Shift peaks of PAH-associated health risks from East Asia to South Asia and Africa in the future

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Key Points:

- Population-weighted global average BaP concentrations under all RCPs consistently exceeded the WHO-recommended limits from 2008 to 2050
- Peaks in PAH-associated ILCR shift from East Asia in 2008 to South Asia and Africa by 2050 mainly due to changes in traditional biofuel use
- Policies that encourage using clean energy and complete combustion technologies will help mitigate health risks from PAHs

Abstract

Polycyclic aromatic hydrocarbons (PAHs), emitted from combustion of biofuels and fossil fuels, are toxic compounds and known to cause lung-cancer. Integrating a global atmospheric chemistry model and plausible future emissions trajectories, we assess how global PAHs and their associated lung cancer risk will likely change in the future. Benzo(a)pyrene (BaP) is used as an indicator of cancer risk from PAH mixtures. From 2008 to 2050, the population-weighted global average BaP concentrations under all RCPs consistently exceeded the WHO-recommended limits, primarily attributed to residential biofuel use. In developing regions of Africa and South Asia, PAH-associated lung-cancer risk increased by 30-64% from 2008 to 2050, due to increasing use of traditional biofuels with population growth. With the stringent air quality policy, PAH lung-cancer risk substantially decreases by ~80% in developed countries. Climate change is likely to have minor effects on PAH lung-cancer risk compared with the impact of emissions.

Plain Language Summary

Polycyclic aromatic hydrocarbons (PAHs) are unavoidably derived from combustion processes, and are contaminants of global concern because they increase the risk of lung cancer and are detrimental to human health and the ecosystem. While high concentrations of PAHs were already measured in 2008, future changes in energy use, land use, and climate policy may alter the PAHs concentrations. In this work, we estimated how future changes in emissions and climate would affect PAH distribution and human health. We found the peaks of PAH-associated lung cancer risks are shifting from East Asia in 2008 to South Asia and Africa by 2050, due to increasing traditional solid biofuel use with rapid population growth. Our work implies that developing efficient combustion technologies and reducing traditional biomass fuels in the future are needed in South Asia and Africa to avoid the deterioration of air quality and human health.

1. Introduction

Polycyclic aromatic hydrocarbons (PAHs) are unavoidable byproducts from combustion processes involving organic matter. They are contaminants of global concern, and several PAHs are persistent organic pollutants in the atmosphere that increase risk of lung cancer in humans [*Boffetta et al.*, 1997; *Perera*, 1997; *Chen and Liao*, 2006; *Muir et al.*, 2019]. As one of the most carcinogenic PAHs, benzo(a)pyrene (BaP) is commonly used as an indicator of cancer risk from PAH mixtures [*Delgado-Saborit et al.*, 2011; *Boruvkova*, 2015; *IARC*]. High concentrations of BaP (>1 ng m^{-3}) have been measured in several megacities across Asia, Africa, Europe, and North America [*EMEP*; *IADN*; *Bostrom et al.*, 2002; *Shen et al.*, 2014; *Mu et al.*, 2018], with biofuel combustion dominating the global BaP emission budget. More than 60% of total atmospheric BaP emissions are from residential indoor biomass burning, while deforestation and wildfires contribute another 14% [*Shen et al.*, 2013].

To mitigate health risks from PAHs, we need to understand how these pollutant concentrations will change in the future. Although changes in energy and land use affect greenhouse gases (GHGs) and PAHs (including BaP), the main drivers of PAHs are predicted to differ from GHGs. For example, while residential biomass burning contributes substantially to BaP emissions, fossil fuel

use is the major contributor to GHGs. In 2010, traditional solid biofuel use was concentrated in developing regions of Africa, South Asia, and China, primarily in households for cooking, heating, and lighting [*O'Neill et al.*, 2014; *Bauer et al.*, 2017; *Tao et al.*, 2018; *Hailu and Kumsa*, 2021]. As the demand for biomass energy is projected to increase to meet increasing energy demand, and perhaps also to satisfy the Paris Agreement [*Rogelj et al.*, 2018], BaP emissions from biofuel use are challenging to reduce by the 2050s, especially in developing regions. Therefore, regulatory control policies aimed at improving air quality, human health, and socio-economic development could greatly change sectoral profiles and spatial distributions of BaP emissions and PAH-associated lung cancer risks.

