Global cancer risk from unregulated polycyclic aromatic hydrocarbons

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GeoHealth

RESEARCH ARTICLE

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Key Points:

- Benzo[a]pyrene is a small contributor to human cancer risk of polycyclic aromatic hydrocarbons (PAHs) worldwide (11%)
- Using benzo[a]pyrene as a surrogate compound leads to erroneous conclusions about high-risk populations and the importance of uncertain chemical processes
- Science and policy could be improved by considering a wider group of both emitted PAHs as well as their degradation products

Global Cancer Risk From Unregulated Polycyclic Aromatic Hydrocarbons

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With Jamie Kelly (former MIT postdoc; now University College London) and others

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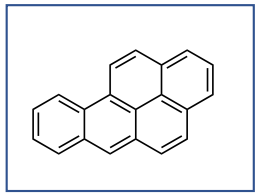
What are PAHs?

Burning of organic matter

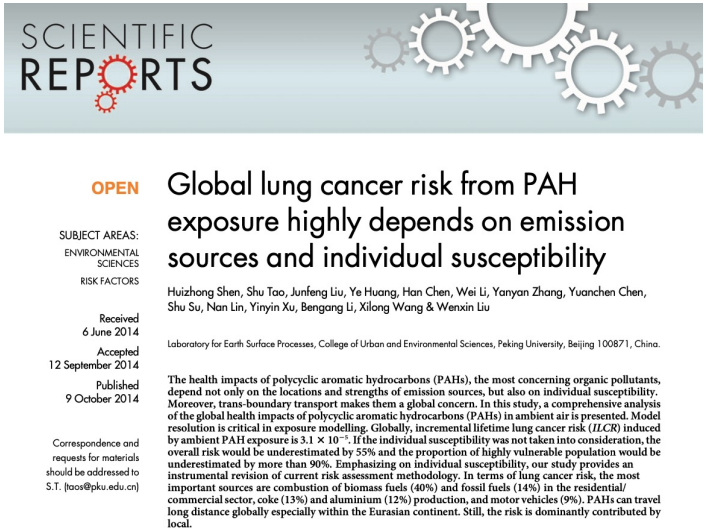
emission



inhalation

DNA damage

cancer



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SUBJECT AREAS:
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Global lung cancer risk from PAH exposure highly depends on emission sources and individual susceptibility

Huizhong Shen, Shu Tao, Junfeng Liu, Ye Huang, Han Chen, Wei Li, Yanyan Zhang, Yuanchen Chen, Shu Su, Nan Lin, Yinyin Xu, Bengang Li, Xilong Wang & Wenxin Liu

Laboratory for Earth Surface Processes, College of Urban and Environmental Sciences, Peking University, Beijing 100871, China.

The health impacts of polycyclic aromatic hydrocarbons (PAHs), the most concerning organic pollutants, depend not only on the locations and strengths of emission sources, but also on individual susceptibility. Moreover, trans-boundary transport makes them a global concern. In this study, a comprehensive analysis of the global health impacts of polycyclic aromatic hydrocarbons (PAHs) in ambient air is presented. Model resolution is critical in exposure modelling. Globally, incremental lifetime lung cancer risk (*ILCR*) induced by ambient PAH exposure is 3.1×10^{-5}. If the individual susceptibility was not taken into consideration, the overall risk would be underestimated by 55% and the proportion of highly vulnerable population would be underestimated by more than 90%. Emphasizing on individual susceptibility, our study provides an instrumental revision of current risk assessment methodology. In terms of lung cancer risk, the most important sources are combustion of biomass fuels (40%) and fossil fuels (14%) in the residential/commercial sector, coke (13%) and aluminium (12%) production, and motor vehicles (9%). PAHs can travel long distance globally especially within the Eurasian continent. Still, the risk is dominantly contributed by local.

