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

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

→ Uncertainties in $m_{BC}$
→ Uncertainties in $MAC_{BC}(\lambda)$
→ Uncertainties in DRF
→ Uncertainties in climate predictions

$m_{BC}$ → DRF

$MAC_{BC}(\lambda) = \frac{b_{abs,BC}(\lambda)}{m_{BC}}$

Climate model:

Optical model: mass absorption cross section ($MAC_{BC}$)
Black Carbon mass concentration

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

Refractory
Insolubility in water

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

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

* Petzold et al., 2013, Reccomandation 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.

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

Thermo-optical transmittance method (TOT)

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 $D_{rBC} = [80 \text{ to } 800] \text{ nm}$
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

Lab experiments

Field Campaigns

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

Sites of the study:
Cabauw, Melpitz, Paris SIRTA, Bologna

* Laborde et al., 2013 Black carbon physical properties and mixing state in the European megacity Paris
Results: SP2 missing rBC mass correction

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

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

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

\[ \frac{m_{rBC}}{m_{EC}} = 0.92 \]

GSD = 1.5

\[ \min \frac{m_{rBC}}{m_{EC}} = 0.53 \]

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

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

2) the SP2 BC particle cut–off is likely between PM\(_1\) and PM\(_{2.5}\)

Upper cut–off related differences contribute to the discrepancies between measured rBC and EC mass, in particular in Melpitz winter and summer campaigns.
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 $\frac{m_{rBC}}{m_{EC_{PM2.5}}}$ 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 $\mu$m 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!