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Stable carbon isotopes - an indicator for heterogeneous aging of organic aerosol?

Ulrike Dusek, Rupert Holzinger, and Thomas Röckmann

Utrecht University, IInstitute for Marine and Atmospheric research Utrecht (IMAU), Utrecht, Netherlands (u.dusek@uu.nl)

Organic aerosol (OA) sources that derive from photosynthesis (such as biomass or fossil fuel combustion) are usually depleted in ^{13}C. Oxidative processing (aging) of the organic aerosol can cause enrichment in aerosol ^{13}C, if a significant amount of the oxidized compounds evaporates from the aerosol.

We expose a series of aerosol samples from Ghent, Belgium to different temperatures in an oven. We measure δ^{13}C values and detailed organic chemistry on sub-fractions of OA that are thermally desorbed at several 50 $^{\circ}$C temperature steps ranging from 50 to 200 $^{\circ}$C. For carbon isotope analysis the compounds released at each temperature step are oxidized to CO_2 using a platinum catalyst at 550 $^{\circ}$C. The CO_2 is then passed on to an isotope ratio mass spectrometer (IRMS) to measure δ^{13}C ratios. A part of the flow is diverted to an aerosol Proton-Transfer-Reaction Mass Spectrometer (PT-RMS). This instrument is able to resolve low volatility and highly oxygenated compounds that are virtually inaccessible to other chemical classification. Here, we use the detailed chemical information to derive O/C ratios for all organic sub-fractions released at different temperatures.

Both δ^{13}C values and O/C ratios increase with increasing oven temperature. Hence, less volatile compounds that are released at higher temperatures contain more O and are enriched in ^{13}C compared to compounds released at lower temperatures. The increase of O/C ratios with oven temperature is plausible, since the addition of an O containing functional group to an organic molecule drastically decreases its vapour pressure. Interestingly, these more oxidized compounds also show higher δ^{13}C values, as could be expected from heterogeneous aging processes. These should increase both the oxygen and the ^{13}C content of the organic fraction. This hypothesis is further substantiated by a strong correlation of the ^{13}C enrichment with the change of O/C ratios between 100 and 150 $^{\circ}$C. At higher T this correlation does not exist, which might indicate sources other than heterogeneous oxidation for the more highly oxygenated compounds.