

Author's response

We thank one Anonymous Referee and Matthew Johnson for taking time to review our manuscript, their positive feedback and their valuable input that allowed us to improve our manuscript. We further thank K. M. Meiners for taking time to perform editor's duties and the motivating words within the Editor decision.

We structured our response in the following way: (1) Referee Comment #1, (2) Referee Comment #2, (3) Additional changes, (4) References. For the Referee Comments, we kept the division into a main and technical part. While the Referee Comments are given in bold letters, our answers are given in common formatting and changes in the text are given within quotation marks in italics. Given page (p.) and line (l.) numbers refer to the marked-up manuscript version highlighting the changes made during the revision.

We want to clarify the change of $ATN_{700-450}$ to $\Delta ATN_{700-450}$ in the revised version of the manuscript and the Author's Response. This was proposed by Anonymous Referee #1. We agreed to and conducted this change, as $ATN_{700-450}$ is the difference between the attenuation at 700 and 450°C (without requiring the incident intensity of the transmitted laser signal).

Referee Comment #1

(1) Main comments

Section 2.2.1 How do you think the non-homogeneous distribution of the LAPs on the filter affects the quantification of OC and EC? In particular, are you able to estimate the related uncertainty?

A non-homogeneous distribution of LAPs on the filter will definitely affect the quantification of OC and EC if only parts of the filter are analysed. To prevent this influence, we concentrated the water-insoluble particles of our filters on circular spots with a diameter of 16 mm, cut the loaded area using a punch of 17 mm and analysed both halves of the filter. Thus, all particles retained by the filter were analysed between 2017 and 2024.

For the year 2016, punches with a diameter of 15 mm were made. This is less than the loaded area and inhomogeneities could affect the result, as discussed in the manuscript. Based on a separate dataset of snow samples collected spatially and analysed temporally close to the samples described within the current manuscript, an average underestimation of TC of 18 % with a standard deviation of 9 % was found for samples with visible inhomogeneities. Based on single samples ($n=9$) the underestimation spanned between 7 and 32 %. This uncertainty already includes the uncertainty of the analysis.

We agree that a quantification of this effect improves the manuscript and consequently performed the following changes:

We added the following information in Section 2.2.1:

"An underestimation of water-insoluble total carbon between 7 and 32 % was found for comparable samples ($n=9$) with an average of 18 % and a standard deviation of 9 %. Thus, we report results for 2016 with an increased uncertainty of 50 %." (p. 4, l. 96-98)

We consequently increased the error bar of WinstTC in Figure 3 to 50 %.

For samples of 2016 containing dust, no inhomogeneities were visible. Thus, the error bar for MD in 2016 in Figure 6 was not increased.

Line 128-136. It would benefit readers if the authors could provide more details about the procedure used to detect the dust layer based on TOA. For instance, does '4' refer to the change in transmittance during the cooling phase from 700 to 450 °C? If so, it would be clearer to denote it as delta ATN (also in Figure 1). The statement “Since the temperature range previously mentioned was not always reached for our set of samples” suggests that a different thermal procedure was used in prior analyses. Additionally, please clarify why the color change observed during the cooling phase indicates the presence of mineral dust.

We agree that the text was not clear. Consequently, we adapted the paragraph as given below.

Still, we want to answer the referee’s questions first: Yes, “4” refers to the change in transmittance during the cooling phase and we agree to denote it as $\Delta ATN_{700-450}$, as it is the difference between the attenuation at 700 and 450°C (without requiring the incident intensity of the transmitted laser signal). All samples were analysed with the same thermal procedure, which defines the temperature setting during the heating period. Calibration takes place after the actual analysis, when heating is stopped and a fan helps with cooling. So the actual analysis, which can be done according to different thermal procedures, is over and the oven just needs to cool down, which is supported by a small fan. This time period is utilized to do the calibration. Data acquisition is continued just long enough to catch the calibration peak. Maintenance at the instrument led to small changes in the position of the insulation material and the efficiency of cooling. Consequently, the oven no longer cooled down to 400 °C before data acquisition stopped. To avoid similar problems in the future, we reduced the temperature interval and evaluated the temperature range between 700 °C and 450 °C only. The color change observed during cooling is most likely induced by hematite, a phenomenon described by Yamanoi et al. (2009) for the analysis of α -Fe₂O₃ powders. Still, we did not manage to link the effect directly to the hematite content of the samples, but to the overall iron content. More details are given in Kau et al. (2022).

“The identification of mineral dust layers allows to retrace events with mineral dust deposition during the accumulation period and is the starting point for mineral dust concentration and deposition assessment. We identified dust-laden samples based on the temperature dependent change in optical properties of residual material seen in thermal-optical analysis. If mineral dust is present this refractory material remains on the filter and shows a reddish colour due to the presence of iron oxides. As described by Kau et al. (2022), the transmitted laser signal ($\lambda=660$ nm) was evaluated in the calibration phase. At this point the actual analysis of carbonaceous compounds is already finished. Oven and filter sample are just cooling down, but data is still logged to record the calibration peak. Kau et al. (2022) evaluated a temperature range between 700 and 400°C. Due to small adjustments at the fan which supports cooling, the lower temperature, i.e., 400°C, given previously was not always reached at the end of the calibration phase for our set of samples. Thus, we now evaluated the change in transmittance observed between 700 and 450°C using the transmitted laser signal I_{700} and I_{450} , respectively. Converted into a dimensionless temperature dependent attenuation ($\Delta ATN_{700-450}$, see Eq. (1)), samples that equalled or exceeded a value of 4.0 were classified as containing mineral dust. For clarification, $\Delta ATN_{700-450}$ corresponds to $ATN_{700-400}$ defined in Kau et al. (2022), however, using the intensity of the transmitted laser signal at 450°C. To account for lower filter loadings, we added high linearity of the relationship between the temperature and the transmitted laser signal (coefficient of determination, R^2 , above 0.9) as a criterion. We refer to this method to identify dust-laden filters as TOA approach.”

$$\Delta ATN_{700-450} = 100 * \ln \left(\frac{I_{450}}{I_{700}} \right) \quad (1)$$

(p. 5-6, l. 143-158)

Line 172. The authors should specify whether the “Laser/Temp Correction Method in the Calc453” refers to the default correction, which does not account for changes in transmittance with temperature due to the presence of dust. Is this the same as the “default method” indicated in Figure 2? Moreover, it would be helpful if the authors could elaborate on the correction based on the linear fit in the methods section, as this information is crucial for ensuring the replicability of the approach. Yes, they are the same. In the revised version of the manuscript, we consistently refer to the two Laser/Temp Correction Methods as either “default option” or “linear option” to make the text more comprehensible. Changes were made in Section 3.2.1 (p. 8, l. 203; p. 9, l. 219, 222, and 228) as well as the label of Figure 2.