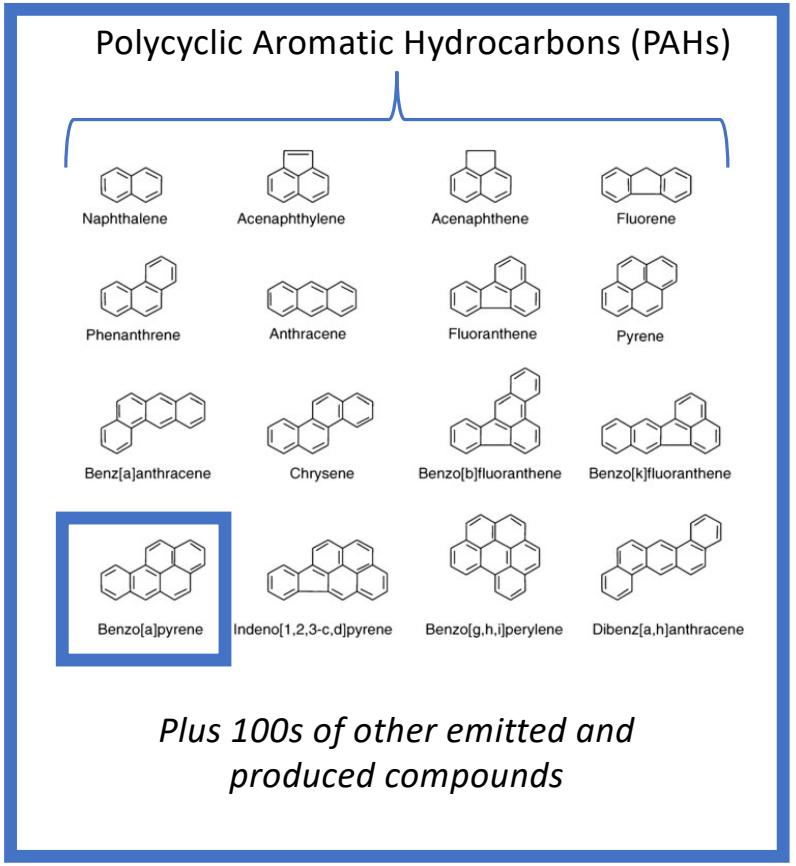
Shen et al., *Scientific Reports*, 2014

Global lung cancer risk results from exposure to PAHs, including local and long-range transport

Highly regulated pollutants nationally and internationally

Challenges of assessing and regulating a class of compounds

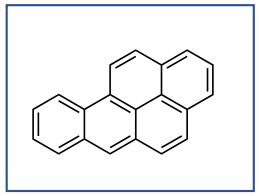
- Hundreds of different PAHs
- **Benzo(a)pyrene** often used as a proxy or marker for the entire mixture
  - WHO, UK, EU, Canada
  - Previous measurement studies estimate it comprises *40-80%* of overall PAH risk
- US EPA prioritizes 16 emitted PAHs
- 4 PAHs used as indicators of emissions for Convention on Long-Range Transboundary Air Pollution (UNECE)



Atmospheric reactions produce additional PAHs

Burning of organic matter

emission



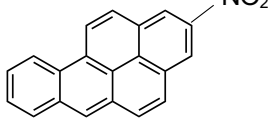
inhalation

DNA damage

cancer

Atmospheric reactions

nitro-PAHs

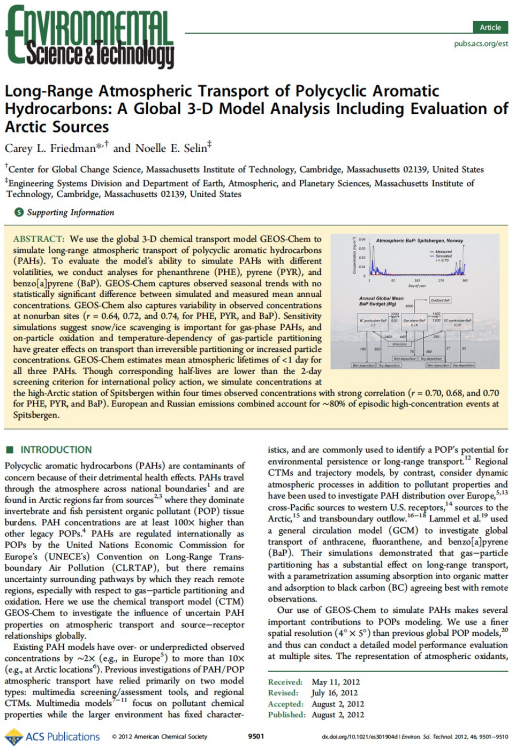


- *Increasing attention in environmental measurement and toxicology to degradation products and PAHs beyond the 16 EPA compounds (including higher molecular weight PAHs)*

- *Oxidation products can be orders of magnitude (10-1000x) more toxic than parent compounds*

What is the relative importance of different PAHs to global cancer risk?

