Multi-Year Source Apportionment of the Central European Organic Aerosol During Wintertime

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COLOSSAL Chemical On-Line cOmpoSition and Source Apportionment of fine aerosoL

ACTRIS

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Motivations

Since 2012, an Aerosol Chemical Speciation Monitor (ACSM, Aerodyne Inc, Ng et al., 2011) has been deployed at the Central European Research Station Melpitz (Germany) as part of the European ACSM network developed within the ACTRIS-Project (European Research Infrastructure for the observation of Aerosol, Clouds and Trace gases, www.actris.eu). This long-term measurements represents a unique chance to identify and quantify the impact of the air quality regulations on the aerosol chemical composition.

The present work is focused on the organic aerosol (OA) source apportionment analysis over 5 consecutive winters (December, January, and February).

Melpitz

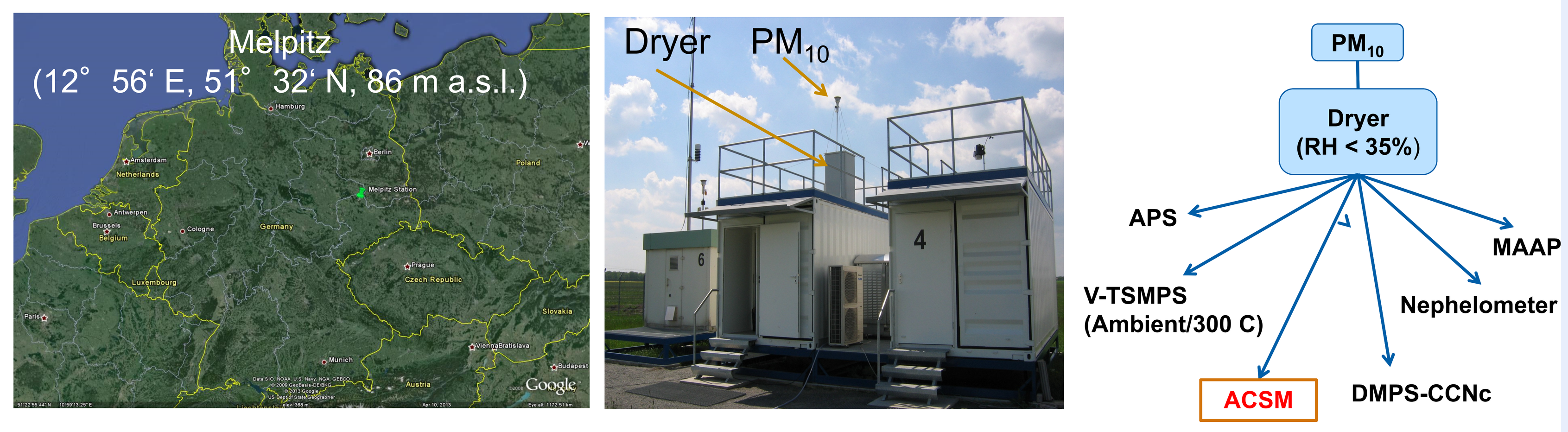

Fig. 1: Localization of the Research Station Melpitz (left), picture of the laboratory container and the sampling inlet (middle), distribution of the on-line instrumentation inside the container (right).

Source apportionment analysis approach

Source apportionment was performed using the Multi-linear Engine (ME-2, Paatero 1999) and the Source Finder SoFi-Pro (Cananaco et al., 2013). The model was run for 5-factors solution (Fig. 2) applying the rolling windows approached after constraining the mass spectra of the following factors:

- Hydrocarbon-like OA (HOA) identified at Melpitz (Crippa et al., 2014)
- Biomass Burning OA (BBOA) identified at Melpitz (Crippa et al., 2014)
- Coal Combustion OA (CCOA) using the reference from Lin et al., 2017.

Trajectory analysis was made based on 96 h backward trajectories for the altitude of 500 m above model ground with HYSPLIT-4 and analyzed using Zefir 3.7 (Petit et al., 2017) for the identification of potential aerosol sources using Concentration-weighted Trajectory (CWT).
Factor identification was made by combining external measurements (Fig. 3 - 4) and trajectory analysis (Fig. 5).

