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Huizhong Shen

Polycyclic Aromatic Hydrocarbons

Their Global Atmospheric Emissions, Transport, and Lung Cancer Risk

Doctoral Thesis accepted by
College of Urban and Environmental Sciences,
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Supervisor's Foreword

As a group of widely spread contaminants with potential impact on human health, polycyclic aromatic hydrocarbons (PAHs) are among the top priority pollutants in many countries especially developing ones where solid fuels are extensively used in industrial and residential sectors. Moreover, pollution of PAHs is a global issue not only because they are emitted everywhere, but also because they can travel among countries or even continents due to strong long-range transport potential. To this stage, quantitative information on sources, fate, and health impact of PAHs on a global scale is not available, resulting in difficulty for policy-makers to formulate abatement strategies. In this thesis, Dr. Huizhong Shen has provided an integrated analysis on emissions, transport, inhalation exposure (ambient air), and subsequently lung cancer risks of PAHs on a global scale. The transport modeling and the exposure risk assessment were conducted using benzo[*a*]pyrene (BaP) as an indicator for PAHs. The emission inventory features in high spatial and temporal resolutions as well as detailed source information, based on a lately compiled fuel consumption data product (PKU-FUEL, which can be freely downloaded together with a series of emission inventories at inventory.pku.edu.cn). By using subnational, instead of national fuel data, the spatial bias caused by conventional spatial disaggregation assuming even per person fuel consumptions within countries was able to be reduced. Meantime, the detailed source information enables the author to distinguish relative contributions of various sectors on the PAH exposure induced lung cancer morbidity. The simulated near-surface air concentrations of BaP were satisfactorily validated after the calculated concentrations were spatially downscaled to match the grid resolution with the observation sites. It was estimated that the PAH exposure induced incremental lifetime lung cancer risk in 2007 was 3.1×10^{-5} globally, with hot spots in South and East Asia. One of the important findings is that the overall risk would be underestimated by approximately 50 % (1.4×10^{-5}) if individual (mostly genetic) susceptibility was not taken into consideration. Moreover, the susceptibility leads to significant increases in the size of both high and low vulnerable populations. Among various emission sources,

combustion of solid fuels including coal, wood, and crop residues in residential sector contributed more than half of the overall risk. Although PAHs can travel long distance globally, majority of PAH exposure induced lung cancer morbidity was due to local emissions.

Beijing Prof. Shu Tao
January 2016

Abstract

Environmental polycyclic aromatic hydrocarbons (PAHs) are mainly emitted from incomplete combustion of fuels, waste burning, open biomass burning, and gas leaking in industrial processes. PAHs are of great concern because of their adverse health effects on human. This study estimated global atmospheric emissions of 16 PAH compounds from 69 emission source types during the period from 1960 to 2030. Using regression analysis and technology split method, country- and time-specific emission factors were derived. Based on high-resolution fuel combustion inventory, historical energy data, and six IPCC SRES scenarios, high-resolution PAHs emission inventory at a resolution of $0.1° \times 0.1°$ and time trend of historical emissions at a country level were developed separately. Using this inventory and the Model for Ozone and Related Chemical Tracers (MOZART-4), global transport of benzo[*a*]pyrene (BaP), one of the high molecular weight PAH compounds, was performed and evaluated with observations. Global near-surface concentrations of BaP were generated following a spatial downscaling method. This concentration distribution was used to evaluate lung cancer risk of global population being induced by inhalation exposure to ambient PAHs. Influence of individual susceptibility, contributions of various emission sources, and transboundary pollution were also quantified.

It is estimated that the global total annual atmospheric emission of 16 PAHs in 2007 was 655 Gg (521−816 Gg, as interquartile range), with residential/commercial biomass burning (52.4 %), petroleum consumption by on-road motor vehicles (16.5 %), and open-field biomass burning (agricultural waste burning, deforestation, and wildfire, 12.5 %) as the major sources. South, East, and Southeast Asia were the regions with the highest PAH emission densities, contributing half of the global total PAH emissions. Among the global total PAH emissions, 7.7 % of the emissions were in the form of high molecular weight carcinogenic compounds and the percentage of the carcinogenic PAHs was higher in developing countries (8.0 %) than in developed countries (6.4 %), due to the differences in energy structures and the disparities of technology. The potential health impact of the PAH emissions was greatest in the parts of the world with high

anthropogenic PAH emissions, because of the overlap of the high emissions and high population densities.

