Response letter

“Spatial variability of Saharan dust deposition revealed through a citizen science campaign” (essd-2023-16)

eferee #2 (minor revision)

The manuscript by Dumont et al. presents interesting and valuable data. It builds on a unique initiative realized at short notice after a transport event of desert dust to the Pyrenees and the Alps in February 2021. Starting with a very simple call to mountaineers a significant number of snow samples could be collected and evaluated. Still a number of points remain unclear and need to be improved. These points are listed below in the more detailed comments. Additionally, some comments about the ‘lessons learned’ for a follow-up citizen science campaign would be interesting to know to provide input for future citizen science campaigns.

We appreciate the positive feedback from the reviewer and the time spent on the evaluation of our manuscript. We are confident that we can address the reviewer suggestions in a revised manuscript as explained below. We thank the reviewer for this careful evaluation of our work and the relevant suggestions to help other researchers make good use of the data. As explained below, we can provide most of the missing information in a revised manuscript. The reviewer's comments are reported in black below, our replies are in blue and the changes in the manuscript are highlighted in bold blue. A detailed point by point response is provided below.

Specific comments

Page 2, Introduction: a short comment should be added why the measurements of radionuclides are performed

Several French news websites stated that the dust plume carried radionuclides from former french nuclear weapons tests in the Sahara. These measurements were made to verify this claim.

Page 2 line 44 now reads : “optical properties of the dust. **Several french news websites stated that the dust plume carried radionuclides from former french nuclear weapons tests in the Sahara. We thus also performed radionuclides analysis on the samples to verify this claim.** This study presents the citizen science campaign and the results of the sample analysis.”

Page 3, lines 62-63: Which distances were covered when the three closest samples were considered? And where were these closest samples situated – this could be important as differences were reported for the different facing slopes. Perhaps it is more suitable to give this information in section 3 (Results) – but it should be given at some point.

We selected the three samples closest to the MOCAGE sites and averaged their measured mass. The distance for the 3 samples ranges between 6 to 10 km for the Queyras location, 0.5 to 4 km for the Chartreuse location, and between 67 to 74 km for the Pic du Midi location.

MOCAGE simulations are performed over a grid size of 10 km (Josse et al., 2004). Therefore, the location chosen is representative of a 100 km2 area. For the Queyras and Chartreuse locations, the 3 samples chosen are located within this 10km grid size. For the Pic du Midi, the 3 samples are located further and results might be taken with more caution.
Otherwise, to ensure the representativity of the grid size, we ensured that samples covered different aspects (i.e. from 126, 258, and 274°N for the Queyras location, 9, 222, and 310°N for the Chartreuse location, and 5, 125, and 180°N for the Pic du Midi location; aspects computed based on a 250m DEM resolution)
Finally, the mass of the 3 samples chosen is 5.2, 4.7, and 6.6 g m^{-2} for the Queyras, 2.1 2.52, and 2.34 g m^{-2} for the Chartreuse and 11.6, 6.2 and 24.4 g m^{-2} for the Pic du Midi. For both Queyras and Chartreuse locations, the 3 samples are close to each other in terms of mass while for the Pic du Midi the values of the 3 samples are more diverse, showing here, again that the MOCAGE evaluation for this point should be taken with more caution.

These informations have been precised in the revised manuscript as follow:
(In section Meteorological conditions):
"**We further evaluated the accuracy of the total dust mass deposition from MOCAGE by comparison with the averaged mass measured of the three closest samples for each site. For the Chartreuse and Queyras locations the three samples are within the grid size resolution of MOCAGE (i.e. 10 km). For the Pyrenees, samples are located 60 to 70 km away and results might be taken with more caution. The three samples taken for each location covered different aspects. The three samples close to Pic du Midi also exhibit a larger spread in mass values that the samples for the two other locations.**"

Page 3, lines 65 to 67: How was the filtration performed, which type of filters was used and how big was the loaded area? Did you actually determine mass concentrations or mass loadings per unit area? How was the analytical uncertainty determined?

