Changes of PM$_{2.5}$ concentrations and their sources in the US from 1990 to 2010

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PM increases the mortality rate

Harvard 6 Cities Study
Dockery, Pope et al. (1993)

Di et al. (2017)
Motivation

• Traditionally epidemiological studies treat all PM$_{2.5}$ with equal toxicity

• How does PM$_{2.5}$ toxicity vary by composition and source?

• CTMs are key to developing necessary exposure estimates
The hypothesis

Epidemiological analysis can connect to specific air pollution sources

There are available health data (death rates, smoking rates, etc.) for 1990-2010, from the U.S. National Center for Health Statistics (NCHS)
The problem

Only a few observations for 1990

More observations for 2000 and 2010 from 2 networks

- Chemical composition is available
- No measurements for sources!
Historical Source-Resolved Simulations (1990-2001-2010)
Application of PMCAMx over US

- **The whole US**
  - 4752 x 2952 km²
  - 36 x 36 km grid resolution
  - 14 vertical layers, up to 14 km

- **Inputs**
  - **Meteorology:** WRF (Skamarock et al., 2005)
  - **Emissions:** Anthropogenic (Xing et al., 2013)
    - Biogenic (MEGAN3, Guenther et al., 2012)

- **CB5 mechanism** (Yarwood et al., 2005)
  - 79 gas phase species
  - 13 radicals
  - 190 reaction in the gas phase
  - 43 aerosol species

PMCAMx Chemical Transport Model

PMCAMx processes:

Emissions → Horizontal Advection → Vertical Advection → Vertical Diffusion → Horizontal Diffusion → Dry/Wet Deposition → Chemistry

- Gas-Phase Chemistry
- Water Chemistry (Clouds)
- Aerosol Chemistry
  - Inorganics
  - Organics
Particulate Matter Source Apportionment Technology

• Utilizes the fact that at a given location and time, all the molecules of a species regardless of their source have the same probability of reacting, being transported or being deposited

• Newly formed secondary species are apportioned based on the apportionment of their precursor species

• Very small computational overhead
Particulate Matter Source Apportionment Technology

- Advection and diffusion
- Wet deposition
- Gas phase chemistry
- Partitioning

Wangstrom et al. (2008)
Skyllakou et al. (2014)
Skyllakou et al. (2017)
Emission sources in this study

**ONROAD:** On-road mobile emissions

**NON-ROAD:** Non-road mobile emissions

**BIOGENIC:** Biogenic emissions

**Electrical Generation Units (EGU):**
Emissions Electricity Generating Units

**NON-EGU:** all other US point sources

**OTHER:** All other area type emissions, including wildfires, restaurants, agriculture, residential wood combustion, on-road mobile emissions outside of the US
Annual Emissions of Elemental Carbon (tn/year) from onroad transport
Annual emissions, contribution (%) of each source

Elemental Carbon
- 0.6 Tg/yr: 46% other, 18% non-egu, 28% non-road, 5% road
- 0.4 Tg/yr: 51% other, 17% non-egu, 22% non-road, 6% road
- 0.3 Tg/yr: 60% other, 17% non-egu, 22% non-road, 7% road

SO₂
- 22 Tg/yr: 18% other, 15% non-egu, 16% biogenic, 5% road
- 15 Tg/yr: 20% other, 14% non-egu, 16% biogenic, 12% road
- 8 Tg/yr: 24% other, 16% non-egu, 18% biogenic, 20% road

NMVOCs
- 155 Tg/yr: 40% other, 17% non-egu, 31% biogenic, 22% road
- 125 Tg/yr: 42% other, 10% non-egu, 21% biogenic, 15% road
- 101 Tg/yr: 41% other, 12% non-egu, 15% biogenic, 14% road
Sources of PM$_{2.5}$ ($\mu$g/m$^3$) for the whole 2010
Sources of PM$_{2.5}$ ($\mu$g/m$^3$) for the whole 2010

Long Range Transport (19%)

Electrical Generation Units (11%)

Non-Electrical Generation Units (5%)

On-road (5%)

Non-road (3%)

Biogenic (5%)

Other (52%)
Predicted annual average PM$_{2.5}$ ($\mu$g/m$^3$)
Annual average contribution of each source