Summary

- Anthropogenic emissions represent close to 30 % of the total OA mass concentration. Anthropogenic emissions are dominated by biomass burning (13 %). Coal combustion represents 10 % of OA and liquid fuel combustion 6 %.
- OOA-1 was associated with SOA formation. It is the only factor that shows an increase during day (Fig. 4). The CWT map indicates that it might be representative of the European background aerosol.
- OOA-2 was associated with long-range transport of processed anthropogenic OA based on its correlation with eBC, levoglucosan. This factor also shows similar special distribution than BBOA and CCOA.
- The general low level of the PBL (< 500 m for 70 % of the time) indicates a significant influence of local/regional OA emissions during winter.

Results

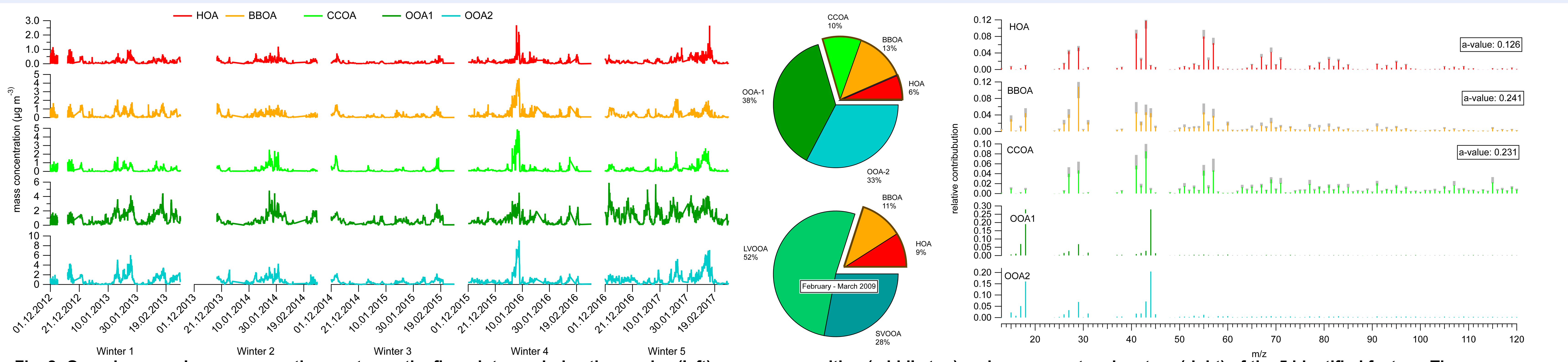

Fig. 2: Organic aerosol source apportionment over the five winter periods : time series (left), mean composition (middle top) and mass spectra signature (right) of the 5 identified factors. The mean composition is compared with previous measurements from Crippa et al. (2014) (middle bottom).

