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# Health and environmental consequences of crop residue burning correlated with increasing crop yields midst India's Green Revolution

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The Green Revolution (GR) enhances crop yields significantly that contributes greatly to the social and economic development of many less developed countries. However, the increasing crop yields might rise crop residue biomass burning, leading to adverse environmental and health consequences. We assess the impact of crop residue burning associated with the GR-induced growing crop yields on benzo[a]pyrene (BaP) pollution, a congener of polycyclic aromatic hydrocarbons with strong carcinogenicity. We find a significant increasing trend of BaP emission and contamination from crop residue biomass burning from the mid-1960s to 2010s in India, coinciding with the growing crop yields occurring during the GR. Our results reveal that agricultural BaP emission driven lifetime lung cancer risk (ILCR) in India increased 2.6 times from the onset of GR in the mid-1960s to 2014 and the 57% population in India was exposed to the BaP level higher than the India national standard ( $1 \text{ ng m}^{-3}$ ). We show that the reduction of open crop residue burning during the rice and wheat residue burning period would be a very effective measure to reduce BaP environmental contamination and health risk.

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## INTRODUCTION

Significant social and economic developments occurring in India and other Southern Asian countries in the past half century may be credited to the Green Revolution (GR), which has transferred these countries from food-grain deficits to surpluses and sustained rapid population growth<sup>1,2</sup>. Great success notwithstanding, negative effects of the GR on the ecological environment have also drawn increasing attention during the past two decades<sup>3–5</sup>. These side effects manifest mostly as environmental degradation consequent to intensive cultivation, such as deforestation from conversion to cropland, soil degradation, and salinization<sup>4,6</sup>. Modern agriculture often accompanies with extensive use of pesticides and synthetic fertilizers<sup>5</sup>, which has been considered an important factor contributing the success of the GR. Many of these chemicals are toxic, persistent, and bioaccumulative, causing adverse effects on human health and the ecosystem<sup>7,8</sup>.

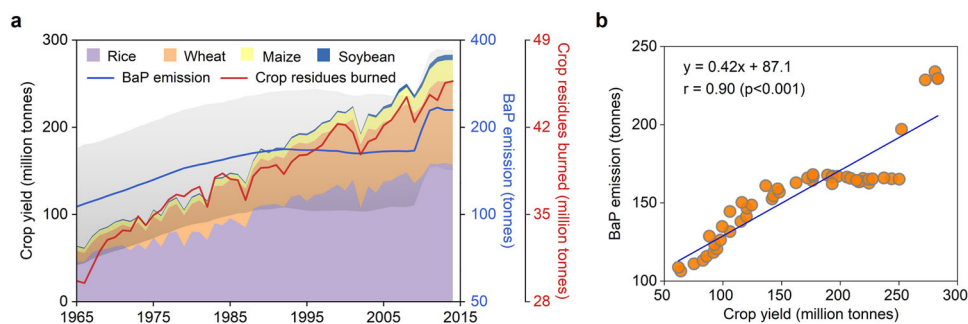
While a number of side effects of the GR on human health have been identified, knowledge gaps remain in the detrimental effects of the GR on the environment and human health. The agricultural GR in India and other Asian countries is featured by a rapid increase in staple crop production, including wheat, rice, maize, and cash crops such as cotton and jute<sup>4</sup>. These crops provide not only food and resources for the sustainable development and a growing population in these Asian countries, but also produce post-harvest crop residues<sup>9–12</sup>. Using the Intergovernmental Panel

on Climate Change (IPCC) national inventory preparation guidelines, the data collected in India showed that over 25% of the total crop residues were burnt on the farms<sup>10,11</sup>. It was also reported that more than 90% of households in rural areas and 32% of households in urban India used biomass as the primary energy source for cooking and heating<sup>13–15</sup>, of which agricultural residue contributed about 16–20% to the total indoor solid biomass burning<sup>15,16</sup>. The crop residue burning in India, especially in the Indo-Gangetic Plain (IGP), known as the South Asia's cereal basket, caused poor air quality<sup>17–21</sup> and health consequences of local residents<sup>22,23</sup>.

Biomass burning is also a major emission source of polycyclic aromatic hydrocarbons (PAHs), a class of organic pollutants that receive great public concern due to their widespread occurrence and toxic effects on ecological safety and human health<sup>24</sup>. India has been ranked the second largest emitter of PAHs in the globe, next to China<sup>25–27</sup>. Biofuel has been considered as the largest source of PAHs in India, accounting for 92.5% of the total source<sup>26,28</sup>. Total emissions of the 16 parent PAH congeners prioritized by the United States Environmental Protection Agency (USEPA) from open and indoor crop residue burning in India account for about 20–25% of all emission sectors, which is considerably higher than that from industries and transportations<sup>26</sup>. The burning of leftover agricultural matter in open fields for crop rotation is a common practice in the northwestern region of IGP in the rice and wheat residue burning period<sup>9–11</sup>, which also

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**Fig. 1** Variation of crop yield, crop residues burned, and BaP emission in India. **a** The yield of wheat, rice, maize, and soybean, crop burning, and agricultural BaP emissions in India from 1965 to 2014. The solid purple line indicates agricultural BaP emissions. The gray shaded area represents uncertainties at 95% confidence intervals. **b** The correlation between total crop yield and agricultural BaP emission during the same period. The agricultural activities include field crop residue burning, indoor crop residue burning, gas/diesel combustion from agricultural activities (about 0.2% of the total emission). The yields of wheat, rice, maize, and soybean, and crop residues burned in India are collected from Food and Agriculture Organization of the United Nations (FAO, <https://www.fao.org/faostat/en/#data>), and BaP emissions from agricultural activities in India are collected from PKU-PAH inventory database (<http://inventory.pku.edu.cn/home.html>).

contributed severe PAH contamination to the IGP. Extensive studies have been conducted by Indian scientists to explore the contamination characteristics and source–receptor relationships of PAH in northwestern India including Delhi and National Capital Region (NCR)<sup>29–32</sup>, and the effect of crop residue burning on PAH pollution in northwestern India<sup>25,27,33</sup>.

In the present study, we extended the investigations of PAH contamination and health effect in India back to the 1960s since the onset of the GR. We propose that indoor and outdoor crop residue burning and other agricultural activities in India markedly contributes to emissions of benzo[a]pyrene (BaP), a representative PAH compound often used as marker for carcinogenic PAHs. We incorporate a coupled atmospheric transport and multi-compartment exchange model<sup>34–36</sup> to quantify BaP contamination in the air from 1960 to 2014 based on a global gridded BaP emission inventory<sup>26</sup>. We then estimate incremental lifetime lung cancer risk (ILCR)<sup>37</sup> using modeled ambient BaP concentrations commencing at the start of the GR in the early 1960s with a focus on India. Extensive model sensitivity simulations were conducted to examine the response of ILCR to BaP emission reductions in agriculture sectors.

