Recent advances in top-down estimates of emissions from human activities, soils, and fires

Jun Wang
Lab for Atmospheric and Environmental Research (AER)
http://arroma.uiowa.edu
College of Engineering
University of Iowa

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Colloquium
Department of Meteorology and Atmospheric Science
The Pennsylvania State University
IPCC AR6: “Observed warming is driven by emissions from human activities, with greenhouse gas warming partly masked by aerosol cooling”

AR-5
- Prior to AR6, IPCC’s summary for policy makers has centered around the climate forcing by forcing agent.
- In AR6, a new paradigm has emerged, in which the climate forcing must be attributed to the sources of these agents, i.e., the emissions.
<table>
<thead>
<tr>
<th>Emitted Compound</th>
<th>Resulting Atmospheric Drivers</th>
<th>Radiative Forcing by Emissions and Drivers</th>
<th>Level of Confidence</th>
</tr>
</thead>
<tbody>
<tr>
<td>CO₂</td>
<td>CO₂</td>
<td>1.68 [1.33 to 2.03]</td>
<td>VH</td>
</tr>
<tr>
<td>CH₄</td>
<td>CO₂, H₂O, O₃, CH₄</td>
<td>0.97 [0.74 to 1.20]</td>
<td>H</td>
</tr>
<tr>
<td>Halo-carbons</td>
<td>O₃, CFCs, HCFCs</td>
<td>0.18 [0.01 to 0.35]</td>
<td>H</td>
</tr>
<tr>
<td>N₂O</td>
<td>N₂O</td>
<td>0.17 [0.13 to 0.21]</td>
<td>VH</td>
</tr>
<tr>
<td>CO</td>
<td>CO₂, CH₄, O₃</td>
<td>0.23 [0.16 to 0.30]</td>
<td>M</td>
</tr>
<tr>
<td>NMVOC</td>
<td>CO₂, CH₄, O₃</td>
<td>0.10 [0.05 to 0.15]</td>
<td>M</td>
</tr>
<tr>
<td>NOₓ</td>
<td>Nitrate, CH₄, O₃</td>
<td>-0.15 [-0.34 to 0.03]</td>
<td>M</td>
</tr>
<tr>
<td>Aerosols and precursors</td>
<td>Mineral Dust, Sulphate, Nitrate, Organic Carbon and Black Carbon</td>
<td>-0.27 [-0.77 to 0.23]</td>
<td>H</td>
</tr>
<tr>
<td></td>
<td>Cloud Adjustments due to Aerosols</td>
<td>-0.55 [-1.33 to -0.06]</td>
<td>L</td>
</tr>
<tr>
<td></td>
<td>Albedo Change due to Land Use</td>
<td>-0.15 [-0.25 to -0.05]</td>
<td>M</td>
</tr>
<tr>
<td>Natural</td>
<td>Changes in Solar Irradiance</td>
<td>0.05 [0.00 to 0.10]</td>
<td>M</td>
</tr>
</tbody>
</table>

**Total Anthropogenic RF relative to 1750**

- 2011: 2.29 [1.13 to 3.33]  (H)
- 1980: 1.25 [0.64 to 1.86]  (H)
- 1950: 0.57 [0.29 to 0.85]  (M)

Radiative Forcing relative to 1750 (W m⁻²)
Air pollution affected by emissions is a leading risk factor that contributes to millions of deaths each year.

<table>
<thead>
<tr>
<th>Risk Factor</th>
<th>Total Number of Deaths (Millions) in 2019</th>
</tr>
</thead>
<tbody>
<tr>
<td>High systolic blood pressure</td>
<td>6.67 million</td>
</tr>
<tr>
<td>Tobacco</td>
<td></td>
</tr>
<tr>
<td>Dietary risks</td>
<td></td>
</tr>
<tr>
<td>Air pollution</td>
<td>6.67 million</td>
</tr>
<tr>
<td>High fasting plasma glucose</td>
<td></td>
</tr>
<tr>
<td>High body-mass index</td>
<td></td>
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<tr>
<td>High LDL</td>
<td></td>
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<tr>
<td>Kidney dysfunction</td>
<td></td>
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<tr>
<td>Malnutrition</td>
<td></td>
</tr>
<tr>
<td>Alcohol use</td>
<td></td>
</tr>
</tbody>
</table>

Source: WHO & Health Effect Institute & State of Global Air

Source: WMO, Air Quality and Climate Bulletin
Importance of emissions: a modeling perspective

