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Interactive comment

Interactive comment on "Firewood residential heating – local versus regional influence on the aerosol burden" *by* Clara Betancourt et al.

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The authors would like to express their thanks and appreciation to the valuable comments from the reviewers. We address below each of the reviewer comments individually.

Anonymous Referee #2 Received and published: 7 January 2021

Summary: The authors reported isotope ratios of the biomass burning tracer levoglucosan with both model simulation and measurement in two sites in Germany. The simulations indicate that the largest part of the sampled aerosol is 1 to 2 days old, and thus originates from local to regional sources. The isotopic ratios of levoglucosan showed high variability in the observation and this reported as a result of different local

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sources instead of aging or transportation. Overall Comments: The article provides new, insightful information regarding how to examine the long range transport and local influence of biomass burning emissions using both simulation and measurements of levoglucosan. The overall completeness of the manuscript is decent and the presentation is clear. I recommend publication if the authors properly address the following comments. Specific Comments: One of the major conclusions here is that a large fraction of biomass burning aerosols were local. However, it was only lightly discussed with several sentences in Line 197- 203 simply as the result of the simulation. An in-depth justification and discussion are needed here since this is fundamental to the manuscript.

Response: the fact that a large fraction of biomass burning aerosols were local was demonstrated not only by FLEXPART simulations but also by the combination between concentration and isotopic ratio observations. Here, the best tool to interpret these measurements is the Keeling plot analysis. We reformulated some sentences in this section to underline the additional information gained by using this approach, now to read: ‘To investigate the chemical stability of levoglucosan for this study, a Keeling plot approach was employed. The test hypothesis was that chemical decay plays here an important role. Accordingly, the Keeling plot describes the mixing of two reservoirs, in this study of fresh - ’isotopically-light’, high-concentrated emissions with aged - ’heavier’, low-concentrated background (Lin, 2013). For this analysis, the measured isotopic ratio was plotted vs. the inverse concentration (Figure 7). A linear regression analysis was carried out. Remarkably, according to the 95% confidence interval analysis, the yielded y-intercept range of -25.3 to -21.4 ‰ theoretically describing the sources, agrees well within error ranges with the published isotopic composition measured in aerosol from the combustion of various C3 plants (Sang et al., 2012). A y-intercept of -23.2$\pm$0.1 ‰ was derived, theoretically representing the mean isotope ratio of fresh emissions. The knowledge gained from the Keeling plot analysis was used to initialize the FLEXPART isotopic runs. Since the information on type of fuels consumed in certain regions is very scarce, a constant source specific d13C0 of -23.2 ‰ was considered to calculate the 13C emissions used in the simulations (Section S5 in the Supporting information). This isotopic ratio is in the range of soft woods (conifers). Since background levoglucosan concentration data were not available, the lowest measured concentration (12.4 ng m-3 at the EIFE station on November 10th, 2015) was considered as constant background value. A corresponding d13C of -24.0$\pm$0.3 ‰ was derived (see Figure 7). The most important outcome was that the slope of the fitted line to the experimental data was found to be slightly negative, contradicting the assumption of an aged heavy background. This analysis thus demonstrates that the initial hypothesis of levoglucosan chemical instability was false. Consequently, the variability in the observed δ-values is likely due to the contribution of local to regional sources that possess different isotopic ratios in the above-mentioned range and not due chemical processing'. (lines 284-301) We've introduced in the conclusion section sentences that emphasize the local origin of the investigated aerosol, now to read: 'Levoglucosan isotope ratios close to emission d13C0 can be either explained by deposition, or by local to regional sources and dispersion. Since dry and wet deposition are insignificant, the latter hypothesis is the most likely. The presented case study shows reasonable agreement between modelled and observed data within error ranges. However, the frequent underestimations, especially at STYR might indicate unidentified sources or flaws in the levoglucosan emission strength. This comparison supports the fact that sources, which are very close to the receptor but not accurately described in the developed emission inventory approach, strongly influence the local aerosol burden, particularly for the STYR site. The Keeling plot analysis proved that an aged background was insignificant for this study, supporting the conclusion that long-range transport minimally impacted the investigated aerosol. This can also explain the few overestimations of the derived concentration at EIFE which might be caused by overestimated background levels rather than underestimated removal (Grythe et al., 2017). Repeated calculations reducing the background and increasing the emissions indicated that in the initial model runs, the source strength is most likely underestimated and the background levels overestimated. The measured δ13C-values show by far higher variability compared with the simulated isotopic ratios. This can be explained by possible individual source-to-source variation (e.g. due to differences in the used fuel). Within the variability of δ13C of emissions, the retro-plume-modelled age of levoglucosan agrees well with the age resulting from the observed isotopic ratios. This agreement demonstrates that the limitation to 7-days backwards calculations does not create any significant bias. Finally, since observations as well as the retro plume analyse show that chemical aging does not play a significant role in the cold season in Central Europe, levoglucosan can be used as a 'conservative' tracer under similar conditions. All these findings demonstrate the FLEXPART fitness to simulate aerosol processes occurring between source and receptor. The sensitivity studies revealed individual factors leading to potential biases, while the comparison between simulated and observed concentration assessed the most probable sources and loss processes for the investigated aerosol. Both sensitivity and case studies unquestionably point out that local/regional domestic heating is the major source contributing to the biomass burning aerosol burden under the investigated conditions.' (lines 346-368)