Global models of PAHs include few species



Environmental Science & Technology

Article

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Long-Range Atmospheric Transport of Polycyclic Aromatic Hydrocarbons: A Global 3-D Model Analysis Including Evaluation of Arctic Sources

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Supporting Information

ABSTRACT: We use the global 3-D chemical transport model GEOS-Chem to simulate long-range atmospheric transport of polycyclic aromatic hydrocarbons (PAHs). To evaluate the model's ability to simulate PAHs with different volatilities, we conduct analyses for phenanthrene (PHE), pyrene (PYR), and benzo[a]pyrene (BaP). GEOS-Chem captures observed seasonal trends with no statistically significant difference between simulated and measured mean annual concentrations. GEOS-Chem also captures variability in observed concentrations at nonurban sites (r = 0.64, 0.72, and 0.74, for PHE, PYR, and BaP). Sensitivity simulations suggest snow/ice scavenging is important for gas-phase PAHs, and on-particle oxidation and temperature-dependency of gas-particle partitioning have greater effects on transport than irreversible partitioning or increased particle concentrations. GEOS-Chem estimates mean atmospheric lifetimes of <1 day for all three PAHs. Though corresponding half-lives are lower than the 2-day screening criterion for international policy action, we simulate concentrations at the high-Arctic station of Spitsbergen within four times observed concentrations with strong correlation (r = 0.70, 0.68, and 0.70 for PHE, PYR, and BaP). European and Russian emissions combined account for ~80% of episodic high-concentration events at Spitsbergen.

■ INTRODUCTION

Polycyclic aromatic hydrocarbons (PAHs) are contaminants of concern because of their detrimental health effects. PAHs travel through the atmosphere across national boundaries[1] and are found in Arctic regions far from sources[2,3] where they dominate invertebrate and fish persistent organic pollutant (POP) tissue burdens. PAH concentrations are at least 100× higher than other legacy POPs.[4] PAHs are regulated internationally as POPs by the United Nations Economic Commission for Europe's (UNECE's) Convention on Long-Range Transboundary Air Pollution (CLRTAP), but there remains uncertainty surrounding pathways by which they reach remote regions, especially with respect to gas−particle partitioning and oxidation. Here we use the chemical transport model (CTM) GEOS-Chem to investigate the influence of uncertain PAH properties on atmospheric transport and source−receptor relationships globally.

Existing PAH models have over- or underpredicted observed concentrations by ~2× (e.g., in Europe[5]) to more than 10× (e.g., at Arctic locations[6]). Previous investigations of PAH/POP atmospheric transport have relied primarily on two model types: multimedia screening/assessment tools, and regional CTMs. Multimedia models[7−11] focus on pollutant chemical properties while the larger environment has fixed characteristics, and are commonly used to identify a POP's potential for environmental persistence or long-range transport.[12] Regional CTMs and trajectory models, by contrast, consider dynamic atmospheric processes in addition to pollutant properties and have been used to investigate PAH distribution over Europe,[5,13] cross-Pacific sources to western U.S. receptors,[14] sources to the Arctic,[15] and transboundary outflow.[16−18] Lammel et al.[19] used a general circulation model (GCM) to investigate global transport of anthracene, fluoranthene, and benzo[a]pyrene (BaP). Their simulations demonstrated that gas−particle partitioning has a substantial effect on long-range transport, with a parametrization assuming absorption into organic matter and adsorption to black carbon (BC) agreeing best with remote observations.

Our use of GEOS-Chem to simulate PAHs makes several important contributions to POPs modeling. We use a finer spatial resolution (4° × 5°) than previous global POP models,[20] and thus can conduct a detailed model performance evaluation at multiple sites. The representation of atmospheric oxidants,

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Friedman and Selin (2012, ES&T): BaP, phenanthrene, pyrene

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Global lung cancer risk from PAH exposure highly depends on emission sources and individual susceptibility

Huizhong Shen, Shu Tao, Junfeng Liu, Ye Huang, Han Chen, Wei Li, Yanyan Zhang, Yuanchen Chen, Shu Su, Nan Lin, Yinyin Xu, Bengang Li, Xilong Wang & Wenxin Liu