- CTM and climate model simulations can only be as good as the emissions.
- Data assimilation (without constraints of emissions) for CTM and ESM:
  - suffers from emission errors that are persistent.
  - may improve forecast when observations of state parameters are available, but such improvement decay quickly with time once obs. are not available.
- For both climate studies and air quality forecast, there is a need to have a holistic interpretation from emissions to observations, and vice versa.
Top-Down vs. Bottom-Up estimates of emissions

**Bottom-up emission estimate**
- Usually has a 2~3 yr lag
- Seasonal/ monthly
- Point or area average
- Chemically speciated
- Lack of constraint on emission above the surface

**Top-down emission estimate**
- Has the potential for near real time
- Daily (polar-orbiting) or higher (geo.)
- Globally with high spatial resolution
- Trace gases & optical thickness
- Reflecting the columnar mass, and thus 1\textsuperscript{st} order of emission

Ground-based network/data
Emission Sources

Primary sources for aerosols: directly from surface

- dust
- smoke
- volcano ashes

Secondary sources:
- Atmospheric chemistry
  - SO$_2$ $\rightarrow$ sulfate
  - NO$_x$ $\rightarrow$ nitrate
  - VOC $\rightarrow$ organic aerosol
  - NH$_3$ $\rightarrow$ NH$_4$
  - NO $\rightarrow$ NO$_x$

Emission sources have large spatial and temporal variations (minutes-hours, meters to kilometers).
Outline

• Satellite constraints of $\text{SO}_2$ & $\text{NO}_2$

• Efficacy of top-down emissions

• Satellite constraints of emission processes
  – Soil NOx
  – Fire emissions

• Summary and outlook
NO\textsubscript{x} is mainly from fossil fuel combustion; limiting precursor for ozone formation
Average OMI SO$_2$ burdens over eastern USA

Image courtesy: Nickolay Krotkov, NASA
Questions to be addressed:

- Separate vs. joint DA of SO\textsubscript{2} and NO\textsubscript{2} from satellite observations
- Implication for AQ forecast at urban scale

(Georgoulias et al., 2019)
Optimize the emissions by iteratively minimizing the cost function that depends on the model error, observation error, and the difference between model and observation.

\[
J(\sigma) = \frac{1}{2} \sum_{\Omega} \left[ H(M(\sigma)) - c_{obs} \right]^T S_{obs}^{-1} \left[ H(M(\sigma)) - c_{obs} \right] + \gamma \frac{1}{2} [\sigma - \sigma_a]^T S_a^{-1} [\sigma - \sigma_a]
\]
Using OMI SO$_2$ to constrain SO$_2$ emissions

April 2008.

prior (bottom-up, 2006)

posterior (top-down)

SO$_2$ emission

Simulated SO$_2$

OMI SO$_2$

Simulation – OMI

Inverse modeling

$$E = E_0 + \frac{\partial E}{\partial C} \Delta C + \varepsilon$$

Adjoint techniques

Observational constraints

model & instrument & retrieval & representation errors
Implication for air quality forecast:
applying posterior emission from last month to forecast AQ in this month

A new approach for monthly updates of anthropogenic sulfur dioxide emissions from space: Application to China and implications for air quality forecasts,

Joint inversion of anthropogenic SO$_2$ and NO$_x$ emissions

Prior (MIX, 2010)  OMPS-constrained Posterior  (Posterior – Prior) / Prior

SO$_2$

Prior: 1166 Gg S/mon

OMPS-constrained: 748 Gg S/mon

(Posterior – Prior) / Prior: -35.8%

NO$_x$

Prior: 714 Gg N/mon

OMPS-constrained: 672 Gg N/mon

(Posterior – Prior) / Prior: -5.8%

Results using GEOS-chem adjoint (V.8) at 2° x 2.5° resolution, Oct. 2013

Y. Wang et al., ACP, 2020a.
There is good correlation between TROPOMI NO\(_2\) vertical column density and VIIRS nighttime light, thus VIIRS nighttime light intensity should be good proxy for downscaling NO\(_x\) emissions.

Y. Wang et al., ACP, 2020b
Apply top-down constraints in present month to improve forecasts in next month

Bias: 44 μg/m³
RMSE: 78 μg/m³

Bias: -7 μg/m³
RMSE: 37 μg/m³

Bias: 45%
R: 0.61

Bias: 25%
R: 0.75

All results are for Nov. 2013 at 0.25x0.3125 degree resolution by using GEOS-chem nested model.
Efficacy of the top-down emissions

Two methods to test it:

1) Compare with the emissions inverted from satellite-based aerosol observations.