## RESULTS

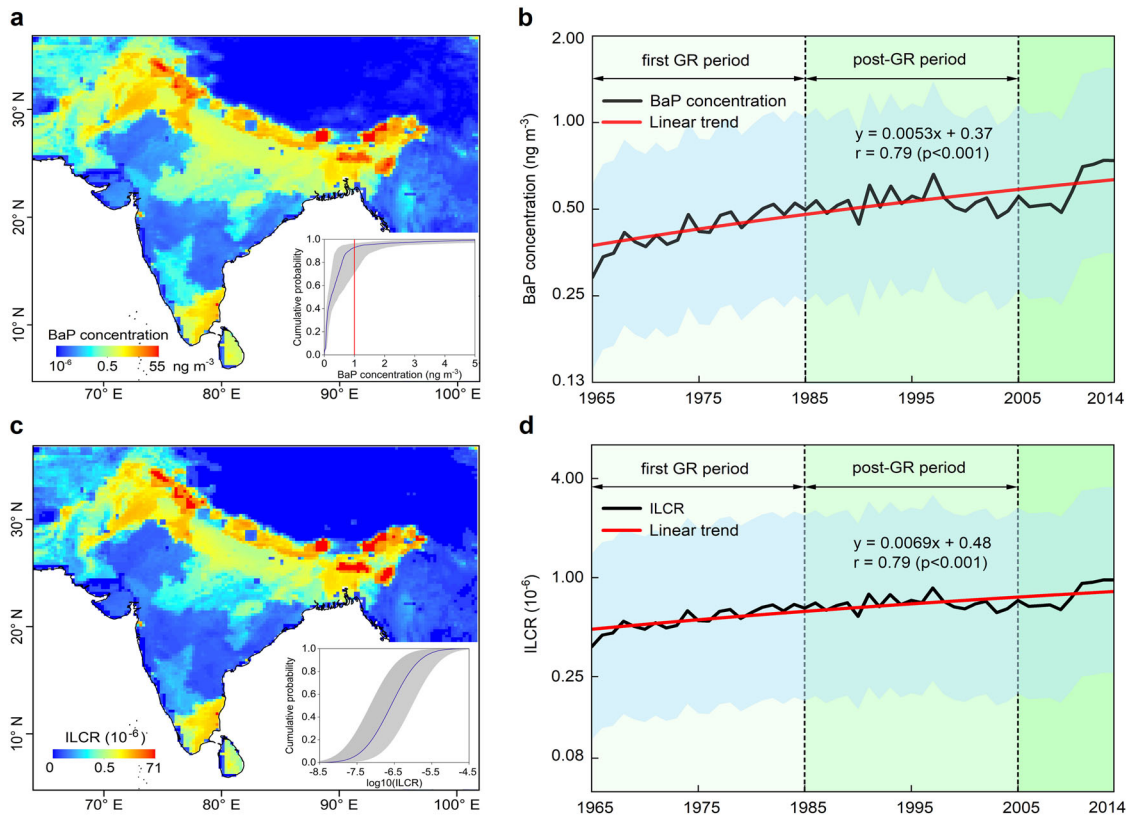
### Crop yield and BaP emission

Figure 1a displays the annual yields of four major crops, crop residues burned, and BaP agricultural emissions from 1965, the year of GR onset, to 2014 in India. The crop yield in India increased from 64 million tonnes in 1965 to 283.1 million tonnes in 2014. Accordingly, the crop residues burned in India increased from 29.7 million tonnes in 1965 to 45.7 million tonnes in 2014. As a result, BaP emission from agricultural sectors, including indoor, and field crop residue burning, and gas/diesel combustion due to agricultural activities (collectively referred to as “agriculture” hereafter), increased from 106.5 (95% confidence interval (CI): 67.0–169.4) tonnes in 1965 to 229.6 (95% CI: 142.1–371.0) tonnes in 2014. Strong, statistically significant correlation at  $r = 0.9$  between BaP agriculture emissions and the total crop yields (the sum of four crops) is illustrated in Fig. 1b. The result suggests that the increasing BaP emission is likely associated with the growing crop productions. Supplementary Fig. 1 shows gridded averaging BaP emission (Supplementary Fig. 1a) and its trend (Supplementary Fig. 1b) from agricultural sectors of 1965 through 2014. High emissions are discerned over the IGP region, in line with high crop yields (Supplementary Fig. 2). As expected, the linear trend of BaP emissions also agreed with the linear slopes of crop yields (Supplementary Fig. 1c), showing strong growing emissions

associated with increasing crop yields in the state of Punjab, Uttar Pradesh, and Bihar across the IGP. These states are reported to produce the highest cereal crop residues and biomass fuels<sup>11,12</sup>.

### Ambient BaP air concentrations and exposure risk from agricultural emissions

We simulate BaP air concentrations spanning 1965–2014 from all emission sectors and from agricultural sectors only across India and its neighbor countries. Ambient BaP air concentrations and exposure risk from all sectors are presented in Supplementary Note 1 and Supplementary Figs. 3–6. We then examine the responses of CanMETOP modeled BaP concentration and cancer risk to the BaP agriculture emissions. Figure 2a shows modeled airborne BaP concentrations from agricultural emissions. Higher BaP air concentrations are seen in the IGP, northwest, northeast, and south India compared to the rest of the country, in line with BaP emissions from agricultural and crop residue burning (Supplementary Fig. 1a) in these traditional crop yield areas (Supplementary Fig. 2), where the GR achieved most success in India. These areas also have higher population densities than the other regions of India (Supplementary Fig. 4). Among these regions, the BaP levels in the atmosphere in Haryana, Punjab, and Himachal Pradesh located in northwest India, and Meghalaya and Manipur Pradesh located in northeastern India exceed the Indian national standard ( $1 \text{ ng m}^{-3}$ )<sup>38</sup>. Northwestern India, known as the “breadbasket” of the country, produces two-thirds of India’s food grains, of which wheat and rice are the principal crops<sup>21</sup>. As a result, this region generates a large amount of crop residues<sup>11,15,21</sup>. The burning of agricultural leftovers in open fields for crop rotation is a common practice in northwestern India<sup>17</sup>. In the states of Haryana, Punjab, and Himachal Pradesh, 80% of its rice straw was burned in situ<sup>39</sup>. Open-field burning of agricultural residues during rice and wheat harvesting periods contributed higher BaP contamination in northwestern India. Given the lack of lands for settled agricultural leftovers, a “slash and burn” form of shifting cultivation, locally known as *jhum*, was widely operated by tribal groups in northeastern India, such as Tripura, Meghalaya, and Manupur<sup>40</sup>. Slash-and-burn agriculture involves the cutting and burning of plants in a forest or woodland to create a field called a swidden<sup>41</sup>. Previous studies have revealed that northeastern India accounts for the highest fire events and biomass burning during spring associated with slash-and-burn<sup>42–44</sup>. This activity occurs before the onset of Indian Monsoon. In the autumn harvest season, a rice-wheat cropping activity occurs in October and November after the Indian Monsoon<sup>45</sup>. Crop residues, together with wood, provide a primary source of energy for domestic cooking and contribute higher BaP air concentrations to