Global total PAH emissions peaked at 869 Gg in 1978 and declined gradually to 624 Gg in 2008. Total PAH emissions from developed countries peaked at 261 Gg in the early 1970s and decreased to 51 Gg in 2008. Emissions from developing countries peaked at 663 Gg in the early 1990s and decreased to 558 Gg in 2008. Simulation of PAH emissions from 2009 to 2030 revealed that PAH emissions in developed and developing countries would decrease by 46−71 % and 48−64 %, respectively, based on the six IPCC SRES scenarios.

Based on the inventory, global chemical transport of BaP was performed, distribution of global BaP concentrations was derived with downscaling method, and lung cancer risk being induced by PAHs exposure was evaluated using BaP as an indicator. The results indicate that global lung cancer risk represented as Incremental Lifetime Lung Cancer Risk (*ILCR*) was 3.1×10^{-5}, individual susceptibility strongly influenced the outcomes of quantitative risk assessment. If the individual susceptibility was not taken into consideration, the risk would be underestimated by 55 %, and the fraction of the most vulnerable population ($ILCR > 3.1 \times 10^{-5}$) would be underestimated by more than 90 %. Further analysis revealed a significantly positive correlation ($p < 0.1$) between the country-specific *ILCR*s and lung cancer incidences. Contributions of individual sources to the overall risk depend not only on the emission strengths but also on the proximities to people.

Globally, biomass fuel burning in residential/commercial sector contributes 40 % of the total *ILCR*, followed by residential/commercial fossil fuel combustion (14 %), coke production (13 %), primary aluminium production (12 %), and motor vehicles (9 %). *ILCR*s also vary dramatically among populations at different risk levels. A small fraction (1.7 %) of the population facing high risk ($ILCR > 3 \times 10^{-4}$) is largely because of exposure to emissions from coke and aluminium productions. Emissions from residential solid fuel combustion contribute mainly to the population at risk levels between 3×10^{-8} and 1×10^{-4}. These results provide a sound scientific basis for abatement strategy formulation. Globally, residential biomass burning causes the largest overall lung cancer risk and should be the top priority in the emission abatement. On the other hand, control of emissions from motor vehicles and residential coal combustion with the highest *SILCR* is the most effective way of reducing risk. If the objective is to protect the most vulnerable people, emissions from coke and aluminium production should be addressed first. However, the specific strategies should be different among countries, depending on local emission and risk. For example, the overall risk in Russia is dominated by primary aluminium production, while motor vehicles are responsible for the risk of vulnerable populations in Indonesia. Of course, abatement costs should also be taken into account before the action plan is formulated.

It appears that interregional transport within the Eurasian continent is active. As a result of the westerly wind movement and lower air lose rate in high-latitude area, the transport of BaP from Western/Eastern Europe to the Former Soviet Union region represents the largest *ILCR* flow, leading to a 4.5×10^{-7} increase to local *ILCR* of the latter. The second largest movement occurs from East Asia to Southeast

Asia owing to the extraordinary emission intensity in the source region. The net exported risk (NER), defined as the difference between the exported and imported ILCR multiplied by regional population, is calculated for each region. Western/Eastern Europe (NER = 145), South Asia (NER = 57), and East Asia (NER = 42) are the main export regions of risk, with positive values of exported risk, while Southeast Asia (NER = −126), the former Soviet Union (NER = −92), and the Middle East (NER = −19) are risk passive recipients. Still, even with the active interregional transport, regional risks are predominantly caused by local emissions. The highest external contribution to local *ILCR* is merely 2.6 % (Southeast Asia).

Keywords Polycyclic aromatic hydrocarbons · Emission inventory · Global chemical transport modeling · Inhalation exposure · Lung cancer risk

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