As indicated in the text, both options for the Laser/Temp Correction Method are made available by the manufacturer in the Calc software. To ensure the replicability of the approach, the software version (Calc453) and the exact option names (“Simple Laser/Temp Corr. – Divide by 2” and “Linear Fit”, respectively) are given in the text. Unfortunately, no description of the Laser/Temp Correction Methods is provided by the manufacturer. Own postprocessing of the raw data indicates that the “linear option” corrects for the linear change of the transmittance signal in the calibration phase.

Line 189. Please explain why this method can only be applied to samples where the optical signal is predominantly influenced by dust, and how this can be inferred. While you can estimate dust presence through calcium measurements or pH data, this does not provide information on the content of elemental carbon (EC) and the relative contributions of these two components to the optical properties of the sample. Additionally, if dust is negligible, I assume that applying the linear fit correction would not result in any significant changes to the results, as illustrated in Figure 2. Is this assumption correct?

The assumption of Anonymous Referee #1 regarding no significant changes using the linear option for the Laser/Temp Correction Method for samples with negligible amounts of mineral dust is comprehensible. Still, we cannot generally suggest using the linear option for all sample types and sites, where various particle types may occur. For samples containing mineral dust, the dust will have a dominant influence on the laser signal. The use of the linear option reduces this bias and is therefore needed. For all other cases it is preferable to stick to the default option, which is the commonly used method. This also increases comparability between different data sets of EC obtained by TOA. Thus, we stick to our previous recommendation and did not make changes in the text.

Line 260-265: The differing behavior of the snowpack with mineral dust (MD) and the lowest snowpack layers shown in Figure 4 suggests that MD from various sources exhibits distinct optical responses. Is this due to the fact that optical properties are not solely determined by iron content? Could the authors elaborate on this point further? Would the authors recommend validating the linear fit method with field measurements to ensure its applicability in different environments or regions?

Yes, optical properties are not exclusively connected to the Fe content. We want to point out that Fe can be present in various compounds, which feature different properties. Mineral dust includes Fe containing minerals exhibiting a temperature dependent change in optical properties, e.g., hematite. In contrast, particulate matter samples collected in railway tunnels do not show this behaviour despite their high Fe loadings. Here, Fe is present in other forms, e.g., Fe metal. This was discussed in Kau et al. (2022).

Attenuation depends on various sample parameters including particle concentration, size, shape and refractive index (e.g., Baker and Lavelle, 1984 and references therein). As discussed for the lowest layers shown in Figure 4, we attribute the differing relationship of the lowest layers to several factors, which include mixture with residual material of the last accumulation period, mixture with local dust and

possible weathering of last accumulation period's dust. These would affect the exhibited attenuation of the dust.

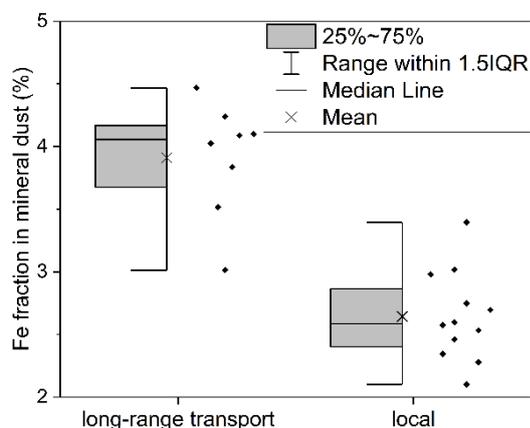
An influence of different source regions may lead to differences in composition and particle size distribution of mineral dust, possibly affecting the fit between the attenuation and the Fe loading. This can be the case for different sampling sites as well, as aerosol properties can change depending on the duration or distance of transport. Before applying the fit calculated for our site for different regions, we advise validation. To emphasize this, we added the following statement in Section 3.3.1:

“Note that the relationship between $\Delta ATN_{700-450}$ and the Fe loading may differ for samples collected at other sites. Thus, the fit obtained here should be validated prior to application.” (p. 13-14, l. 301-303)

Fig 5. Presenting the statistical distribution of eight measurements using a box-whisker plot can be misleading, as this plot summarizes a dataset consisting of only eight data points with just six numbers. I would suggest simply reporting the median and providing the range in brackets within the text.

We agree that the use of boxplots in this case can be misleading. We decided to use the boxplots in the submitted version of the manuscript, as both the variation of the data points in the two groups and measures of central tendency, i.e., median and average values, can be directly compared.

To consider the comment of Anonymous Referee #1, we included the data points in the boxplots. Thus, the number and distribution of samples in each group are easily visible and possible misinterpretation of the graph is avoided. The positive aspects, i.e., visual comparison of the groups, is still kept. The new Figure 5 is shown here:



Page 15 discusses previous studies that recommended quantitatively measuring dust using polycarbonate filters (Kuchiki et al., 2015). Why did the authors choose not to pursue this method? Did they plan to use the same filter for both gravimetric measurements and thermal optical analysis? Furthermore, does the comparison between thermal optical analysis (TOA) and gravimetric dust quantification suggest that the TOA approach should be applied to all reddish filters, and should the defined threshold for triggering measurements be modified?

Yes, due to the limited amount of sample and low analyte concentrations one filter needs to be used for the gravimetric measurements and thermal-optical analysis. Even if higher sample masses were available, preliminary tests showed that melting the sample, homogenizing it and splitting it to several filters does not give satisfactory reproducibility. Taking another depth profile for gravimetry would introduce the uncertainty of spatial variability, as discussed in Section 3.1 for the TOA- and IC-profile. Polycarbonate filters are not suitable for thermal-optical analysis, thus, gravimetry had to be conducted with quartz fibre filters. This is now clarified in the manuscript in Section 3.3.3:

“Gravimetric data of insoluble particles are available for 2017-2022 and 2024 based on the quartz fibre filters subsequently used for TOA.” (p. 17, l. 384-385)

The second question addresses the sensitivity of the TOA approach, which is capable of identifying mineral dust loaded samples if $\Delta ATN_{700-450}$ equals or exceeds 4.0. Our data do not support a reduction of the threshold, as we observed poor correlation between the transmittance and the temperature ($R^2 < 0.9$, usually around 0.98 for samples with mineral dust) resulting in random $\Delta ATN_{700-450}$ values from noise. Thus, we cannot quantify small dust loadings. The TOA instrumentation is not optimized for the assessment of mineral dust, but an interference is utilized to assess an additional parameter. In our study covering nine years, we found coloured filters with mineral dust loadings low enough not to trigger the TOA approach in two years only. In 2018, the division of a mineral dust layer during sampling led to two samples with lower mineral dust concentrations each ($\Delta ATN_{700-450} < 4.0$). While no interference on the OC and EC results is expected, they would be overlooked in the mineral dust approximation if a critical check of the colour of the filters after TOA was dismissed. The same is true for the filters of 2021, where four separate layers showed low mineral dust loadings. Each of these filters would contribute only little to the mineral dust approximation, but multiple filters with low loadings add up. As we recommended checking the filters' colour after TOA for quality assurance in the submitted version of the manuscript (line 340-341) already, we did not make changes.

