Source apportionment of carbonaceous aerosol in urban environments through single particle mass spectrometry

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Background & Aims

- Single particle mass spectrometers are well suited to assigning particles to their respective sources in urban environments
- However, attempting to quantify and therefore apportion chemical species at the single particle level is much more challenging.

The **aims** of this study were to:

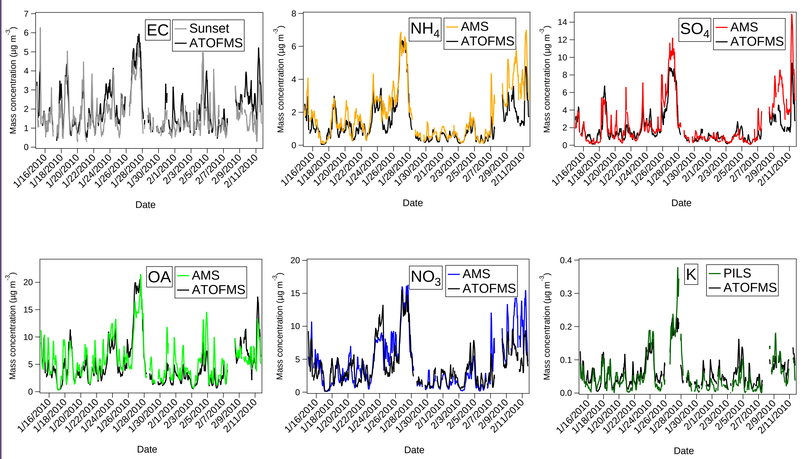
(i) Develop a methodology for **estimating mass concentrations** of organic aerosol (OA), elemental carbon (EC) and inorganic ions at the **single particle level**

(ii) Use this information to perform **source apportionment** at the single particle level for each chemical species and PM_1 mass in Paris, France

Methods

1. ATOFMS particle number concentrations were **scaled** using simultaneous tandem differential mobility sizer (TDMPS) measurements.

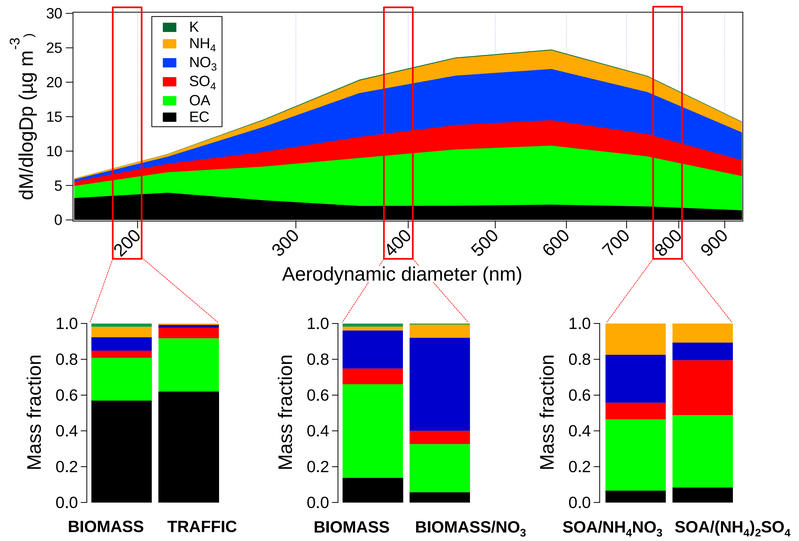
2. Arbitrary **relative sensitivity factors** (RSF) were calculated for each chemical species by comparing the relative peak area (RPA) of chosen marker ions in the total average ATOFMS mass spectrum of all particles with mean mass concentrations from external measurements:



ATOFMS reconstructed mass for each species vs concurrent measurements

3. ATOFMS mass spectra were clustered (*K*-means) to produce **10 carbonaceous classes**. The average mass spectra of each class was then queried to estimate the **mass fraction** of each chemical species in each class using the relevant RPA and RSF values.

