PAHs from fossil fuels and biomass burning: regional and global atmospheric transport perspectives

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Jianmin Ma
Peking University
Lanzhou University

Credits to Huang Tao, Zhao Yuan, Chen Kaijie, and Lian Lulu
Why PAHs/BaP?

- Primary sources are fossil fuel combustion (anthropogenic) and wildfire biomass burning (natural)

- Listed by the UNCEC Convention on Long Range Transboundary Air Pollution Protocol in Persistent Organic Pollutants (POPs)

- Significant health effect with mutagenic properties and carcinogenicity, major toxic component in PM$_{2.5}$

- BaP is listed in ambient air quality standards like PM$_{2.5}$, Pb…
Outlines

➢ Models and emission inventories

➢ PAH emissions: wildfires, fossil fuel combustion, and others

➢ Impact: wildfires vs fossil fuel combustion from global to local

➢ Model intercomparison and validation
Models and emission inventories

Models:
- CanMETOP (regional and global, PKU and Lzu)
- WRF-CMAQ (local and regional, NCAR and USEPA)
- GEOS-Chem (global, Harvard Uni)

Emission inventories:
- PKU-FUEL (global 0.1° × 0.1° lat/lon monthly, 1960-2017, http://inventory.pku.edu.cn/)
- Lzu-WILDFIRE (global 0.25° × 0.25° and 1° × 1° lat/lon, 2001-2020, http://kleppc.lzu.edu.cn/)
PAH emissions: wildfires, fossil fuel combustion, and others

Emission types

- **Anthropogenic emission** from fossil fuel combustion
- **Natural emission** from wildfire biomass burning
- **Secondary emission** from environmental reservoirs where chemicals are accumulated from the past
- **Virtual emission** embodied in trade
Gridded global BaP emission inventories from fossil fuel and forest fire

Mean BaP annual emissions (kg/year) from fossil fuel combustion averaged from 2001 to 2014 (PKU fuel)

Mean BaP annual emissions (kg/year) from forest biomass burning averaged from 2001 to 2014

- Russian emissions were highest, followed by Canada and Europe

Luo, et al., EP, 2020
Wu, et al., EI, 2022
Huang et al., NPJ Clim Atmos., 2022 (under review)
Annually averaged BaP emissions from forest fire (a) and grassland fire (b) from 2001 to 2020

Global BaP emissions from forest fire and grassland fire from 2001 to 2020

Monthly averaged BaP emissions from forest fire and grassland fire from 2001 to 2020

From updated carbon stocks and burned areas till 2020
Virtual emission and emission flows embodied in trade

Black carbon virtual emissions embodied in global trade

Lian et al. ES&T, 2021; Du et al., 2022, to be submitted
Impact: wildfires vs fossil fuel combustion from global to local

I. African wildfires
II. Arctic and boreal forest fires
III. Arctic and southward shift of global supply chain
IV. India’s Green Revolution
Global and regional BaP contamination from all sectors

BaP concentrations averaged from 1991 to 2014

Regional BaP concentrations from 1991 to 2014

Huang et al., Environ. Sci. Ecotech. 2022 (under review)
Modeled global annual mean BaP air concentrations from PKU Fuel and wildfires

- Annual mean BaP air concentrations from 2001 to 2014 subject to BaP emission from PKU-FUEL
  - In Africa, BaP emission from natural sources (wildfire) was high than anthropogenic emission
  - Wildfire emitted BaP alone contributed ~50% to its global total from all sectors

- Annual mean BaP air concentrations from 2001 to 2014 subject to wildfire biomass burning

Wu et al., Environ. Int. 2022
I. African wildfires-emitted BaP

- **definite risk** with the LCR level $\geq 1 \times 10^{-4}$ regime under the WHO standard

- **high risk** ($10^{-5} < \text{LCR} \leq 10^{-4}$) in Sub-Saharan Africa

Particle phase BaP in Africa averaged from 2001 to 2014

High BaP level in Sub-Saharan Africa
Source-receptor relationships

Source apportionment approaches in a chemical transport model:
- Tagging method
- Brute-Force/Zero Out methods
- Tracer method (CMAQ)

- African BaP emissions from wildfires contribute ~18% to southern Europe in autumn
- ~3% to South America in winter

Percentage contribution of modeled BaP concentrations from African wildfire biomass burning to different regions in different seasons of 2010 (zero out approach)

Zhao et al., ES&T, 2019; Luo et al., EP, 2020; Wu et al., Environ. Int., 2022
Percent contribution (%) of the boreal forests in Canada, European, and Russia to the mean BaP air concentrations averaged over the period from 2003 to 2014 at the three Arctic sampling sites.

\[ C_{ij} = \frac{A_{ij}}{A_j}, \quad A_j = \sum_i A_{ij}. \]

- northern boreal forest biomass burning contributed over 90% to the modeled BaP at the high Arctic Alert site. At the low Arctic site of Pallas, wildfires contributed 7% to the mean BaP.
- the effect of northern boreal forest biomass burning on BaP concentrations was enhanced toward higher latitudes in the Arctic.

MODIS monitored total fire points from 2000 to 2016.

_Luo et al., EP, 2020_
Factors contributing to Arctic PAH pollution using deep learning and CTM

- Forest fires played a significant role in the changes in PAH concentrations in the high Arctic, followed by air temperature and meridional wind fields.
- Russia was identified to be a major source, mostly from wildfires.

Zhao et al., ES&T, 2019
III. Global industry relocation reduces Arctic PAH contamination virtual emission impact

- Southward shift of supply chain reduced the contribution of European and North American emission to Arctic PAH pollution and increased the contribution of Asian emission

- Southward shift of supply chain reduced Arctic BaP pollution by up to 60% from industrial and fossil-fuel combustion sources

Lian et al. ES&T, 2021
IV. BaP pollution and human risk associated with India’s Green Revolution (1965-2015)

- GR promotes crop yields
- Crop yields enhances indoor and outdoor crop residue biomass burning
- Crop residue BB results in significant growing PAH/BAP emissions and environmental contamination
- Causing increasing lifetime lung cancer risk (ILCR)

Death rates, age-adjusted lung cancer incidence rates (LCIRs), and ILCR (1990-2015)

Huang et al., 2022, NPJ Atmos & Clim., under review
Model inter-comparison and validation

1. Source – receptor relationship from all emission sectors (Tagging method)
   Assessing contribution from source locations (local vs long-range transport)

2. Source – receptor relationship from selected emission sectors (fossil fuel combustion, wildfire biomass burning, crop biomass burning, industry – (Brute Force/zero out methods)
   Assessing contribution from source sectors (local vs long-range transport)
Emission and model validation

Sampled ambient PAH concentration data are available for share

Modeled and measured BaP air concentration (ng/m^3) across the globe from 1991 to 2014

Modeled and measured BaP air concentration (ng/m^3) across Europe from 2001 to 2019

China and India
Thank you!