In the conclusions section, while summarizing the study's results, the authors should also emphasize the caveats and limitations of the described method, as well as the implications of their findings, as outlined in the author guidelines (https://www.atmospheric-chemistry-and-physics.net/policies/guidelines_for_authors.html).

We appreciate the thorough review of the manuscript regarding the author guidelines. The manuscript was submitted to The Cryosphere and we rechecked the information given for this journal. In contrast to Atmospheric Chemistry and Physics, we could not find requirements for the conclusions section. As we still find our conclusions section suitable, we did not make changes to the text.

(2) Technical corrections

Line 44. Please add references to support this sentence:” and is commonly applied to filters loaded with insoluble particles from snow samples.”

We added references, which were already cited at other positions in the submitted manuscript already. Thus, no changes were necessary in the References section. The text now reads:

“The latter is the reference method for the quantification of elemental carbon in ambient air samples (DIN e.V., 2017) and is commonly applied to filters loaded with insoluble particles from snow samples (e.g., Zdanowicz et al., 2021; Tuzet et al., 2019; Forsström et al., 2013).” (p. 2, l. 45-48)

Line 46. “A comprehensive discussion of the impact of this bias on elemental carbon results in seasonal snow covers is still lacking.” This sentence is correct when referring specifically to thermal optical method. I suggest revising this sentence to clarify it refers to one specific measurement technique.

The text now reads:

“The co-occurrence of mineral dust leads to an interference in the quantification of elemental carbon in thermal-optical analysis and adapted evaluation of measurement data was proposed without quantifying the impact on elemental carbon concentrations (Wang et al., 2012; Gul et al., 2018).” (p. 2, l. 48-50)

Pag9. I suggest reporting the concertation of OC and EC from previous studies in a table. This would make it easier for the reader to compare the previous results with the ones reported in this work.

We agree that a table is a preferred type of presentation for data giving an overview. However, the data referred to within our work comprises concentration and deposition data presented in different ways (ranges, averages, medians) by authors for different sample types at various sites and points in time. As the resulting table would not be clear, we did not include the data in a table.

Line 296-297: please define uncertainty units

We are thankful for the comment. While the first parameter describing the share of Fe in mineral dust is dimensionless, the unit (%) for the second parameter was missing.

The share of Fe in mineral dust is based on the mass fraction of Fe in mineral dust (median: 4 %). For the calculation of uncertainty, its reciprocal value ($1/0.04 = 25$) and respective standard deviation is used, as the factor of 25 is used to estimate the mineral dust loading from the Fe loading (line 282-283). To consider the spread around the fit to calculate Fe from $\Delta ATN_{700-450}$, we calculated the difference between the calculated and measured Fe loadings and related it to the measured Fe loadings (line 254-255). The average and standard deviation in % were used for error propagation.

We added the unit of the latter parameter in Section 3.3.3 (p. 15, l. 337). To increase comprehensibility, we added the average of this parameter (11 %) in Section 3.3.1 (p. 13, l. 290; previously only the span and the median were given).

Line 299. Please specify that the indicated size range refers to dust particles in snow (that might have been subject to deposition post-processing). In fact atmospheric dust particles show a smaller diameter range (SCHWIKOWSKI et al., 1998).

We included the comment. The text now reads:

“We assume the error in retention of the quartz fibre filter to be negligible, as the mode of deposited mineral dust particle sizes in snow was between 4.5 and 13 μm (determined with Mastersizer 2000, Malvern Panalytical), which is in good agreement with dust-laden snow samples collected in the Italian Alps showing modes of 7.9 and 8.5 μm (Di Mauro et al., 2019).” (p. 15, l. 339-341)

Referee Comment #2

(1) Scientific

The paper presents an impressive nine-year continuous dataset and introduces a TOA-based method for estimating mineral dust deposition using Fe as a proxy. These are significant contributions. However, the introduction never fully clarifies what gap in the literature this study directly fills. The authors mention missing comprehensive discussions (e.g., mineral-dust bias in EC quantification), but don't articulate: 1) Why earlier studies (e.g., Cerqueira 2010; Tuzet 2020) were insufficient. 2) How this work advances monitoring practice or radiative forcing estimates. I recommend adding a short “This study addresses three gaps...” paragraph at the end of the Introduction to anchor the novelty.

We thank Matthew Johnson for the positive evaluation of our work and agree that an overview of novelty is beneficial for the manuscript.

Firstly, we definitely do not describe earlier studies mentioned in the comment, i.e., Cerqueira et al. (2010) and Tuzet et al., (2020), as insufficient. Apparently, they did not encounter problems with the quantification of organic and elemental carbon via thermal-optical analysis due to lower mineral dust loadings of their samples. Problems with mineral dust were found by Wang et al. (2012) and Gul et al. (2018) and they propose an adapted evaluation method for thermal-optical analysis. To improve the connection between the studies by Wang et al. (2012) and Gul et al. (2018) and our work, we adapted the text, which now reads:

“The co-occurrence of mineral dust leads to an interference in the quantification of elemental carbon in thermal-optical analysis and adapted evaluation of measurement data was proposed without quantifying the impact on elemental carbon concentrations (Wang et al., 2012; Gul et al., 2018).” (p. 2, l. 48-50)

Regarding the second point, we adapted the last paragraph of the Introduction section to highlight the novelty of our study. It now reads:

