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Atmospheric Chemistry and Physics Discussions

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 #1 Received and published: 19 December 2020

General comments: This manuscript presents the simulated isotope ratios of the biomass burning tracer levoglucosan using Lagrangian Particle Dispersion Model (LPDM) FLEXPART. The authors combine the model results with observed levoglucosan concentrations and d13C to evaluate the sources of residential burning emission. The simulations indicate the aerosol is 1 to 2 days old aerosol, likely from local

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to regional sources. The low OH concentrations in winter likely lead to limited levoglucosan photochemical aging. The isotope analysis shows that the observed d13C is in the range of -25.3 to -21.4 ‰ , which is agreed with previous studies on levoglucosan source specific isotopic composition in biomass burning aerosol. The authors present scientific results well using FLEXPART model and isotope analysis and statistical analysis. The overall quality ofthe manuscript is good, although some areas will need improvement. I recommend that the manuscript be published after making the following revisions.

Specific comments: 1. The authors compared the difference between ECMWF and GFS dataset. However, the meteorological data (e.g., cloud fraction, low cloud cover, precipitation, temperature, wind speed) is not shown and compared with observed levoglucosan. Please provide some details for correlation between meteorological factors and observed levolucosan concentrations.

Response: For the first time, we've introduced isotopic ratios additionally to concentration in a Lagrangian model and compared simulation results to observations, with the concrete goal to differentiate between local and remote sources of biomass burning aerosol. In the preparatory sensitivity studies, we varied some of model parameters, which either are considered as main sources of uncertainties (such as meteorology as a whole), or have a strong impact on the calculated source-receptor relationships (such as emission strength or loss processes). Due to the specific cold season conditions for this study, such as higher atmospheric stability or reduced photo-chemical activity, these variations didn't induce significant differences for the simulation results. Therefore, we didn't see the necessity to deepen our investigations with such detailed correlations. We consider that these are beyond the scope of our study, which was designed to scrutinize sources by using this novel approach, and not for model development. To make this clear in the manuscript, we've introduced the following sentence in the lines 171-173: 'Following the overall goal of this study to separate local from remote sources of firewood domestic heating aerosol, changes in output depending on

selected input parameter modifications were evaluated for consistency.' We agree that isotopes might contribute to test and develop models, but we consider that a summer study would be the right framework for that in a specifically dedicated publication.

2. Line 140: Why the in-cloud scavenging is not considered? Cloud/fog scavenging might occur at lower PBL. Have you checked the cloud bottom layer height? Do you have vertical cloud data with relative humidity? Any vertical profile of in-cloud scavenging, blow-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 (lines 227-231), 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.' A detailed analysis to justify the reason for the negligible impact of scavenging is beyond the scope of this study.

3. Line 140-141: The OH-decay rate constant of levoglucosan, 2.67 10-12 cm3 molecules -1 s-1 is one order of magnitude lower than the 1.1 10 11 cm3 molec-1 s-1 reported by Hennigan et al. (2010). Please explain why you choose 2.67 10-12 cm3 molecules -1 s-1.

Response: The comparison of model results with the observations indicates that use of the kinetic data from the isotopic study by Sang et al. 2016 (also including the kinetic isotope effect of the reaction levoglucosan + OH) is most appropriate to the purpose of this work. For the concentration, the model predominantly underestimates the sources. A four-time reduction of the lifetime would make the underestimation stronger. Conversely, much higher emissions must be considered, to explain the measured levoglucosan. As for the isotopic analyses, the lifetime reduction to two days would translate in lighter emissions by 2 to 3‰ moving to the lowest end of the hard wood source specific delta. An explanatory section is given in the lines 318-321, now to read: '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 ïĄď13C0.'

4. Line 189-190: Please explain more on why higher residence time of model particles can lead to higher derived levoglucosan concentration. Wind speed, turbulence condition, levoglucosan decay rate can also affect levolucosan concentration.

Response: The term 'residence time (t_res)' in the FLEXPART retro plumes designates the receptor sensitivity to a source, which is of course impacted by wind speed and turbulence conditions. To avoid confusion, we reformulated the sentence, now to read: 'For the latter case, higher GFS Hmix are derived. Lower receptor sensitivity to emissions, expressed as tres (Supporting Information, Section S5), is expected due to dilution. The tres from ECMWF runs are smaller though. A possible explanation is that due to shallow mixing layers, a large part of the emitted particles likely leaves the investigated footprint layer and does not contribute to the receptor (Hueser et al., 2017).' (lines 188-192)

5. Line 193: "vertical mixing parameterizations". Please specify which parameter.

Response: For clarification, the sentence was changed (lines 194-198), now to read:

‘Differences in the modelled levoglucosan concentration and isotopic composition are expected, mainly due to the different parameterization of sub-grid processes in the atmospheric boundary layer in the GFS and ERA5 models (Berrisford, 2011;Han, 2011). Mixing influences the concentration of a model tracer that is released in the planetary boundary layer. Yet, during the more stable cold season over Europe, the differences are expected to be small.’

