Response to reviewer

We would like to thank referee#2 for the detailed review of our manuscript. The comments made by the referee were appreciated and helped improve the manuscript. Please find in the attached pdf our responses to the comments (in blue) and our proposed changes to a potential revised manuscript (in blue and in italic):

RC2: <u>'Comment on tc-2022-259'</u>, Anonymous Referee #2, 22 Mar 2023 reply

The manuscript presents nitrate records obtained from three different ice cores collected at nearly the same location on Col du Dome, Mont Blanc in the years in 1994, 2004 and 2012. Using the records of nitrate and the radionuclides 3H and 210Pb together with a 3D ice flow model the authors argue that there are discontinuities in the depth-age relation of the ice cores drilled in 2004 and 2012, which were caused by the presence of an upstream crevasse. This is an interesting hypothesis. Although it is common knowledge that areas with upstream crevasses should be avoided as ice core drilling sites, it could be valuable to demonstrate what the effects of such a crevasse are. However, I find the argumentation rather speculative and not well supported by the data, which are often inconclusive. Further, I miss in some part scientific rigorousness as outlined below. Considering all my concerns as outlined below, this manuscript requires major revisions.

Major comments

210Pb data presented in Fig. 4 have very different time resolution. It is scientifically not sound to compare such data. For instance, the peak at 1970 in CDK would disappear if the same averaging period as in the upper part of the record would be applied. This peak is most likely due to the strong 210Pb seasonality (Eichler et al., 2000). When using the same temporal averaging the postulated anomaly in the 1960s and 1970s in the CDK and CDM cores will be much smaller and may be due to an increased input of 210Pb in the 1970s. Such an increase has been observed already at other glaciers in the Alps, e.g. at Silvretta and Adamello (Festi et al., 2021), at Colle Gnifetti (Gäggeler et al., 1983) and at Grenzgletscher (Eichler et al., 2000) and was attributed to enhanced vertical transport related to the maximum in sulphate aerosol particles acting as transport vehicles (Eichler et al., 2000).

We regret the ambiguity in the presentation of the original version of Fig. 4. Since we were focused on the depth range corresponding to the depth-age anomaly which was sampled in relative high resolution, we only sampled the upper part for of CDK and CDM at lower frequency and interpolated the measurements. This will be changed in the revised version (showing raw data). Samples of CDK and CDM now are shown in Fig, 4 as measured and the depth intervals of the individual measurements (typically 0.7 to 1 m) are assigned clearly. In addition, as requested, we discuss whether or not the re-increase of 210Pb in the 1970s in CDK and CDM might be of atmospheric origin.



Figure 4: ²¹⁰Pb profiles of the three CDD ice cores. The decay-corrected ²¹⁰Pb activity is shown using the drilling year of the respective ice cores as reference. *For CDK and CDM the depths covered by the samples are plotted with thick black lines, whereas the thin integrating line is given to guide the eye and was used to calculate the ²¹⁰Pb inventories. Blue dashed vertical lines indicate the approximate boundaries of the anomaly. Where available, absolute time markers detected over the ²¹⁰Pb perturbed depth zones are also reported. <i>C10 data are from Vincent et al. (1997) and this study.*

The discussion whether or not the re-increase of 210Pb in the 1970s in CDK and CDM might be of atmospheric origin was revised as follows in Section 3.2:

" As a consequence, a strong seasonal cycle with ²¹⁰Pb concentrations three to four times higher in summer than in winter is observed at high altitude Alpine sites. As expected, this also is observed in the snow deposition at CDD and shown in Fig S1 of the Supplement for summer 2004 and the outstanding hot summer 2003, for which an extremely enhanced upward transport was already reported previously (Legrand et al., 2005). Whereas in 2004 a summer to winter ²¹⁰Pb ratio of 2 was found, this ratio reached a factor of 7 in 2003. Together with the systematic decrease of the winter to summer layer thickness ratio with increasing core depth at the drill site (see Section 3 and Preunkert et al., 2000), this pronounced ²¹⁰Pb seasonality counteracts the expected ²¹⁰Pb decrease from radioactive decay.

