

Interactive comment on “Radiocarbon analysis in an Alpine ice core: record of anthropogenic and biogenic contributions to carbonaceous aerosols in the past (1650–1940)” by T. M. Jenk et al.

T. M. Jenk et al.

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First of all we thank the reviewer for the very careful reading of the manuscript and for providing numerous valuable comments, which helped us to improve the quality of our manuscript.

R. The paper addresses a topic (organic aerosols, their sources) is of the highest relevance, since their climatic forcing is a major source of uncertainty in climate modelling. Many related papers have been published in ACP. The use of sub-tropic glaciers as an archive of past aerosols is a promising new method. By pushing the limits of radiocarbon determination with AMS towards lower sample sizes, the authors have for the first time obtained a record covering the industrial era, with a reasonable time-resolution.

The authors present a thorough interpretation, and also freely discuss the shortcomings, which naturally occur for this first series: limits in resolution and precision, and gaps in the data. Thus, the interpretation must remain partly vague, still it is valuable, since it might guide the interpretation of future measurements. After identification of local events, the trends in the data are in reasonable agreement with expectations. The paper convinces the reader that the method is suitable to investigate historic aerosols. A major finding which does not agree with expectations is why EC does not approach $fM = 1$ for samples before 1800. Future investigations will show whether this is related to a methodical problem or whether this is a surprising property of natural EC aerosol. A disadvantage for the understanding is that the technical description, in a complementary publication submitted to Nuclear Instruments and Methods in Phys. Res. B, is not yet available. This is, however, not the fault of the authors or the present paper. The paper is well structured and clear. The authors have compiled a comprehensive bibliography, which is valuable by itself. A table of the 33 samples showing total mass, OC/EC concentrations, fM , and $\delta^{13}C$ should be added, maybe in the supporting online material. Considering the scientific potential, I expect that this is only the first such record of a whole series.

A. We are thankful that not only our results but also discussion of the shortcomings and limitations of this first ice core record are acknowledged. We share the opinion that our observations might help for future measurements. The mentioned paper is now accepted and in press. Instead of supplying a table with the concentration of each sample we will add a table with averaged OC and EC concentrations, fM (as per cent biogenic contribution) and resulting source apportioned OC and EC concentrations to the online supporting material. We think the averages are more useful, since they contain well defined time information. We did not include the $\delta^{13}C$ results because most of them are discussed and values are given in the text.

Specific comments

R. * Abstract, line 16: "Before 1800, OC was of pure biogenic origin...": I do not un-

derstand why the number 1800 is given. Figure 3 suggests a biogenic origin till year 1870.

A. We agree and we will change the according text.

R. * Abstract, line 17: "21+/-2 microgram/kg": The error of the mean provides little information. The standard deviation should be given, since the variability is a relevant climatic information: " 21 microgram/kg with a standard deviation of ? ..."

A. We agree and will accordingly follow this suggestion in the manuscript.

R.* Abstract, line 20: Why do you show especially mention 1940? This year is rather untypical, since it has the highest EC and 2nd highest OC in the whole record.

A. We think these high values rather indicate a strong increased atmospheric concentration. Besides, they seem to be comparable to recent aerosol samples in terms of fossil contribution and a comparison with the biogenic background is thus interesting. However, the high OC value might indeed be untypical and caused by the high variability observed in this fraction.

R. * Page 5912, line 5: for what amount of ice are these process blanks valid? The blank will probably depend on that.

A. This blank is valid for samples of 0.1-1.5 kg. Investigation of the blank showed, that within this range not the amount of ice is the main contributor to carbon mass but rather the procedure of cutting ice, filtration and the filter itself.

R. * Page 5912, line 22: The formula $pMC = fM * 100$ is either wrong, or fM is used in the paper different from this definition. As far as I understand, fM in the paper indicates the ^{14}C content of a sample relative to the ^{14}C content of a purely biogenic sample from the same year. Thus, fM is both normalized to the calibration curve, and corrected for decay. If I'm right, "... and can easily ...Polach (1977)" should be deleted.

A. Fraction of modern carbon (fM) can be transferred into percent of modern carbon

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(pMC) as defined in Stuiver and Polach (1977) using the formula: $pMC = fM * 100$. Accordingly, fM includes correction for the decay (reference year 1950 until the year of measurement in 2004 or 2005) and correction for the chronological age of the sample. Different from this definition, results of fM used in this study additionally include normalization to the IntCal04 Northern Hemisphere radiocarbon calibration curve (Reimer et al., 2004) which considers the fact that the atmospheric $^{14}C/^{12}C$ ratio is varying over time. Hence, fM then directly indicates the ^{14}C content of a sample relative to the ^{14}C content in the living biosphere at the corresponding time. In summary, a sample with $fM = 1$ consisted 100% of contemporary carbon at the time of snow deposition on the glacier, and a sample with $fM = 0$ contained pure fossil carbon. The text will be changed accordingly.