“We present a multi-year time series of snowpack sampling and analysis at a glacier field close to Sonnblick Observatory in the Austrian Alps starting in 2016. Using thermal-optical analysis as the central analytical technique, we report concentrations of elemental carbon and water-insoluble organic carbon. Besides the presentation of this data set, the novelty of the paper is its methodological focus. Applying different laser correction methods, the bias in elemental carbon measurements introduced by mineral dust can be quantified and a more reliable method for general monitoring practice of elemental carbon is introduced. A particular advantage is that the laser correction method can also be applied to existing data sets via post-processing. The interference itself, a temperature dependent change in transmittance of the laser applied in thermal-optical analysis, is used to identify samples containing mineral dust and to approximate the mineral dust load in the snowpack. Thus, thermal-optical analysis can be used for both the quantification of elemental carbon and the approximation of mineral dust concentrations.” (p. 2, l. 58-67)

Section 3.3 introduces an Fe-based approximation for mineral dust loading from TOA data. While elegant and practical, several limitations appear under-discussed. First, the limited calibration dataset. Only 14 samples were used to derive the ATN–Fe fit (Eq. 1) and the valid range is stated as $ATN = 4–13$. Yet the paper applies the relationship throughout nine years of snowpacks, without showing how often values fall outside this range. Please provide a histogram of $ATN_{700–450}$ values for all mineral dust layers to show how representative the calibration domain is.

We agree that the occurrence of ATN -values within and outside of the fitted range should be given for mineral dust samples of all years. Thus, we plotted the data in a histogram as suggested, using a bin width of 1 for the $\Delta ATN_{700-450}$ values. The samples used for the fit are hatched. Three samples exceeded the covered range ($\Delta ATN_{700-450} > 13$). For one of these samples (collected in 2024, in the highest $\Delta ATN_{700-450}$ bin), the Fe loading from ICP-OES analysis was available and was compared to the Fe loading based on $\Delta ATN_{700-450}$ and the fit. The Fe loading and consequently the mineral dust loading would be underestimated by 30 % for this sample based on $\Delta ATN_{700-450}$, as linearity is no longer given. For this sample, the mineral dust approximation used in the time series presented in the manuscript was based on the Fe loading from ICP-OES. For the other two samples exceeding an $\Delta ATN_{700-450}$ of 13 (both from 2019), no ICP-OES measurements were done. Lacking an alternative, we based the mineral dust approximation on $\Delta ATN_{700-450}$, knowing that it will result in an underestimation. This likely contributes to the poor agreement of the mineral dust deposition in 2019 with gravimetry. This was previously described insufficiently in the manuscript.

We added the histogram in the new Appendix C as Figure C1 to make the information easily accessible while not disrupting the reading flow. We added the following text:

“Appendix C: Further information on the fit between the Fe loading and $\Delta ATN_{700-450}$

As described in Sect. 3.3.1 and 3.3.2, the fit to approximate the Fe loading based on $\Delta ATN_{700-450}$ was calculated using 14 samples from the GOK site and a nearby site, Kleinfleißkees. The fit covered a range of 4.0 to 13 and was applied to all samples from the GOK site between 2017 and 2024. Most samples were in the range of the fit, as seen in Fig. C1Figure . Three samples are outside the linear range ($\Delta ATN_{700-450} > 13$). For one of these samples from 2024 ($\Delta ATN_{700-450}=34$), the Fe loading obtained via ICP-

OES was available and was compared to the Fe loading based on its $\Delta ATN_{700-450}$ value and the fit. Using the latter would underestimate the Fe loading by 30 %. For this sample, the approximation of mineral dust was based on the Fe loading obtained via ICP-OES. This leads to a decreased uncertainty of 23 % for the mineral dust approximation of 2024. For the other two samples exceeding an $\Delta ATN_{700-450}$ of 13 (both from 2019, $\Delta ATN_{700-450}$ of 21 and 35), no Fe loading from ICP-OES was available. Lacking an alternative, we based the mineral dust approximation for these samples on $\Delta ATN_{700-450}$ and consequently increased the error bar for 2019 in Fig. 6 to 87 % to consider the underestimation of these samples.”

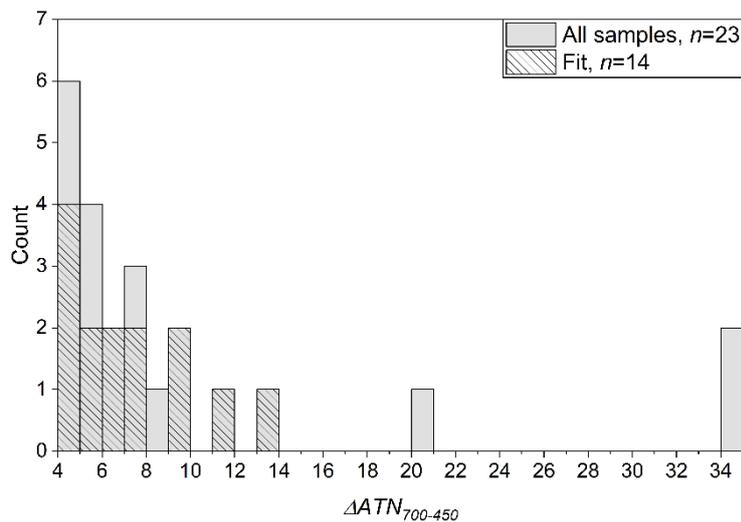


Figure C1: Overview of samples the fit to estimate the Fe loading from $\Delta ATN_{700-450}$ was based on and applied to.

(p. 20, l. 466-478)

We adapted the error bars for mineral dust in 2019 in Figure 6a and b, considering the underestimation by an estimated uncertainty of 30 % for the two samples, obtaining a resulting uncertainty of 87 % for 2019. We reduced the uncertainty for the year 2024 to 23 %, as mineral dust was approximated from ICP-OES for one out of three samples.

Second, the assumption of constant Fe fraction. The method assumes 4% Fe by mass in all long-range dust, but literature and the authors' own comparison (Figure 5) show Fe can vary from ~2% to >11% depending on source and transport history. Please discuss how source-region variability (e.g., Saharan vs. Middle Eastern dust) could bias the long-term trend analysis.

Scheuvens et al. (2013) did an extensive compilation of mass fraction data found in literature and discussed various elemental ratios for source apportionment. Compared to other elements found in mineral dust, they found Fe contents, Fe/Al ratios and Fe enrichment factors to be rather constant and homogeneously distributed in northern African dusts and sediments. For an “average northern African dust”, they find Fe contents comparable to the composition of the upper continental crust. Based on the substantial dataset, significant trends in the distribution were found only for Ca and Mg in northern African dusts.