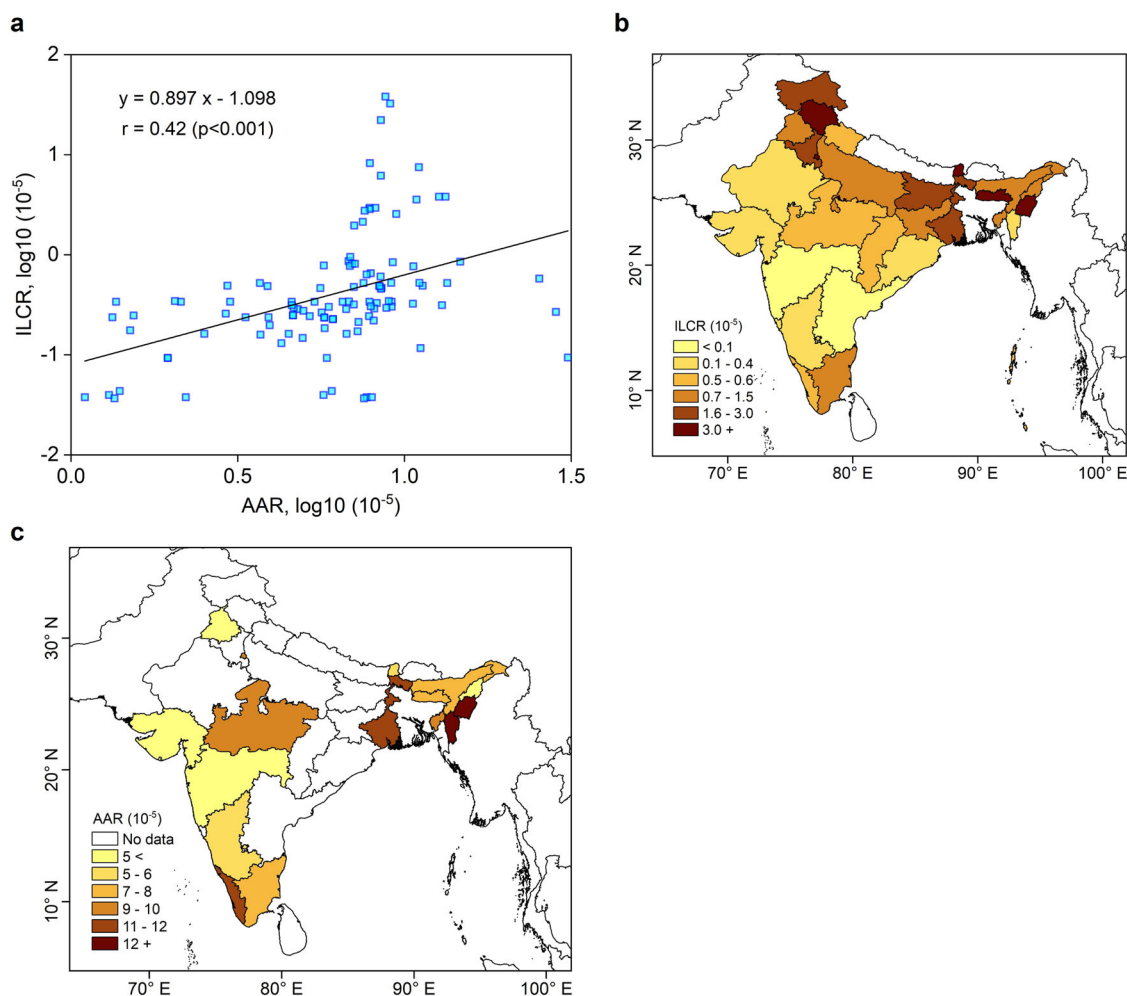
**Fig. 2** BaP air concentrations and ILCR from agricultural emission across India from 1965 to 2014. **a, c** Shows annually averaged gridded BaP air concentrations and ILCR from agricultural emission averaged over India from 1965 to 2014. The inset panel in the lower right corner in **a** and **c** shows cumulative distributions of gridded BaP concentrations and ILCR in India, and the uncertainties at 95% confidence intervals are shown by gray shaded areas. The solid red line in **b** and **d** indicates the linear trend of BaP concentrations and ILCR, and the uncertainties at 95% confidence intervals are shown by light blue shaded areas.

rural households in this region<sup>16,46</sup>. Our modeling results reveal that approximately 6.94% (95% CI: 3.28–29.41%) of the land area and 6.42% (95% CI: 2.33–44.57%) of the Indian population are exposed to BaP concentrations released from agricultural emissions, that exceed the Indian national standard ( $1 \text{ ng m}^{-3}$ ). However, due to the short period (about 20 days) of field crop residue burning in spring and autumn harvest seasons<sup>45</sup>, the open field burning yields low BaP emission on an annual basis. Supplementary Fig. 7 shows MODIS satellite remote sensing 8-day fire mask and land types, as well as the change in fire mask number in 2014 (“Methods”). Only fire marks over cropland are illustrated. The fire mask number reaches the first peak in May and the second peak in October, which corresponds well to the post-harvest wheat and rice residue burning periods during the spring and autumn seasons when massive agriculture wastes are burned right after harvest. The spatial pattern of 8-day fire mask also agrees well with that of BaP agriculture emission and concentration, featured by higher airborne BaP in northwestern and northern India (Fig. 2a).