We determined the total mass of dust on the filter, which was then divided by the snow area sampled, to provide dust mass per snow unit area. The overall uncertainty results from the precision weighting scale uncertainty on one hand and from the uncertainty in the sampled snow area on the other hand. The precision of the weighting scale amounts to less than 5% uncertainty for all but three samples, thereby reported as the "analytical uncertainty". The uncertainty on the sampled area was not formally determined. We are aware that this is a weakness of our study but we do not know how to estimate the uncertainty given the variety of recipients, sample volumes, and expertise of the participants…

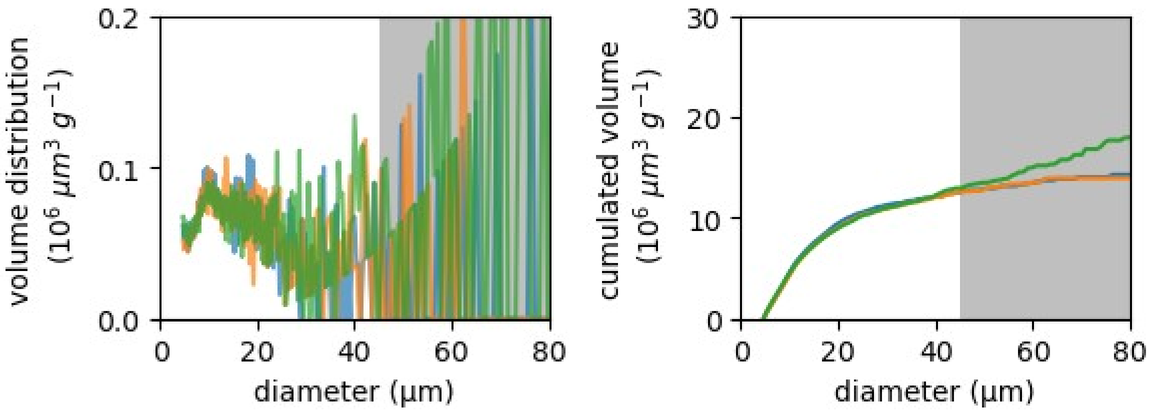
P3 lines 65-67 has been modified as follows and now reads:

"**First, the samples were homogenised, and 5~mL of each sample was kept for size distribution analysis. The rest of the sample was vacuum filtered on 47~mm diameter pre-weighted polycarbonate membranes (pore size 0.45~μm), resulting in a $\approx$12.5 cm^{2} loaded area. Those membranes were then dried in a dessicator, and weighted to provide the filtered dust mass. The analytical uncertainty of the measured dust mass is well within 5$\%$ (lower than 2$\%$ for 80$\%$ of samples). The dust surface concentration then results from dividing the measured mass by the snow sampled area, for which the uncertainty was not formally determined, so that 5$\%$ is a lower estimate of the overall uncertainty on that measurement.**"

Page 3, lines 68 ff: The method described in Delmonte et al (2004) covers the size range of 0,7 – 20 µm. This is quite different to the size range given in Figure 2. Furthermore, I do not understand the approach to differentiate between the two parts of the size distribution (i.e. the well measured part and the tail of the distribution being more uncertain). Why is the cut diameter set at the lowest diameter where none of the three measurements detected any particle. Variations seem to get really large when at least one of the three individual measurements does not detect any particle. How did this cut diameter vary when all samples are considered. Is it possible to draw some general conclusions (either here or in section 3.3.2)? Some information about the cutoff diameter is available in Figure S3, but it is never mentioned in the text.

We agree that the description of the Coulter method was misleading. Indeed, the size range analyzed with the Coulter counter solely depends on the size of the aperture tube used in the instrument. We selected a different aperture compared to Delmonte et al, as our size range of interest is shifted towards higher diameters. Other aspects of the protocol used are directly derived from Delmonte et al, thus the reference to it. We rewrote the method's description to make this clear.

Concerning the definition of the cutoff diameter: we actually also used the definition you suggest, which resulted in lower values of the cutoff, often excluding what seemed like an exceedingly large fraction of the distribution. We agree that on the selected exemple, there is a large variability on the size distribution before the cutoff. Yet this dispersion does not show on the cumulative distributions, which do spread away approximately at the cutoff we finally chose. In order to avoid confusion we will replace Figure 2 by the following one, which reflects more the previous discussion.