Additionally, future climate changes may alter the atmospheric transport and lifetime of PAHs depending on PAH volatility and degradation. Although a recent study showed that the volatilization of particle-bound PAHs (such as BaP) is not significantly affected by climate warming [*Yu et al.*, 2019], atmospheric degradation is highly uncertain and may alter regional BaP distributions. Assuming black carbon (BC) captured most PAHs to prevent degradation, Friedman et al. (2014) reported a 3% decrease in global BaP concentrations due to climate change. However, recent laboratory studies have shown that viscous secondary organic aerosol (SOA) coatings can effectively protect BaP from rapidly ozone oxidation [*Ringuet et al.*, 2012; *Zhou et al.*, 2012; *Berkemeier et al.*, 2016]. Therefore, our previous work implemented a new modelling approach in a three-dimensional global atmospheric chemical transport model that accounts for the shielding of BaP by viscous SOA coatings [*Shrivastava et al.*, 2017]. Since the effectivity of SOA coatings is sensitive to changes in temperature and humidity [*Zelenyuk et al.*, 2012; *Shrivastava et al.*, 2017], it is unclear how the climate change will affect the future BaP concentrations.

RCP scenarios are multimodel global scenarios of greenhouse gases and air pollutants to span a range of future climate forcing levels [*Taylor et al.*, 2012]. Although previous modelling studies have investigated the relative importance of future emissions and future climate influence on air pollutants, they mainly focus on the aerosols and ozone concentrations [*Rogelj et al.*, 2014; *Silva et al.*, 2016; *Silva et al.*, 2017; *Nolte et al.*, 2018; *Y Zhang et al.*, 2018; *Fenech et al.*, 2021]. However, primary PAHs (including BaP) are not included in RCP. Using the novel treatment that shields PAHs by viscous SOA under cool/dry conditions, we examine how BaP concentrations could change in the future due to emissions/climate-change scenarios represented by various RCPs. In addition, this study helps to understand how future changes in climate policy, energy structure, and land use would affect future PAH-associated health risks.

2. Methods

2.1. Emissions

For 2008, we used the PKU 0.1° × 0.1° global BaP emission inventory [*Shen et al.*, 2013]. The PKU BaP emission inventory is divided into residential biofuel, residential fossil fuel, industry, transportation, agricultural waste burning (AWB), and open-fire biomass burning sectors (Table S1, Text S1), varying monthly by sector and region.

For each grid, future BaP emissions were generated assuming the same spatial and temporal trends as OC emission changes projected by different RCP scenarios [*van Vuuren et al.*, 2011] from 2008 to 2020 and 2050 (Figure S1, Text S1). We assumed that the ratio of BaP to OC, which varies by

sector and region, will be constant from 2008 to 2050 (Table S2). Based on the changes in OC emissions, we reversed the variations in BaP emissions from 1960 to 2014, and compared them with the realistic interannual BaP emissions. The global modified normalized mean biases of -3.4% (Figure S2) gives us more confidence to project future BaP emissions.

Additional details about emissions, including BC, OC, SO_2, NO_x, NH_3, and non-methane volatile organic compounds, are described in Text S1.

2.2. Model Overview and Simulation Design

We used the global Community Atmosphere Model, version 5.2 (CAM5), with a new PAH representation [*Shrivastava et al*., 2017] to simulate the global distribution of BaP. Gas-phase chemistry was represented by the Model for Ozone and Related Chemical Tracers chemical mechanism [*Emmons et al*., 2010]. The properties and processes of aerosol species for mineral dust, BC, primary organic aerosol, SOA, sea salt, and sulfate are included in the Modal Aerosol Module (MAM3) [*Liu et al*., 2012], with an updated chemical mechanism for SOA [*Shrivastava et al*., 2015]. The model includes gas-phase reactions of BaP with hydroxyl radicals (•OH) and heterogeneous reactions of particle-phase BaP with ozone and •OH radicals. Gas-particle partitioning of BaP is calculated by the poly-parameter linear free energy relationship model, which includes both absorption into organic aerosols and adsorption onto soot surfaces [*Shahpoury et al*., 2016]. Viscous SOA can significantly slow the heterogeneous oxidation of PAHs by shielding them from ozone oxidation, but this shielding is less effective over warm and/or humid locations [*Shrivastava et al*., 2017; *Mu et al*., 2018]. Wet and dry deposition of particle BaP is treated similar to those of other aerosol species in CAM5 [*Liu et al*., 2012].