The spatial distribution of modeled mean BaP concentrations averaged over the period of 1965 through 2014 subject to all emission sources show higher BaP levels in northern India (Supplementary Fig. 3a). The maximum concentration ( $55 \text{ ng m}^{-3}$ ) from the agriculture emission accounts for about 40% of the maximum BaP level ( $140 \text{ ng m}^{-3}$ , Supplementary Fig. 3a) from all emission sources, in line with the fraction of the agriculture emission to the total BaP emission. Figure 2b shows the variation of annual mean BaP air concentrations averaged over India from 1965 to 2014 from the agricultural emissions. The mean BaP concentrations from the crop residue burning increased from 0.29

(95% CI: 0.13–0.65)  $\text{ng m}^{-3}$  in 1965 to 0.66 (95% CI: 0.29–1.52)  $\text{ng m}^{-3}$  in 1997, but showing no significant change thereafter until 2008. From 2010 to 2014, the mean BaP concentrations from the crop residue burning in India increased markedly from 0.57 (95% CI: 0.27–1.22)  $\text{ng m}^{-3}$  to 0.74 (95% CI: 0.35–1.55)  $\text{ng m}^{-3}$ . Pingali distinguishes the first GR period extending from 1966 to 1985 and the post-GR period as the next two decades<sup>4</sup>. The first GR symbolizes a significant increase in agricultural productivity resulting from the introduction of high-yield varieties of grains, use of chemicals, and improved management techniques, which have contributed to rapid growth in India food production. Cereal yields, including rice, wheat, maize, and soybean, have increased by 130% compared to that in 1965 in India (Fig. 1a). This rapid growth of crop products drives, to a large extent, growing BaP air concentrations from the agricultural emissions. In the post-GR period, along with the decreasing crop prices, the investment in Indian agriculture dropped<sup>47,48</sup>, resulting in a reduction of agricultural products. As aforementioned, decreasing BaP emissions from crop residue burning during this period are also attributable partly to the implementation of agricultural emission control across India. It should be noted that BaP air concentrations from agricultural emission markedly increased after 2009, which may be due to the rapid growth of crop yields (Fig. 1a).

Figure 2c shows annually averaged ILCR from agricultural emissions from 1965 to 2014. In line with BaP air concentrations from agricultural emissions (Fig. 2a), higher ILCR is predicted in Punjab to the northwest India and Meghalaya to the northeast India where the estimated ILCRs are as high as  $5 \times 10^{-5}$  and  $6 \times 10^{-5}$ , respectively. As a result, 11.48% (95% CI: 1.97–63%) and 0.11% (95% CI: 0.03–2.32%) of the residents in these regions are

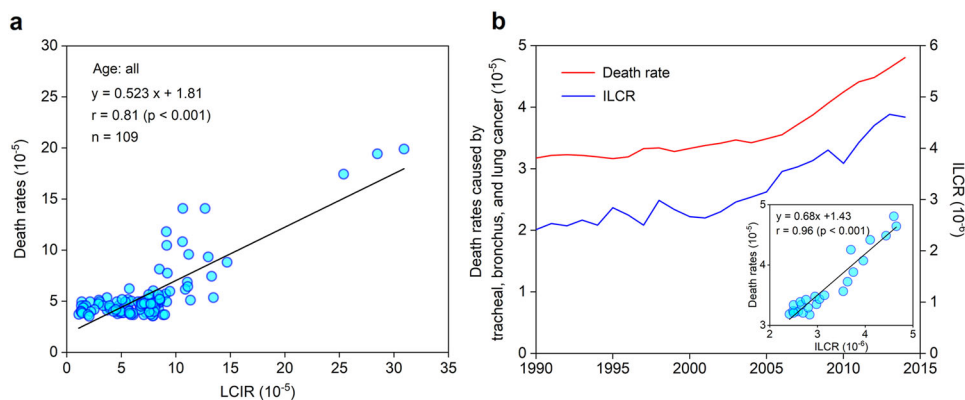


**Fig. 3 Comparison between ILCRs induced by BaP exposure and age-adjusted lung cancer incidence rates (LCIRs).** **a** Correlation between the state-averaged ILCRs and LCIRs, including 13 states in India from 1990 to 2012. **b** Annually averaged ILCRs over each state of India from 1990 to 2012. **c** Annual mean age-adjusted LCIRs in each state in India from 1990 to 2012, collected from the India Council of Medical Research ([https://ncdirindia.org/NCRP/Annual\\_Reports.aspx](https://ncdirindia.org/NCRP/Annual_Reports.aspx)).

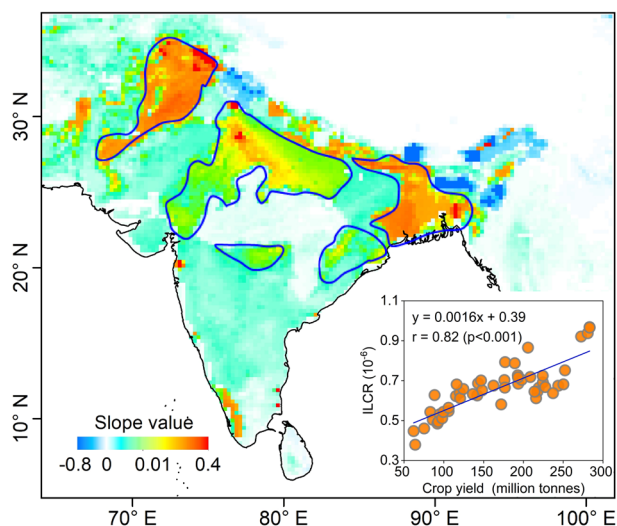
under  $10^{-6}$  and  $10^{-5}$  ILCR induced by BaP contamination from the agricultural emission. As shown in Supplementary Fig. 8, the percentage of ILCR from agricultural emission accounted for 40–60% of the ILCR from all emission sources in northeastern and northwest India as seen in the gridded crop yield (Supplementary Fig. 2). The annual mean ILCR averaged over India subject to BaP emissions from agricultural sectors increases from  $3.8 \times 10^{-7}$  (95% CI:  $1.06 \times 10^{-7}$ – $1.37 \times 10^{-6}$ ) in 1965 to  $9.66 \times 10^{-7}$  (95% CI:  $2.63 \times 10^{-7}$ – $3.54 \times 10^{-6}$ ) in 2014 at an annual increase rate of 2.59%. The vulnerable population, exceeding  $10^{-6}$  and  $10^{-5}$  ILCR subject to agriculture and crop residues burning emissions, increases from 2.56% (95% CI: 0.7–48.59%) and 0.08% (95% CI: 0.01–1.01%) in 1965 to 25.75% (95% CI: 6.38–71.35%) and 0.1% (95% CI: 0.03–0.58%) in 2014, respectively, manifesting one order of magnitude enhancement for the past half century. It can be seen from Supplementary Fig. 8 and Fig. 2d that the ILCR averaged over India from agricultural emissions accounts for about 30% of all BaP emission sources. Both annual BaP concentration and ILCR subject to indoor and outdoor crop residue burning exhibit statistically significant increasing trends from 1965 to 2014.