**PM$_{2.5}$**
- 1990: 51% (other), 6% (non-egu), 16% (egu), 8% (biogenic), 13% (road), 5% (LRT)
- 2001: 51% (other), 15% (non-egu), 6% (egu), 7% (biogenic), 15% (road), 19% (LRT)
- 2010: 52% (other), 11% (non-egu), 5% (egu), 5% (biogenic), 19% (road), 6% (LRT)

**OA**
- 1990: 45% (other), 9% (non-egu), 6% (egu), 16% (biogenic), 21% (road), 6% (LRT)
- 2001: 44% (other), 10% (non-egu), 6% (egu), 11% (biogenic), 25% (road), 6% (LRT)
- 2010: 43% (other), 15% (non-egu), 6% (egu), 6% (biogenic), 27% (road), 6% (LRT)

**Sulfate**
- 1990: 13% (other), 13% (non-egu), 6% (egu), 49% (biogenic), 22% (road), 40% (LRT)
- 2001: 14% (other), 12% (non-egu), 6% (egu), 44% (biogenic), 28% (road), 33% (LRT)
- 2010: 15% (other), 11% (non-egu), 6% (egu), 44% (biogenic), 28% (road), 40% (LRT)
Evaluation for annual average PM$_{2.5}$

**1990**
- FBias: 19%
- Ferror: 27%

**2001**
- FBias: 7%
- Ferror: 25%

**2010**
- FBias: 7%
- Ferror: 26%

**Equations**

\[
FBIAS = \frac{2}{n} \sum_{i=1}^{n} \frac{(P_i - O_i)}{(P_i + O_i)}
\]

\[
FERROR = \frac{2}{n} \sum_{i=1}^{n} \frac{|P_i - O_i|}{(P_i + O_i)}
\]

**Legend**
- IMPROVE: rural sites
- STN: urban sites
Evaluation for annual average OM

1990

IMPROVE: OM = 1.8*OC
STN: OM = 1.4*OC

fbias = 12%
ferror = 27%

2001

IMPROVE: OM = 1.8*OC
STN: OM = 1.4*OC

fbias = 6%
ferror = 28%

FBIAS = \[ \frac{2}{n} \sum_{i=1}^{n} \frac{(P_i - O_i)}{(P_i + O_i)} \]

FERROR = \[ \frac{2}{n} \sum_{i=1}^{n} \frac{|P_i - O_i|}{(P_i + O_i)} \]
Evaluation for annual average PM$_{2.5}$ Sulfate

**1990**

**IMPROVE**

**2001**

**IMPROVE**

**STN**

fbias$= 18\%$

ferror$= 21\%$

\[
FBIAS = \frac{2}{n} \sum_{i=1}^{n} \frac{(P_i - O_i)}{(P_i + O_i)}
\]

\[
FERROR = \frac{2}{n} \sum_{i=1}^{n} \frac{|P_i - O_i|}{(P_i + O_i)}
\]

fbias$= 16\%$

ferror$= 28\%$

fbias$= 17\%$

ferror$= 27\%$
PM$_{2.5}$ reduction from 1990 to 2010

Absolute reduction ($\mu$g/m$^3$)

Percent reduction (%)
Evaluation of predicted of PM$_{2.5}$ reductions

Pearson’s r
1990-2001: 0.76
1990-2010: 0.81
2001-2010: 0.60
Separation of US domain in different parts
Sources of annual average PM$_{2.5}$
US population inhabitants/km$^2$

1990, total 247 million

2010, total 307 million

2000, total 280 million

https://www.census.gov/
Population exposure to annual average PM$_{2.5}$

$$L_{i,j} : \text{population exposure}$$

$$L_{i,j} = C_{i,j} \times P_{i,j}$$

annual concentration

from Walker et al. 1999
Conclusions

• The model reproduces the annual average concentrations of PM$_{2.5}$ with errors less than 30% for all the years.

• PM$_{2.5}$ Concentrations are reduced from 20%-50% between 1990 and 2010.

• These differences are captured by the model.
• The main sources of OA are **onroad transport** and **biomass burning** and other

• The contribution of the onroad transport decreases almost 3 times from 1990 to 2010,

• The main source of sulfate is the **Electrical Generation Units** source

• The emissions of SO$_2$ were reduced 3 times from 1990 to 2010

• The reduction of human exposure to PM$_{2.5}$ was attributed almost 20% to road transport and 30% to ‘EGUs’