We performed the following model simulations: (1) standard BaP simulations at 2008 level (2008_CTRL); (2) same as 2008_CTRL, except turn on different BaP source sectors (Table S1) one at a time (2008_sector); (3) future BaP simulations in 2020 and 2050 under RCP scenarios (RCPs_emis); (4) same as RCPs_emis, but turn on different BaP source sectors one at a time (RCPs_sector). All simulations above were performed for two years, with the first year for spin-up. Winds and temperature are nudged toward ERA-Interim data from 2007 to 2008.

RCP8.5 simulations were also performed to investigate the relative effects of emissions and climate change on BaP concentrations, including (5) BaP simulations at 2008 level (RCP8.5_2008); (6) same as RCP8.5_2008 except under future meteorological field (RCP8.5_2050_Clim); (7) BaP simulations at 2050 level (RCP8.5_2050). All RCP8.5 simulations were performed for four years, using CCSM4-simulated meteorology nudge from 2007 to 2010 and 2047 to 2050 [*NCAR*, 2011], representing past (2008) and future (2050) climatic conditions, respectively.

By comparing model-simulated results of 2008_sector and 2008_CTRL, RCP_sector and RCP_emis, we derive contributions from each BaP emission sector in 2008 and 2050, respectively. Differences between 2008_CTRL and RCP_emis simulations yield effects of future emissions on BaP concentrations. Furthermore, the difference between RCP8.5_2050 and RCP8.5_2008 represents the combined effect of future climate and emission changes on BaP concentrations, where the impact of climate change is reflected by comparing RCP8.5_2050_Clim and RCP8.5_2008 simulations.

2.3. Incremental lifetime cancer risk (ILCR)

The ILCR induced by exposure to PAHs in ambient air is calculated with the cancer slope factor (CSF), lifetime average daily dose (LADD), and a factor SUS describing individual susceptibility, respectively, depending on age, gender, ethnicity and geographic region as follows [*Shen et al.*, 2014]

$$ILCR = CSF \times LADD \times SUS = CSF \times \frac{C \times IR \times y}{BW \times LE} \times SUS$$

ILCR in this study is a population-weighted average and represents the maximum likelihood estimate; the unit for ILCR is one death per 100,000 persons.

3. Results

3.1. Variation in BaP concentrations due to changes in emissions

In this study, we estimated urban population exposure to BaP by downscaling global model BaP concentration estimates from ~200 km horizontal grid spacing to a higher resolution of ~10 km, to resolve strong gradients and high BaP concentrations near urban areas (Text S2). The World Health Organization (WHO) suggests that lifetime exposure to 0.1 ng m^{-3} of BaP would theoretically lead to one extra lung cancer death in 100,000 exposed individuals [*Bostrom et al.*, 2002]. However, the simulated population-weighted global average (PWGA) exposure of 1.28 ng m^{-3} greatly exceeds the WHO-recommended limit in 2008, with high levels of BaP exposure over large portions of East Asia, South Asia, Southeast Asia, Europe, Russia, Africa, North and South America (Figure 1a). Model predictions were evaluated at 69 background/remote sites (before downscaling) and 294 urban sites (after downscaling) around the world (Figure S3). The current model (with BaP shielded by viscous SOA coatings) agrees with field measurements, with normalized mean biases of +24.7% and +15.9% at background and urban sites, respectively.