Similarly, the lack of discussion of the source-specific isotopic composition of levoglucosan also undermines the completeness of the manuscript.

Response: At the moment, there is no accurate information on the used fuels in the domestic heating during the cold season. Therefore, a distinction of the initialized source specific isotope ratios among different categories would make no sense. We decided to use an average d13C0, which we derived from the Keeling plot. Theoretically, the y-intercept of the line fitted to the experimental data gives the isotopic ratio of the source. The negative 'isotopic' age shows yet that this might be heavier than the real mean delta value. We introduced explanatory sentences, now to read: 'A y-intercept of -23.2±0.1 ‰ was derived, theoretically representing the mean isotope ratio of fresh emissions. The knowledge gained from the Keeling plot analysis was used to initialize the FLEXPART isotopic runs. Since the information on type of fuels consumed in certain regions is very scarce, a constant source specific d13C0 of -23.2 ‰ was considered to calculate the 13C emissions used in the simulations (Section S5 in the Supporting information). This isotopic ratio is in the range of soft woods (conifers).' (lines 290-294) and: 'This yields a 'negative' age for the observations (Supporting Information, Table S7.3), probably a consequence of the inaccuracy of the used emission isotope ratio, which is in the upper range of source specific d13C0. Here is to be noted that a four-time shorter levoglucosan lifetime, when using the kOH reported by (Hennigan et al., 2010) would translate in even stronger emissions, characterized by isotopic ratios 2 to 3 ‰ lighter. Then again, these are at the lowermost end of the reported d13C0.' (lines 317-321)

Line 26: " -25.3 to -21.4 ‰Í . These numbers are different from " -26.3 ‰ and -21.3 ‰ in Line 265. Why are they different?

Response: The isotopic ratios in the initially submitted abstract represent the values derived from the linear regression analysis (Keeling plot section). This was not our intention therefore, we've revised the numbers, now to read: 'The high variability in the observed δ13C implies that the local levoglucosan emissions are characterized by very different isotopic ratios in the range of -26.3 to -21.3 ‰.' (lines 29-30)

Line 29: " These findings demonstrate that the aerosol burden from home heating in residential areas is not of remote origin and thus it can be mitigated by reducing local emissions. " I find this statement too general for the scope of the paper.

Response: since we don't further handle that, we've removed the sentences both from the abstract and conclusions.

Line 139: "Since anthropogenic biomass burning aerosol is emitted into the lower mixing layer, in-cloud scavenging is not likely." More justification needed here to eliminate in-cloud scavenging. It is not rare for particles emitted in the lower mixing layer going through in-cloud scavenging.

Response: Certainly, the in-cloud scavenging was considered in the wet-deposition runs. The wet-deposition module by Grythe et al. (2017) delivered the share of below- and in-cloud scavenging in the model output. In-cloud scavenging simply did not occur in this study, even if we set the in-cloud scavenging parameters to maximum (Cloud Condensation Nucleus effectiveness = Ice Nucleus effectiveness = 1.0). We agree that the sentence 'Since anthropogenic biomass burning aerosol is emitted into the lower mixing layer, in-cloud scavenging is not likely' is misleading at this stage of the manuscript. Therefore, we've removed it in the revised manuscript. Instead, we've changed the wet-deposition paragraph, now to read: 'Further, the existent simulations show that wet deposition removed minimal amounts of the emissions. This might be explained by a short exposure of aerosol to weak precipitation of less than 5mm in 6h in the investigated periods. Moreover, there were no in-cloud-scavenging events for the investigated periods. Possible explanations are absence of fog or spreading of emissions in layers lower than the cloud bottom layer height. Similarly to the low impact on concentration, wet deposition had no significant influence on the isotopic composition of the sampled aerosol either.' (lines 227-231)