We also collected lung cancer incidence rates (LCIRs) reported by the Indian Council of Medical Research ([https://ncdirindia.org/NCRP/Annual\\_Reports.aspx](https://ncdirindia.org/NCRP/Annual_Reports.aspx)), which were used to compare with modeled ILCR due to human inhalation exposure to ambient BaP

concentrations in India. Statistically significant correlation ( $p < 0.001$ ) between the state mean ILCRs and non-smoker lung cancer incidence rates as shown in Fig. 3a manifests that the estimated ILCR explains 17.6% ( $R^2$ ) of lung cancer incidences in India. The spatial similarity between ILCRs and lung cancer incidence rates suggests that BaP contamination is an important factor contributing non-smoker lung cancer incidence rates. However, the officially reported long-term lung cancer incidence rates in the IGP, the region mostly contaminated by BaP, is not available. In 2012, The Indian Council of Medical Research released the India state-level disease burden initiative (<https://vizhub.healthdata.org/gbd-compare/india>), which includes state level specific diseases and risk factors to the overall health loss from 1990 to 2010. Based on this data source, we collected the state-level death rates caused by tracheal, bronchus and lung cancer from 1990 to 2012, and compared these death rates with state-level age-adjusted lung cancer incidence rates in 13 states of India from 1990 to 2012 as shown in Fig. 3c. Results are illustrated in Fig. 4. As seen, the death rates caused by tracheal, bronchus and lung cancer match well with the lung cancer incidence rates at a coefficient of  $r = 0.81$  ( $p < 0.001$ ), suggesting that the death rates caused by tracheal, bronchus and lung cancer can be used to replace lung cancer incidence rates and evaluate the association between lung cancer risk and modeled ILCR due to inhalation exposure to ambient BaP concentration in IGP. Likewise,



**Fig. 4** Comparison between the death rates caused by tracheal, bronchus and lung cancer and age-adjusted lung cancer incidence rates (LCIRs) as well as ILCR. **a** Correlation between state-level death rates caused by tracheal, bronchus and lung cancer and state-level age-adjusted LCIRs in 13 states from 1990 to 2012 in India as shown in Fig. 3c. Note that three larger LCIR values ( $>25 \times 10^{-5}$ ) were reported by Mizoram India (<https://health.mizoram.gov.in>). **b** Comparisons between state-level death rates caused by tracheal, bronchus, and state-level ILCR induced by exposing BaP contamination in IGP from 1990 to 2015. The death rates caused by tracheal, bronchus, and lung cancer are collected from the Indian Council of Medical Research (<https://vizhub.healthdata.org/gbd-compare/india>).



**Fig. 5** Gridded linear trend (slope) of ILCR in India. Slopes of linear regression of modeled ILCR subject to BaP agricultural emission from 1965 to 2014. Inset panel in the low right corner of the figure is correlation diagrams between total crop yield and ILCR in India subject to agricultural emission. The statistically significant regression at greater than or equal to the 90% confidence level is encircled by the blue contour.

Fig. 4b shows that the annually averaged ILCR agrees well with the death rates caused by tracheal, bronchus and lung cancer over IGP from 1990 to 2014, at a correlation coefficient of  $r = 0.96$  ( $p < 0.001$ ). Both ILCR and death rates caused by tracheal, bronchus, and lung cancer in IGP increased from 1990 to 2014.

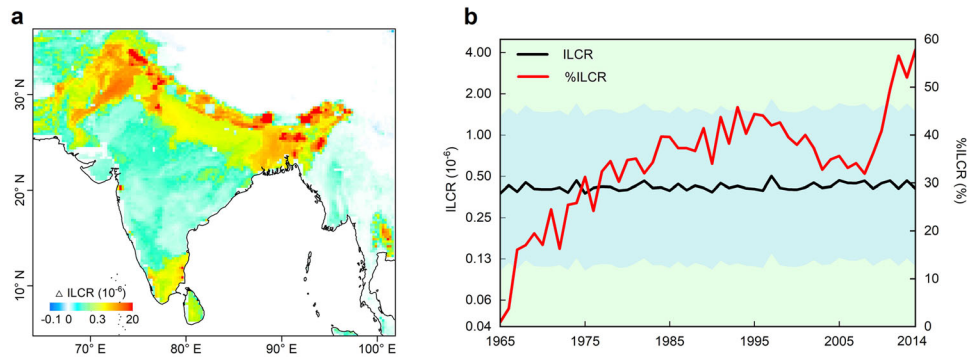
#### Trend of airborne ILCR induced by agricultural BaP emission

The gridded linear trend (slope) of modeled ILCR subject to agricultural emissions of 1965 through 2014 are illustrated in Fig. 5. As expected, the linear trend of ILCR also agree with the linear slopes of agricultural BaP emissions (Supplementary Fig. 1b), strong positive trends of ILCR subject to BaP agricultural emissions occur in northwestern, northeastern, and eastern India, the regions, where the GR took place. The greatest success of GR was achieved in Punjab, Haryana, and Uttar Pradesh in the mid-1960s, characterized by the use of a high-yielding variety (HYV) of seeds in wheat cultivation. By 1983, the GR had incorporated rice

and coarse cereals such as maize, sorghum, and soybean, and extended its range to Bihar, Andhra Pradesh, and Tamil Nadu to the east, and Karnataka and Maharashtra to western India<sup>49</sup>. We note that the most statistically significant trends of crop yields, BaP concentrations, and ILCR in the IGP superimposes with the regions having most severe air pollution in India, which has been attributed to a large extent to crop residue burning<sup>45,50</sup>, particularly during the period of the rice-wheat cropping (Autumn) and slash and burn (Spring) seasons. The graphic plotted in the inset panel of Fig. 5 shows that ILCR significantly related with the total crop yields (the sum of four crops) at a correlation coefficient of  $r = 0.82$  ( $p < 0.001$ ). Supplementary Fig. 9 further compares the slopes (trends) of the ILCR and crop yields (wheat, rice, maize, and soybean) during the GR Phase I (1965–1985) and Phase II (1986–2014). The spatial pattern of the ILCR trend during the two GR phases is, to a large extent, in line with the trend of crop yields during the same period, particularly in the IGP where the GR was initiated in Punjab, Haryana, and Uttar Pradesh in the mid-1960s and subsequently extended to eastern and western India<sup>49</sup>. During the GR Phase II, however, negative trends of crop yields and ILCR are seen in northwestern India, indicating the reduction of the crop yields in this part of India, which further reduces lung cancer risk of local residents to BaP owing to declining emissions from crop residue burning. As aforementioned, this is likely a consequence of the declining investment in agricultural development in India.