Simulations using meteorological characteristics of 2008, but with changing emissions project that PWGA BaP concentrations will exceed 0.1 ng m^{-3} between 2008 and 2050 (Figure 1b-e), although global BaP concentrations will decrease by 50-100% in 2050 compared to 2008 (except RCP6). The decrease in BaP concentrations coincides with strong OC emission reductions projected by three of the RCPs in many regions of the world, especially in some developed and moderately developed countries (such as Europe, Russia, China, and the United States) [*van Vuuren et al.*, 2010; *Masui et al.*, 2011; *Riahi et al.*, 2011; *Thomson et al.*, 2011]. However, in rapidly developing regions of the world, including South Asia and Africa, BaP emissions are projected to increase by 2050 due to local increases in cropland and pasture related to more agricultural waste burning (AWB) and deforestation fires, and more primary energy consumption [*Liousse et al.*, 2014]. As a result, high levels of BaP exposure are likely to persist from 2008 to 2050 under the four RCPs (Figure 1b-e) in East Asia, South Asia, and Africa.

Figure 2 shows the population-weighted average BaP concentrations at three points in time in each RCP simulation (2008, 2020, and 2050). Relative to 2008, PWGA BaP concentrations are estimated to decrease by ~9% in 2020 and 41% in 2050 (Figure 3a) under the RCP4.5 and RCP8.5

scenarios. However, BaP concentrations are projected to increase in Africa (Figure 2i) due to increasing biofuel use, changes in land use [*van Vuuren et al*., 2011] and rapid industrialization throughout the 21st century [*Liousse et al*., 2014]. With the fastest population growth in the world, the increasing traditional biomass use in households for cooking and lighting will largely offset the reduction in emission intensity [*Meng et al*., 2019; *Hailu and Kumsa*, 2021], consequently enhancing BaP emissions in Africa. Moreover, shifts in land use increase both AWB and forest fires in Africa, increasing BaP emissions. For example, under RCP8.5, increasing deforestation is projected from 2008 to 2050 to meet agricultural demand for food [*Riahi et al*., 2011]. On the other hand, RCP2.6 projects major changes in land use from the forest and agricultural land in 2008 to land clearing for cultivation of bioenergy crops in 2050 [*van Vuuren et al*., 2010] that will increase deforestation and abandoned agricultural land [*Fargione et al*., 2008; *Searchinger et al*., 2015]. Similarly, many developing countries in South Asia, including India, could significantly increase BaP emissions, at least from 2008 to 2020 (Figure 2h), due to increases in AWB and residential primary energy consumption [*van Vuuren et al*., 2011]. In portions of the United States, Europe, Russia, South America, East Asia, and Southeast Asia, RCPs project strong decreases in BaP concentrations from 2008 to 2020 and 2050 (Figure 2b-g). In these regions, emission control regulations yield the adoption of better combustion technologies, such as improved cooking stoves, and a shift from coal and oil to renewable energy sources such as wind, solar, and nuclear energy. In the RCP8.5 scenario, decreases in domestic solid biomass fuels and AWB emissions, along with broader air pollution controls [*Riahi et al*., 2011] are the main drivers for reducing BaP emissions in developed and moderately developed countries (Figure S4b-g).

