 (25)	0 - 3 (18)	4 - 8 (18)
Summer	Median	3.81	6.78	15.35	19.53
	Threshold exceedance	6.8 %	3.8 %	20.8 %	14.7 %
	Min. events - max. events (total events)	2 - 5 (44)	1 - 2 (26)	0 - 8 (24)	0 - 9 (34)
Autumn	Median	2.27	7.38	9.66	22.17
	Threshold exceedance	6.3 %	12.5 %	21.4 %	33.3 %
	Min. events - max. events (total events)	1 - 2 (32)	0 - 5 (24)	3 - 3 (14)	5 - 8 (21)

Further, the reviewer underlined how particle density may change as function of particle composition.

We are aware, that the particle density also depends on the particle composition. In the case of dust transport events the particles in the coarse size range are dominated by dust aerosol which has a density around 2.6 g cm^{-3} . This confirms the choice of the particle size dependent density which has values between 2.1 g cm^{-3} and 2.6 g cm^{-3} . During background conditions, the aerosol composition is different and is in the super micrometer range a mixture of mainly organics, ammonium sulphate and an unknown component (Putaud et al., 2004). Therefore, Putaud et al. (2004) suggested a density of 1.77 g cm^{-3} . This density is subject to a high uncertainty, as their results are based on 1 month of measurements in summer, and thus other seasons are not considered. Furthermore, it is technically challenging to measure the density of super micrometer particles. In the estimation of the uncertainty for the density we used 1.77 g cm^{-3} to estimate the lower uncertainty for background conditions. The coarse particle concentration during background conditions is typically very low, such that the uncertainty of the OPC measurements dominates over the uncertainty of the particle density. Given all this, we are aware that the PMcoarse concentration during background conditions has some asymmetric bias, but it is challenging to fully account for it. We included a sentence in the new Sec.2.7 of the revised manuscript.

On the other hand, this paper is an extension of Duchi et al (2016), but there is no comparison of the current results with those of the previous article, nor an explanation of the new results.

Furthermore, this citation is very important for the paper, and is not accessible via the indicated DOI. The authors are suggested to make an effort on it as well.

We thank the reviewer for pointing out that the DOI of Duchi et al. (2016) is not working. We contacted the editorial board of the journal without success and thus decided to provide the URL in the bibliography.

We further agree with the reviewer that a part dedicated to the comparison to Duchi et al. (2016) is missing. While both studies give a general overview of the fraction of dust transport days and its seasonal cycle, they differ in the subsequent analysis of these events. Next to the identification of dust events, Duchi et al. (2016) analyze the source origin of the dust and the relation to the coarse particle number concentration. The use of number concentration, however, limits the comparability with other studies. Hence, in our work we converted the size distribution into PMcoarse mass concentration, to better compare our data to previous studies based on in-situ measurements. Considering the comments of both reviewers, the updated version of our manuscript includes a detailed evaluation of uncertainties, which helps quantifying the reliability of the current approach, which was not detailed in Duchi. Overall, the present manuscript provides a more complete phenomenological context and uncertainty evaluation of dust events in Europe, representing an evolution and not only an extension of Duchi. To point this out we added a paragraph in L.192, which reads as:

Comparison to the study of Duchi et al. (2016)

Duchi et al. (2016) analyzed the dust transport at Mt. Cimone between 2002 and 2012. The dataset in this paper extends this analysis until 2023. Both studies observed an overall fraction of DTDs of 15.7 % or 15.8 %, indicating that the annual fraction of DTDs did not change significantly. Also, the seasonal cycle of DTDs was consistent in both studies, with a broad maximum in spring/summer, a second maximum in October/November, and a minimum in winter. When looking at the duration of DTEs, the highest fraction was always the 1 day duration events with 44 % for Duchi et al. (2016) and 42.2 % in this study. For Duchi et al. (2016) the second highest fraction with 28 % were the 2 day events and further they only report that 8 % of the DTEs lasted more than 5 days. In this study, the fraction of the 2 day events was reduced to 22.3 %. The further duration classification differed slightly, as we categorized differently the DTEs based on their duration. After the discussion of the occurrence of DTDs and the seasonal cycle, Duchi et al. (2016) focused their work on the changes in the coarse particle concentration during DTDs and the source origin from the various parts of the Saharan desert. In our work we discuss the interannual variability and the seasonal cycle of the PMcoarse concentration instead of the coarse particle concentration, so that our results can be more comparable to other studies. Furthermore, we give an estimate of the uncertainty related to this analysis.

