Emission, source identification and evolution of black carbon in China

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Emission inventory of BC in China

Bond et al., *Global biogeochemical cycles* 2007; Wang et al., *EST* 2012
Why focusing on BC particle?

BC is the light-absorbing aerosol particle, identified as the second-most important climate forcing agent after CO$_2$, and as one of the most important so-called short-lived climate forcers.

- BC radiative forcing: +0.64 W/m² (IPCC, 2013)
- Uncertainty source: Emission, life time, mass absorption cross-section (MAC), absorption forcing efficiency (Bond et al., 2013)
Why focusing on BC particle?

BC plays the key role in modifying the PBL meteorology and hence enhancing the haze pollution.

Ding et al., *GRL* 2016
High carbonaceous aerosols in PM$_{2.5}$

Beijing 1999-2008

Comparison with other cities

- High OC and EC concentrations in Beijing compared with other cities
- High OC/EC ratio indicates the large fraction of SOC

It is necessary to investigate the sources and formation of organic aerosols

Min Hu and Song Guo, *IGAC Newsletter*, 2010
PM$_{2.5}$ and its major chemical speciation at different locations in China

Jing Zheng, Min Hu* et al, *Chemosphere*, 2016, 159, 480-487
Comparison of PM$_{2.5}$ and major chemical species for urban and regional sites

The size of each cycle is proportional to correlation values between the temporal trends of each species.

EC, stronger local contribution urban reached 222% higher than regional sites.
For most chemical species, urban similar to regional sites, uniform regional pollution.

Jing Zheng, Min Hu* et al, *Chemosphere*, 2016, 159, 480-487
Work done by PKU group

- Emission factor and characteristics of BC
**Fine Particle Emissions from On-Road Vehicles in the Zhujiang Tunnel, China**

**TABLE 1. Average Speciated PM$_{2.5}$ Emission Factors (mg vehicle$^{-1}$ km$^{-1}$) in Zhujiang Tunnel and Comparisons with Other Tunnel Studies**