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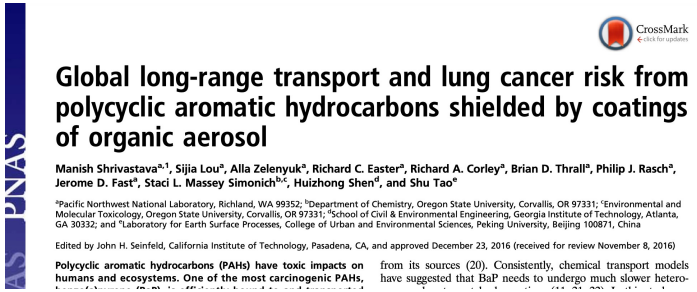
The health impacts of polycyclic aromatic hydrocarbons (PAHs), the most concerning organic pollutants, depend not only on the locations and strengths of emission sources, but also on individual susceptibility. Moreover, trans-boundary transport makes them a global concern. In this study, a comprehensive analysis of the global health impacts of polycyclic aromatic hydrocarbons (PAHs) in ambient air is presented. Model resolution is critical in exposure modelling. Globally, incremental lifetime lung cancer risk (*LLCR*) induced by ambient PAH exposure is 3.1×10^{-5}. If the individual susceptibility was not taken into consideration, the overall risk would be underestimated by 55% and the proportion of highly vulnerable population would be underestimated by more than 90%. Emphasizing on individual susceptibility, our study provides an instrumental revision of current risk assessment methodology. In terms of lung cancer risk, the most important sources are combustion of biomass fuels (40%) and fossil fuels (14%) in the residential/commercial sector, coke (13%) and aluminium (12%) production, and motor vehicles (9%). PAHs can travel long distance globally especially within the Eurasian continent. Still, the risk is dominantly contributed by local.

Shen et al. (2014, Sci Rep): BaP only



PNAS

CrossMark

Global long-range transport and lung cancer risk from polycyclic aromatic hydrocarbons shielded by coatings of organic aerosol

Manish Shrivastava[a,1], Sijia Lou[a], Alla Zelenyuk[a], Richard C. Easter[a], Richard A. Corley[a], Brian D. Thrall[a], Philip J. Rasch[a], Jerome D. Fast[a], Staci L. Massey Simonich[b,c], Huizhong Shen[d], and Shu Tao[e]

[a]Pacific Northwest National Laboratory, Richland, WA 99352; [b]Department of Chemistry, Oregon State University, Corvallis, OR 97331; [c]Environmental and Molecular Toxicology, Oregon State University, Corvallis, OR 97331; [d]School of Civil & Environmental Engineering, Georgia Institute of Technology, Atlanta, GA 30332; and [e]Laboratory for Earth Surface Processes, College of Urban and Environmental Sciences, Peking University, Beijing 100871, China

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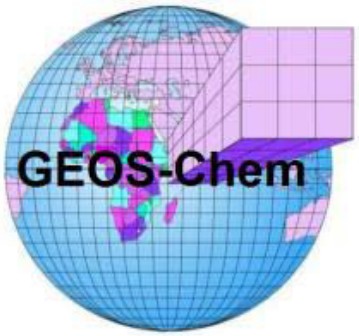
Polycyclic aromatic hydrocarbons (PAHs) have toxic impacts on humans and ecosystems. One of the most carcinogenic PAHs,

from its sources (20). Consistently, chemical transport models have suggested that BaP needs to undergo much slower hetero-