Specific comments

- Please, consider to include information about the backward-trajectories source within Section 2.

We followed the suggestion of the reviewer and included a subsection in the methods, which is entitled ‘FLEXTTRA backward trajectories’, where we describe the meteorological input data, the model setup and the output. Furthermore, we explain in more detail the definition of the geographical box used to verify the overpass on the Saharan desert and how it differs from Duchi et al., 2016. The added section is as follows:

2.2 FLEXTTRA backward trajectories

3D-backward trajectories were retrieved from the FLEXTTRA model (Stohl et al., 1995), which performs the calculations based on the vertical wind. Meteorological data were provided by ECMWF with a $1.25^\circ \times 1.25^\circ$ grid resolution on 60 vertical levels, derived from a combination of observations with numerical models. In this study, a 7-days long backward trajectory was calculated every 6 h (00, 06, 12, 18 UTC). The initializing height was set to 2200 m a.s.l. and every 3 h the calculation provided several parameters, among which the location and the altitude of the air parcel. Stohl and Seibert (1998) indicate an accuracy in terms of travel distance around 20 %.

From the location of the air parcels, it can be assessed whether the trajectories traveled over the Saharan desert before reaching Monte Cimone. Therefore, we divided northern Africa into 4 boxes (Fig. 2 c) with the following boundaries:

- Box 1 (Western Sahara): 15°N to 35°N and -17°E to -7°E
- Box 2 (Central Sahara): 15°N to 37.5°N and -7°E to 15°E
- Box 3 (Eastern Sahara): 15°N to 33°N and 15°E to 34°E
- Box 4 (Sahel zone): 10°N to 15°N and -17°E to 34°E

This grid presents a modified version compared to the one applied in Duchi et al. (2016), where they used one large box ranging from 10°N to 35°N and -15°E to 30°E . With the new division we fully incorporate the northern part of central Africa and enlarge the included part of the eastern Sahara.

- Figure 1 – the legend about (a), (b) and (c) is not corresponding to the text in paper. Please, revise.

Thank you for noticing this mistake. We corrected the figure caption accordingly.

Figure 2. (a) Fraction of dust transport days (brown) and the number of non-dust transport days (grey). (b) Duration of dust transport events divided into 1 day (beige), 2 days (orange), 3 days (light brown) and 4 and more days (dark brown). (c) Grid box extension for the four boxes used to confirm dust transport days. The percentage values give the fraction of back-trajectories that passed over each box. Map made with Natural Earth (naturalearthdata.com).

- Lines 162-163: it is commented that there was no significant temporal trend (slope of 0.063) in the fraction of DTDs obtained from the trend analysis. But dashed line shows an increasing trend. Please, consider to delete the line, because this may lead to misinterpretation.

We agree with the reviewer that the trend shown in the figure can be misleading for the reader. We therefore removed the trend in Fig. 2a as suggested.

- Line 235: opposing -> opposite

We corrected opposing to opposite as suggested.

- Line 247: in the majority of the years was -> in most years was

We corrected the text as suggested.

- Line 249: longer of shorter -> longer or shorter

We corrected the typo

- Lines 262-263: dust mobilization in summer -> dust injection into the atmosphere during the summer.

We corrected the text as suggested.

- Lines 269-271: care must be taken because the results may have a high uncertainty. It is important to solve this issue.

We agree with the reviewer that the PMcoarse enhancement is subject of uncertainty. As already pointed out in the general comments we added more information on that throughout the manuscript. We also specifically addressed this part and refer to the made changes to our response to the general comments.

Literature

Putaud, J.-P., Van Dingenen, R., Dell'Acqua, A., Raes, F., Matta, E., Decesari, S., Facchini, M. C., and Fuzzi, S.: Size-segregated aerosol mass closure and chemical composition in Monte Cimone (I) during MINATROC, Atmospheric Chemistry and Physics, 4, 889–902, <https://doi.org/10.5194/acp-4-889-2004>, 2004.