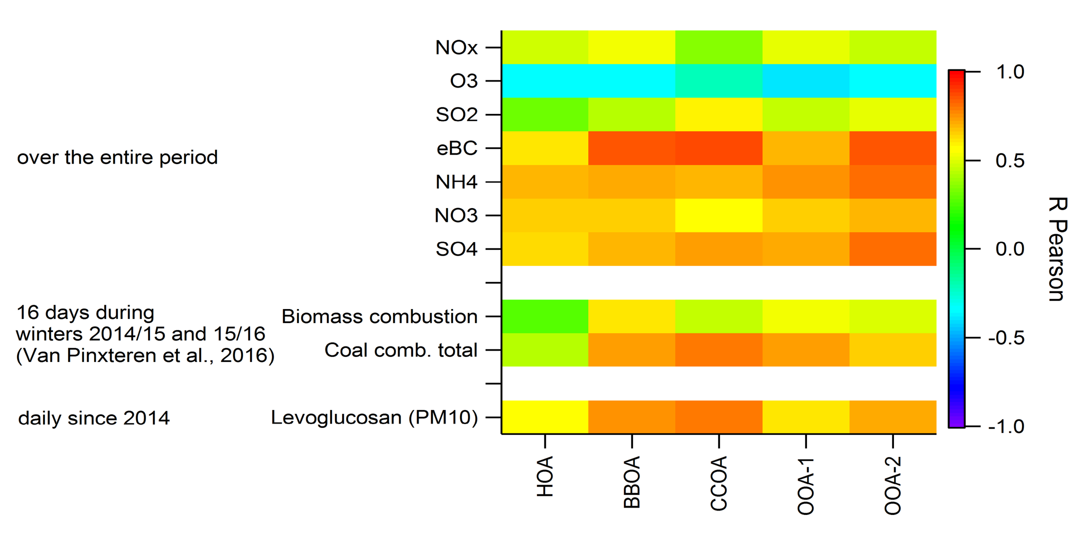

Fig. 3: Correlation matrix between the identified factors and external references.