<table>
<thead>
<tr>
<th>species</th>
<th>Zhujiang Tunnel*</th>
<th>Sepulveda Tunnel$^b$</th>
<th>Kilborn Tunnel$^c$</th>
<th>Howell Tunnel$^c$</th>
</tr>
</thead>
<tbody>
<tr>
<td>mass</td>
<td>110 (4)</td>
<td>52</td>
<td>24.1</td>
<td>39.3</td>
</tr>
<tr>
<td>EC</td>
<td>49.6 (1.9)</td>
<td>25.50</td>
<td>6.87</td>
<td>10.8</td>
</tr>
<tr>
<td>OC</td>
<td>24.3 (0.93)</td>
<td>19.27</td>
<td>6.41</td>
<td>12.9</td>
</tr>
<tr>
<td>SO$_4^{2-}$</td>
<td>3.87 (0.61)</td>
<td>1.77</td>
<td></td>
<td></td>
</tr>
<tr>
<td>NO$_3^-$</td>
<td>1.37 (0.59)</td>
<td>3.27</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Cl$^-$</td>
<td>0.98 (0.16)</td>
<td>0.67</td>
<td></td>
<td></td>
</tr>
<tr>
<td>NH$_4^+$</td>
<td>0.80 (0.25)</td>
<td>1.61</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Na (23)</td>
<td>0.37 (0.07)</td>
<td>0.30</td>
<td>0.98</td>
<td>0.038</td>
</tr>
<tr>
<td>Mg (24)</td>
<td>0.22 (0.02)</td>
<td>0.26</td>
<td>0.077</td>
<td>0.044</td>
</tr>
<tr>
<td>K (39)</td>
<td>0.14 (0.04)</td>
<td>0.08</td>
<td>0.13</td>
<td>0.065</td>
</tr>
<tr>
<td>Ca (44)</td>
<td>0.64 (0.09)</td>
<td>0.30</td>
<td>0.31</td>
<td>0.43</td>
</tr>
<tr>
<td>Ti (47)</td>
<td>0.027 (0.004)</td>
<td>0.09</td>
<td>0.010</td>
<td>0.19</td>
</tr>
<tr>
<td>V (51)</td>
<td>0.0015 (0.0008)</td>
<td>0.05</td>
<td>0.00068</td>
<td>0.00040</td>
</tr>
<tr>
<td>Cr (52)</td>
<td>0.0054 (0.0004)</td>
<td>0.02</td>
<td>0.012</td>
<td>0.0018</td>
</tr>
<tr>
<td>Mn (55)</td>
<td>0.019 (0.001)</td>
<td>0.02</td>
<td>0.0058</td>
<td>0.0060</td>
</tr>
<tr>
<td>Fe (57)</td>
<td>1.12 (0.09)</td>
<td>2.79</td>
<td>0.51</td>
<td>0.55</td>
</tr>
<tr>
<td>Co (59)</td>
<td>0.00013 (0.00004)</td>
<td>0.00</td>
<td>0.00</td>
<td>0.0004</td>
</tr>
<tr>
<td>Ni (60)</td>
<td>0.0034 (0.0014)</td>
<td>0.01</td>
<td>0.00</td>
<td>0.0009</td>
</tr>
<tr>
<td>Cu (63)</td>
<td>0.034 (0.002)</td>
<td>0.17</td>
<td>0.014</td>
<td>0.012</td>
</tr>
<tr>
<td>Zn (66)</td>
<td>0.078 (0.010)</td>
<td>0.14</td>
<td>0.023</td>
<td>0.028</td>
</tr>
<tr>
<td>As (75)</td>
<td>0.0020 (0.0006)</td>
<td>0.00</td>
<td>0.0021</td>
<td>0.00045</td>
</tr>
<tr>
<td>Mo (98)</td>
<td>0.0014 (0.0001)</td>
<td>0.01</td>
<td>0.0020</td>
<td>0.0013</td>
</tr>
<tr>
<td>Cd (114)</td>
<td>0.00049 (0.00022)</td>
<td>0.02</td>
<td>0.0004</td>
<td>0.00005</td>
</tr>
<tr>
<td>TI (205)</td>
<td>0.00004 (0.00002)</td>
<td>0.00</td>
<td>0.00002</td>
<td>0.00001</td>
</tr>
<tr>
<td>Pb (208)</td>
<td>0.014 (0.003)</td>
<td>0.03</td>
<td>0.0031</td>
<td>0.0010</td>
</tr>
<tr>
<td>U (238)</td>
<td>0.00004 (0.00001)</td>
<td>0.00</td>
<td>0.00001</td>
<td>0.00</td>
</tr>
</tbody>
</table>

$^a$ Values in parentheses indicate associated uncertainties (SD/$\sqrt{n}$). $^b$ Data from ref 21. $^c$ Data for summer weekday tests in the Supporting Information of ref 22.

**FIGURE 1. Comparison of distributions of $\alpha$-alkanes (a), PAHs (b), and hopanes (c) in PM$_{2.5}$ emissions in the Zhujiang Tunnel, the Van Nuys Tunnel, and the Wutong Tunnel.**

Platform of vehicle emission experiments

Engine

A KB-2 Sampler
TEM-EDX
Morphology

OC-EC Aerosol Analyzer

POM

PM

Ion Chromatography System

Ion

6890/5973 GC-MS

GC-MS/FID

VOCs
Vehicle Emission Factor of BC

**Diesel**

- Emission factor (mg kg\(^{-1}\))
- Engine load
- Engine speed

**GDI gasoline**

- Engine load
- Engine speed

Yanhong Qin, Min Hu* et al. *China Environ. Sci.*, 2016
Vehicle Emission Factor of BC

EC emission factor (mg kg\(^{-1}\))

L-diesel  M-diesel  Gasoline  Tunnel

OC emission factor (mg kg\(^{-1}\))
Platform of biomass burning experiments

- Lower moisture content
- Higher moisture content

- Corn straw burning
- Wheat straw burning

- PM10 sampling channel
- PM2.5 sampling channel

- Zero gas supply equipment
- K-type thermocouple
- Electronic scale
Biomass Burning Emission Factor of BC