The cutoff diameter spans from 35 to 70 µm and generally varies with the mean volumic diameter (see Fig S4).

Page 3 lines 68 now read (please see also response to referee #1 on this matter):

"Dust size distributions were measured **with a Coulter counter (multisizer IIe) following protocols adapted from Delmonte et al. (2004)**. **The main adaptation concerns the measured size range, which was set to 4 – 120~μm by choosing a 200~μm measuring aperture. However, above $\tilde$40-60 μm, depending on the sample's concentration, the size distributions measured with the Coulter counter suffer from poor counting statistics, as shown in Figure ~\ref{cut_diameter}. We thus determined for each sample a so-called cutoff diameter, as the lowest diameter where no particle was detected in any of the three individual replicate measurements (See Fig.~\ref{cut_diameter} for an example). This cutoff diameter (see Fig. ~**

ref{small_and_big} for values distribution) separates a lower part of the distribution, well measured (typically better than 10\% uncertainty), and a tail, which is highly uncertain although it may represent a significant portion of the measured distribution (Fig. ~\ref{cut_diameter})."

The following sentences were added in section 3.3.2. after line 211, to comment further on the cutoff diameter and the distribution tail, and precise a sentence (line 207), which appeared misleading in this context:

"The distribution tails show **the regular occurrence of particles larger than the cutoff diameter (35 – 70~μm , Fig. ~\ref{small_and_big}, central right panel), whereas the mean diameter of the well-measured fraction of the distribution is much lower, around 19~μm for the volume distribution and 12~μm for the surface distribution. The cutoff diameter varies with the mean volumic diameter (Fig. ~\ ref{small_and_big}, right panel) : detecting enough big particles to have a good counting statistics implies a larger mean volumic diameter and a higher cutoff diameter. The cutoff diameter should therefore be seen as an estimate of a potential measurement bias. Adding the distribution tail into the calculation of the average diameters slightly influences the average volume diameter.**

The volume distribution can be converted into a mass estimation using a dust density of 2.5 g cm^{-3}, **which can be compared to the gravimetric measurement on the filters (Fig. ~\ref{small_and_big}, left panel). This comparison shows that the distribution tail, poorly measured with the Coulter counter, actually represents most of the dust mass.**"

Page 4, line 80: give a range of mass loadings on the filters

The range of mass loadings was added as follows:"The direct analysis of the filters, with relatively thin dust layers **(mass loads from 0.001 to 0.3 g)**, required the development of a specific calibration procedure **provided with the dataset (see Data availability)**."

Page 5, line 85: add a reference where the calibration function can be found

The sentence was modified to include the reference (see our response to the previous comment). This information was also added in the Data availability statement that now reads:

"Data presented in this study are available at https://doi.org/10.5281/zenodo.7464063 \ citep{dumont_2022_data}. **The dataset contains the mass and size data for each sample, as well as the exact coordinates for each sample. It also contains the elemental composition of the samples, and the calibration function of XRF. A data file is also provided for the optical properties (MEE and extinction) and for the radionuclides analysis.**"

Page 5, line 90: As far as I understand the samples collected within the citizen science campaign were not used to determine the optical properties. This should be mentioned more clearly.

Yes this is correct, this is now clearly indicated in new Tab. 1 shown below :

Analysis	Total analyzed	Regions	Source	Reference	Comments
Dust mass	113	87 locations Pyr., French Alps, Swizterland	Citizens (95) Res. Labs (19)	circles on Fig. 4 Sec. 2.3, Sec. 3.3.1	152 samples collected 138 of the first dust event 27 with problems
Size distribution	95	87 locations Pyr., French Alps, Swizterland	Citizens (79) Res. Labs (16)	circles on Fig. 4 Sec. 2.3, Sec. 3.3.2	taken among the 113 samples for mass
Elemental composition	70	70 locations Pyr., French Alps, Swizterland	Citizens (54) Res. Labs (16)	circles on Fig. 4 Sec. 2.4, Sec. 3.4	taken among the 113 samples for mass
Additional samples					
Radionuclides	3	Pyr., French Alps.	Res. Labs (2) Citizens (1)	Triangles on Fig. 4 Sec. 2.6, 3.6	need higher mass than the common samples
Optical Properties	2	Pyr., French Alps	Res. Labs (2)	Sec. 2.5, Sec. 3.5	need higher mass than the common samples

Table 1. Overview of all the dust samples analyzed in this study

Page 6, line 102: It is somewhat surprising to find a reference to Fig S5, as the references to Figures S1-S4 are only given later, in the Results section.