Shrivastava et al. (2017, PNAS): BaP only

Our approach: A global model to examine the relative impacts of different PAHs

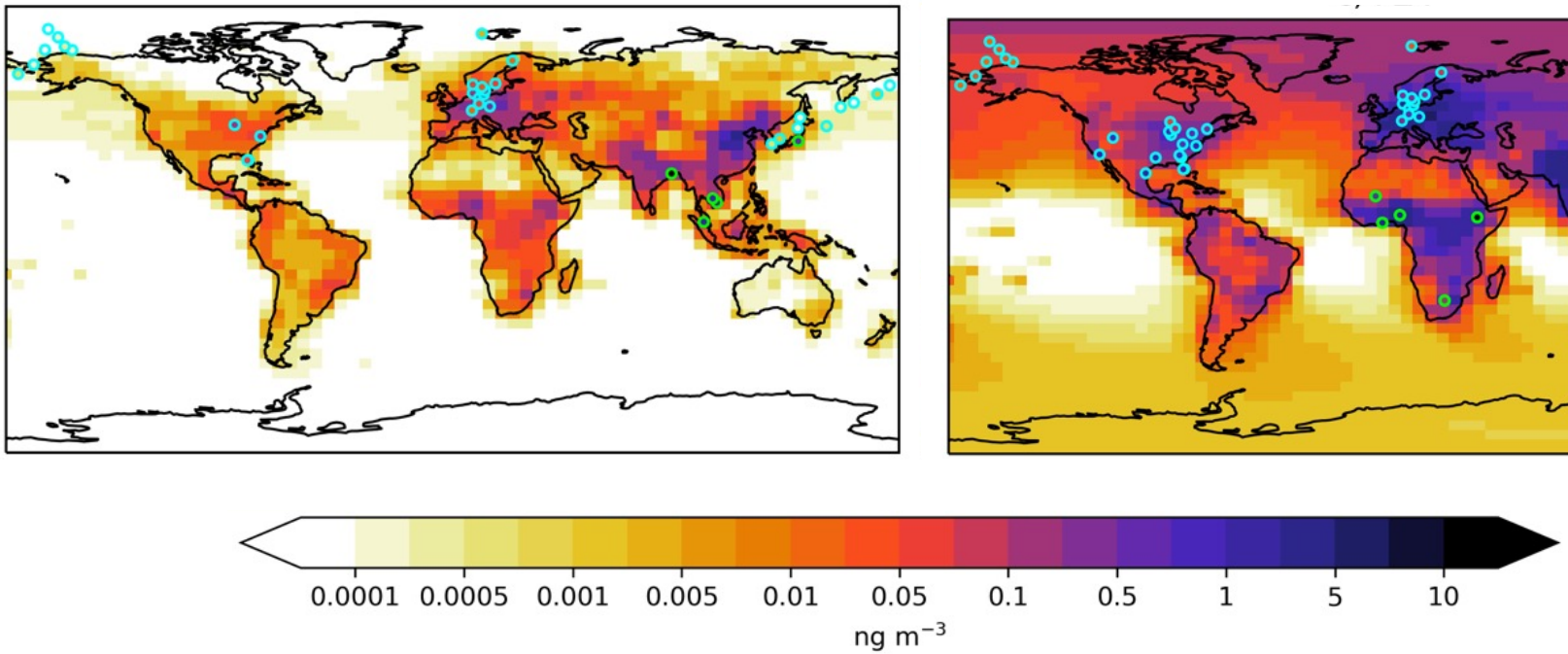


- Global-scale, 3-dimensional atmospheric chemistry and transport model
- Global emissions inventory for 16 PAHs (from Shen et al. 2013)
- Developed chemical mechanism for nitro- and dinitro-PAH formation and included in model (**48 species**)
- Evaluated vs. global database of atmospheric measurements (plus extensive uncertainty analysis)
- Used animal-based toxicity approach to avoid "double counting" cancer causes from multiple PAHs
  - Compared with epidemiological estimates of cancer risk

Finding #1: BaP is a poor indicator compound

BAP – the PAH usually used to represent the entire mixture

Fluoranthene – another emitted PAH



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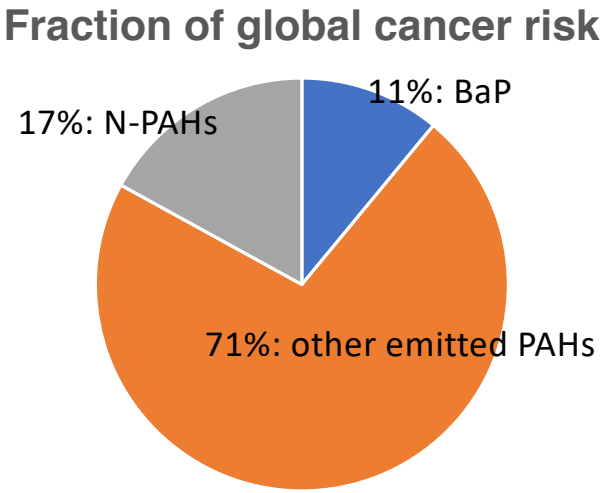
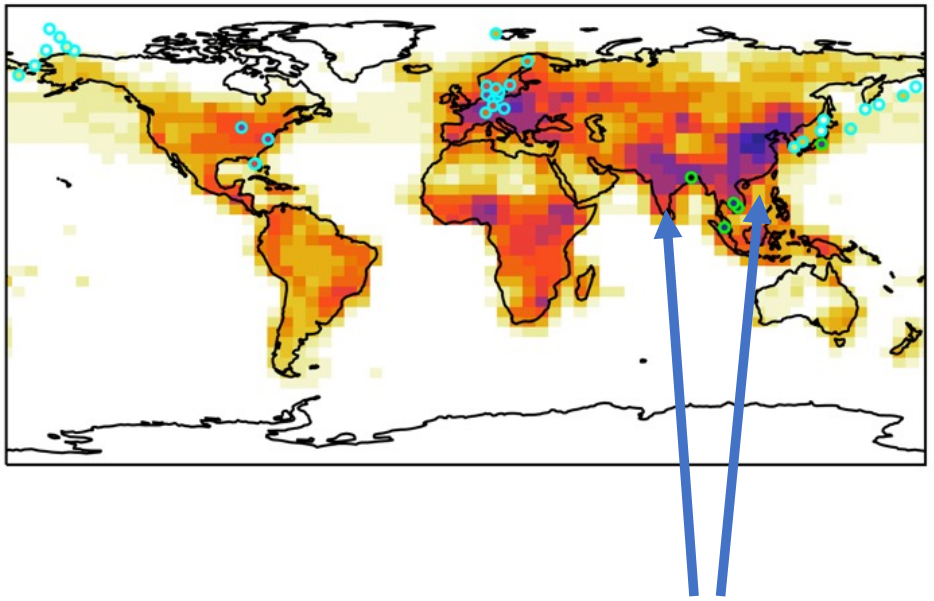