Comparing dusts originating from a bigger regional scale, Kandler and Scheuvens (2019) report that the local variation in composition of dust can exceed the differences between Asian and African dust. The data referred to in the comment spanning between 2 and 11 % was collected during a citizen science project by Dumont et al. (2023). The study covers a big input area with samples collected from the Pyrenees to the Swiss Alps and the authors describe a gradient in the Fe mass fraction along the dust plume.

In our work, dusts from long-range transport analysed were collected between 2020 and 2024, covering several transport events. Thus, a variety of dust arriving at the site is covered. The observed variation of the Fe mass fraction is much smaller than the range found in literature, which we attribute to the fact that sampling was always conducted at the same site. As described in line 291, the variability of the mass fraction of Fe found for the samples collected between 2020 and 2024 at our site is reflected in the given uncertainty (line 295-297 in the submitted manuscript).

Third, the uncertainty is stated as 65% for mineral dust estimates (Section 3.3.2). This is substantial. A sensitivity test showing whether interannual variability is still statistically meaningful under such large uncertainty would strengthen confidence in the conclusions.

We agree that the estimation of mineral dust has a substantial uncertainty, which we do not want to hide. The comparison to other studies giving the spatial variability in ion concentrations puts this substantial uncertainty into context.

Here, we also want to refer to a study by Putaud et al. (2010). They report five approaches for mineral dust calculations based on various tracers used at different sampling sites in the supplementary information. They applied the different calculations to data of three sites and conclude that the uncertainty of mineral dust concentrations can range up to 150 %, while assuming it is much lower at several sites.

Regarding the interannual variability, comparing pairs of values for mineral dust input will not give significant differences. This must be considered in future trend analysis, where data from a longer period is necessary. Thus, no changes in the text were made.

Figure 1 provides a qualitative visual comparison of the TOA vs IC method comparison but the text states “very good agreement” without any numerical metric. Please include agreement statistics e.g. percentage of overlapping layer identifications, Cohen’s kappa, number of false positives/negatives per method. This would make the comparison more rigorous.

We appreciate the input regarding the quantification of agreement with a numerical metric. We calculated Cohen’s kappa as suggested. Using the depth resolution of the TOA-profile for the evaluation by the raters (IC approach, TOA approach), we obtained a Kappa Statistic of 0.48. Considering that the two neighbouring profiles may show slight displacement and the suspicious coloured filters, a Kappa Statistic of 0.65 was obtained. According to the classification given in Landis and Koch (1977), considering the limitations of the comparison increases the agreement from moderate to substantial. We added the results in Section 3.1 and the corresponding text reads:

“The agreement of the two approaches was quantified calculating Cohen’s kappa based on the depth resolution of the TOA-profile and evaluating the result based on the classification given by Landis and Koch (1977). While a strict comparison of the layers using the criteria ($\Delta ATN_{700-450} \geq 4.0$ for TOA approach and $pH \geq 5.6$ and Ca^{2+} concentration $\geq 8.5 \mu eq L^{-1}$) showed only moderate agreement, the consideration of the aforementioned limitations (two neighbouring profiles may show slight displacement, layers with coloured filters) increases the agreement from moderate to substantial.” (p. 7, l. 188-193)

We added the publication of Landis and Koch (1977) to the References section.

Light-absorbing impurities (LASI) strongly affect albedo and melt timing. The Introduction opens with this motivation, but the Discussion does not return to these impacts -- the paper remains methodologically focused. Please add a paragraph estimating or contextualizing the radiative significance of the observed impurity levels (e.g., based on Di Mauro 2019).

The mentioned effects of LASI are a strong motivation to collect and present concentration and deposition data and thus these basics were included in the Introduction section. However, modelling

these effects is not the focus of our current work. While we cannot offer a direct estimation, we expect a bigger influence on the melt timing for years where mineral dust layers occur closer to the surface. There, it can accumulate faster during the melt season, while dust in the lowest layers is covered by snow for the main part of the melt season. We added the following text in Section 3.1:

“In years where mineral dust layers occur close to the surface, e.g., 2018, we expect an increased effect on the duration of the snow cover due to direct and indirect forcing. In contrast, if the mineral dust layers occur close to the bottom layers and are covered by snow for most of the melting season, e.g., in 2017, we expect smaller effects. No further quantification of these effects is conducted within this work.” (p. 6, l. 184-187)

The manuscript discusses the issues arising from incomplete snowpack sampling in 2019 (3.2.2 and 3.3.2). However, results for that year are still plotted without any visual cue of lower confidence and results from 2019 are included in averages. Please mark 2019 values in plots as low-confidence (distinct color or hatching) and consider excluding 2019 from interannual statistics, or provide results with/without 2019.

We agree that there is room for improvement regarding the data of 2019 and are thankful for the input. To raise awareness of the lower confidence for the results of 2019, we marked them with a * for both carbon and mineral dust concentrations and depositions in Figure 3 and 6 and gave an explanation in the label of the graphs. Apparently, the mark was not visible enough. We find using another shade of grey or hatching, which was used in Figure 6 for another differentiation, unfavourable and thus stucked with the previous mark, but increased its size markedly.

Regarding the results, we agree that providing results with and without 2019 is beneficial. Thus, we added the results of the interannual statistics without 2019 in Table 1 in brackets. We adapted the label of Table 1 and added a clarification in the text:

“Table 1: Minimum, maximum, average concentrations and depositions and their standard deviations for the accumulation periods 2016-2024 (WinsTC) and 2017-2024 (WinsOC and EC). Results excluding data of 2019 is reported in brackets.” (p. 9, l. 245-246)

“Results excluding data of 2019, where the snow cover was sampled incompletely, are provided in brackets.” (p. 9, l. 240-241)

Appendix A is very informative and somewhat buried. It explains convincingly why coagulants were rejected, but some conclusions are speculative without quantitative data. Please summarize key quantitative changes (e.g., how much EC or TC deviated in the test aliquots) to support the conclusions.