#### BaP lung cancer risk during the GR

We also simulated the ILCR across India subject to fixed BaP emissions from the agricultural sectors in 1960, hereafter referred to as “NO-GR” scenario simulation (“Methods”). Supplementary Fig. 10 shows the spatial patterns of ILCR from 1965 to 2014 from “NO-GR” simulation. Similar to the ILCR subject to annual changes in BaP emissions from the agricultural sectors from 1965 to 2014 (hereafter referred to as “GR” scenario simulation, Fig. 2c), the modeled ILCR from “NO-GR” scenario in northern India is higher than that in the south because northern India is the major crop production area in the country and hence the largest BaP emitter from crop residue burning. To see more clearly the extent of BaP contamination and exposure risk occurring during the GR, we illustrate the ILCR differences between “GR” and “NO-GR” scenario simulations in Fig. 6a (defined as  $\Delta\text{ILCR} = \text{ILCR}_{\text{GR}} - \text{ILCR}_{\text{NO-GR}}$ ). Accordingly, BaP concentration differences ( $\Delta\text{BaP}$ ) between “GR” and “NO-GR” scenario simulations are shown in Supplementary Fig. 11a. Both  $\Delta\text{BaP}$  and  $\Delta\text{ILCR}$  exhibit almost identical spatial

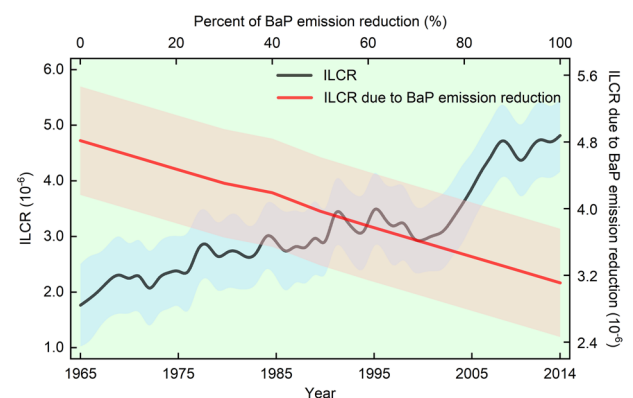


**Fig. 6** Difference in ILCR between GR and NO-GR model scenarios and fractional contribution of GR to ILCR. **a** The ILCR difference, defined by  $\Delta\text{ILCR} = \text{ILCR}_{\text{GR}} - \text{ILCR}_{\text{NO-GR}}$ , where  $\text{ILCR}_{\text{GR}}$  and  $\text{ILCR}_{\text{NO-GR}}$  stand for the ILCR subject to GR and NO-GR model scenario simulations (“Methods”). Positive  $\Delta\text{ILCR}$  indicates an enhancement of lung cancer risk due to BaP. **b** Annual mean ILCR (the solid blue line) averaged over India subject to “NO-GR” scenario simulation from 1965 to 2014, and the percent contributions of GR to the ILCR, defined as  $\% \text{ILCR} = (\Delta\text{ILCR} \times 100) / \text{ILCR}_{\text{GR}}$ , marked by the red solid line. The light blue shaded area in **b** represents uncertainties at 95% confidence intervals.

patterns, showing large positive values in the north and small values in the south, suggesting that the GR might act as a driver to enhance BaP contamination and lung cancer risk due to higher ambient BaP levels in northern India or, typically, in the northern “grain belt”. Higher  $\Delta\text{ILCR}$  is discerned in northwestern and northeastern India, indicating that the populations in these regions have suffered from increasing lung cancer risk due to inhalation exposure to ambient BaP over the past half century. The association of the annual ILCR averaged during the GR is further quantified by the ILCR fraction, defined as  $\% \text{ILCR} = (\Delta\text{ILCR} \times 100) / \text{ILCR}_{\text{GR}}$  (Fig. 6b). Our model results show that the lung cancer risk increases from 1% in 1965 to 45.8% in 1993 but decreases after 1993 up to 2008. The  $\% \text{ILCR}$  grows again from 2009 to 2014, increasing from 36.2% in 2009 to 57.8% in 2014. Likewise, annual BaP concentrations averaged over India, defined by  $\% \text{BaP} = (\Delta\text{BaP} \times 100) / \text{BaP}_{\text{GR}}$  (Supplementary Fig. 11b), exhibits a similar annual variation with  $\% \text{ILCR}$  (Fig. 6b).

## DISCUSSION

Despite the success of the GR in India, our modeling evidence shows an accompanying enhancement of BaP contamination, which is associated with increased lung cancer incidents. The lessons learnt from the GR operations over the past decades are that actions should be taken to reduce, or better still, eliminate the adverse side effects characterized by PAH emission and lung cancer risk and thereby fulfill the sustainability of the GR. By employing the multivariate regression model (“Methods”), we have further examined the response of BaP ILCR to different model scenarios for the IGP, which has the highest crop yields and highest BaP emissions in India during the two phases of GR in India. Using 2014 as the base year, we performed 10 model scenario simulations by reducing annual BaP emissions from post-harvest crop residue burning (field + indoor burning) from 10 to 100%, respectively. Figure 7 shows modeled annual ILCR (scaled on the left Y-axis and bottom X-axis) and its declining trend (scaled on the right Y-axis and upper X-axis) as the responses to the emission reduction scenarios over IGP. The ILCR in IGP exhibits strong increasing trend from 1965 onward, in line with its trend over entire India during the same period (Fig. 2b) but with significant larger ILCR values, indicating higher lung cancer risks in this region. The ILCR declines linearly with the percent reduction of BaP emission from crop residue burning. Although mean BaP agriculture emission averaged over 1965 to 2014 only accounts for 23% of the total emission, the elimination (100% reduction) of crop residue burning could reduce 35% cancer risk. Our multi-regression analysis (Methods) between modeled ILCR and BaP