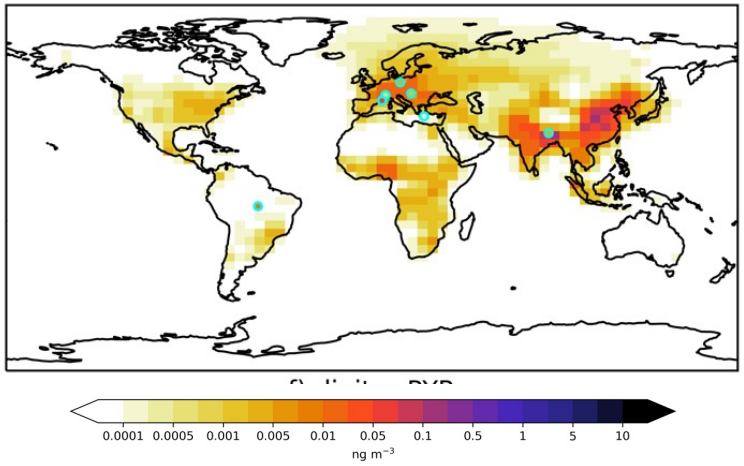
Compare with 40-80% estimate for BaP from measurement studies



*Relative risk differences: BaP suggests **3.5x** difference in risk, our method suggests **12x***

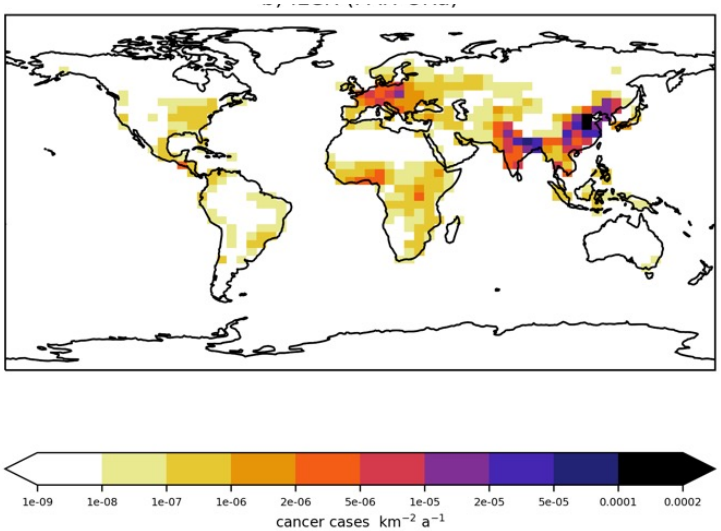
Finding #2: PAH degradation products contribute substantially to cancer risk (≳BaP)

Nitro-PYR – the oxidation product of PYR



- *Likely even more than we calculate here, as we don't account for oxy-PAHs and other degradation products, and limited info on toxicity for those included (12 out of 32)*
- *Unregulated, and largely unmonitored*
- *Different distribution than parent compounds*

Implications: What are the best ways to reduce overall risk?



- Increased scientific and regulatory attention to degradation products and PAHs other than BaP – at global scale
- More measurements needed to quantify exposure
- Monitoring changes in BaP will not be an effective indicator of overall change in risk
- Changed identification of high priority source reductions?
- Better understanding of exposure to mixtures and their impacts