We are happy to include more detailed information for interested readers. The text now reads:

“We added $\text{NH}_4\text{H}_2\text{PO}_4$ to a limited number of three types of samples (ultrapure water to be considered as blank; sample containing mineral dust; sample not affected by mineral dust) and compared it to untreated aliquots to assess the applicability for our background site ($n=5$ for each sample type and treatment). Addition of the coagulant led to a decrease in pH (ultrapure water: 5.6 to 4.4; sample containing mineral dust: 8.2 to 4.6), which can alter the sample composition, e.g., by removing carbonate carbon. We observed changes in the signal of the flame ionisation detector in various OC and EC fractions (positive and negative) and obtained blanks with doubled TC loadings (untreated: $2.5 \mu\text{g cm}^{-2}$; $\text{NH}_4\text{H}_2\text{PO}_4$: $4.8 \mu\text{g cm}^{-2}$). Addition of $\text{NH}_4\text{H}_2\text{PO}_4$ led to a decreased TC loading for the sample containing mineral dust (untreated: $114 \pm 2 \mu\text{g cm}^{-2}$; $\text{NH}_4\text{H}_2\text{PO}_4$: $80.5 \pm 1.75 \mu\text{g cm}^{-2}$), presumably dominated by carbonate carbon. The removal of substances leading to a broad shoulder in OC4 was comparable to filters treated with HCl, which will quantitatively remove carbonate carbon. The untreated sample without mineral dust showed comparable TC loadings for filters loaded with

untreated sample and addition of $\text{NH}_4\text{H}_2\text{PO}_4$ ($17 \pm 3 \mu\text{g cm}^{-2}$ and $19 \pm 2 \mu\text{g cm}^{-2}$, respectively), while only the untreated aliquots showed EC ($0.23 \pm 0.11 \mu\text{g cm}^{-2}$). In agreement with Kuchiki et al. (2015), no influence of an aqueous solution of the substance pipetted on a clean filter on the laser signal was observed. Still, no automatic split point could be set for any replicate of the sample not containing mineral dust treated with $\text{NH}_4\text{H}_2\text{PO}_4$, while the untreated sample showed EC." (p. 19, l. 427-438)

Line 14, 'Using the interference introduced by mineral dust, we identify mineral dust layers and find very good agreement with a complementary method based on calcium concentrations and the pH.' I would like some numbers to show the degree of agreement e.g. correlation coefficient. Please present these in the Abstract. Is there an accepted standard for the 'right' value, to which these two methods could be compared? What do the differences between the results of the two methods tell us about the methods or the samples?

As described in a previous comment, Cohen's Kappa was calculated to quantify the agreement of the two methods. While this showed substantial agreement of the complementary methods, it does not conclude whether layers are correctly classified as containing mineral dust. However, the substantial agreement of the two methods based on different mineral dust tracers (IC approach: pH, Ca^{2+} concentration; TOA approach: change in optical properties of Fe containing compounds) is evidence for a correct classification. There is no accepted standard for a "right" value.

In the Abstract, the text now reads:

"Using the interference introduced by mineral dust, we identify mineral dust layers and find substantial agreement with a complementary method based on calcium concentration and the pH." (p.1, l. 14-16)

Overall, I suggest adding more specific quantitative findings to the Abstract (e.g., "EC median = 11.1 ng g^{-1} ; mineral dust deposition up to 2100 mg m^{-2} ").

In addition to the quantitative findings already given in the abstract (e.g. averages, standard deviations and upper ranges), we use the opportunity to shed more light on the results of the single layers. We included the maximum concentrations of WinsOC and EC as well as the highest mineral dust concentrations in the Abstract. While the latter was already given in the text in Section 3.3.3, we added the WinsOC and EC concentrations of single layers in Section 3.2.2.

The corresponding text in the Abstract now reads:

"Average concentrations for elemental carbon and water-insoluble organic carbon were 11.1 ± 2.5 and $458 \pm 215 \text{ ng g}^{-1}$, respectively, and ranged up to 86.3 and 3260 ng g^{-1} in single layers, respectively." (p. 1, l. 13-14)

"Based on thermal-optical analysis and an average iron fraction in mineral dust of 4 %, the approximated mineral dust concentrations ranged up to $25 \mu\text{g g}^{-1}$ in single layers. The approximated mineral dust input during the accumulation period agrees well with gravimetric results and ranged up to 2100 mg m^{-2} ." (p. 1, l. 17-19)

We added the following text in Section 3.2.2:

"For single layers, WinsOC and EC concentrations ranged up to 3260 and 86.3 ng g^{-1} , respectively." (p. 9, l. 243-244)

Samples are analysed for the period 2016 to 2024. The data is presented. But I would like more commentary to understand the significance of the results. Are there meaningful trends in the data? How do the data from these years fit into what has been found for the preceding years, are there patterns, trends, anomalies? I think there is a lot more context to the measurements that could usefully be added.

We agree that trend analysis would be interesting. However, the limited number of years covered so far (2017-2024 for EC, 2016-2024 for mineral dust) is insufficient for a robust statistical evaluation. As no earlier quantitative data for LASI deposited in the seasonal snow cover at the site is available, an extension to preceding years for further evaluations is not possible. For contextualization, an overview of EC and mineral dust concentration and deposition data from other studies (line 219 - 238 and 320 - 334, respectively) is already presented in the submitted manuscript. At present, we are happy to provide a starting point for future trend analysis with our data.

Lines before 283, do you think the decrease in Fe fraction is due to preferential deposition of particles with a high iron content (density? size?)

This is an interesting question. Dumont et al. (2023) observed decreasing deposited dust mass, decreasing particle size distributions and decreasing total elemental Fe mass along the dust plume path from the Pyrenees to the Swiss Alps. Kandler and Scheuven (2019) report a tendency to downwind-fining with largest particles close to the source and a quick depletion of quartz (mass) from the aerosol due to its particle size. Kandler et al. (2007) describe variations in the mineralogical composition of mineral dust sampled in Izaña, Spain, for different size ranges. Based on these and other studies, changes of the dust composition during transport are evident.

However, these investigations exceed the scope of our work and are not accessible through the data we collected. We did not make modifications in the manuscript regarding this topic.

I like the discussion lines 302 to 310. But when you say 'the uncertainty of the mineral dust approximation is 65%', which result does this refer to? It is unclear, and rather than uncertainty, I think you may mean variability?

The uncertainty of 65 % results from the variability of the share of Fe in mineral dust (25 ± 3) and the spreading of data points around the fit to calculate Fe loading from $\Delta ATN_{700-450}$ (lines 295-297). We agree that the inserted information about the two parameters may be confusing but cannot offer an alternative text with increased comprehensibility while keeping all the information we want to give. As the result is a mixture of different variabilities and describes the resulting uncertainty within which the actual result is expected, we did not change the term.

line 324, why can they only be underestimated?

This referred to the incomplete sampling of the snow cover in 2019. If mineral dust layers occurred in the unsampled part of the snow cover, the reported deposition will miss them and an underestimation results. On the contrary, the concentration in this case can be positively or negatively biased, depending on the occurrence of dust and the water equivalent of the unsampled layers as discussed in line 316 to 317. To clarify, we modified the text in Section 3.3.3, which now reads:

"As mineral dust deposition in 2019 may be underestimated due to incomplete sampling of the snow cover, it may be the second highest or highest year regarding mineral dust input." (p. 17, l. 366-367)

Line 416, 'Data used in this work will be uploaded to TU Wien Research Data and the doi will be added here.' I think that at this stage in publication, it is time to upload the data and make the doi. For readers, reviewers, etc. The data must now be in final form if you have analysed it and written the paper, correct?