2) Use these emissions for the models that are different from the host model that is used in the top-down estimates.
Use MODIS AOD/Radiance to constrain aerosol primary & secondary emissions

Xu et al., JGR, 2013

(a) GC prior $SO_2$ Column at OMI

(b) GC posterior $SO_2$ Column at OMI

(c) OMI Retrieval of $SO_2$ Column

(d) OMI $SO_2$ loading vs. GC-modeled $SO_2$

OMI $SO_2$ loading moles cm$^{-2}$

 Apr. 2008 over China
Two different sensors (one for gas and one for aerosols) telling nearly the same results about SO$_2$ emissions!
The top-down approach using OMI SO$_2$ and global GEOS-Chem adjoint modeling can timely update anthropogenic SO$_2$ emission for regional AQ modeling.

Efficacy is shown to be robust for four different AQ models (chemistry schemes).
Here in U.S.
Has the NOx emission reduction been slow down since 2009?

Jiang et al., 2016
Soil NOx emissions

- ~1/4 of global NOx production is derived from soils, mostly from fertilized agriculture; however, estimates of global soil NOx emissions vary widely (9–27Tg per year).
- Fertilization and N deposition are known to increase soil NOx emissions; however, the majority of studies are conducted at temperatures below 35°C.
- Strong pulse NOx emission responses to rewetting of soils in high-temperature regions are important, yet understudied in managed systems.
Observation-based insights of emission process dependence of soil NOx emission on temperature

Wang et al., 2021
Pulse of NOx emission after re-wetting

Tong et al., 2021, *Environ. Sci. & Tech.*
Improved simulation of soil NOx emission

• Soil and lightning NOx combined emissions trends change from −3.95% a⁻¹ during 2005–2009 to 0.60% a⁻¹ from 2009 to 2019, thereby rendering the abrupt slowdown of total NOx emissions reduction.

• Non-linear inter-annual variations explain 6.6% of the variance of total NOx emissions.

• Inter-annual variations of either soil or lightning are comparable (slightly larger than anthropogenic sources).

Wang et al., 2021, ERL
At regional scale

- SNO\textsubscript{x} exceed anthropogenic sources over croplands which accounts for 50.7% of NO\textsubscript{x} emissions

- Such considerable SNO\textsubscript{x} enhance the monthly mean NO\textsubscript{2} columns by 34.7% (53.3%) and surface NO\textsubscript{2} concentrations by 176.5% (114.0%), leading to an additional 23.0% (23.2%) of surface O\textsubscript{3} concentration in California (cropland).

Tong et al., 2021. EST
Next frontier in remote sensing of fires

Fire combustion efficiency?

FRP T, fire area
Fire Phase often described as Modified Combustion Efficiency, $\frac{CO_2}{(CO+CO_2)} \rightarrow$ Emission Factor

\[
\begin{align*}
    \text{Liu et al., [2017]} \\
    \text{Pokhrel et al., [2016]}
\end{align*}
\]
How emission factor is treated in fire emission estimates?

- Static for the same type; no consideration of wind speed, relative humidity, ..
- Schemes for surface/biome types are oversimplified and vary in different emission algorithms