R. * Page 5914, line 11: "EC and OC are not correlated": If I judge this statement from Figure 2 by eye, I think that there is a correlation between EC and OC. If a statistical analysis shows that this impression is not true, the result of the statistical analysis should be given.

A. If excluding the extremely high OC value around 1850 a correlation is found. We will change the text in the manuscript accordingly.

R. * Page 5914, line 12: "... concentration trend obvious for EC.": In this case I see no "obvious" trend in Figure 2. The trend is only detectable considering the following discussion (there, the higher values are attributed to mineral dust events). The word "obvious" should be omitted.

A. We agree and will omit "obvious".

R. * Page 5915, line 6: "A significant contribution of anthropogenic (fossil) sources after around 1870 can be clearly identified in both fractions...": for me, this is only clear for OC in Figure 3. In EC, the small number of points with large uncertainties do not justify the attribute "clearly".

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A. We agree and will omit “clearly”.

R. * Page 5915, line 6: see Abstract, line 16.

A. see above

R. * Page 5916, line 19: Quite a few samples seem affected! The authors should comment how many samples affected and unaffected by Sahara dust exist.

A. The according section will be condensed and changed (see also reply to the comments of reviewer 1) as we realized that the discussion of the dust influence received too much attention. Only three samples in the EC concentration record and 2 samples in the EC 14C record are probably affected by dust, numbers will be given in the text.

R. * Page 5918, line 8: "No correlation between OC_b and EC_b was found. This seems reasonable ..". I do not understand the argument. There should be a partial correlation since biomass burning contributes to both OC_b and EC_b?

A. OC_b reflects gaseous and particulate emissions from vegetation as well as gaseous precursors from biomass burning, whereas EC_b exclusively represents particulate emissions during combustion of biomass. Thus one would expect at the most a weak correlation. We will change the text in this sense.

R. * Page 5918, line 12: What is the "natural background level" and how was it determined?

A. The biogenic background level was defined as the averaged value for the time period before 1800, excluding the 1670 period (probably reflecting forest fire events) and the period 1680-1730 (probably influenced by Saharan dust layers) for OC and EC, respectively. The text will be changed accordingly.

R. * Page 5918, line 16: "The OC_f peak in the 1840s can be assumed as an artefact derived from the very high OC concentration in the according sample." - So you mean this increased OC_f is not real? This would suggest a cross-talk OC_b -> OC_f !? Please

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explain how this can happen (maybe in the methodical section).

A. This sentence will be removed as it seems to be only confusing and not really relevant for the main discussion. Just a short explanation here: If you measure a high OC concentration and for the same sample a biogenic contribution of less than 100% you automatically derive a relatively high absolute value for the OCf concentration.

R. * Page 5919, line 4: I had difficulties to understand this sentence. What I understand is: "For the OC/EC ratio we observe a base line value of 1.5 +/- 0.3 (1 sigma). This value is significantly higher in the 1670s ...". Is this true?

A. Yes, this is right. We will change the sentence as suggested.

R. * Page 5920, line 1: "The decreased consumption of fossil fuels during the 1920s due to the world economic crisis is clearly visible in ECf, ...": Formulated like that, this is in contradiction to the next sentence. Probably you mean: "At the time of decreased consumption of fossil fuels during the 1920s due to the world economic crisis, a decrease is clearly visible in ECf., whereas ...".

A. We agree and changes will be made as suggested.

R. * Table 1: From where is the "natural background" taken?

A. see above

R. * Table 1: See Abstract, line 20: Why do you show especially year 1940?

A. see above

Technical corrections

R. * Abstract, line 4: "in the past": should be more specific to indicate the time range. e.g. "before 19??"

A. Changes will be made as suggested.

R. * Abstract, line 7: The first part of the sentence "The combination of ... CP allows ..."

repeats the statement of the previous sentence. Should be modified e.g. "This allows a distinction and quantification ..."

A. Changes will be made as suggested.

R. * Introduction, line 8: explanatory insert "... negative aerosol forcing (cooling) ..." suggested.

A. Changes will be made as suggested.

R. * Page 5908, line 14: "effect on" ? "response to" ?

A. We choose "effect on" as they have effect on climate.

R. * Page 5910, Footnote: why is this a footnote, and not a Reference? - However, this might be ACP style for yet unpublished manuscripts.

A. Yes

R. * Page 5913, line 12: Replace "Failure" by "The failure rate".

A. Changes will be made as suggested.

R. * Page 5914, line 26: "... concentrations were measured significantly higher ..." means for me that the concentrations were not actually higher, but just measured higher. The authors probably mean "... concentrations measured were significantly higher ..."

A. Changes will be made as suggested.

R. * Page 5922, line 32: paper Lavanchy et al. 1999b is not referenced in the text.

A. This is not correct, it is referenced on page 5908, line 5.

Interactive comment on Atmos. Chem. Phys. Discuss., 6, 5905, 2006.

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