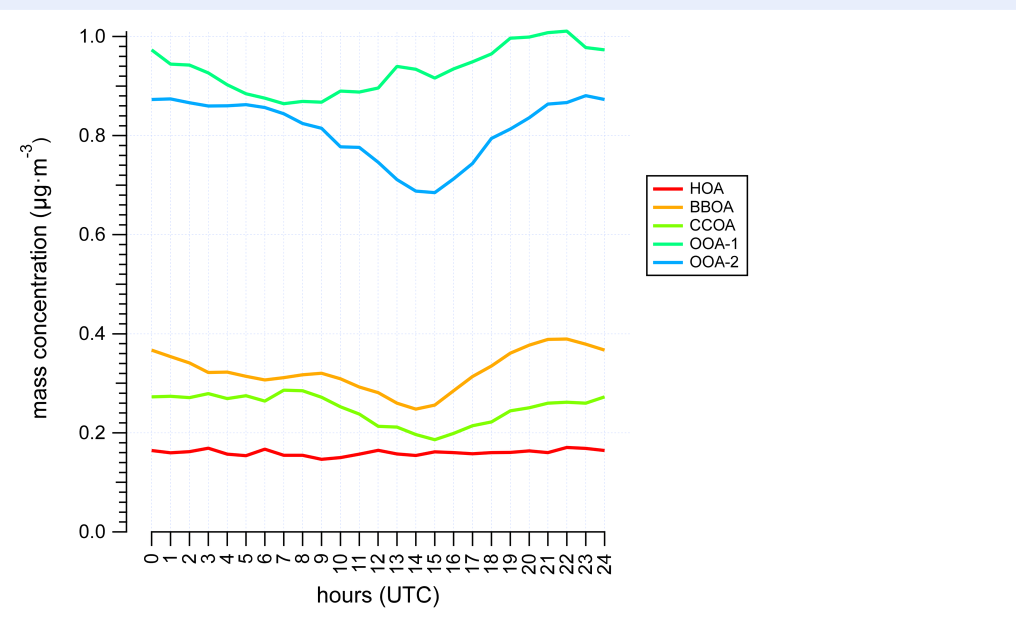

Fig. 4: Mean diurnal pattern of the identified factors

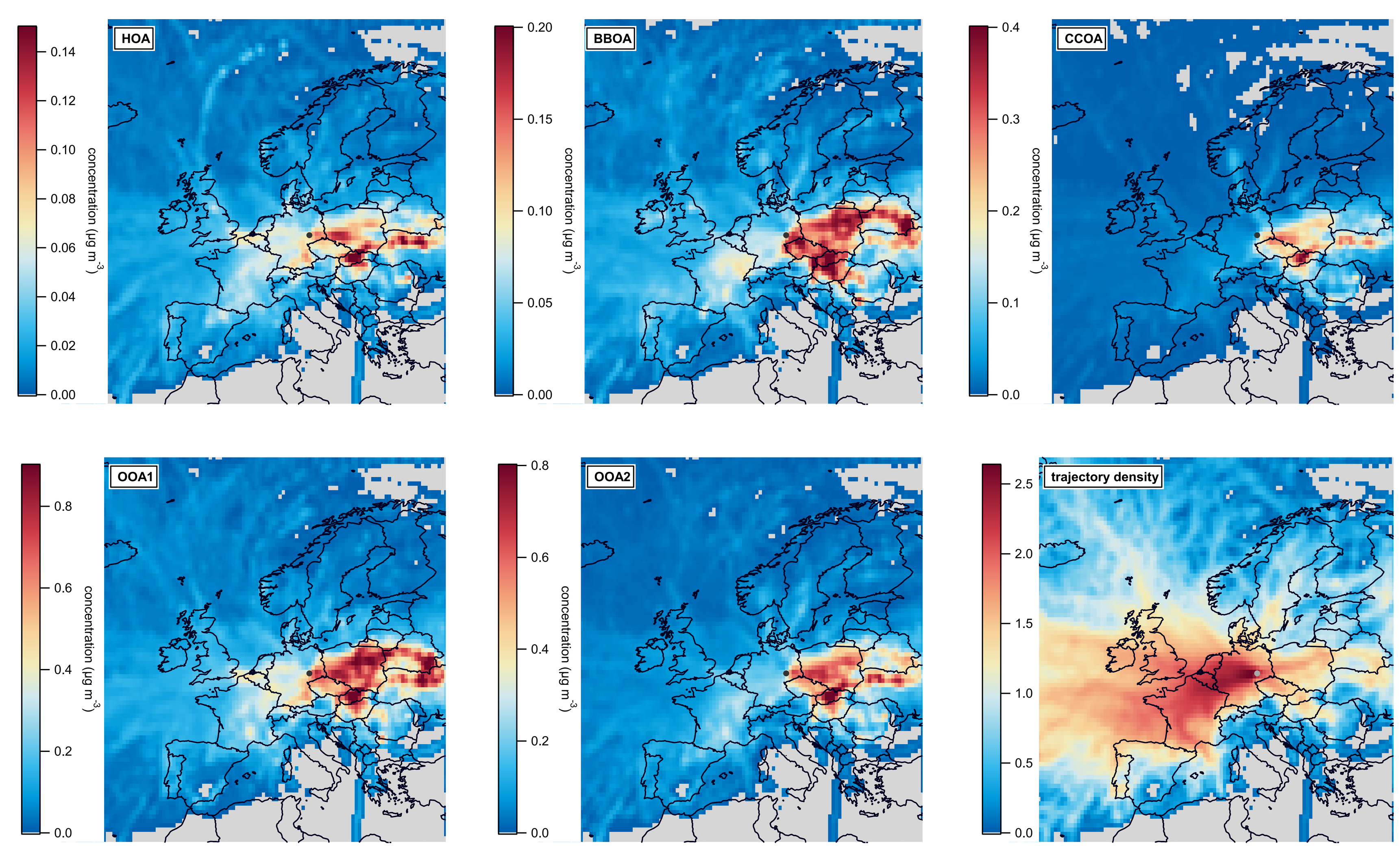

Fig. 5: CWT analysis on the identified factor time series based on the 96 h Back trajectories from HYSPLIT. The following meteorological conditions as available in HYSPLIT were considered to cut off the trajectories : PBL above 500 m (70 % of the trajectories), precipitation rate of > 1 mm h^{-1} (11 %), and an altitude < 3000 m (5 %). The trajectory density map represents the occurrence of back trajectory endpoints falling into a particular cell (log (τ + 1), no unit).

References: **Canonaco, F.** et al. Atmos. Meas. Tech., 6, 3649-3661, 2013; **Crippa, M.** et al. Atmos. Chem. Phys., 14, 6159-6176, 2014; **Lin, C.** et al., *Environ. Sci. Technol.,* 51,18,10624-10632, 2017; **Ng., N.L.** et al., Environ. Sci. Technol., 45, 910-916, 2011; **Paatero, P.** J Comput. Grah. Stat., 8, 854-888, 1999, **Petit, J.-E.**, et al., Environ. Modell. Softw., 88, 183-187,2017;**van Pinxteren, D.** et al.,Faraday Discuss., 189, 291-315, 2016.

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