Straw moisture content, %

(a) Corn

(b) Wheat wangdu

Wheat Zhengzhou
### Emission factor of BC for power plane and steel plant

<table>
<thead>
<tr>
<th></th>
<th>PM$_{2.5}$ conc. (mg/m$^3$)</th>
<th>OC (%)</th>
<th>EC (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Power plant #1</strong></td>
<td>4.35</td>
<td>1.05±0.44</td>
<td>0.34±0.39</td>
</tr>
<tr>
<td><strong>Power plant #2</strong></td>
<td>0.94</td>
<td>1.88±0.87</td>
<td>0.33±0.15</td>
</tr>
<tr>
<td><strong>Sintering</strong></td>
<td>0.68</td>
<td>12.96±11.49</td>
<td>1.95±1.96</td>
</tr>
<tr>
<td><strong>Electric furnace</strong></td>
<td>5.33</td>
<td>0.54±0.12</td>
<td>0.02±0.03</td>
</tr>
</tbody>
</table>

*Note: PM$_{2.5}$ conc. (mg/m$^3$) represents the concentration of PM$_{2.5}$ in the environment.*
Work done by PKU group

- Emission factor and characteristics of BC
- Source identification of BC particles
- Evolution of BC particles during aging process in the atmosphere
\( f_M = \frac{(^{14}C/^{12}C)_{\text{sample}}}{0.95(^{14}C/^{12}C)_{\text{OXI}}} \)

where \((^{14}C/^{12}C)_{\text{sample}}\) is the \(^{14}C\) to \(^{12}C\) ratio observed in the sample, and \((^{14}C/^{12}C)_{\text{OXI}}\) is a ratio from the NIST Oxalic Acid Standard I (NBS OXI). The values of blanks ranged from \(4 \times 10^{-3}\) to \(5 \times 10^{-3}\).

Due to above-ground nuclear testing during 1950s and 1960s, the ambient \(^{14}C\) excess from nuclear bomb tests needs to be corrected. For biogenic emissions a value of 1.04 was used, taken from measurements performed by Levin in 2007 (Levin et al., 2008). Szidat et al. (2009) recommended that the ratio was 1.16 for 30–50-year-old trees. In this study, a correction factor of 1.08, as suggested by Heal et al. (2011), was chosen. Therefore the fraction of contemporary carbon \((f_c)\) in the samples was estimated using \(f_c = \frac{f_M}{1.08}\) and the fraction of fossil carbon \((f_f)\) using \(f_f = 1 - f_c\).

\( f_c = \) fraction of contemporary carbon

- Fossil fuel emission contribute about 80-87% EC in both seasons.
- More contemporary carbon in winter due to biomass burning.

Xuesong Sun, Min Hu* et al., *Atmos. Environ.*, 2012
Source apportionment of PM$_{2.5}$ during the harvest season in eastern China’s agricultural regions

- High-level PM$_{2.5}$, OC, EC, K and Cl were observed in local wheat burning period.
- Wheat residue burning contributed over 50% of PM$_{2.5}$, OC, EC, K and Cl.
- Relative humidity and aging process of PM$_{2.5}$ might affect the fate of Cl in PM$_{2.5}$.

Jianfeng Li, Yu Song*…Min Hu*, Atmos. Environ. 2014
The identification of source regions of BC at a receptor site of the eastern coast of China

Source regions were identified by total potential source contribution function (TPSCF).

TPSCF values were consistent with BC emission rates in the emission inventory.

Qingfeng Guo, Min Hu* et al., *Atmos. Environ.*, 2015
The identification of source regions of BC at a receptor site of the eastern coast of China

Residential emission is the main BC source during heating period

Qingfeng Guo, Min Hu* et al., Atmos. Environ., 2015
The modified EC tracer method was adopted to estimate SOC. \((OC/EC)_{pri}\) was calculated for the data points in day and night, respectively. At the rural site in the PRD SOA is 47\% of OC, up to 80\%. Good correlations between estimated SOC with OOA and WSOC, and estimated POC with HOA, proved the results is reliable.
Work done by PKU group

- Emission factor and characteristics of BC
- Source identification of BC particles
- Evolution of BC particles during aging process in the atmosphere
Aging of BC particles from clean to haze in Beijing