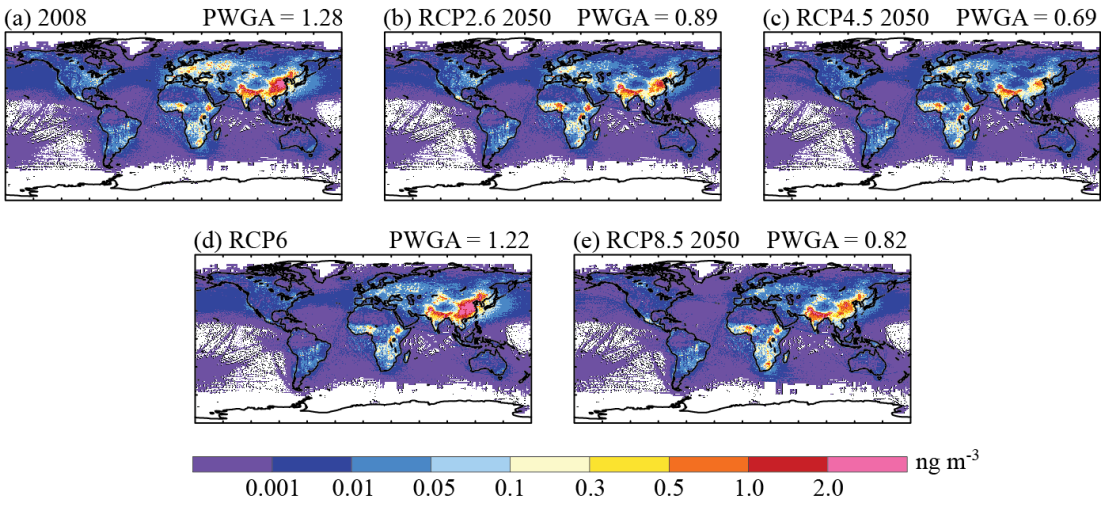

Figure 1. Spatial distribution of near-surface BaP concentration (after downscaling, ng m^{-3}) in (a) 2008 and (b-e) 2050 under RCP2.6, RCP4.5, RCP6, and RCP8.5, respectively. White areas are grid cells with BaP concentrations < 10^{-5} ng m^{-3}.

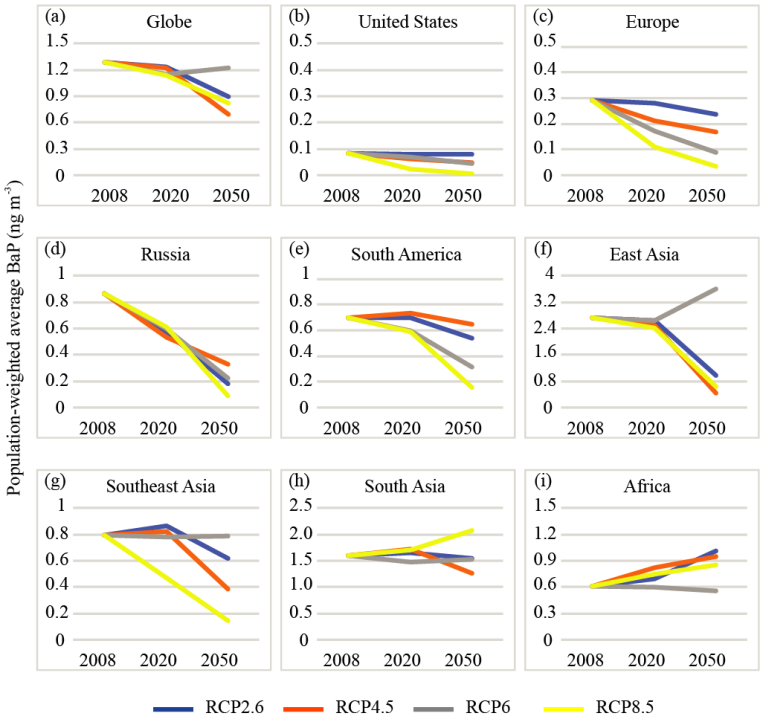

Figure 2. Time series of population-weighted average BaP concentrations. Line charts show the RCP-projected, population-weighted, downscaled BaP (ng m^{-3}) for individual regions.

In general, socio-economic developments promote cleaner fuels and improvements in combustion technologies. Globally, air pollution controls are expected to become more stringent with rising income, but trends differ for specific regions and particular times. For example, in 2008, 55% of global BaP emissions were from residential use of firewood and crop residues. BaP emissions from such biofuel use could either remain similar to 2050 levels if energy access gains are insufficient to outpace population growth, or decrease dramatically if modern energy transition accelerates, e.g., replacing traditional stoves with improved stoves that burn more efficiently or shifting to modern fuels.

3.2. Variation in PAH-associated ILCR due to changes in emissions

We use BaP as an indicator of lung cancer risk caused by exposure to all PAH mixtures (not just BaP), using a method based on epidemiological data [*Shen et al.*, 2014]. On a global population-weighted basis, ILCR is projected to exceed the WHO-acceptable guideline limit (1 death per 100,000 persons) in 2050 under all RCP scenarios (Figure 3a). The PAH-associated ILCR is projected to increase the most in Africa by 37-64% under all RCP scenarios except RCP6, followed by ~30% in South Asia under RCP8.5 (Figure 3b-c). Although the rate of increase of ILCR is projected to be the greatest in Africa, the absolute ILCR in South Asia (~ 3 deaths/100,000 persons) is projected to be significantly higher than in Africa (~2 deaths/100,000 persons) even in 2050.

