The Fate of Black Carbon in the Atmosphere: Rapid Removal by Wet Deposition after Aging

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Radiative forcing by Tropospheric aerosol

Direct Effect: Scattering and absorption of incoming sunlight by aerosol particles

Indirect Effect: The number of CCNs influences the cloud droplet size and thereby changes the cloud albedo and lifetime

Organics
SO₂
BC
Dust
Sea salt

Aerosol particles

Wood
Industries
Traffic
Biomass burning
Ocean

CCN and IN
Indirect effect of carbonaceous particles: Ship tracks

Ship tracks on the East Atlantic

Aerosol particles emitted by ships (soot particles with a high sulfur content) act as CCN and form clouds and enhance cloud reflectivity.
The global mean radiative forcing of the climate system for the year 2000, relative to 1750.

Source: www.ipcc.ch
Pathways of the Traditional Warm Indirect Aerosol Effect and the Glaciation Indirect Aerosol Effect

Cloud droplets → Cloud cond. nuclei → Aerosol particles → Human activity

Cloud albedo

Cloud cover and lifetime

Precipitation

Mixed phase cloud hydrometeors

Ice crystals

Ice nuclei

1st Twomey effect

2nd Twomey effect

Lohmann, GRL, 2002
Radiative forcing by BC

**Direct effect:**
Absorption of incoming sunlight

**Indirect effect:**
- Incorporation of BC into cloud droplets and ice crystals
  (wet deposition of BC → decreasing absorption)
  (modification of cloud optical properties)

**Semi direct effect:**
Absorption of solar radiation by soot may cause evaporation of cloud droplets
Jungfraujoch 3580 m a.s.l.

- GAW station
- Few local emissions
- Good infrastructure
- Free troposphere
- Aged aerosol
- 40% cloud occurrence
**BC seasonality**

**Winter** (November-December-January)  **Summer** (June-July-August)

<table>
<thead>
<tr>
<th>Season</th>
<th>BC Concentration at 880 nm (ng/m³)</th>
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</thead>
<tbody>
<tr>
<td>Winter (Nov-Dec-Jan)</td>
<td>BC = 18.9 ng/m³</td>
</tr>
<tr>
<td>Summer (Jun-Jul-Aug)</td>
<td>BC = 145 ng/m³</td>
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<tr>
<td></td>
<td>BC (Jun-Jul-Aug) = 101.7 ng/m³</td>
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</tbody>
</table>
**BC mass fraction in PM1**

*OM = 1.9 OC for summer and winter*

**Winter & Spring 2004 PM1**
- **OM:** Predominantly from anthropogenic sources
- **SO4:** Mainly anthropogenic
- **NO3:** Mainly anthropogenic
- **NH4:** Anthropogenic
- **BC:** Anthropogenic
- **None determined**

PM1 mass concentration = 1.4 µg/m³
BC mass concentration = 84 ng/m³

**Summer 2005 PM1**
- **OM:**
- **SO4:** Mainly anthropogenic
- **NO3:** Mainly anthropogenic
- **NH4:** Anthropogenic
- **BC:** Anthropogenic
- **None determined**

PM1 mass concentration = 3.4 µg/m³
BC mass concentration = 89 ng/m³
Atmospheric aging processes change the mixing state, important for e.g. modeling the radiative forcing of black carbon.

**External Mixture**
- BC particles are separated from scattering particles

**Coated Internal Mixture**
- BC particles are coated with scattering material

Internal Mixture
- \((\text{NH}_4)_2\text{SO}_4\)
- SOA

Coated Internal Mixture
- Coated BC particles
- Scattering material
Inlets

Ice CVI inlet:
removes:
- droplets
- int. particles
- large ice crystals
(Size: 5-30 μm)

Interstitial inlet:
(no activated particles)
removes:
- droplets
- ice crystals
(Size < 2μm)

Total inlet:
(all particles, including activated ones)
heated inlet

Ice residuals
Laboratory (dry aerosol)

Free particles
All particles
**BC measurements:**
- **MAAP** = Multi Angle Absorption Photometer
- **PSAP** = Particle Soot Absorption Photometer

**Chemical composition measurements:**
- **AMS** = Aerosol Mass Spectrometer

**Cloud microphysics:**
- **PVM** = Particulate Volume Monitor
- **CPI** = Cloud Particle Imager

**Size distribution:**
- **SMPS** = Scanning Mobility Particle Sizers
Scavenging of Black Carbon in liquid cloud

Fraction of BC aerosol that is incorporated into a cloud droplet or an ice crystal

\[
\text{Scavenged fraction} = \frac{C_{\text{cloud}}}{C_{\text{total}}} = \frac{C_{\text{tot}} - C_{\text{int}}}{C_{\text{total}}}
\]
Scavenging of Black Carbon in mixed phase cloud

Fraction of BC aerosol that is incorporated into a cloud droplet or an ice crystal

\[
\text{Scavenged fraction} = \frac{C_{\text{cloud}}}{C_{\text{total}}} = \frac{C_{\text{tot}} - C_{\text{int}}}{C_{\text{total}}}
\]

Nearly no scavenged fraction
Scavenged BC fraction evolution with temperature

< -20°C: cloud exists mainly of ice crystals (low scavenging)

> -20°C: ~ of liquid droplet number (~ of BC scavenging)

- BC scavenged fraction is 61% at T>-5°C
Evolution of particles in cloud: Bergeron-Findeisen process

Saturation Vapor Pressure (SVP) difference: $\text{SVP (ice)} < \text{SVP (liquid)}$

$\Rightarrow$ Flux of water vapor from liquid droplets to ice crystals
**Ice CVI inlet:**
removes:
- droplets
- int. particles
- large ice crystals

(Size: 5-30 μm)

**Total inlet:**
(all particles, including activated ones)
heated inlet

**Laboratory (dry aerosol)**
Ice residuals mainly consisted of BC and refractory material (mineral dust, ...)

**Ice nuclei chemical composition**

AMS data from Max-Planck Institut Mainz

**Ice residuals**

**Total**

<table>
<thead>
<tr>
<th>Component</th>
<th>AMS</th>
<th>SMPS</th>
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<tbody>
<tr>
<td>Ammonium</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Organics</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Nitrate</td>
<td></td>
<td></td>
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<tr>
<td>Sulphate</td>
<td></td>
<td></td>
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<tr>
<td>BC</td>
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</tbody>
</table>

AMS data from Max-Planck Institut Mainz
Enrichment of BC in small ice crystals (most points above line 1:1)
Conclusions

Aging processes result in coating of BC with soluble components
✓ Internal mixture of JFJ aerosol
✓ Influence on hygroscopic properties of soot particles

In liquid clouds
✓ BC is incorporated into cloud droplets as bulk aerosol
✓ 60% of BC mass is incorporated into cloud droplets and ice crystals (wet deposition of BC increases)

In mixed-phase clouds
✓ Incorporation of BC is considerably lower (Bergeron-Findeisen process)
✓ BC is enriched by 20% in the ice phase (influence on cloud optical properties)
✓ Ice nuclei mainly consist of BC and refractory material

Summary:
Incorporation of BC into cloud droplets and ice crystals for an aged aerosol
✓ Increases the wet deposition of BC (influence on lifetime of soot particles)
✓ Influence the optical properties of cloud by possibly increasing the number of CCN and by acting as IN
Thank you for your attention

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