PM2.5 modelling: research and policy challenges

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PM modelling : Why?

- Impact of emission reduction scenarios on PM concentrations; assessment of the compliance with regulatory target values or thresholds; integrated assessment modelling

- Particulate episodes forecasting and integrated air quality monitoring systems (maps and spatialized information); complementary tools to measurements

- Better understanding of the phenomenology: conditions under which episodes develops, role played by precursors

- Source apportionment and emission inventories assessment

  - As for ozone, high interest of policy makers but for PM it is not always compatible with the science limits
Few difficulties inherent to PM2.5 modelling

- Physico-chemical processes knowledge for implementing relevant parametrisations in chemistry transport models
  - Secondary organic aerosols formation from VOC
  - Aqueous chemistry (sulfates)
  - Physical processes that impact the particle size (nucleation, aggregation)

- Quantification of the emissions of primary particles and precursors (VOC)
  - Carbonaceous particles: residential heating, wood combustion, traffic
  - Agricultural sources: nitrogen compounds
  - Natural and anthropogenic VOCs
  - Temporal profiles, dependency on meteorological conditions

- Model validation
  - Lack of speciated data (species and size-distribution)
  - Lack of collocated data (nitric acid and nitrate for instance)
  - Measurement systems diversity and lack of standards
Example: the CHIMERE model (CNRS/INERIS)

Meteorological Data
- ECMWF, GFS/MM5

Boundary conditions
- Average monthly climatologies GOCART, MOZART, LMDzINCA

Emissions
- Anthropogenic (EMEP)
- Biogenic

CHIMERE
- Gaseous chemistry
  - MELCHIOR [Lattuati, 1997]
  - 44 species, 116 reactions
- Aerosol module
  - Bessagnet et al., 2004
- Transport
  - Horizontal (PPM)
  - Vertical diffusion
- Deposition
  - Dry [Seinfeld and Pandis, 1998]
  - Wet [Guelle et al., 1998]

Air pollutant forecasting (D, D+1, D+2)
- PREVAIR

Simulation for emission reduction studies
- EURODELT-A - CITYDELT-A

Optical properties computation
- Hodzic et al., 2004 + new PhD.

- Free download at http://euler.lmd.polytechnique.fr/chimere
- Development at IPSL/LMD (CNRS), LISA and INERIS
Aerosol modelling in CHIMERE

Composition:
- PPM (primary particle material)
- OCAR (organic material)
- BCAR (black carbon)
- DUST (from erosion and resuspension)
- SIA (NH4, NO3, SO4)
- SOA (1-4 or 9 species)
- SALT (sea salts)
- WATER
**SOA modelling in CHIMERE**

<table>
<thead>
<tr>
<th>Surrogate precursor</th>
<th>Oxidant</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>OH</td>
</tr>
<tr>
<td>Toluene</td>
<td>L</td>
</tr>
<tr>
<td>Trimethylbenzene</td>
<td>M</td>
</tr>
<tr>
<td>α-Pinene</td>
<td>L</td>
</tr>
<tr>
<td>β-Pinene</td>
<td>L</td>
</tr>
<tr>
<td>Limonene</td>
<td>L</td>
</tr>
<tr>
<td>Terpinene</td>
<td>M</td>
</tr>
<tr>
<td>Terpineol</td>
<td>M</td>
</tr>
<tr>
<td>Humulene</td>
<td>L</td>
</tr>
</tbody>
</table>

Note: L: product data from laboratory experiments; M: product data from theoretical mechanism; --: no reaction; NA: data unavailable for that reaction pathway.

\[
K = \frac{A_i / \left( M_0 + \sum_j A_j \right)}{G_i}
\]

\[
G_i + A_i = C_i
\]

\[
K = \frac{760 \cdot RT}{10^6 \cdot MW_{OM} \gamma_i P_{i, sat}}
\]

**4-9 SOA scheme**

**Gas/particle partitioning**

Pun et al., 2006
Performances of CHIMERE at background sites

Error statistics based on daily mean values (last model version)

<table>
<thead>
<tr>
<th>*Correlation</th>
<th>Bias</th>
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<tbody>
<tr>
<td>Sulfate</td>
<td>none</td>
</tr>
<tr>
<td>Nitrate</td>
<td>Negative (coarse nitrate)</td>
</tr>
<tr>
<td>Ammonium</td>
<td>none</td>
</tr>
<tr>
<td>PM10</td>
<td>Negative (other sources?)*</td>
</tr>
<tr>
<td>PM2.5</td>
<td>Positive (secondary production)</td>
</tr>
<tr>
<td>Sea salts</td>
<td>?</td>
</tr>
<tr>
<td>BC (« black carbon »)</td>
<td>« correct »</td>
</tr>
<tr>
<td>OC (« organic carbon »)</td>
<td>« difficult »</td>
</tr>
<tr>
<td>SOA</td>
<td>?</td>
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</table>