<table>
<thead>
<tr>
<th>Specie</th>
<th>GFED4s Savanna</th>
<th>GFED4s Boreal forest</th>
<th>GFED4s Temperate forest</th>
<th>GFED4s Tropical forest</th>
<th>GFED4s Peat</th>
<th>GFED4s Agriculture</th>
<th>FLAMBE Light Grasses/tundra</th>
<th>FLAMBE Grasslands/Savannah</th>
<th>FLAMBE Cerrado/Woody Shrub</th>
<th>FLAMBE Crops</th>
<th>FLAMBE Temperate/Boreal-Low</th>
<th>FLAMBE Temperate-High</th>
<th>FLAMBE Tropical Forest</th>
<th>FLAMBE Wetland</th>
<th>FLAMBE Boundary regions</th>
</tr>
</thead>
<tbody>
<tr>
<td>CO₂</td>
<td>1686</td>
<td>1489</td>
<td>1647</td>
<td>1643</td>
<td>1703</td>
<td>1585</td>
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<td>Grasslands/Savannah</td>
<td>Cerrado/Woody Shrub</td>
<td>Crops</td>
<td>Temperate/Boreal-Low</td>
<td>Temperate-High</td>
<td>Tropical Forest</td>
<td>Wetland</td>
<td>Boundary regions</td>
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<td>CO</td>
<td>63</td>
<td>127</td>
<td>88</td>
<td>93</td>
<td>210</td>
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<tr>
<td>CH₄</td>
<td>1.94</td>
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<td>H₂</td>
<td>1.7</td>
<td>2.03</td>
<td>2.03</td>
<td>3.36</td>
<td>3.36</td>
<td>2.59</td>
<td>Light Grasses/tundra</td>
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<td>Cerrado/Woody Shrub</td>
<td>Crops</td>
<td>Temperate/Boreal-Low</td>
<td>Temperate-High</td>
<td>Tropical Forest</td>
<td>Wetland</td>
<td>Boundary regions</td>
</tr>
<tr>
<td>NOₓ (as NO)</td>
<td>3.90</td>
<td>0.90</td>
<td>1.92</td>
<td>2.55</td>
<td>1.00</td>
<td>3.11</td>
<td></td>
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<tr>
<td>N₂O</td>
<td>0.20</td>
<td>0.41</td>
<td>0.16</td>
<td>0.20</td>
<td>0.20</td>
<td>0.10</td>
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<tr>
<td>PM2.5</td>
<td>7.2</td>
<td>15.3</td>
<td>12.9</td>
<td>9.1</td>
<td>9.1</td>
<td>6.3</td>
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</table>

6 biome types

van der Werf et al., 2017.
If the combustion happens heterogeneously at the surface of solid fuels (vegetation and wood), the combustion is smoldering producing incomplete-oxidized products.

If oxidation happens homogeneously between oxygen in the air and the gas pyrolysate, combustion products are soot and complete-oxidized gases. These products absorb enough energy during the combustion process leading them to emit visible radiation as a flame (Rein 2009; Sato et al. 1969).

While fire emits radiation at all wavelengths, it is the visible intensity that indicates the strength of flaming.
Fire Light seen by VIIRS

12 June 2012

Source: Google Maps
Insights for Fire MCE Climatology as revealed by Visible Energy Fraction (VEF)

\[ VEF = \frac{VLP \times \Delta t}{FRP \times \Delta t} = \frac{VLP}{FRP} \]

Our research algorithm: Firelight Detection Algorithm (FILDA)

MODIS Land Cover Types

Annual mean 2017

Wang et al., 2020
Rem.. Sen. Environ.
VEF is indicative of MCE

- VEF spatial distribution clearly shows the impact of biome types on fire MCE
- FRP has difficulty to describe MCE variation, such as shrubland vs. evergreen forests

Wang et al., 2020, RSE.

\( y = 0.017x + 1.072 \)
VEF & MCE variations show meteorological impact on combustion

High & dry winds lead to increase of flaming on 11 Nov. 2018.

Camp Fire
Diablo Winds

10 am local each day

Synoptic map at 700 mb.

Diablo Winds
VEF has a potential to better predict fire growth
High VEF $\rightarrow$ flaming $\rightarrow$ predicting movement of fire lines
All the three future GEO satellite will provide hourly retrievals of SO$_2$ and NO$_2$. 
Geostationary and Extended Orbits (GEO-XO)

Recommended GEO-XO Constellation

GEO-West
Vis/IR Imager
IR Sounder
Ocean Color

Host Sat
Lightning Mapper

Host Sat
Atmospheric Composition Instrument

GEO-East
Vis/IR Imager
IR Sounder
Ocean Color

Host Sat
Lightning Mapper

https://www.nesdis.noaa.gov/GEO-XO

Now in formulation
Summary

- Satellite data can provide timely insights on the change of emissions from different sources and in some cases, reveal process-level of understanding of emissions.

- Top-down method offers unique opportunity to improve the regional AQ forecast via data assimilation (with timely update of emissions, e.g., 4D-VAR).

- In U.S., as anthropogenic emissions decrease, the background emissions (including those from agricultural activities, soils, and fires) are increasingly important for the air quality prediction.

- Climate predictions and mitigation of climate change requires accurate knowledge of the emissions from different sectors. Future use of TEMPO, GEMS, and others multi-sensor data toward rapid update of emissions for improving urban scale air quality forecast and mitigation of climate change all look promising!
Thank you!