Line 204: " The simulations thus show that the major part of the sampled aerosol originates from local sources being emitted during the sampling day and the day before." Is this a common way to define local? In figure 2, the two-day-old region are pretty far away from the measurement site.

Response: We agree. The actual goal of the study was to distinguish between local/regional and far-away sources. Therefore, we've replaced 'local' to 'local to regional' for the results throughout the manuscript. We've also changed the title from 'Firewood residential heating – local versus regional influence on the aerosol burden' to 'Firewood residential heating – local versus remote influence on the aerosol burden'

Editorial Comments: Line 258: Please add figure number Response: done (262) Line 264: Please add figure number Response: done (267) Line 268: "rations". Typo Response: done (300) Line 353: "we" Why bold? Response: no reason; changed (368) Figure 7: The legends and the markers don't match. Response: done

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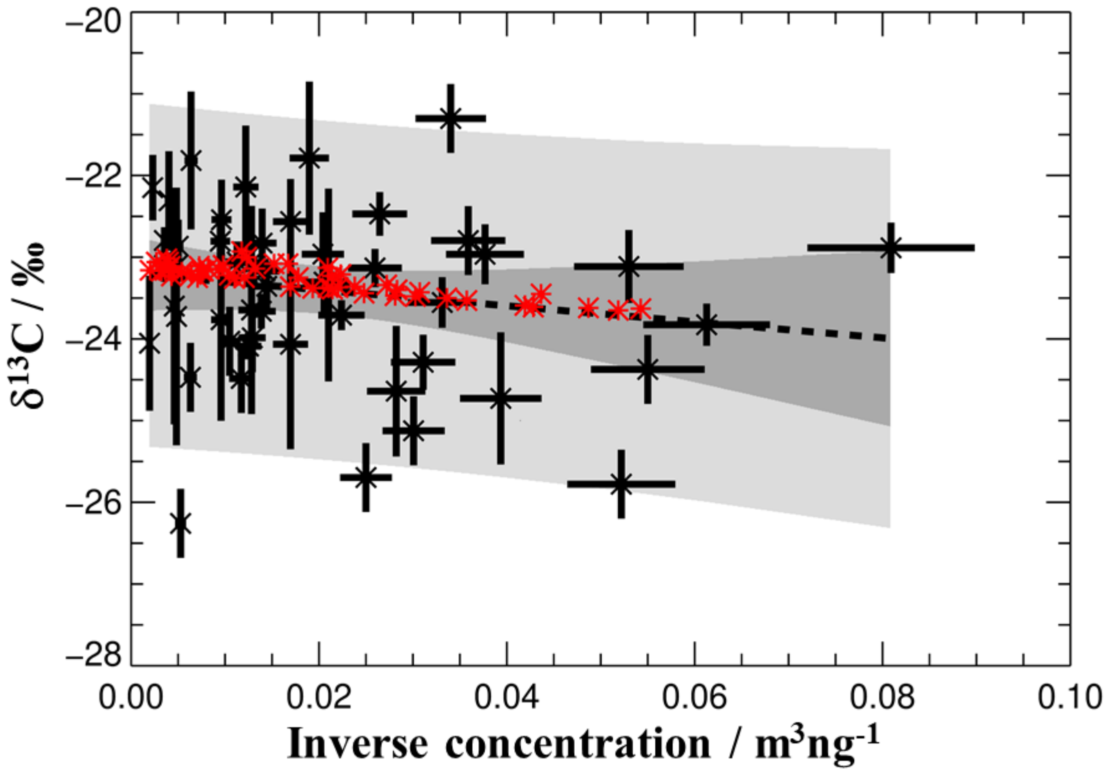

Fig. 1. Figure 7 Keeling plot depicting the observed levoglucosan d13C vs the inverse concentration (black symbols). Model results are given (red symbols) as well as the line fitted to the experimental data (

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