Quantitative Mixing State Results

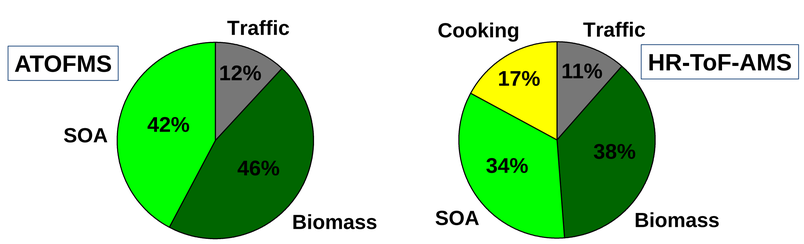


ATOFMS-derived size-resolved bulk composition of Paris aerosol and dominant single particle mixing states

- Quantification approach enabled the estimation of the **chemical composition** of the single particle classes.
- The mixing state of carbonaceous particles in Paris was found to be heterogeneous.
- EC-rich fresh combustion particles associated with **local traffic** and **biomass burning** dominate the smallest sizes. **Aged regional SOA** particles dominate the larger sizes

Organic Aerosol Apportionment

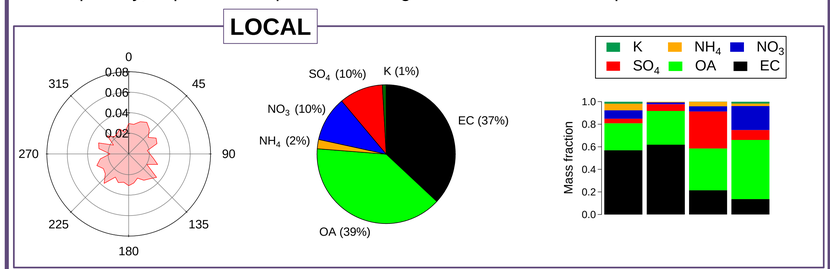
- ATOFMS OA mass fractions for each particle class were assigned either to **traffic, biomass or SOA** and compared with AMS OA PMF analysis[1].
- Good agreement observed, but **cooking** OA was **not detected** by the ATOFMS.

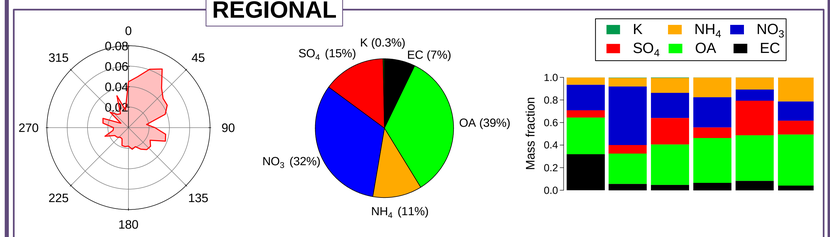


OA apportionment using ATOFMS (left) and HR-ToF-AMS data (right)

Local vs Regional Contributions

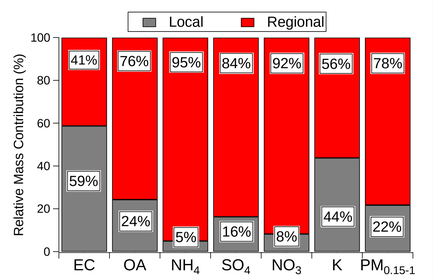
- Particle classes were grouped into two categories, **local** and **regional**, based upon their temporality, dependence upon meteorological conditions and composition.





Bulk composition (left) particle mixing states (middle) and wind dependence (right) for local and regional particles detected in Paris

- **Local** emissions contribute most of the **EC** mass
- **Regional** emissions contribute most of the **OA** and i**norganic ion** mass.
- Only **22%** of PM_1 is associated with local activities, highlighting the impact of **transboundary** emissions in Paris
- This new approach to single particle apportionment is directly applicable to other urban environments.



Local vs regional contributions

Acknowledgements

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[1]Crippa et al., Atmos. Chem. Phys. **13**, 961–981, 2013