**Fig. 7** CanMETOP predicted annual BaP ILCR in the IGP from 1965 to 2014 (solid black line, scaled on the left Y-axis and bottom X-axis) and the moving averaged declining trend of ILCR by reducing BaP indoor and outdoor crop residue burning from 10% to 100% (solid red line, scaled on the right Y-axis and upper X-axis) based on 2014 emissions. Red and blue shaded area indicate  $\pm 1$  standard deviation.

non-agriculture and agriculture emissions reveal that, while the BaP emission from outdoor crop residue burning accounts for only 0.3% in the total emission on an annual basis, it could contribute 3% to the modeled ILCR. Given multiple and complex sources of non-agriculture emissions (=total emission minus agriculture emission), such as emissions from non-agriculture biomass (forest) burning, industrial, and many other sources, it is not straightforward to reduce these emissions at a low cost. Rather, our finding suggests that the reduction of BaP emission from indoor and outdoor crop residue burning might be a very effective measure to reduce BaP contamination and health risk. A recent modeling investigation suggests that complete elimination of precursor emissions from household sources could reduce  $\text{PM}_{2.5}$  below its Indian national standard ( $40 \mu\text{g m}^{-3}$ )<sup>51</sup>. Given the lack of sufficient data for the incidence of lung cancer in India and the indoor BaP concentrations are not taken into account separately in the estimate of ILCR, we are not able to establish a statistically significant relationship between BaP concentrations from indoor crop residue burning and lung cancer risks. We did not take wet deposition, driven largely by precipitation, into consideration either in our modeling investigation but the annual precipitations could contribute 11% to the predicted BaP concentration if they were taken into account in the regression model (Methods). On the other hand, the annual air temperature and wind speed only contribute 1.4 and 0.4% to the forecasted BaP concentrations by

the statistical regression model. The result manifests that precipitation washout occurring during the Indian monsoon season (June–September) would likely buffer the outdoor emissions during that period whereas the climate change seems not to play a significant role in the long-term trend of BaP concentrations and ILCR.

It is arguable that India's Green Revolution could also lead to the changes in its local economic conditions, food consumption patterns and individual behavior, and exposure to other sources of carcinogenic emissions. For instance, the growing income due to better agricultural yields could have led to increased vehicle ownership and the use of transportation services, which are also BaP sources<sup>52</sup>. The firewood and crop residue burning in India contribute 86.9% to the total BaP emission in 2007 in which the agriculture emissions account for 31%, whereas the fossil fuel burning (6.4%) and transportation (1.8%) associated with industry and the human lifestyle made little contribution to the country's BaP emission<sup>26</sup>. The spatial inconsistency between the India's agriculture output value and its gross domestic product (GDP) in 2014 (Supplementary Fig. 12), as compared to the state-level industrial and vehicle population output value which agree nicely with the state-level GDP, also suggests that India's agriculture does not make a primary contribution to its GDP. Given that the largest BaP emission source, the indoor firewood burning, which contributes 45.8% to the total emission, does not directly connect to agriculture activities, the crop residue burning can, therefore, be considered as a primary source associated agriculture activities.

However, we wish to emphasize that crop residue biomass burning and resulted PAH emissions are not caused by the GR in India directly, because "slash and burn" is a traditional agricultural activity in Northern India before the GR. In light of this, crop residue burning is, in reality, an issue of proper agriculture management. The governments and decision-makers of India have already taken actions by investing in economically viable no-burn alternatives and subsidizing farmers to reduce field crop residue burning. The government of India has encouraged the use of advanced biomass stoves<sup>53,54</sup>, cleaner energy<sup>55</sup>, and improving crop residue management practices<sup>56,57</sup>. Two consecutive programs, the National Project on Biogas Development (NPBD) and the National Biogas and Manure Management Program (NBMP) during the past decades have been implemented to promote the use of cleaner cooking stoves fueled by biogas which can be produced from cow dung and biomass waste to replace biomass fuels<sup>58,59</sup>. Several measures, such as returning biomass to soil, producing biogas from biomass, and rice straw mulching, have been taken to decrease crop residue burning<sup>60</sup>. The reduction of PAH emissions from crop residue could be also achieved, to some extent, by the "Happy Seeder" system in India at a relatively low cost<sup>47</sup>. In the case of indoor crop residue burning for domestic cooking and heating, rapid transition from rural residential solid fuel combustion to clean energy might provide another measure to eliminate post-harvest crop residue burning.

## MATERIAL AND METHODS

### Modified CanMETOP model

Ambient air BaP concentrations and atmospheric transport from 1965 to 2014 were predicted by a modified version of the CanMETOP (Canadian Model for Environmental Transport of Organochlorine Pesticides)<sup>34</sup>. The model, originally developed for modeling environmental fate and atmospheric transport of persistent organic pollutants (POPs) on a regional scale, was subsequently extended to a global scale to investigate the intercontinental long-range transport of POPs with a spatial resolution of 1° latitude by 1° longitude<sup>35</sup>. The model has previously been applied extensively to simulate atmospheric transport and deposition, source-receptor relationships, and

cycling in multi-environmental compartment (air, soil, water, snow/ice, vegetation) exchange of POPs<sup>61</sup>. The model employed in this study was the same as that adopted in the global version of the CanMETOP<sup>35,36</sup> but the model domain has been reduced to cover India and surrounding regions (Supplementary Fig. 13) and horizontal spatial resolution was reduced to 1/4° latitude by 1/4° longitude. The model was integrated from 1960 to 2014 in which 1960 to 1964 was taken as model spin-up time. Modeled BaP air concentrations from 1965 to 2014 were used and analyzed in this study. Detail model description and input parameters are presented in Supplementary Note 2 and Supplementary Table 1.

To discern and quantify the contribution of GR to the BaP fluctuations and lung cancer risks, we set two model scenarios. The first is "NO-GR" scenario, which uses the fixed BaP emission from agriculture sectors in 1960. We then integrate CanMETOP model from 1960 to 2014 subject to the constant BaP agriculture emissions in 1960 only. The other model scenario, namely the "GR" scenario, uses monthly varied BaP agriculture emissions from 1960 to 2014. The differences and fractions of modeled BaP concentrations between the two scenarios are estimated to assess quantitatively the GR contribution to BaP contamination and lung cancer risks.