Thanks for noticing it, the supplementary figures have been renumbered.

Page 6, line 123: It seems that just a limited number of the filters of the citizen science campaign (just one?) could be evaluated for radionuclide analyses. This should be mentioned more clearly.

This is now explicitly written in **new Tab.1** (see response just above) that sums up all the analysis and all the samples. The reason why only a single sample cloud be used for radionuclide analysis is the limited mass of dust available in most of the samples. Only the one with the highest dust load (0.3 g) could provide robust results. This is now explained more explicitly as follows :

Page 6, line 120 **"The filters with the highest dust load (ca. 0.1-0.3 g)** were analysed using ultralow background Germanium HyperPure gamma spectrometry detectors installed in the underground facilities at University Paris-Saclay (Gif-sur-Yvette) and Modane (Underground Lab of Modane, France). To obtain sufficient counting statistics, filters were analysed for ca. $2 x 10^5$ s, approximately two days. The results obtained on the most heavily loaded filter (ALP-34-FE; 0.3 g of dust) and those obtained on a filter prepared with a similar quantity (0.3 g) of IAEA-444 soil-certified material were compared."

Page 9, lines 171-172: 152 samples and 85 participants – I guess that some participants brought a lot of samples and others just one. Could you give some details here? This could help to get a feeling about the variability of the results.

This section has been extended with Tab. 1 to clarify this point and several modifications have been done in the text (please see also response to general comments 1-5 from reviewer 1 for the list of modifications). The 114 samples finally analyzed (138 - 27 samples that had leaks, see page 9 line 189) correspond to 70 different locations. Over these 113 samples, 20 participants collected one sample, 24 participants two samples, and the rest collected 3 to 6 samples.

Page 9, lines 171-172 was modified as follows:

“Among these 152 samples, 138 were taken within days after the first event in February, and 16 (2 in the French Alps and 14 in the Swiss Alps) were collected in March, thus combining the effect of two dust events. The 152 samples were collected by more than 85 individual participants. In the remainder of the text, the analysis was restricted to the 136 samples that contained only dust from the first event. **Table \ref{tab:recap} indicates the number of samples used for each type of analysis. Among the 136 samples, only 113 could be analysed due to problems on the remaining 25 (labelling problems, mass outliers, leaks and information missing). For these 113 samples, 20 participants collected one sample, 24 participants two samples, and the rest of the participants collected 3 to 6 samples.”**

Page 9, lines 195-196: This remains unclear. I guess this is the comparison done for the ‘three closest sites’ (see page 3, line 62). The actual number of the deposited dust mass per unit area (in-situ measurements) should be given and compared to the modelled data. In Fig. 5 (as well as Fig. S1 and S2) the cumulative mass depositions are reported as ‘computed as MOCAGE outputs and corrected according to the observations’. Does this correction already reflect the underestimation reported here?

Yes, the comparison was done by comparing the MOCAGE depositions flux at the end of the event with the averaged mass deposition from the 3 closest samples of each location (see also response to your comment number 2). We added the mass deposition fluxes from MOCAGE and the measurements as follow:

“These measurements allow us to estimate that the MOCAGE simulation underestimates the deposition fluxes. **The modeled deposition fluxes are 0.31, 0.11 and 0. 52 g m^{-2}, while the mass measured from the three closest samples are 14.2, 5.6 and 2.1 g m^{-2}, for the Pic du Midi site, Queyras site and Chartreuse site respectively. This means that MOCAGE simulation underestimates the deposition fluxes** by a factor equal to 45.7 for the Pic du Midi site, 10.8 for the Queyras site and 19.1 for the Chartreuse site (Sect.~\ref{par:res:met}).”