Second, a well-marked anomaly characterized by ²¹⁰Pb enhancements (including ²¹⁰Pb peaks up to 10 times higher ²¹⁰Pb than expected from atmospheric deposition) is observed in the three cores. The anomaly extends from ~83 to 108 m depth (i.e., ~26 to 54 years) in C10, ~85 to 108 m (i.e., ~32 to 70 years) in CDK, and ~82 to 102 m (i.e., ~33 to more than 58 years) in CDM ice. The ²¹⁰Pb re-increase observed in CDK and CDM, however, is less pronounced than in C10. In addition, the starting depths of the CDK and CDM ²¹⁰Pb re-increases correspond to the 1970s, for which ²¹⁰Pb enhancements have been reported at other ice core sites (Eichler et al.,2000) and attributed to an enhanced vertical transport related to the temporal maximum of atmospheric sulfate aerosol acting as transport vehicle. To check whether these atmospheric conditions also could be responsible for the enhancement seen in CDK and CDM, we report exemplarily the CDK ²¹⁰Pb activity, corrected for its respective deposition date together with the corresponding sulfate concentration in Fig. S1 of the Supplement. As mentioned above, a strong seasonality was detected in the uppermost part of the CDK core for a few years where ²¹⁰Pb samples are available in seasonal resolution (Fig. S1a of the Supplement). If atmospherically derived, mean ²¹⁰Pb concentrations of ice layers from 60 to 85 m depth (i.e., from 1988 to 1972), i.e., in the period for which the sulfate aerosol maximum was observed at CDD (Preunkert et al., 2001), would correspond to around $130 \pm 60 \text{ mB kg}^{-1}$ of ²¹⁰Pb in freshly deposited snow, which is comparable to the atmospherically derived ²¹⁰Pb further upward in the core. However, from 85 to 108 m depth, this connection between sulfate levels and ²¹⁰Pb activity no longer holds. Whereas sulfate concentrations strongly decrease, ²¹⁰Pb at the time of deposition (decay-corrected) would be strongly enhanced (mean of 600 mBq kg⁻¹) and far above what is expected from atmospheric ²¹⁰Pb contributions. Thus, the mechanism proposed by Eichler et al. (2000) cannot be invoked in this part of the CDD core. For CDM (not shown) a similar picture appears. While from 80 to 90 m surface decay-corrected ²¹⁰Pb (160 \pm 70 mBg kg⁻¹) would not have been significantly enhanced compared to the atmospherically derived ²¹⁰Pb concentrations seen further up in the CDM core, this is not the case between 90 and 103 m depth. As for CDK, mean values at the time of deposition would have been around 650 mBq kg⁻¹ and thus far too high to what would be expected from atmospheric transport.

Third, below the anomaly, a decrease

Revised Fig. S1 of the Supplement now is as follows:



Figure S1: 210Pb profiles of the CDK ice cores (left y-axis, black) together with corresponding SO42- concentrations (right y-axis, red). The decay-corrected 210Pb activity is shown using the from the ice core chronology estimated snow deposition date. A thin integrating line is given to guide the eye, whereas the depths covered by the samples are assigned with thick lines. The blue dashed vertical lines in (b) indicate the approximate boundaries of the anomaly.

The entire depth records of 210Pb should be shown and not only the interval between 40 and 130 m in Fig. 4. Without the upper part, it is impossible to see if there is a decrease of 210Pb with depth at all and if the surface activity is in the range expected for glaciers in the Alps. Done. The full records will be shown in revised Fig. 4 (see above).