Hongya Niu, Min Hu* et al, STOTEN, revised after review
Growth of BC particles with comparison between clean and haze period

- Higher shell diameter in haze day
- No obvious change in core diameter

Hongya Niu, Min Hu* et al, STOTEN, revised after review
Growth of BC particles with comparison between clean and haze period

Core-shell ratio = \frac{\text{core diameter}}{\text{shell diameter}} (0 < \text{ratio} < 1)
QUasi-real Atmosphere aerosoL evolutIon sTudY Chamber: QUALITY Chamber

<table>
<thead>
<tr>
<th>Condition</th>
<th>Discrsipion</th>
</tr>
</thead>
<tbody>
<tr>
<td>Temp. &amp; RH</td>
<td>Same as in the ambient air, Difference of Temp. &lt;1°C, Difference of RH &lt;3%</td>
</tr>
<tr>
<td>Radiation</td>
<td>Same trends as in the ambient air, but 50% lower</td>
</tr>
<tr>
<td>Gaseous species</td>
<td>Same as in the ambient air, Difference &lt; 10%</td>
</tr>
<tr>
<td>Particles</td>
<td>Controlled in the chamber, lower than in the ambient air</td>
</tr>
</tbody>
</table>
Chamber Experiment design

Measured parameters and relative instruments
- Chemical composition: AMS, SP2
- Diameter & density: SMPS, APM
- Optical properties: PAX
- Hygroscopicity: HTDMA, CCN, VTDMA

Outdoor

CCN Channel-A
CCN Channel-B

Indoor

Soot generator
Neutralizer
RH
DMA
PAX
SP2
APM
AMS
CPC
Dryer
DMA
Humidifier

Measurement
Injection
Growth of BC particles in different environment

- Fast increase of diameter, density and mass
- Total growth of $D_{me}$ in each experiment: 32-152 nm
- Growth rate: 26 (11-47) nm/h

Jianfei Peng, Min Hu*…Renyi Zhang*, PNAS, 2016
BC morphology change

Jianfei Peng, Min Hu*...Renyi Zhang*, PNAS, 2016
Evolution of hygroscopicity

- HTDMA RH=88%
- CCN 100.2%-100.8%

HGfd: hygroscopicity growth factor in diameter
Enhancement of light absorption

Jianfei Peng, Min Hu*…Renyi Zhang*, PNAS, 2016
**BC aging and direct radiative forcing**

- The timescales to achieve complete morphology modification and an absorption amplification factor of 2.4 for BC particles are estimated to be 2.3 h and 4.6 h, respectively, in Beijing, compared with 9 h and 18 h, respectively, in Houston.
- BC under polluted urban environments could contribute significantly to both pollution development and large positive radiative forcing, implying that reduction of BC emissions achieves a co-benefit in simultaneously controlling air pollution and protecting climate, especially for developing countries.

Jianfei Peng, Min Hu*…Renyi Zhang*, *PNAS*, 2016
Future work

- Aging of BC particles from vehicles & biomass burning, properties changing and impacts on climate change
- Evaluation of the aging time scale and absorption enhancement of BC particles in the ambient air with DMA-SP2 system

Look forwards to cooperating with you
Acknowledgements

National Natural Science Foundation of China (21025728, 21190052, 41121004), the National Basic Research Program, the Ministry of Science and Technology (2013CB228503).

PKU Atmospheric Chemistry Team

International cooperation with

✓ Prof. Ren-Yi Zhang, Department of Atmospheric Science, Texas A&M University, USA
✓ Prof. Ali Wiedensohler, Birgit Wehner, Leibniz Institute for Tropospheric Research (Tropos), Germany
✓ Prof. Michael Boy, Department of Physics, University of Helsinki, Finland
✓ Prof. Kondo, Univ. of Tokyo, Japan
✓ Prof. James J. Schauer, Univ. of Wisconsin, USA
✓ Prof. Jose Jimenez, Univ. of Colorado, USA
✓ Prof. Douglas Worsnop, John Jayne, Aerodyne Res. Inc., USA…..
Thank you for your attentions
Your comments are welcome!

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