Firewood residential heating – local versus regional influence on the aerosol burden

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Keywords: levoglucosan isotopic analysis, domestic heating, source apportionment, FLEXPART modelling
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As a particular form of biomass burning (BB), domestic heating with firewood is a major source of fine dust in the cold season. Understanding its impact on air quality requires reliable aerosol source apportionment and assessment of prevailing loss processes. Further, to establish effective mitigation policies, it is necessary to accurately quantify the contribution of local vs. remote sources to the aerosol burden. To this end, source-receptor modelling is employed, such as chemical mass balance or Lagrangian techniques, to calculate concentrations of the BB specific tracer in aerosol, levoglucosan (Fine et al. 2002, Chunmao et al. 2019). In the last decades, it has been shown that combining stable isotope ratios with concentration measurements allows for separating the impact of chemical degradation from changes linked to source strength or atmospheric transport. Based on that, Gensch et al. (2018) developed a numerical approach, comparing stable carbon isotopic ratio and concentration measurements with back trajectory analyses by the Lagrangian particle dispersion model FLEXPART (https://www.flexpart.eu/) to investigate chemical aging processes in BB aerosol.

In the present study, stable carbon isotopes were implemented in the full dispersed output of FLEXPART by explicitly tracking of the levoglucosan fraction containing ^{13}C. Further, sensitivity studies were carried out to examine the simulation responses to the uncertainties of the governing atmospheric processes described in FLEXPART and thus, to determine the model performance for given conditions. Finally, the set of selected modelling routines were applied in a case study with the goal to assess the contribution of local vs. remote sources of biomass burning emissions from residential heating to the particulate matter sampled at two measurement stations of the North Rhine-Westphalia Environmental Agency, LANUV. Thereby, the measured levoglucosan concentration and isotopic composition in 50 selected aerosol samples taken at an urban background station in Mülheim-Styrum and at a rural background station in the Eifel, in the cold seasons of 2015 – 2017 were compared with the model results.

The simulations indicate that the biggest fraction of the sampled aerosol is 1 to 2 days old. Chemical aging, also limited by low mean OH concentrations in the cold season, has thus a minor influence on the observed levoglucosan concentration and δ^{13}C (Fig. 1). The experimental data, interpreted as a two end-member mixing series between low-concentration/isotopically-heavy back-ground and high-concentration/isotopically-light fresh emissions, support the model outcome, showing similar isotopic ratios for the two constituents. The high variability in the observed δ^{13}C implies that the local levoglucosan emissions are characterized by very different isotopic ratios in the range of -25.3 to -21.4 % (Fig. 1). These values are in good agreement with previous studies on levoglucosan source specific isotopic composition in BB aerosol (Sang et al. 2012).

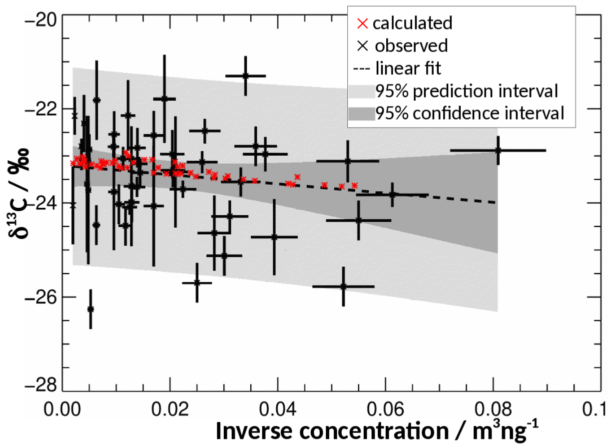

Figure 1: Keeling plot (for details, see text)

These findings demonstrate that the aerosol burden from residential heating in living areas is of local origin and thus, mitigation is possible through reduction of local emissions. In this work we show that combining Lagrangian modelling with isotope ratios is valuable to obtain additional insight in source apportionment. There is, though, a need for a better isotopic description of sources. Moreover, studies investigating long range transport of BB aerosol from large-scale fires in the dry season are essential to examine the role of aging among other loss processes.

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