Our results demonstrate that the greatest PAH-associated lung cancer risk in the 21st century will be localized over South Asia, Africa, and East Asia.

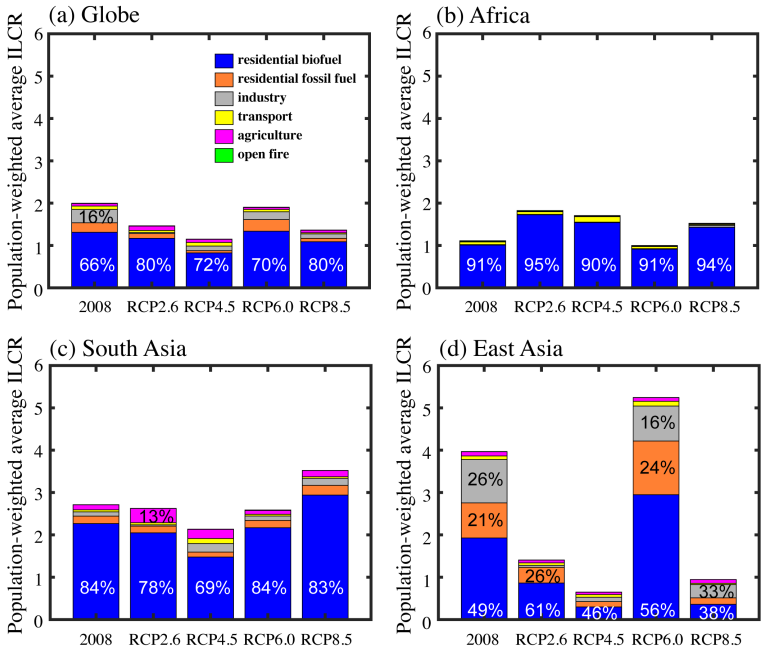

Figure 3. PAH-associated ILCR in 2008 and 2050. Bar charts show the global and regional ILCR (deaths per 100,000 persons).

Residential biofuel use dominated the global lung-cancer risk in 2008 (66%) (Figure 3a) due to the following factors: (1) lower combustion efficiency of biomass burning compared to other fuel types; (2) higher human exposure due to co-location of population and residential biofuel emissions. The contribution of residential biofuel use to global lung-cancer risk is projected to increase in 2050 (~70-80%). In developing regions of South Asia and Africa, increasing use of residential biofuels with low combustion efficiency co-occurs with rapidly growing populations, exacerbating human exposure to PAHs (Figures 3 and S4).

In East Asia, PAH-associated ILCR is projected to decrease under all RCPs, except RCP6 (Figure 4d), due to the use of cleaner fuels that accompany socio-economic development in China [*Tao et al.*, 2018]. Similarly, PAH-associated ILCR in Russia is projected to decrease substantially in 2050 (compared to 2008) due to the assumed implementation of air quality regulations [*Rafaj et al.*, 2010; *Riahi et al.*, 2011]. In other developed countries and regions, e.g., the United States and Europe, PAH-associated ILCRs were already much lower in 2008 than in developing regions and are expected to decrease further by 2050, resulting from the decline in residential consumption (Figure S5a-b). In contrast, the contribution from transportation and AWB will most likely increase.

3.3. Variation in PAH-associated ILCR associated with climate and emission

To investigate the effect of climate change on BaP concentrations, RCP8.5 was chosen as the reference scenario because it represents the most severe future global warming scenario among four RCP scenarios, significantly impacting air pollutants. In addition, RCP8.5 assumes a fragmented world that restricts international trade in energy and technology and describes an energy-intensive, fossil-based economy [*Riahi et al.*, 2011]. These assumptions are consistent with current global realities and fossil fuel production plans (https://productiongap.org/2021report/).

