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Interactive comment on "Towards a quasi-complete reconstruction of past atmospheric aerosol load and composition (organic and inorganic) over Europe since 1920 inferred from Alpine ice cores" *by* S. Preunkert and M. Legrand

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Responses to Anonymous Reviewers

Response to Anonymous Reviewer #1

Thanks to reviewer n°1 (denoted "R1" in the following) for his/her recommendations

and constructive comments on the manuscript. We (denoted as "Au" in the following) will respond in the following point per point to his/her comments.

R1: The manuscript presents a comprehensive set of impurity records from 3 ice cores obtained from Col du Dôme (CDD). The dataset covers the period 1920 to present and the authors aim at obtaining an almost complete set of both organic and inorganic compounds. To a large degree the manuscript presents previously published results by the same authors (that is reflected in the publication list where 16 out of 35 references are first- authored by the two authors). The present MS does, however, contain new records and I think the review is useful for a large community including modellers of recent atmospheric conditions. The records presented in the MS are of relevance to the journal, the conclusion con- cerning increased biogenic emissions since the 1950s is compelling, and, in general, the data are well presented and the MS is well written. I have only minor comments and suggestions.

R1: General comments:

R1: The approach of separating summer and winter signals appears sound and relevant, but it does of course put high demands on the dating of the ice cores that needs to be truly annual. Based on the material presented in the figures it is hard to judge if the dating is really sufficiently good in all three cores. The comparison in Figure 1 looks rather convincing, but we don't see annual layers here. An example showing the annual layering in, say, a decade and how they correlate in all three cores would be useful.

Au: We agree and will add a figure to the manuscript, which shows NH4+ raw data in the three cores over the time period of around 2 decades (see Figure 1)

R1: Likewise, the separation of samples or sample fractions heavily influenced by Saharan dust is very reasonable, but we are not introduced to the criteria applied to identify the Saharan dust in this manuscript. Please repeat the principles here for completeness, and maybe show an example of records with/without strong influence of Saharan dust.

Au: You are right. We have forgotten to summarize the Saharan dust criteria. We will insert in section 3.1 (page 7,line 21) the following: “As detailed by Preunkert et al. (2001a), summer samples containing more than 100 ng g-1 of calcium and lying below the 25% quartile of a robust spline [Bloomfield and Steiger, 1983] through the raw acidity profile, are considered as significantly affected by Saharan dust input. In addition, over the last 4 decades (after 1960), because of enhanced acidity, samples containing Saharan dust input can remain acidic [Maupetit and Delmas, 1994]. Such samples were characterized by Maupetit and Delmas [1994] as having typical sulfate to calcium and nitrate to calcium weight ratios close to 2.6 and 1.2, respectively. Winter samples, containing more than 20 ng g-1 of calcium and having an alkalinity lying below the 25% quartile of the robust spline, were also considered as contaminated by Saharan dust input. With that 98 summer (on a total of 762 samples) and 4 winter (on a total of 337 samples) samples are suspected to be influenced by Saharan dust." Since we show however the impact of a Saharan dust event on the ionic budget (Figure 2 of the manuscript) we think that it may be not necessary to add an additional graph here.

R1: Whereas the CDD dataset presented in the MS is very comprehensive and complete, I do miss some comparisons to comparable records form the region, such as the Colle Gnifetti ice cores. For the future, it would be most useful for the paleo community if the authors of this and other alpine related records would join forces and cross-compile their records to provide a more general picture of the past regional climate. Looking forward to see such a compilation in a future publication.

Au: Yes we fully agree, as a further step it would be a very useful work for the ice core people community.

R1: Specific comments:

R1: P. 1102: Dating: Do you see Katmai 1912AD in your CDK sulphate? It would make a nice reference point.

Au: Unfortunately we see no clear signal, which corresponds to the Katmai event. Actually we see two enhanced SO42- layers (in 1913 and 1922 based on our dating), but both horizons are accompanied by Ca2+ and are alkaline. Therefore, since no unambiguous identification was possible, we did not assign Katmai to any ice layer.

R1: P. 1103: Thevenon et al., JGR, 2009 report on a 1000 yr ice core from the Colle Gnifetti glacier. They date it among others by identifying ‘four visible yellowish dust horizons attributed to Saharan dust events of 1977, 1947, 1936, and 1901’, and by the volcanic layer of Katmai 1912. Are those Saharan dust events also observed in your cores? If not, why is that?

Au: Thanks for this comment, since we have forgotten to mention this result: As detailed in Preunkert et al. (2000) (see figure 2) we could identify the dust horizons of 1977, 1947, and 1936, which were already identified at Colle Gnifetti by Haeberli et al. (1983) and Jung (1993), within the Col du Dôme ice. We will add this information in the text of our manuscript. Concerning Katmai, please see the answer above.

R1: P. 1103, l. 26: ‘. . .using an electric plane device’ - what is that? Please be a bit more specific.