In the C10 core, 210Pb was determined by gamma-spectrometry (Vincent et al., 1997), whereas for the CDK and CDM cores 210Pb was analyzed by alpha-spectrometry of its decay product 210Po after chemical enrichment, which is the much more sensitive method. Gamma-spectrometry is rather insensitive due to the high conversion of the low energy gamma-line at 46 keV (96% in the form of electron and only 4% in the form of gamma-emission) and the rather low efficiency of gamma-detectors. This method is normally used for samples with high activity concentrations of 210Pb, e.g. from lake sediments. For low-activity ice samples, the

uncertainty is high (more than 50%, Vimeux et al., 2008). Especially in the region, where the anomaly was observed in the C10 core, also 137Cs activity concentrations are high due to the fallout from nuclear tests. This must have resulted in a high background in the gamma-spectrum. These uncertainties need to be discussed.

Thanks for this comment. The uncertainties of the gamma spectrometry used will be now discussed in the manuscript and respective error bars of the 210Pb data are reported. In fact, this specific gamma spectrometry method was developed at the Laboratoire de Glaciologie et Géophysique de l'Environnement, now Institut des Géosciences de l'Environnement (Delmas and Pourchet [1977], Pinglot and Pourchet, 1995) and applied in the past for many ice core studies in the Alps, sub-Arctic, South America, and also at polar sites (see e.g. Pourchet et al., 2000, Pinglot et al., 2003, Vimeux et al., 2008, Pourchet et al 2003).

Note that, since the energy of the 210Pb (46.54 keV) and the 137Cs (661 keV) is rather different (resolution of the detector is between 1.3 and 1.7 keV) we assume that the radioactive fallout from the nuclear tests should not have led to significant downgrading of the quality of the 210Pb measurements. Anyway, the adopted detection limit for C10 was assigned originally for ice core measurements which included also the time period in which 137Cs activities were high.

The following text will be added in section 2:

"" Previously reported ²¹⁰Pb measurements in C10 ice (Vincent et al. 1997) analyzed at the Laboratoire de Glaciologie et Géophysique de l'Environnement, now Institut des *Géosciences de l'Environnement (IGE), were complemented by two samples measured for* this study. The analytical technique was high-resolution gamma-ray spectrometry, designed to detect very low levels of radioactivity using a 20% high-purity Ge (N-type) detector, with an anti-Compton scintillation detector (Pinglot and Pourchet, 1995) for which snow and ice samples were filtered previously through ion-exchange papers (Delmas and Pourchet, 1977). This method is less sensitive than α -spectrometry and Vincent et al. (1997) did not assign uncertainties to their analyses. Here we estimate the uncertainty based on what has been reported in other studies using this detection method developed at *IGE. Pinglot et al., 2003 reported a detection level of 10 mBq at a 97.5% confidence level* for 3 days of counting on ice core samples with a typical ²¹⁰Pb activity of 20 – 50 mBq kg⁻¹. These measurements included Chernobyl fallout in sub-Arctic glacier sites, and the levels were similar in range to the background activities of 50-100 mBg/kg found in our cores. *On the other hand, detection levels of 13 and 25 mBq were calculated at 97.5 % confidence* when peak interferences where neglected or considered, respectively, for a 10 g sediment sample containing 1000 times higher ²¹⁰Pb activities as found in ice cores (~70 Bq kg⁻¹) that was measured for 63 hours (Pinglot and Pourchet, 1995). Vimeux et al. (2008) reported a lower detection limit of 4 mBq kg⁻¹ for ²¹⁰Pb measurements (activities between 20 and 100 mBq kg⁻¹) on relatively small (150-250 g) ice core samples from Patagonia. The ²¹⁰Pb activities in C10 ranged from 50 – 700 mBq kg⁻¹, with the measurements done on the C10 drilling chips merged over 3 to 5 m, allowing to obtain sample weights of up to \sim 3 to 5 kg. Since these sample masses, type (ice core sample) and geometry (filter) are comparable to those used in the Pinglot et al. (2003) study but are very different from the sediment sample in Pinglot and Pourchet (1995), we assume in the following a detection level of 10 mBq and an uncertainty of 30 mBq for the C10²¹⁰Pb measurements. Note that, the dataset from Vincent et al. (1997) was supplemented by two additional samples for which ²¹⁰Pb analysis and quality control were not yet finished in 1997.