Yes, the data is prepared. Obviously, we misunderstood that the data should be uploaded before the review. We thought the uploaded file should already include the input of referees. The data was uploaded to TU Wien Research Data and the text in the Data availability section changed to:

"Data presented within this work are available at TU Wien Research Data (<https://doi.org/10.48436/yb10b-yfc83>)." (p. 21, l. 480-481)

(2) Technical

Advisory bodies (SI; IUPAC) advise that the symbols used for variables should be italicized. Please apply this convention at multiple locations e.g. 'n', 'R²' and so on.

We are thankful for the remark and applied the changes to the manuscript at several positions for the symbols "n", "R²", "λ", and "ΔATN₇₀₀₋₄₅₀" in the text and the figures.

There are a lot of abbreviations. LAI, LASI, LAP, GOK, GAW, TOA, WinsTC, WinsOC, EC, OC, ICP-OES, IC, WMO, FLK. They are often a barrier to understanding for non-experts. Some are used rarely. Many (ICP-OES, LAI, LAP, WMO and RF) are only used once. TOA is defined at the fifth use, not the first use. RF is undefined. Recommend to use best practice - define an abbreviation on first use and only define an abbreviation if it is used multiple times. Some (WinsOC & WinsTC) are defined twice. Avoid abbreviations in Abstract whenever possible since it should function as a standalone summary.

We agree that the use of abbreviations may be a barrier to understanding the text. We critically reviewed the text. If an abbreviation is only used once, it was only kept when the abbreviation is better known than the written-out word. This includes analytical techniques as ICP-OES or organisations as GAW or WMO. We defined these abbreviations to explicitly give their meaning but kept the abbreviation for best comprehension. The use of different nomenclature for light-absorbing impurities or particles (in snow) - LAI, LASI, LAP - is inconvenient and hinders literature search for the topic. Thus, we included all forms suitable for our analytes including their abbreviations.

We now define TOA and IC at their first use. The corresponding sentence in Section 2.1 now reads:

"At the end of the winter accumulation period (end of April or early May), snowpacks were sampled in increments of 20 cm for carbonaceous compounds (quantification via thermal-optical analysis (TOA), referred to as TOA-profile) and 10 cm for Ca²⁺ concentration analysed via ion chromatography (IC) and pH (referred to as IC-profile)." (p. 3, l. 73-75)

We are not sure what the last sentence in the comment refers to. In the Abstract, no abbreviations were used. In case the referee referred to the Conclusions section: We agree that it should function as a standalone summary. Thus, we defined abbreviations again here, risking double definition of WinsTC, WinsOC, EC and TOA throughout the manuscript. All abbreviations are used more than once in the Conclusions section.

There are times when the meaning is unclear, please rewrite to improve clarity. Examples:

Line 131, 'Since the temperature range given previously was not always reached for our set of samples,'

We agree that a more detailed description is beneficial, especially because both referees noted it. To clarify, we added an equation for the calculation of the parameter and adapted the text, which now reads:

“As described by Kau et al. (2022), the transmitted laser signal ($\lambda=660$ nm) was evaluated in the calibration phase. At this point, the actual analysis of carbonaceous compounds is already finished. Oven and filter sample are just cooling down, but data is still logged to record the calibration peak. Kau et al. (2022) evaluated a temperature range between 700 and 400°C. Due to small adjustments of the insulation material, the lower temperature, i.e., 400°C, given previously was not always reached at the end of the calibration phase for our set of samples. Thus, we now evaluated the change in transmittance observed between 700 and 450°C using the transmitted laser signal I_{700} and I_{450} , respectively. Converted into a dimensionless temperature dependent attenuation ($\Delta ATN_{700-450}$, see Eq. (1)), samples that equalled or exceeded a value of 4.0 were classified as containing mineral dust. For clarification, $\Delta ATN_{700-450}$ corresponds to $ATN_{700-400}$ defined in Kau et al. (2022), however, using the intensity of the transmitted laser signal at 450°C.”

$$\Delta ATN_{700-450} = 100 * \ln \left(\frac{I_{450}}{I_{700}} \right) \quad (1)$$

(p. 5-6, l. 146-158)

Line 153, 'Considering layers with coloured filters would match the two approaches in 2018,'

We changed the text for better comprehensibility. It now reads:

“For 2018, the two approaches show the same number of mineral dust layers when the layers with coloured filters are considered. Here, the mineral dust layer observed within the IC-profile is divided in two samples in the TOA-profile.” (p. 6, l. 175-177)

Line 197, 'EC concentrations for single layers and changes for layers including mineral dust are shown in Figure 3 exemplary for 2020 and 2024.' -- 'exemplary' is unclear, used in a way that doesn't align with it's meaning -- do you mean 'EC concentrations for single layers and changes for layers including mineral dust are shown in Figure for 2020 and 2024, chosen to exemplify the

The text now reads:

“EC concentrations of single layers and changes for layers including mineral dust are exemplified in Figure 3 using the data of 2020 and 2024.” (p. 9, l. 229-230)

Line 75, 80, 119 and elsewhere, 'filtrated' is not a word, use 'filtered'

We thank Matthew Johnson for the correction and changed the text accordingly. (p. 3, l. 88 and 93; p. 5, l. 134)

Line 79, replace 'Contrary,' with 'On the contrary' or 'In contrast'

Thank you again, we changed the text to “In contrast”. (p. 3, l. 93; p. 13, l. 285-286)

Check x-axis label in Figure 4, remove '()'

$\Delta ATN_{700-450}$ is a dimensionless quantity. To prevent confusion, we adapted the suggestion to leave out the previously given “()” in Figure 4 and added the information of the parameter being dimensionless in the text in Section 3.1:

“Converted into a dimensionless temperature dependent attenuation ($\Delta ATN_{700-450}$, see Eq. (1)), samples that equalled or exceeded a value of 4.0 were classified as containing mineral dust.” (p. 5, l. 152-154)

Additional changes implemented independent of referees' comments

For a more accurate description, we changed “share of Fe in mineral dust” to “Fe mass fraction in mineral dust”.

We adapted the text and now correctly state the lower limit for $\Delta ATN_{700-450}$ as “ ≥ 4.0 ”. This has no impact on the evaluations presented within this work but may be of relevance for future evaluations.

