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14C-based Source Apportionment of Carbonaceous Aerosols in Switzerland for 2008 – 2012

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Carbonaceous particles (total carbon, TC) are a major fraction of the fine aerosol and affect climate and human health. TC is classified into the sub-fractions elemental carbon (EC) and organic carbon (OC). EC originates only from fossil fuel combustion and biomass burning. OC can be emitted directly as primary organic aerosol from biogenic emissions, wood burning and fossil fuel combustion or can be formed in-situ in the atmosphere (secondary organic aerosol) (Szidat et al. 2006). Radiocarbon (14C) analysis is a direct and quantitative tool for distinguishing fossil and non-fossil sources, since 14C in fossil fuels is completely depleted whereas other sources have a contemporary 14C level.

This study presents source apportionment results from the winter season over a time period of 5 years (2007/2008–2011/2012) using 14C measurements on aerosol filters collected simultaneously at 16 air quality monitoring stations across Switzerland. For every year 5 winter smog episode days were selected from which filters from all stations were analyzed. To resolve a good spatial variability 11 stations north and 5 stations south of the Alps were selected. This 14C data set is unique around the world concerning the number of analyzed filters and the duration.

The filter sampling was conducted using high volume samplers with PM10 inlets and a time resolution of 24h. Separation of OC and EC was carried out using the THEODORE system (Szidat et al. 2004) and a Sunset EC/OC analyzer (Zhang et al. 2012), respectively. The resulting CO_2 was cryo-trapped and sealed in glass ampoules for 14C measurements, performed with the Mini Carbon Dating System MICADAS (Ruff et al. 2007) at the Swiss Federal Institute of Technology (ETH) Zürich.

The results for non-fossil (NF) OC (5 year average) are 81% $\pm$ 10% for north and 85% $\pm$ 8% for south of the Alps. ECNF values range from 31% to 53% north and from 36% to 66% south of the Alps. Both, the OCNF and ECNF show higher values south of the Alps. The highest values were found in alpine valleys with OCNF of max. 100% and ECNF of max. 87%. The station-to-station variation north of the Alps is low, whereas in the south a spatial trend was found with an increase of the non-fossil values towards the north showing the influence of more fossil air masses advected from the Po-valley. No real time trend over the 5 winters was found. The high ECNF and OCNF values together with a good correlation with levoglucosan show that wood burning is the major source of TC in Switzerland during winter smog episodes.

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