It is unclear, which 210Pb decay correction was made. In Fig. 4 it is stated that for the C10 core the 1994 activity is shown. However, 210Pb activity concentration are much higher than in the original publication (Vincent et al., 1997). For a comparison between the cores, the activity should be corrected to the same reference date.

Thanks for this comment. In Fig 4, the same reference date (1994) is applied for C10 as in Vincent et al., 1997. C10 data seem to be higher in our Fig. 4 compared to the one from Vincent et al., 1997, since two additional samples were added to the data set in our study. C10 data from Vincent et al., 1997 were complemented with two additional samples for which additional measurements and quality check were done after 1997. This will be stated in the revised manuscript in section 2:

"...Note that, the dataset from Vincent et al. (1997) was supplemented by two additional samples for which ²¹⁰Pb analysis and quality control were not yet finished in 1997."

I cannot follow the argument how the presence of the crevasse caused such a large 210Pb anomaly in the C10 core, but did not affect the stratigraphy, while in the other two cores the stratigraphy was disturbed, but the 210Pb anomaly was much smaller if present at all. This is a contradiction to me.

Essentially there are two states of the crevasse – one for which the crevasse is open to bedrock and sealed by a snow bridge, and a second in which it is at least partly open to the atmosphere. Whereas in the first state the 222Rn emitted from the granite in the bedrock will accumulate, diffuse in the surrounding firn and produce 210Pb in excess (this would correspond to what is observed in C10), in the second state the excess 222Rn gas can escape from the crevasse to the atmosphere, thus 210Pb production will be strongly limited (this would correspond to what is observed in CDK and CDM). To clarify that, these two states will be reported in Fig 5b and 5c in the revised manuscript.



Figure 5: (a) Thickness changes between 1993 and 2017. The contour lines of surface topography correspond to the 1993 surface (adapted from Vincent et al., 2020) overlain by a modelled flow line (color scale on top) which reports the calculated arrival depth at the drill site of C10, CDK, and CDM (black star) (Gilbert et al., 2014). The crevasse location (blue line) is based on the 30th June 2004 aerial photo from IGNF (see Fig.1) (b and c) Schematic representation of the origin of the ²¹⁰Pb anomalies found at the drill site following the ice flow model of Gilbert et al., 2014, extracted along the flow path reaching the drill site. Isochrones are marked in red, flowlines in green (see also Section 4). The grey shaded zone indicates firm, the dotted zone indicates the snow bridge over the crevasse. Two states of the crevasse are reported: (b) the crevasse is open to the bedrock but sealed from the atmosphere by a snow bridge. In this state ²²²Rn and ²¹⁰Pb accumulate to reach concentrations well above atmospheric conditions in the crevasse and the surrounding firn (c) the crevasse is at least partly open to the atmosphere. In this state ²²²Rn and ²¹⁰Pb concentrations in the crevasse and the surrounding firn are strongly reduced compared to (b). The formation of missing or doubling ice layers is indicated by the orange and pink arrows.

In addition, we will reword the discussion of this point in Section 4 as follows:

"... "... A partial opening of the crevasse to the atmosphere would allow the bedrock-derived ²²²Rn in the crevasse to mix with the much lower atmospheric ²²²Rn concentrations (Pourchet et al., 2000). This would have led to a strong reduction of additional ²²²Rn accumulation and ²¹⁰Pb production in the crevasse and in the snow and firn around the crevasse, starting from the moment of the opening to the atmosphere. This would explain ²¹⁰Pb inventories of 70 and 55% in CDK and CDM compared to C10, because of the radioactive decay of ²¹⁰Pb accumulated before the opening of the crevasse to the atmosphere, over 10 and 18 years, respectively....."

The agreement between the nitrate records obtained at a nearly identical location (please add coordinates to support this statement) is not as good as I would expect. Maybe plotting them against a m water equivalent scale would make it easier to identify common features. Generally, I find Fig. 2 difficult and confusing. What are the 250 ppb and 400 ppb levels?

