Evaluating the Impact of Future Emissions and Climate on Global PAH Transport

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Background

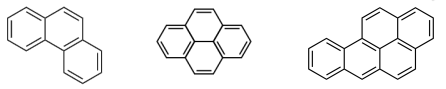
Polycyclic aromatic hydrocarbons (PAHs) are mutagenic byproducts of the combustion of organic material. PAHs can travel long distances in the atmosphere to remote locations, such as the Arctic, where they deposit and pose health risks. In this study, we evaluate the relative influences of a changing climate and anthropogenic emissions on global and Arctic atmospheric PAH concentrations. We use an atmospheric chemical transport model to simulate transport under the following conditions:

	Anthropogenic Emisisons	
Climate	*Present*	*Future*
Present	**Control**	**"PCFE"**
Future	**"FCPE"**	**"FCFE"**

In addition, we simulate an additional scenario (**"FCFE_Ship"**) similar to FCFE but including PAH emissions from projected future Arctic shipping activity.

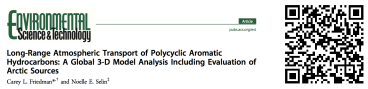
We simulate the transport of three PAHs that have different gas-particle partitioning to capture a range of behaviors:

100% gas ⟷ 100% particulate



Phenanthrene (PHE) | Pyrene (PYR) | Benzo[a]pyrene (BaP)

For a full background on the development, evaluation, and use of our model, please refer to the following publication:



In addition to processes described in the paper, our model includes re-volatilization of previously deposited PAHs from soil and vegetation. The importance of the sum of these "secondary emissions" is greater for PAHs that exist primarily as gases:

	PHE	PYR	BaP
Secondary/Total Emissions	16%	8%	0.01%

Revolatilization fluxes are impacted by temperature changes, primarily via changes in equilibrium partition coefficients (*K*) following the van't Hoff equation:

$$K_{soil-air}(T_2) = K_{soil-air}(T_1) \times e^{\frac{-\Delta H}{R}\left(\frac{1}{T_2}-\frac{1}{T_1}\right)}$$

In this example, *K* is the partition coefficient between soil and air (unitless), T_1 and T_2 are reference and changed absolute temperatures (K), respectively, ΔH is the enthalpy of phase change (kJ/mol) and *R* is the ideal gas constant (kJ/mol/K).

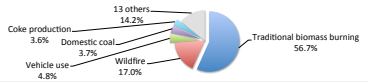
REFERENCES: [1]Zhang, Y.; Tao, S. 2009. Global atmospheric emission inventory for polycyclic aromatic hydrocarbons (PAHs) for 2004. *Atmos. Environ.*, 43:812-819. [2]World Energy Outlook 2010. International Energy Agency. [3]Shen et al. 2011. Global time trends in PAH emissions from motor vehicles. *Atmos. Environ.* 45:2267. [4]World Energy Outlook 2011. International Energy Agency. [5]Liu et al. 2009. Trends in global wildfire potential in a changing climate. *Forest Ecol. Manag.* 259:685. [6]Malevsky-Malevich et al. 2008. An assessment of potential change in wildfire activity in the Russian boreal forest zone induced by climate warming during the twenty-first century. *Climatic Change*. 86:463. [7]Peters et al. 2011. Future emissions from shipping and petroleum activities in the Arctic. *Atmos. Chem. Phys.* 11:5305.

Methodology

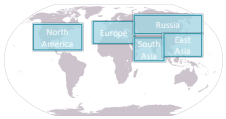
- Control

We use the 2004 global emissions inventory of Zhang and Tao[1] and present day (mean of 1997-2003) climate calculated by the NASA GISS GCM.