Fig 5, S2 and S3 show the mass flux deposition from MOCAGE, corrected according to the observations, meaning that the factors computed between the MOCAGE flux deposition and the measured mass sample indicated above have been applied to the MOCAGE flux in this figure. Therefore the underestimation is not shown in this figure as data have been corrected. As it was unclear, this information has been added in the caption of the figures 5, S2, and S3 as follow:

“(A) Temporal evolution of the hourly dry (yellow) and wet (red) dust loads during the event at Pic du Midi (2100 m a.s.l.). The total cumulative mass deposition is shown by the black line. Depositions were computed based on MOCAGE outputs and corrected according to the observations **(i.e. the measured mass of the three closest samples of the site, see Sect.~\ref{subsubsec:massres})**. The dry vs. wet deposition was determined based on SAFRAN precipitation data (see section \ref{atmo}). Temporal evolution of the hourly wind speed (B) and direction (C) and the hourly precipitation (D) from SAFRAN reanalysis data. The yellow (red) shaded area represents the dry (wet) deposition of the event, according to SAFRAN precipitation.”

Page 12, line 206: Why is the number of samples analysed with the Coulter counter smaller than the number of samples filtrated?

Thanks for pointing this out. The missing samples result from labels mishandling, ambiguous labeling or other lab mishaps. Line 206 (now 230) has been corrected correspondingly and now reads :

“ A total of 95 samples were analysed with the Coulter counter, providing 95 size distribution values over the Alps and Pyrenees (Fig. \ref{small_and_big}). **The missing samples result from labels mishandling, ambiguous labeling or other lab mishaps.**”

Page 12, line 211: At least a short discussion about this comparison should be added.

Yes, this was indeed missing. This was however discussed page 19 line 291 - 295. We added a short comment on the figure page 12, line 211:

“The volume distribution can be converted into a mass estimation using a dust density of 2.5 g cm^{-3}, which can be compared to the gravimetric measurement on the filters (Fig. \ref{coulter_vs_mass}). **Fig. S5 shows that the dust mass measured with the Coulter is lower than the mass weighed on the filters. This underestimation is likely related to large particles (diameter larger than 50 ~μm) as shown on the second panel in Fig. S5.** ”

Page 12, lines 212 to 216: This section needs clarification. What is the 'mean dust diameter' mentioned here? Furthermore, I cannot relate the numbers given in the text to Fig 8A. Fig 8 needs some clarification as well. What means ‘Size-Surf. Distrib.’?

Thanks for noticing this mismatch.The numbers in the text refers to the mean dust volumic diameter while the figure presents the mean dust surface diameter (mean diameter when the average is weighed by the surface of each particle). This has been clarified as follows :

In the legend of Figure 8 : “**Distribution of the surface average diameter (proxy of the particle size)** of the dust deposition against the massif (A), the distance to the source (B) and the aspect computed with a 30 m resolution DEM (C) and a 500 m resolution DEM (D).“

In the text that now reads: “The dust size distribution depends on the location, with larger sizes generally observed closer to the source (Fig. \ref{fig:size_box}B). In the Pyrenees samples, the size spans a range of 15.4-27~μm (**with a volume average diameter of 21~μm**) and varies between 9.8 and 38~μm (**volume average diameter = 21~μm**) and 8.2 and 19.6~μm (**volume average diameter = 15~μm**) for the French and Swiss Alps, respectively. **Figure \ref{fig:size_box}A shows the same gradient for the surface average diameter.** “

Page 12, line 213: the reference to section 3.3.2 is strange – this is section 3.3.2

Thanks for noticing the typo, the reference was changed to 2.3.

Page 19, line 298: Is it possible to give reasons for this decrease in the mass fraction of iron

The possible reasons for the decrease in the mass fraction of iron are briefly discussed p20 line 330-332, likely a preferential loss of mineral along transport.

Page 36, Figure S3 legend: obviously the surface distribution is given on the right and the volume distribution on the left

Thanks for noticing it, the legend of the figures was modified accordingly.