Due to climate change from 2008 to 2050, BaP concentrations change substantially in different regions. In tropical areas of South Asia, Southeast Asia, and Africa, BaP concentrations are reduced by 7–10% with increasing temperature (Figure 4a, Figure S6b). The shielding effectiveness (from SOA coatings) declines with increasing temperature in these regions (Figure S6b), leading to a much faster future BaP oxidation. Additional factors (including minor increases in precipitation and ozone concentrations) also contribute to a decrease in BaP concentrations (Figures S6f, S7b) over Southeast Asia and Africa. In contrast to warm and moisture regions, BaP concentrations increase by 2–3% from 2008 to 2050 in East Asia and Europe, where the shielding effectiveness is not sensitive to a 1-2 K increase in temperature under cold conditions. In these mid-high latitudes, decreasing O_3 concentrations and wind-field convergence changes (Figures S6 and S7) contribute to future increases in BaP concentrations.

In this study, the effect of increasing temperature and moisture on BaP concentrations are likely upper-bound estimates [*Shrivastava et al.*, 2017] since the heterogeneous reaction of particle-bound BaP is assumed to be completely shut off under cool and dry conditions [*Zhou et al.*, 2012]. Nonetheless, the impact of climate change on BaP concentrations is still lower than that of changes in emissions. Thus, "climate benefit" will partially offset increases in BaP concentrations in developing regions of South Asia and Africa, but emission changes will dominate future BaP trends (Figure 4a-c).

We also estimate how PAH-associated health risks will change due to variations in climate and emissions (Figure 4d-g). Overall, the global average PAH-associated ILCR is predicted to decrease by 37% due to future emissions reductions, while climate change contributes another 2% decline (Figure 4d). However, in developing regions of South Asia and Africa, the PAH-associated ILCR is projected to increase by ~20% from 2008 to 2050 (Figure 4e-f). As mentioned in section 3.2, incomplete combustion of fossil fuels and traditional biomass for cooking, burning associated with deforestation as cropland expands are projected to increase human health risks from these toxic components of fine particulate matter.

Compared to 2008, PAH-associated ILCR is also expected to decrease by 70-95% in developed and moderately developed regions such as the United States, Europe, Russia, Southeast Asia, and South America by 2050 (Figure S8). These considerable declines are attributed to strong regional emission reductions from the residential and industrial sectors (Figures S4, S5). Similarly, the PAH-associated ILCR peaked in East Asia in 2008, twice as high as in South Asia, and will decline

by 76% in 2050, primarily attributed to a decrease in residential biomass consumption in rural areas [*Riahi et al.*, 2011; *Shen et al.*, 2013].