6. Line 194-195: “Due to the higher vertical resolution and ability to more accurately account for topography, ECMWF meteorology was chosen to initialize the model for the future runs.” Did you consider to use WRF meteorological data for comparison?

Response: For this study, we didn’t consider using WRF meteorology. FLEXPARTv10 can be run with ECMWF and GFS only. For the scope of this study, we consider that the presented comparison is sufficient (q.v. response to question 1, RC#1).

7. Line 213-214: “... levoglucosan is relatively stable during winter due to the low OH concentration.” Is this also due to the lower temperature in winter? What’s the average temperature during sampling periods? The reference from Busby et al. (2016) seems does not point out the levoglucosan reactivity in winter is due to the low OH concentration. Please check the reference.

Response: To our knowledge, neither the temperature dependence of (levoglucosan + OH) rate constant, nor parametrizations of kinetic barriers for the heterogeneous reaction at lower temperatures were hitherto reported. The only information we could implement into FLEXPART was the levoglucosan lifetime, derived from kOH and OH average concentration. Nevertheless, while the average OH concentration in the retro plumes is prone to uncertainties, it is well accepted that the photo-chemical activity is reduced in the dark winter months. Therefore, we’ve revised the section (lines 270-272), now to read: ‘Simulations showed that most contributing emissions are only one day old. As a consequence, levoglucosan degradation is not expected to be significant for this study. This assumption is supported by the reduced photo-chemical activity

in the dark winter months (Busby et al., 2016).' Additionally, Table S1.2 was included in the Supporting Information to deliver the average temperature during the sampling periods.

8. Line 255: What are the two major types of sources?

Response: Regional and near-by sources are meant here. The sentence was completed (lines 259-260), now to read: 'According to this, there are two major categories of sources contributing to the sampled aerosol, i.e. regional and close-by sources.'

9. Figure 4: Please explain the possible reason for the two outlier points at STYR site. The standard deviation is also higher than others.

Response: One of our findings was that close-by sources impacted the urban area receptor. Here, weak ventilation due to presence of buildings in combination with inversion ("Street canyon situation") can lead to high pollution events. For levoglucosan concentration measurements, the error propagation analysis resulted in a 11% overall precent error for the measurement set. Therefore, the error bars at higher concentrations are bigger.

Technical corrections: 1.Line 184: Figures 6.1 and 6.2 should be Figures S6.1 and S6.2. Response: done (180) 2.Line 258: Figure " " is Figure 4? Response: done (262) 3.Line 264: Figure " " is Figure 5? Response: done (267) 4.Line 302: Table 8.3 should be Table S8.3. Response: done (312) 5.Line 307: Table 7.3 should be Table S7.3. Response: done (317) 6.Supporting Information line 143, 146, 150, 153, 154, 157, 161, 164, 166, 169, page 9: Some equations and words are missing. Response: equations went lost during pdf document merging. Repaired 7.Supporting Information Table S7.2: and Table S7.3: Some words are missing for the captions. Basic statistics for ": : :: : :". Response: equations went lost during pdf document merging. Repaired 8.Table S7.4: Some words are missing for the captions. Basic statistics for the difference between ": : :" and ": : :". Response: equations went lost during pdf document merging. Repaired 9.Supporting Information P.19 Table S8.3 should be Table S8.4.

Response: done 10.Figure 1: The position of tm /s legend is too close to the latitude “-30”. Please move the legend to a clear region. The unit of tm can be consistent with the figures of height vs. time/h. Response: done 11.Figure 5: The color of light green and dark green is too similar, hard to distinguish it. Response: done