Top global source activities for U.S. EPA Σ16PAH (2004):

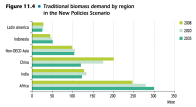


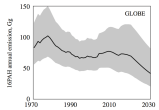
- PCFE: We scale 2004 anthropogenic emissions to 2050 in the following control regions:



Emissions from each top source are scaled according to a related activity. Overall, **emissions in each region decline**:

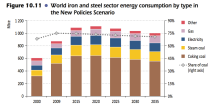
Traditional biomass and Domestic coal burning } Scale with the IEA's[2] projections for traditional biomass demand





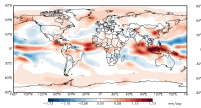
Vehicle use } Scale with Shen et al.[3] projections of vehicular PAH emissions

Coke production } Scale with the IEA's[4] projections for energy consumption in the iron and steel production sector

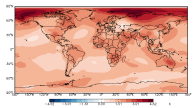


- FCPE: We use NASA GISS GCM projected meteorology (mean of 2047-2053 assuming IPCC A1B emissions) and scale wildfire emissions according to wildfire index change projections:

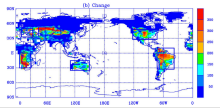
Precipitation ↑ 5% compared to present climate



Temperature ↑ 1.6 C compared to present climate

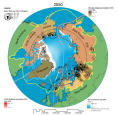


Wildfire } Scale according to global and regional fire index change projections (Liu et al.[5] and Malevsky-Malevich et al.[6])



- FCFE: We combine anthropogenic emissions scaling from PCFE and the climate and wildfire emissions scaling from FCPE for an overall future scenario

- FCFE_Ship:



Projected black carbon emissions from increased shipping from transit and oil/gas extraction in the Arctic (Peters et al.[7]) are scaled with PAH emission factors to estimate future BaP emissions. These emissions are added to the FCFE scenario.

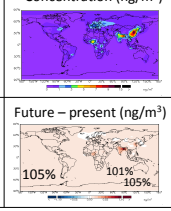
ACKNOWLEDGMENTS: Leading Technology and Policy Initiative (MIT); Yanxu Zhang (U. Washington); Eric Leibensperger (MIT); NSF Atmospheric Chemistry Grant 1053648

Results

Future emissions decrease global PAH concentrations, but climate impacts depend on gas-particle partitioning behavior

For **PHENANTHRENE**, we see a **decrease in mean global concentrations from future emissions and an increase from future climate**

	Emissions	
Climate	*Present*	*Future*
Present	Concentration (ng/m³)	Future – present (ng/m³)
Future	Future – present (ng/m³)	Future – present (ng/m³)



RED: Increasing concentrations
BLUE: Declining concentrations

Percent global mean concentration of control

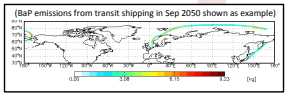
Percent Arctic mean concentration of control

Percent mid-latitude mean concentration of control

For **PYRENE** and **BENZO[a]PYRENE**, we also see **decreases in global mean concentrations from future emissions** (even more so than for PHE).
In a **future climate**, we see balanced increases and decreases for **PYR** that result in a **negligible net change**; for **BaP**, **future climate causes a small decrease** in the global mean:

	Emissions			
	PYRENE		BENZO[a]PYRENE	
Climate	*Present*	*Future*	*Present*	*Future*
Present	Mean concs (ng/m³):	% of control mean:	Mean concs (ng/m³):	% of control mean:
Global	0.048	77%	2.4e-2	67%
Arctic	0.036	89%	7.8e-3	86%
Mid-latitude	0.097	73%	5.2e-2	65%
Future	% of control mean:	% of control mean:	% of control mean:	% of control mean:
Global	102%	80%	97%	66% → 66%
Arctic	101%	91%	99%	86% → 89%
Mid-latitude	102%	76%	98%	64% → 64%

Applying **future Arctic shipping BaP emissions** to the FCFE simulation leads to a **4% increase in Arctic concentrations** and a **0.1% increase in the global mean**



Global concentrations in a future climate are a balance between increasing deposition and secondary emissions

	BaP	PYR	PHE
Change in deposition	↑ 4%	↑ 5%	↑ 5%
Change in % secondary of total emissions	0 % (0.01% to 0.01%)	↑2% (8% to 10%)	↑3% (16% to 19%)

Projected future emissions will reduce global PAH concentrations, but there will be a "climate penalty" that lessens gas-phase reductions

	BaP	PYR	PHE
Future climate penalty (reduction in expected decrease	-1 %	+13%	+30%