The GPS coordinates of the ice cores were checked. Differences are at most 10m distance. We will provide the mean GPS coordinates of the three cores in the Introduction:

"...Underpinning these efforts are three ice cores all drilled to bedrock within maximal 10 m of each other (mean geographic location of 45.842195° N, 6.84675° E) in 1994 ..."

The fact that the agreement of the NO3 depth profile is not as good as expected by the reviewer is likely due to the fact that the corresponding layers were not deposited at the same location along the flowline since the cores were not drilled at the same time. E.g., the layer of 1990 is in C10 at 25 m depth and in CDM at 67 m depth. As shown in Fig. 5a, the deposition locations were different by around 50 m. Taking in account the changing accumulation and winter to summer deposition ratio upstream the core (see section 3) this would result in stratigraphic differences in the NO3 (and all ion) depth profiles.

To point this out we added in the caption of Fig. 2 the following sentence:

"...Note that the chronological changes of the NO3 concentrations are offset in depth relative to each other due to the different years the cores were drilled."

We prefer to keep m scale in Fig. 2 since with that the reader can directly compare the depths in the core with the modelled surface deposition sites of the ice layers (see Fig. 5a). In any case, since ice core sections reported in Fig. 2 (except for the upper part of C10) are below the close off, which is around 50 m depth at the drill site, overplotting the m water equivalent scale would not change the picture.

The bars of 250 and 400 ppb were removed in Fig. 2 since they are not used in the discussion.

What is also puzzling is that 14% of the nitrate values (and even 30% of the ammonium data) were discarded. What is the basis for that? Which criteria did you use to identify contaminated values?

Thanks for this remark. After careful consideration we found that we made an error in the original manuscript. In fact much less data were discarded due to contamination. This is explained now in more detail.

In section 2 it will read:

"....Despite the undersized core section available for the CFA analyses at CEP, 86% of the ice core could be analyzed. The nitrate profile obtained at DRI and CEP (covering 97% in this depth range), were compared from 45 to 86 m depth. Both datasets are in very good agreement (Fig. 2). After having additionally discarded very high peaks in NO₃- values (1.5% of CEP data), which were not present in the DRI dataset and could be attributed easily to contamination, mean NO₃- values from 45.3-86.0 m were 263 ppb (CEP) and 255 ppb (DRI). The agreement is somewhat weaker for NH₄+ likely because only 80% of the depth range is covered by the CEP measurements. After discarding additionally 8 % of the CEP NH₄+ data consisting of high NH₄+ peaks which were not present in the DRI dataset, the mean NH₄+ values of 101 ppb (CEP) and 95 ppb (DRI) were in good agreement." Why are the tritium records not continuous? With a discontinuous record it is difficult to identify the 1963 maximum. In the case of the CDK core the maximum might be at 86 m. The CDK and CDM records were mainly dated first using the depth stratigraphy of major ions and by comparison with C10, and only the depth range over which the 3H bomb test maximum was expected was analyzed for tritium. This is common approach when searching the 1963 bomb maximum since the rest of the 3H depth profile is rather uninteresting scientifically. In the case of CDK, ice layers at 86m depth could be clearly assigned to the years 1970 (see Legrand et al., 2013 and Figs. 2 and 3). Thus, in our opinion there is no reason to search for the 1963 bomb horizon at this core depth.

Minor comments

Bachelor thesis's cited (Waldner, Zipf) are not publicly available. Include information in supplement.

The references for the two Bachelor thesis were initially put to credit the work of the two students, but finally both became co-authors. The references were deleted, and their work (analyses of the samples) will be credited in the author contribution section. Analytical details on measurements are already published and referenced in the manuscript (see Section 2). The detection limit appropriate to the method used in the CDK and CDM 210Pb analyses will be given Section 2 in the revised manuscript:

"²¹⁰Pb samples of CDK and CDM ice were analyzed at IUP by α -spectrometry for its decay product ²¹⁰Po. Typical blank values of (5.7 ± 2.5) 10⁻⁵ Bq for ²¹⁰Po and (3.8 ± 1.6) 10⁻⁵ Bq for ²⁰⁹Po were subtracted from the sample counts (see Stanzick, 2001, and Elsässer et al., 2011 for further working analytical conditions)."