### Incremental lifetime cancer risk

The lung cancer risk at each 1/4° × 1/4° latitude/longitude grid cell was assessed by the ILCR induced by exposure to modeled ambient BaP concentrations. Following risk assessment methodology from US EPA<sup>37</sup>, the ILCR can be calculated as follows:

$$\text{ILCR} = \frac{1}{n} \sum_{i=1}^n \sum_a \left[ \text{CSF}_i \times \frac{(C_i \times \text{IR}_{a,g} \times Y_{a,g})}{\text{BW}_{a,g,i} \times \text{LE}} \right] \quad (1)$$

where  $a$  and  $g$  represents age and gender, respectively. CSF is the cancer slope factor,  $C$  ( $\text{mg m}^{-3}$ ) is ambient BaP air concentration, IR ( $\text{m}^3 \text{day}^{-1}$ ) is the human inhalation rate,  $y$  (year) is the exposure duration, BW (kg) is the body weight, and LE (70 years) is the average life expectancy of population. Monte Carlo (MC) simulation was used to obtain the probability distribution of BW, IR, and CSF.  $n$  is the total number of replications in  $i$ th MC simulation (10,000 times).

Body weight information by age and gender in India is presented in Supplementary Table 2, and the body weight distributions of the Indian population are generated using MC simulation; the results are illustrated in Supplementary Fig. 14. The inhalation rates are calculated by  $\text{IR} = \text{BMR} \times A \times H \times \text{VQ}$ , where BMR is the basal metabolic rate ( $\text{MJ day}^{-1}$ ),  $A$  is the ratio of energy expenditure rate to basal metabolic rate,  $H$  is the volume of oxygen consumed in the production of 1 MJ of energy ( $\text{m}^3 \text{MJ}^{-1}$ ), and VQ is a ventilatory equivalent. The parameters and detailed calculating method can be found in Shen et al.<sup>62</sup>. Since the variation of inhalation rates is associated with body weight, age, and gender, the distribution of inhalation rates of the Indian population can be calculated based on body weight distributions of Indian population by age and gender. CSF for BaP is taken as 26.6 kg (body weight)-day  $\text{mg}^{-1}$  as the maximum likelihood estimate based on epidemiological data from studies on coke oven workers, and log-transformed standard deviation is 0.38<sup>62</sup>. Male to female ratio is assumed to be 1:1 in India based on the 2011 Census of India (<https://www.censusindia2011.com>).

### MODIS 8-day fire mask and land cover types

Remote sensing data of Moderate-resolution Imaging Spectroradiometer (MODIS) including 8-day fire mask MOD14A2 and land cover type MCD12Q1 are used to explore the open fire on croplands in India (<https://adsweb.modaps.eosdis.nasa.gov/search/>). Fire mask data under three confidence levels (low, nominal, and high) are all taken into consideration.

## Model evaluation and uncertainty

To establish the level of confidence in the present model investigation, we further carried out model performance evaluations and validations via statistical error analysis between modeled and measured BaP air concentration data. Overall, the results show good agreements between modeled and measured data. Details are presented in Supplementary Note 3, Supplementary Fig. 15, and Supplementary Table 3. Our model results were also subject to uncertainties from various input variables, including BaP emission inventory, BaP concentrations, and multiple factors (Eq. 1) used in simulating lung cancer risk. BaP emission from PKU-PAH inventory in less developed countries varied with factors of 1.5–1.7 from 1965 to 2014<sup>26</sup>. The simulated BaP air concentrations are subject to errors in physicochemical properties of chemicals of concern<sup>63</sup>. The confidence factors for physicochemical properties of BaP presented in Supplementary Table 1. The uncertainties of input parameters used to calculate ILCR, including body weight and CSF, are described in the previous section. Since Monte Carlo and other sensitivity models are computationally prohibitive, we used a first-order error propagation approach to calculate the model uncertainties<sup>36,64</sup>. Further details of uncertainty analysis are presented in Supplementary Note 4.

## Emission-based BaP ILCR prediction

Annual mean BaP ILCR for the 50 year period (1965–2014) derived from the CanMETOP model are significantly associated with emissions either in the national average case or gridded spatial distribution. Without chemistry, the CanMETOP model suggests that BaP ILCRs should be linearly correlated with its emissions. These facts enable us to develop a set of regression models to predict BaP air concentrations based on emissions, as defined below:

$$\log(\text{ILCR}) = \sum_i a_i \log(E_i) + b \quad (2)$$

where ILCR is the annual mean BaP ILCR averaged over a region of concern,  $E_i$  are annual emissions from the  $i$ th emission sector,  $a_i$  and  $b$  are regression coefficients. To validate the regression models, we use BaP emissions from non-agriculture, indoor and outdoor crop residue biomass burning, and other agricultural activities from 1965 to 2013 in the IGP to establish the regression model. The model is then applied to predict BaP ILCR in 2014. The results are compared with the CanMETOP modeled BaP ILCR in the IGP in 2014, as illustrated in Supplementary Fig. 16. The good agreement ( $r = 0.98$ ,  $p < 0.001$ ) of modeled BaP ILCR by the two models suggests that the regression models could be used to predict annual BaP ILCR with reasonable accuracy and applied in the emission reduction scenario analysis.

## DATA AVAILABILITY

Source data are provided with this paper include gridded total and agricultural (field crop residue burning, indoor crop residue burning, gas/diesel combustion) BaP emission inventory from 1960 to 2014, which can be obtained from PKU-PAH inventory database (<http://inventory.pku.edu.cn/home.html>); gridded wind speed, atmospheric pressure, temperature, and precipitation, which are available from the National Center for Environmental Prediction (NCEP) reanalysis (6-hourly objectively analyzed data) (<https://www.esrl.noaa.gov/psd/data/gridded/reanalysis/>); gridded crop yields from 1965 to 1980 which are available from GRASP database11 (<https://zenodo.org>), and the data from 1981 to 2014 can be downloaded from GDHY database12 (<https://doi.org/10.1594/PANGAEA.909132>), respectively. The raw data that generate all figures in main text and Supplementary Information and support the findings of the study are available on request from corresponding authors.

## CODE AVAILABILITY

The codes that support the findings of this study are available from corresponding authors on request.

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## AUTHOR CONTRIBUTIONS

T.H. and J.M. designed the study. T.H. performed model simulations. S.S., Z.L., and L.L. collected the measured data to validate the model. T.H., J.M., and R.W.M. analyzed the data, interpreted the results, and drafted the manuscript. All authors contributed to interpreting the results and helped to improve the manuscript.

## COMPETING INTERESTS

The authors declare no competing interests.

## ADDITIONAL INFORMATION

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