Too few datasets
Statistical scores for the PM2.5 concentrations
AIRBASE data; 2003

<table>
<thead>
<tr>
<th></th>
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<tbody>
<tr>
<td>DJF</td>
<td>14.8</td>
<td>14.2</td>
<td>0.67</td>
<td>74.7</td>
<td>48.7</td>
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<tr>
<td>MAM</td>
<td>17.6</td>
<td>14.9</td>
<td>0.72</td>
<td>54.3</td>
<td>37.8</td>
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<tr>
<td>JJA</td>
<td>15.3</td>
<td>13.6</td>
<td>0.71</td>
<td>36.8</td>
<td>21</td>
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<tr>
<td>SON</td>
<td>14.9</td>
<td>11.1</td>
<td>0.67</td>
<td>69.8</td>
<td>57.7</td>
</tr>
<tr>
<td>Year</td>
<td>15.7</td>
<td>13.3</td>
<td>0.68</td>
<td>58.8</td>
<td>41.4</td>
</tr>
<tr>
<td>Year PM10</td>
<td>18.9</td>
<td>20.7</td>
<td>0.71</td>
<td>40.3</td>
<td>8</td>
</tr>
</tbody>
</table>
Validation for carbonaceous species on background stations in 2003: elemental carbon

Figure 1: Comparisons between observed (plus symbols) and simulated (circle symbols) EC concentrations in 2003 for the CARBOSOL and EC/OC EMEP campaign dataset
Validation for carbonaceous species on background stations in 2003: organic carbon

Figure 1: Comparisons between observed (red squares) and simulated OC concentrations (filled area) in 2003 for the CARBOSOL and EC/OC EMEP campaign datasets. Carbon in the secondary organic aerosols (SOA-C) is represented by crosshatched area.
Variability among models: the Eurodelta exercises

Area: Germany-France-UK-Spain
Mean concentrations in µg/m³

- CHIMERE
- RCG
- EMEP
- MATCH
- LOTOS

SIA2.5 PPM2.5 PM2.5 NO₃ NH₄ SO₄ HNO₃ NH₃ NOₓ H₂O₂ SO₂
PM2.5 modelling issue: evidence of secondary aerosols episodes (ammonium nitrate)

15th March 2007
PM10, moyenne journalière en µg/m³
Prévision du 15/03/2007 pour le jour-même

100 µg/m³ in Bordeaux and Lyon areas

29th March 2007
PM10, moyenne journalière en µg/m³
Prévision du 29/03/2007 pour le jour-même

70-100 µg/m³ in the North

15th April 2007
PM10, moyenne journalière en µg/m³
Prévision du 15/04/2007 pour le jour-même

70-90 µg/m³ in the North-western part of the country

14, 28 March and 15 April 2007
PM2.5 modelling issue: Ammonium nitrate episodes

Data issued from chemical analyses and the TEOM-FDMS monitoring network and chemical analyses.
PM2.5 modelling issue: sensitivity to emission data

Too simple seasonal profiles for ammonia emissions

Temperature and soil dependent emission processes

\[ \text{NH}_4^+ + \text{OH}^- \xrightarrow{\text{-----}} \text{NH}_3^g + \text{H}_2\text{O} \]

INERIS
PM2.5 modelling issue: SOA formation

Most of physical and chemical processes are uncertain:

- Terpenes and isoprene emissions
- SOA chemistry (need of more complex mechanism with heterogeneous chemistry)
- Gas to particle formation (nucleation and/or condensation)
- Deposition velocity of semi-volatile organic compounds in the gas phase

Average SOA concentrations computed with CHIMERE: Summer 2003
PM2.5 modelling issues: impact of climate change

Present = [1985-1990]

Ammonium nitrate:
- Evaporation
- Over forest areas: Increasing SOA formation
- Increase of Dust emissions
- Over arable lands

[2070-2075]^{A2} - Present

Over Paris and Milan:
- Increasing BLH
Conclusions

Recent and encouraging progress:
- Improvement of model performances and consistent results provided by the existing models
- At the European level, use of models has been recommended for regional studies of emission control scenarios (EMEP PM assessment report, 2007 - http://www.nilu.no/projects/ccc/reports.html)
- PM2.5: secondary episodes with a long range transport component
- Still lot of work need on the processes description

Lot of expectations from emission inventories improvement
- Temporal profiles, VOCs, heavy metals, wood combustion

Need for further model evaluation data
- Chemical data for aerosol species, size-distribution
- Collocated measurement with gaseous species (nitrous acid, NOx, NH3)
- Reference methodologies (EC/OC)
- Spatial representativeness (2D, 3D) - field campaigns, research networks (EMEP, EUSAAR, EUCAARI, EARLINET...)
- Satellite data?