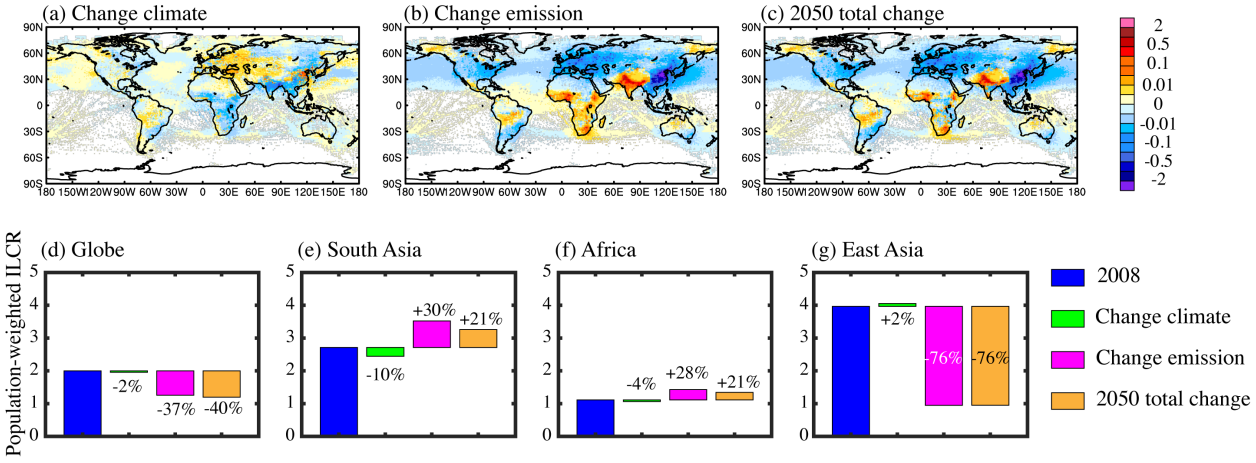

Figure 4. Impacts of changes in climate and emissions on (a-c) BaP concentrations and (d-g) PAHs-associated ILCR (deaths per 100,000 persons) from 2008 to 2050 under RCP8.5.

4 Conclusions and Discussions

In this study, we integrate a state-of-the-art global atmospheric chemistry model and a lung cancer risk model to assess how global PAH concentrations and their associated lung cancer risks may change with respect to several plausible future emissions trajectories. We project that the global population-weighted exposure to BaP will significantly exceed the WHO-recommended limit from 2008 to 2050 under all RCP scenarios. PAH-associated lung cancer risks, which peaked in East Asia (mostly China, 4 deaths per 100,000 persons) in 2008, are likely to shift to South Asia (mostly India, ~3 deaths per 100,000 persons) and Africa (~2 deaths per 100,000 persons) in 2050. The increment in residential energy demand in households for cooking, heating, and lighting accompanied by rapid population growth in India and Africa, as well as the continued use of traditional biomass use, increases in agricultural waste burning, and forest fires, could lead to the increase in health risks from 2008 to 2050. Although future climate change may be beneficial for reducing PAH concentrations and their associated health risks, future PAH-associated ILCR will strongly depend on socio-economic developments and air pollution policies.

Here, we focus on variations in BaP emissions under RCP scenarios. More recently, the Shared Socio-economic Pathways (SSPs) have offered a broader range of future air pollution developments in different regions over the 21st century [*Kriegler et al.*, 2012; *Rao et al.*, 2017; *Gidden et al.*, 2019]. Therefore, we use RCP scenarios for assessing future emissions changes and discuss our results within the context of SSPs. The SSPs provide five possible future development trajectories designed to address air pollutant emissions with strong, medium, and weak pollution control goals over the 21st century [*Rao et al.*, 2017; *Gidden et al.*, 2019]. Globally, RCP-based BaP projections used in this work generally represent the middle to upper range of the SSP

projections (Figure S9a-c). An exception is East Asia, where the RCP projections come closer to spanning the range of SSP-based BaP emissions (Figure S9d).

RCP8.5 projects high BaP emissions by 2050 in developing regions of Africa and South Asia, representing weak air pollution control scenarios (SSP3/4, Figure S2b-c), which appears realistic as emission control regulations in these regions remain weak [*Liousse et al*., 2014; *Rao et al*., 2017; *Kurokawa et al*., 2020]. In addition, current emission inventories show BaP emissions from Africa and South Asia are estimated to remain constant or slightly increase from 2008 to 2014 (Figure S9b-c). All these estimates are consistent with the RCP data for Africa and South Asia in the near-term.

In contrast, based on observed trends between 1992 and 2017, we find that East Asia will see stronger emission controls in the future [*Tao et al*., 2018; *Zheng et al*., 2018; *Q Zhang et al*., 2019]. A recent study reported a 5% per year reduction in residential biomass fuel use in rural China from 2008 to 2012 [*Tao et al*., 2018]. If residential biofuel emissions in China continue to decrease at a 5% per year rate up to 2050, the projected BaP emissions in East Asia would be 11% and 28% lower than projections in RCP4.5 and RCP8.5 by 2050, respectively.

Our analysis suggests a great range of possibilities for PAH emission changes in the future driven by variations in air quality policies across different regions. Since residential biofuel dominates the PAH-associated ILCR in India and Africa, more stringent controls on residential fuel use appear critical to avoid deterioration of air quality and human health. To achieve environmental targets, our study suggests that policies that encourage a shift from traditional solid biomass-based technologies to those using higher temperatures and more complete combustion will be important.

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