The given WinsTC concentration of the snowpack collected in 2016 slightly differed from the actual concentration (385 and 345 ng g⁻¹, respectively) due to a calculation error. We apologize for the mistake in the submitted version of the manuscript and corrected the average and standard deviation values of WinsTC given in Table 1 (455 and 207 ng g⁻¹ instead of 459 and 204 ng g⁻¹). We also updated the WinsTC concentration of 2016 in Figure 3. The WinsTC deposition of 2016 and any conclusions made in the submitted version are not affected.

In Section 3.3.2, we noticed an error in the classification of the dust-laden samples to snow and rime. Previously, 7 snow and 1 rime sample were reported, while it should read 5 snow and 3 rime samples. We are sorry for the error in the submitted version of the manuscript and corrected it. This change does not affect any conclusions made.

The legend in Figure 1 previously only showed “ATN” instead of “ $\Delta ATN_{700-450}$ ”. We clarified the parameter used here.

References

Baker, E. T. and Lavelle, J. W.: The effect of particle size on the light attenuation coefficient of natural suspensions, *J. Geophys. Res.: Oceans*, *89*, 8197-8203, <https://doi.org/10.1029/JC089iC05p08197>, 1984.

Cerqueira, M., Pio, C., Legrand, M., Puxbaum, H., Kasper-Giebl, A., Afonso, J., Preunkert, S., Gelencsér, A., and Fialho, P.: Particulate carbon in precipitation at European background sites, *J. Aerosol Sci.*, *41*, 51-61, <https://doi.org/10.1016/j.jaerosci.2009.08.002>, 2010.

Dumont, M., Gascoin, S., Réveillet, M., Voisin, D., Tuzet, F., Arnaud, L., Bonnefoy, M., Bacardit Peñarroya, M., Carmagnola, C., Deguine, A., Diacre, A., Dürr, L., Evrard, O., Fontaine, F., Frankl, A., Fructus, M., Gandois, L., Gouttevin, I., Gherab, A., Hagenmuller, P., Hansson, S., Herbin, H., Josse, B., Jourdain, B., Lefevre, I., Le Roux, G., Libois, Q., Liger, L., Morin, S., Petitprez, D., Robledano, A., Schneebeli, M., Salze, P., Six, D., Thibert, E., Trachsel, J., Vernay, M., Viallon-Galinier, L., and Voiron, C.: Spatial variability of Saharan dust deposition revealed through a citizen science campaign, *Earth Syst. Sci. Data Discuss.*, *15*, 3075-3094, <https://doi.org/10.5194/essd-15-3075-2023>, 2023.

Gul, C., Puppala, S. P., Kang, S., Adhikary, B., Zhang, Y., Ali, S., Li, Y., and Li, X.: Concentrations and source regions of light-absorbing particles in snow/ice in northern Pakistan and their impact on snow albedo, *Atmos. Chem. Phys.*, *18*, 4981-5000, <https://doi.org/10.5194/acp-18-4981-2018>, 2018.

Kandler, K. and Scheuvs, D.: Asian and Saharan dust from a chemical/mineralogical point of view: differences and similarities from bulk and single particle measurements, *E3S Web of Conferences*, *99*, 03001, <https://doi.org/10.1051/e3sconf/20199903001>, 2019.

Kandler, K., Benker, N., Bundke, U., Cuevas, E., Ebert, M., Knippertz, P., Rodríguez, S., Schütz, L., and Weinbruch, S.: Chemical composition and complex refractive index of Saharan Mineral Dust at Izaña, Tenerife (Spain) derived by electron microscopy, *Atmos. Environ.*, *41*, 8058-8074, <https://doi.org/10.1016/j.atmosenv.2007.06.047>, 2007.

Kau, D., Greilinger, M., Kirchsteiger, B., Göndör, A., Herzig, C., Limbeck, A., Eitenberger, E., and Kasper-Giebl, A.: Thermal–optical analysis of quartz fiber filters loaded with snow samples–determination of iron based on interferences caused by mineral dust, *Atmos. Meas. Tech.*, *15*, 5207-5217, <https://doi.org/10.5194/amt-15-5207-2022>, 2022.

Landis, J. R. and Koch, G. G.: The measurement of observer agreement for categorical data, *biometrics*, *33*, 159-174., <https://doi.org/10.2307/2529310>, 1977.

Putaud, J. P., Van Dingenen, R., Alastuey, A., Bauer, H., Birmili, W., Cyrus, J., Flentje, H., Fuzzi, S., Gehrig, R., Hansson, H. C., Harrison, R. M., Herrmann, H., Hitenberger, R., Hüglin, C., Jones, A. M., Kasper-Giebl, A., Kiss, G., Kousa, A., Kuhlbusch, T. A. J., Löschau, G., Maenhaut, W., Molnar, A., Moreno, T., Pekkanen, J., Perrino, C., Pitz, M., Puxbaum, H., Querol, X., Rodriguez, S., Salma, I., Schwarz, J., Smolik, J., Schneider, J., Spindler, G., ten Brink, H., Tursic, J., Viana, M., Wiedensohler, A., and Raes, F.: A European aerosol phenomenology–3: Physical and chemical characteristics of particulate matter from 60 rural, urban, and kerbside sites across Europe, *Atmos. Environ.*, *44*, 1308-1320. <https://doi.org/10.1016/j.atmosenv.2009.12.011>, 2010.

Tuzet, F., Dumont, M., Picard, G., Lamare, M., Voisin, D., Nabat, P., Lafayasse, M., Larue, F., Revuelto, J., and Arnaud, L.: Quantification of the radiative impact of light-absorbing particles during two contrasted snow seasons at Col du Lautaret (2058 m asl, French Alps), *Cryosphere Discuss.*, *14*, 4553-4579, <https://doi.org/10.5194/tc-14-4553-2020>, 2020.

Wang, M., Xu, B., Zhao, H., Cao, J., Joswiak, D., Wu, G., and Lin, S.: The influence of dust on quantitative measurements of black carbon in ice and snow when using a thermal optical method, *Aerosol Sci. Technol.*, *46*, 60-69, <https://doi.org/10.1080/02786826.2011.605815>, 2012.

Yamanoi, Y., Nakashima, S., and Katsura, M.: Temperature dependence of reflectance spectra and color values of hematite by in situ, high-temperature visible micro-spectroscopy, *Am. Mineral.*, *94*, 90–97, <https://doi.org/10.2138/am.2009.2779>, 2009.