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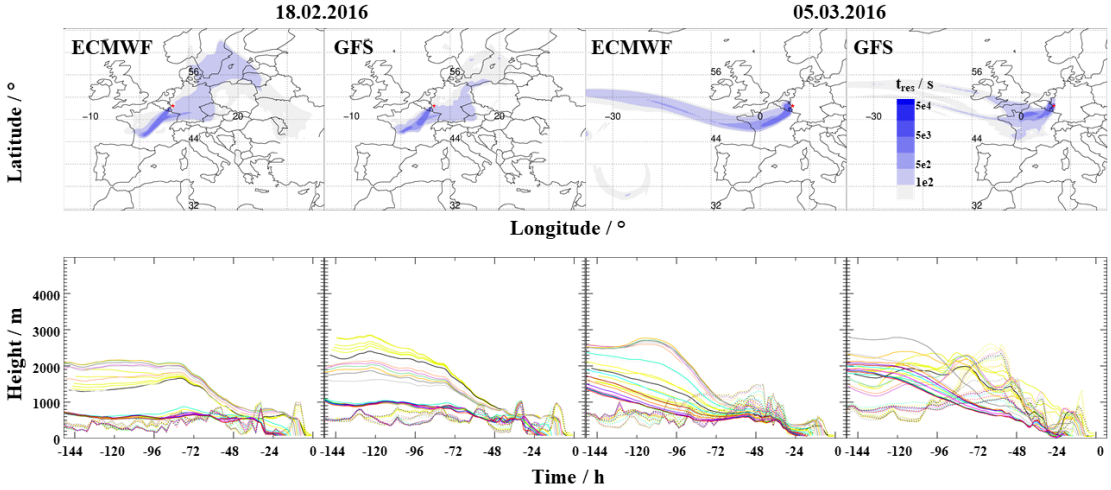

Fig. 1. Simulated retro plumes (top, for the colour code see legend) as well as centroid-back-trajectory (bottom, solid lines) and the corresponding mixing-layer heights (bottom, dashed lines) for ...

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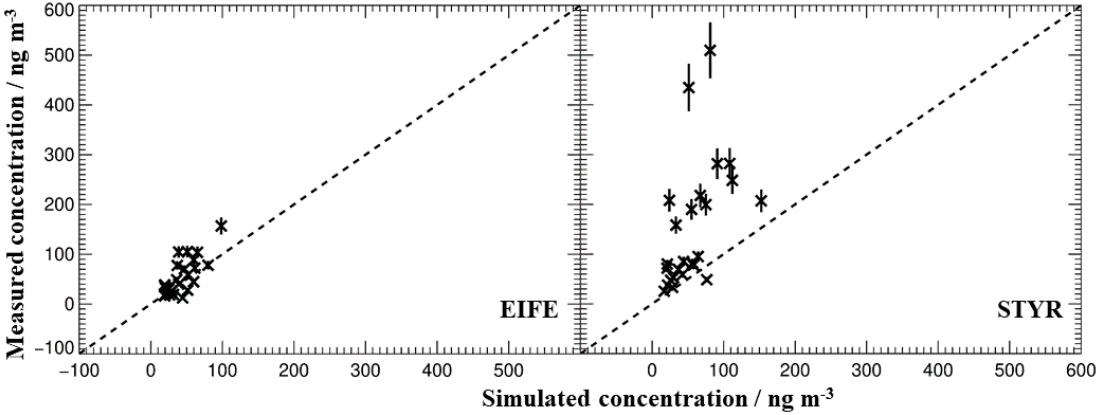

Fig. 2. Comparison between observed and simulated levoglucosan concentration at the EIFE and STYR sites. Also the 1:1 line is given (dashed line).

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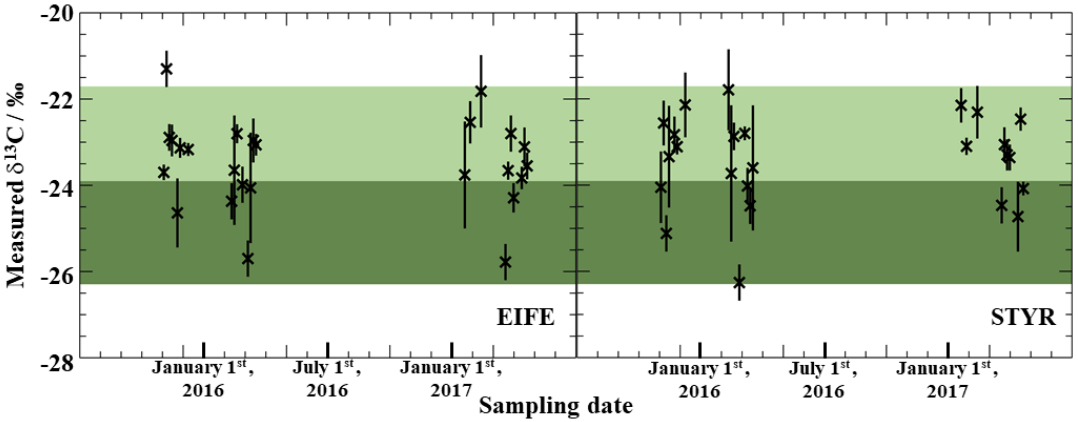

Fig. 3. Figure 5: δ13C of the sampled levoglucosan at EIFE and STYR sites. The shaded areas represent ranges of observed levoglucosan source specific isotope ratios in aerosol formed during the combustion of

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