Au: Sorry the wording might be misleading. We should have written “electric plane tool” or “power planer”. In more detail: we use a portative electric plane tool that is mounted in a way that it works like a bench plane. Pre cut in lamella with a band saw, the ice is then slid over the laces of the electric plane tool. We propose the following wording: . . ..using an adapted and pre-cleaned electric plane tool which is mounted to be used as a sort of bench plane over which the ice is slid.

R1: P. 1105, l. 24; p. 1106. l. 5: You talk about ‘contamination’ of the records by Saharan dust. Strictly speaking, the ‘natural’ Saharan dust input is not contamination?

Au: You are right, we will change the wording in “influenced by Saharan dust”

R1: Figure 6: The figures are very small and hard to read. Hopefully, they can be somewhat expanded?

Au: This is true, and we hope that it is only due to the fact that the discussion paper is in “landscape” format. We hope that the “portrait” format of CP will improve the size of this figure. If not we will separate figure 6 in two independent figures.

R1: Hopefully, the authors will make their important datasets public available somewhere? They should be of interest to a large community including atmospheric modellers.

Au: You are right we did not give our data set to a database yet. We will think about that, but anyway we would like to take the opportunity to invite all interested colleagues to contact us directly for the data until the data will be on a public site.

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R1: Reference:

Thevenon, F., F. S. Anselmetti, S. M. Bernasconi, and M. Schwikowski (2009), Mineral dust and elemental black carbon records from an Alpine ice core (Colle Gnifetti glacier) over the last millennium, J. Geophys. Res., 114, D17102, doi:10.1029/2008JD011490.

Au: References:

Haeberli, W., Schotterer, U., Wagenbach, D., Haeberli- Schwitter, H. and Bortenschlager, S. 1983. Accumula-tion characteristics on a cold, high-alpine firn saddle from a snow-pit study on Colle Gnifetti, Monte Rosa, Swiss Alps. J. Glaciol. 29, 260–271.

Jung, W. 1993. Hundertja ÌĹhrige Zeitreihen der Aerosolde- position auf einem hochalpinen Gletscher. MS thesis. Institut für Umweltphysik der Universität Heidelberg, 71 p.

Maupetit, F. and Delmas, R. J. 1994. Snow chemistry of high altitude glaciers in the French Alps. Tellus 46B, 304–324. Preunkert, S., Wagenbach, D., Legrand, M., and Vincent, C.: Col du Dôme (Mt Blanc Massif, French Alps) suitability for ice core studies

in relation with past atmospheric chemistry over Europe, Tellus, 59, 993–1012, 2000.

Preunkert, S., Legrand, M., and Wagenbach, D.: Causes of enhanced fluoride levels in Alpine ice cores over the last 75 years: Implications for the atmospheric fluoride budget, J. Geophys. Res., 106, 12619–12632, 2001a.

Response to Anonymous Reviewer #2

Thanks for your comments (denoted as “R2” in the following) that were useful since at some places the manuscript was indeed not clear enough.

R2: The authors review previous geochemical data from alpine ice cores recovered from Mt. Blanc, and summarize what is known about various aerosol deposition trends in central Europe from prior to World War II to present. This portion of the paper is comprehensive, which is not a surprise given that the authors have been involved in most of the data collection over the past 15 years. Given the extensive and excellent publication record resulting from these datasets, I have no real concern about the quality of data, the interpretations that are summarized in the paper, or the figures that are adapted from previously published work. If the authors intend simply a review paper, then I imagine they could leave the summary as is and provide a few key insights into where knowledge is still lacking and how it might be addressed in future studies. Unfortunately, the objectives of this paper in terms of providing a quantitative analysis are not clear to me. The authors attempt a semi-quantitative inversion of snow chemical concentrations to atmospheric aerosol concentrations, but this analysis is based on some rather poorly constrained assumptions in section 5. I don’t see any new data collection, or numerical analysis here, but rather the application of more previous work to previously collected data. In the end, it is not clear to me how the conclusions reached from the semi-quantitative inversion are any different that what has already been published without the inversion. Perhaps I am missing a major piece of the paper, but if so I would appreciate the authors documenting in much clearer terms in the introduction what the paper is intended to do. Is it primarily a review, or is the main point the inversion?

Au: Clearly, it is not a review but a work which aims (and it is not obvious) to extract the aerosol mass and composition and their change over the past from the various available chemical ice records of the Col du Dôme glacier site. Well before doing the inversion of ice data in the paper, we select relevant information for discussing exclusively aerosol. As far as we know this work is done here for the first time along an ice core. As an example, the discussion made on the contribution of nitrate to the aerosol mass it is not an obvious question. Note also that the long-term trend of calcium in the CDD ice was never presented (Figure 3) and discussed prior to this present study. Finally, the presentation of the ionic budget (including carboxylates) and its changes over the past (Figure 4) is also presented for the first time for an alpine ice core. But we agree that our wording was not clear enough and we propose to rewrite the end of the introduction as follows: "In this paper, among the array of chemical (organic and inorganic) ice core records available from the high-elevated CDD Alpine site, we select and use those that contain relevant aerosol information in view to reconstruct the past atmospheric change of aerosol load and composition over Europe from 1920 to the recent decades. That permits to reconstruct the mass concentrations of key aerosol components as nitrogen and sulfur related aerosol, carbonaceous aerosol, sea salt and mineral dust and their changes over the past."