Thus, we feel that no supplement information is necessary.

103: Despite the undersized core section available at CEP, the nitrate profile obtained at DRI and IUP are in very good agreement (Fig. 2). Do you mean DRI and CEP? Thanks, CEP was meant, this was corrected.

177: How was the winter to summer layer thickness ratio obtained?

The winter to summer layer thickness ratio was calculated on the basis of the NH4 depth stratigraphy. Details can be found in Preunkert et al., 2000. This now is assigned clearly in the revised manuscript when the term "winter to summer layer thickness" first appears (Section 3.1).

"...and the winter to summer layer thickness ratio, calculated on the basis of the ammonium depth stratigraphy (see details in Preunkert et al., 2000), decreases from 1 at the surface to 0.5 at 100m depth. "

216: Result of annual layer counting, what do you mean with that? Thanks for this remark, we clarified this sentence in section 3.1.

".... As a consequence, annual layer thicknesses of only 0.7 and 0.2 mwe are observed at 100 m and 118 m depth (Preunkert et al., 2000) and the winter to summer layer thickness ratio, calculated on the basis of the ammonium depth stratigraphy (see details in Preunkert et al., 2000), decreases from 1 at the surface to 0.5 at 100 m depth....."

239: For 210Pb seasonality include reference Eichler et al., 2000. ok done

"...CDM ²¹⁰Pb re-increases correspond to the 1970s, for which ²¹⁰Pb enhancements have been reported at other ice core sites (Eichler et al.,2000) and attributed to an enhanced vertical transport related to the temporal maximum of atmospheric sulfate aerosol acting as transport vehicle..;"

245-250: A zero 210Pb level can only be seen if the values are blank corrected and if the ice does not contain any supported 210Pb from mineral dust (see e.g. Gäggeler et al., 2020). Did you do a blank correction and what was the blank?

Thanks for your remark, the data are blank corrected. The "non-zero" term will be changed in "above detection limit" and the detection limit will be added in the manuscript in Section 2. "²¹⁰Pb samples of CDK and CDM ice were analyzed at IUP by α -spectrometry for its decay product ²¹⁰Po. Typical blank values of (5.7 ± 2.5) 10⁻⁵ Bq for ²¹⁰Po and (3.8 ± 1.6) 10⁻⁵ Bq for ²⁰⁹Po were subtracted from the sample counts (see Stanzick, 2001, and Elsässer et al., 2011 for further working analytical conditions)...."

and in Section 3.2 it will read:

"...However, it is worth noting that, especially in the case of the CDM and CDK cores, ²¹⁰Pb activity (after blank correction) is above detection limits even in the bottommost core sections, while in C10 levels are below the detection limit..."

Figure 3: C10 was drilled in 1994. Why do the records of annual layer thickness and nitrate concentration continue to the year 2000?

Thanks for this remark. We agree that the Fig. and caption needed improvement. C10 and CDM annual layer thickness data are shifted in time to compensate for the different drilling dates of the three ice cores. The revised Fig. 3 is as follows:



Figure 3: (a) Annual layer thickness of C10 (Preunkert et al. 2000) and CDK (Legrand et al., 2013) compared to CDM. To compensate for the different drilling dates of the three cores, annual layer thickness data of C10 and CDM were shifted for +10 and -8 years, respectively. For CDM, the annual layer thickness is estimated via the ammonium stratigraphy back to 1980 and via the nitrate (and ammonium) stratigraphy further back in time (Section 3.1.3). (b) comparison of nitrate summer half-year means of C10 (Preunkert et al., 2003), and CDK (Legrand et al., 2013) with CDM. The thick solid lines for C10 and CDK refer to the smoothed profile (single spectrum analysis, see Legrand et al., 2013). CDM depth intervals for which the dating is uncertain (Section 3.1.3), are marked with dashed lines.

References

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