Comparison of collocated rBC and EC mass concentration measurements during field campaigns at several European sites

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Introduction: Why is BC mass concentration important?

→ BC direct radiative forcing (DRF) is particularly uncertain

\[ \text{MAC}_{BC}(\lambda) = \frac{b_{\text{abs},BC}(\lambda)}{m_{BC}} \]

IPCC Fifth Assessment Report (AR5) reported BC DRF with a medium to low level of scientific understanding.

→ Uncertainties in \(m_{BC}\)
→ Uncertainties in \(\text{MAC}_{BC}(\lambda)\)
→ Uncertainties in DRF
→ Uncertainties in climate predictions
**Black Carbon mass concentration**

- **Strong absorber of short- and long-wave radiation**
- **Composed primarily of graphene-like sp2-bonded carbon**
- **Insolubility in water**

A unique definition does not exist! ... BC mass is typically defined operationally...

| Method/Technique                        | Quantity*                  | Instruments                                      |
|-----------------------------------------|----------------------------|--------------------------------------------------|
| Thermal Optical Analysis (evolved carbon method) | Elemental carbon (EC mass) | Sunset thermal-optical OC-EC analyzer            |
| Laser-Induced Incandescence             | Refractory black carbon (rBC mass) | Single Particle Soot Photometer ARTIUM LII-300 |
| Absorption Photometers                  | Equivalent black carbon (eBC mass) | Filter based: Aethalometer, MAAP, PSAP, COSMOS, MWAA, ... Photoacoustic: PAS |

* Petzold et al., 2013, Recommandation for reporting “black carbon” measurements
Thermal Optical Analysis and the Sunset EC/OC analyzer

Analysis based on the different evolution characteristics of EC and organic carbon (OC) as a function of T and type of atmosphere.

Thermo-optical transmittance method (TOT)

Possible Biases:
Pyrolysis, Brown carbon, Inorganics
Different thermal protocols (EUSAAR-2, NIOSH, IMPROVE)

Laser Induced Incandescence Technique and Single Particle Soot Photometer (SP2)

Laser-induced incandescence (LII): measurement of the black body thermal radiation emitted by BC particles when heated up by a laser (SP2: continuous-wave laser $\lambda = 1064$ nm).

Information at single particle level!

- **rBC mass concentration**
- **rBC mass size distribution**

Mixing state
(quantitative: LEO-fit, qualitative: delay-time method)

Thinly or moderate coated
Thickly coated

Possible biases:
Detection range $D_{rBC} = [80 - 800]$ nm
Calibration material interference with some refractory and light-absorbing materials

Chow et al., 1993
Birch and Cary, 1996

Stephens et al., Appl. Optics, 2003
Gao et al., Aerosol Sci. Technol., 2007
Moteki and Kondo, J. Aerosol Sci., 2008
Existing literature

Other works:
- Corbin et al., 2019: $rBC = 1.04$ EC
- Laborde et al., 2012: $rBC = 1.10$ EC
- Sharma et al., 2017: $rBC = (0.66 \pm 0.04)$ EC
- Zhang et al., 2016: $rBC = (0.70 \pm 0.02)$ EC
- Miyakawa et al., 2016: $rBC = (1.08 \pm 0.03)$ EC

Lab experiments

Field Campaigns
**Aim of the study**

Open question: Intercomparison between LII and TOA techniques

**How to reach the aim/methods:**

We compared co-located measurements of EC and rBC mass concentrations from field campaigns performed at several European sites.

**Harmonization of the applied methods:**

- **TOA analysis:**
  - EUSAAR-2 protocol and transmittance correction
- **SP2 calibration:**
  - fullerene soot

**Comparison with previous literature studies.**

* Laborde et al., 2013 Black carbon physical properties and mixing state in the European megacity Paris

**Sites of the study:**

Cabauw, Melpitz, Paris SIRTA, Bologna
Results: SP2 missing rBC mass correction

\[ m_{rBC,corr}^{\text{extrap}} = m_{rBC,meas} + \Delta m_{rBC,<\text{LDL}} \]

\[ \Delta m_{rBC,<\text{LDL}} = 3\text{–}25\% \]

The presence of an additional mode of small particles below the lower detection limit of the SP2 can not be excluded.
Results: Time-resolved intercomparison – this work

\[ \frac{m_{rBC}}{m_{EC}} = 0.92 \]
\[ \text{GSD} = 1.5 \]

\[ \text{min } \frac{m_{rBC}}{m_{EC}} = 0.53 \]
\[ \text{max } \frac{m_{rBC}}{m_{EC}} = 1.29 \]
Results: Discussion of level of agreement/disagreement between the rBC and EC mass concentration measurements

1) m_{EC} ≈ 30-40\% between 1 and 2.5 \, \mu m

Upper cut–off related differences contribute to the discrepancies between measured rBC and EC mass, in particular in Melpitz winter and summer campaigns.

2) the SP2 BC particle cut–off is likely between PM_{1} and PM_{2.5}
Results: Systematic EC and rBC bias due to the presence of particular types of aerosols

The variation of BC sources implied by AAE variability may contribute to $m_{EC} - m_{rBC}$ discrepancy.

Increase in the relative difference between $m_{EC} - m_{rBC}$ with increasing AAE

$AAE(\lambda_1, \lambda_2) \approx 1$

Light absorption of traffic emissions is dominated by BC

Traffic-dominated samples potentially contain a greater fraction of small BC particles that are potentially below the LDL of the SP2

$m_{rBC} < m_{EC_{-PM2.5}}$

Possible source effect as cut-off effect
Results:
Time-resolved intercomparison and comparison with other studies

The TOA and the SP2 techniques both provide a consistent measurement of BC mass within the uncertainties of either technique.
Conclusions

The geometric mean of the ratio $m_{rBC}/m_{EC\_PM2.5}$ of all data points from all campaigns is 0.92, with a geometric standard deviation of 1.50.

However, this ratio differed systematically the campaigns with geometric mean values ranging from 0.53 to 1.29.

The main reason of the discrepancies between $m_{EC\_PM2.5}$ and $m_{rBC}$, was found to be the upper size limit of the SP2 and 2.5 µm as well as the possibility of the presence of a BC mode under the SP2 detection limit.

TOA and LII methods quantify the same BC mass, whereas systematic differences in measured absolute values by up to a factor of 2 can occur.

For future $m_{rBC}$ and $m_{EC}$ comparison works:
- same well-defined size range (e.g. PM1)
- same line (tubing length and dryer presence)
Thank you for your kind attention!