Au: Also at the beginning of section 4 we can specify: "In this section, among species investigated in CDD ice we discuss those that contribute to aerosol and calculate their corresponding mass contribution and changes over the past."

R2: If the inversion is indeed the central point, then a much more rigorous treatment of the inversion techniques and assumptions needs to be presented to be able to evaluate it. So in essence I suggest that the authors either restrict the paper to a comprehensive review, or scale the length of review back considerably and focus instead on the data inversion, with a view towards conclusions that provide a clear addition to the knowledge of this important topic.

Au: No, as stated above the inversion is not a central point of the paper. The inversion is used at the end after having estimated the mass concentrations of key aerosol components and their change over the past (Table 2). However you are right, to be clearer we propose to start this part of the discussion with a first examination of the ice aerosol load from which it appears clearly that the aerosol load change is mainly driven by the increase of ammonium and sulfate together with water-soluble organic matter. Only with the aim to discuss further the change of the other aerosol component an inversion step is needed. We therefore propose to write prior the discussion on the data inversion: "Based on discussions in Sect. 4, Table 2 summarizes the estimated mass concentrations of the different aerosols present in winter and summer CDD ice layers corresponding to the years 1921-1951 and 1971-1988. The total mass concentrations related to the different aerosol fractions together had increased by a factor of 2 in winter (from 183 ppb in 1921-1951 to 333 ppb in 1971-1988) and a factor of 3 in summer (from 570 ppb in 1921-1951 to 1622 ppb in 1971-1988). A large part of these changes are due to the increase of ammonium and sulfate together with water-soluble organic matter. Though dominating the change of ice aerosol load, these ice core changes of ammonium, sulfate and water soluble organic matter can however not be compared to those of other aerosol fractions like dust or water insoluble organic matter in terms of atmospheric changes. Indeed, while for submicron water soluble aerosols as ammonium, sulfate and water-soluble organic we may expect a similar relationship between concentrations in air and snow, other aerosol fractions may have different relationships. For instance, other submicron aerosols like water insoluble organic matter or black carbon being less water-soluble than the preceding components or dust having a super-micron contribution would have different air-snow relationships compared to sulfate. Though, the difference in the air-snow relationships for the different aerosol fractions is not accurately known, a better estimate of the past atmospheric change of aerosol load and composition can be derived by inverting the ice core concentrations to corresponding ambient air concentrations. Furthermore, doing that we can also compare the inverted ice data that cover the years 1921-1951 and 1971-1988 with atmospheric data gained at the Vallot site in summer 2004, a recent period over which DOC and WSOC snow deposit data are not available (see Sect. 2). "

Interactive comment on Clim. Past Discuss., 9, 1099, 2013.

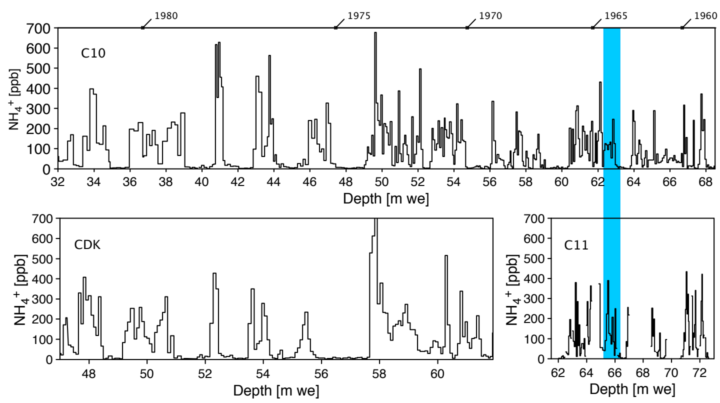

Fig. 1. NH4+ raw data of the C10 (top) CDK (bottom left) and C11 (bottom right) ice cores, illustrating the resemblance of ionic depth profiles of the CDD ice cores. The blue zone reports the 1963 horizon

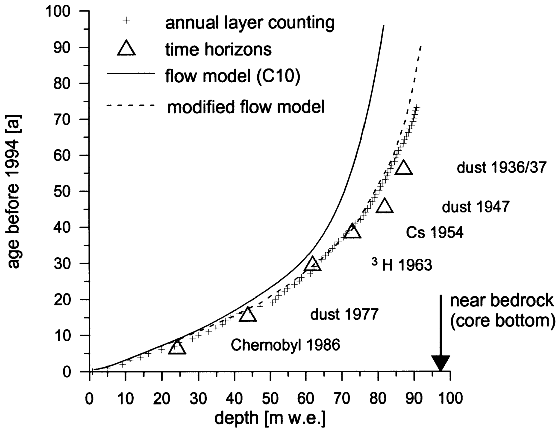

Fig. 8. Comparison of different depth-age relation in the upper 119 m of the C10 core (see text).

Fig. 2. Dating of the C10 ice core (Figure 8 from Preunkert et al. (2000))