Environmental Science & Technology
Article
Cite This: Environ. Sci. Technol. 2019, 53, 11337−11344
pubs.acs.org/est

$PM_{2.5}$-Associated Health Impacts of Beehive Coke Oven Ban in China

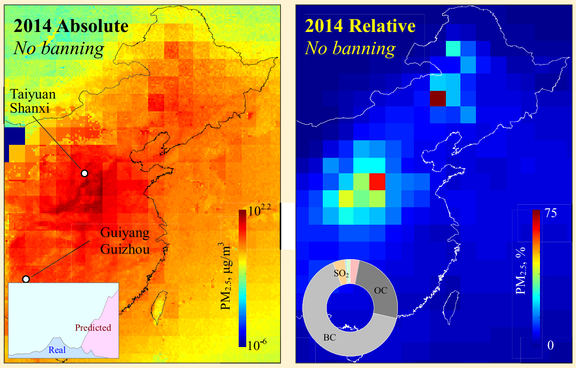
Yang Xu,[†] Qirui Zhong,[†] Xiao Yun,[†] Huizhong Shen,[†] Guofeng Shen,[†] Junfeng Liu,[†] Jianmin Ma,[†] Jianying Hu,[†] Yi Wan,[†] Xuejun Wang,[†] Chongguo Tian,[‡] and Shu Tao*[,†]

[†]College of Urban and Environmental Sciences, Laboratory for Earth Surface Processes, Sino-French Institute for Earth System Science, Peking University, Beijing 100871, China

[‡]Yantai Institute of Coastal Zone Research, Chinese Academy of Sciences, Yantai, Shangdong 264003, China

Supporting Information

ABSTRACT: Historically, beehive coke ovens (BCOs) were extensively operated in China and emitted large quantities of pollutants, including primary $PM_{2.5}$ and secondary $PM_{2.5}$ precursors, and other climate forcers. Although these ovens were legally banned in 1996 by the Coal Law, the process of phasing them out took over a decade to accomplish. Based on historical operation data derived from remote sensing images, temporal trends and the spatial distribution of the emissions of various pollutants from BCOs were compiled and used to model the resulting perturbation in ambient $PM_{2.5}$, population exposure, and $PM_{2.5}$-associated adverse health impacts. Historically, $PM_{2.5}$ originating from BCOs affected a vast region across China, which peaked in approximately 1996 and decreased afterward until the ovens' final elimination in 2011. According to the results of a supply−demand model, emissions from the BCOs would have continued to increase after 1996 if they had not been banned. As a result, national average $PM_{2.5}$ attributable to BCOs in 2014 would have been more than three times as high as that in 1996. It was estimated that the cumulative number of premature deaths associated with BCO-originating $PM_{2.5}$ from 1982 to 2014 was as high as 365 000 (95% confidence interval 259 000−402 000). The number would have nearly tripled if BCOs had not been banned and halved if the ban had been implemented immediately after the regulation was in force, suggesting the importance of legislation implementation.



INTRODUCTION

Historically, a large number of beehive coke ovens (BCOs) were operated in rural China over several decades.[1] Because of poor combustion conditions and the total absence of emission control facilities, BCOs were once among the most polluting industrial processes, and the emission factors (EFs, quantity of a pollutant emitted per unit of fuel consumed) of most air pollutants from BCOs were much higher than those from industrial coke ovens (ICOs) and most other industrial processes.[2] Although severe environmental damage caused by BCOs was well known for decades, a large gap between the capacity of ICOs and market demand in the context of soaring iron and steel production became the major driving force for the rapid expansion of BCOs in rural China. Additionally, the high profit of BCO coke production was very attractive to poor farmers.[3]

In 1996, the Coal Law was promulgated, making BCOs illegal in China.[4] Unfortunately, the ban was not fully implemented at that time. Although local environmental protection administrations launched a series of campaigns to demolish BCOs, BCOs were illegally rebuilt by local farmers and were scattered in remote mountain areas.[5] The major reasons for illegal production included a shortage of coke in the market and the low income of farmers.[6] Such a cat-and-mouse game went along for more than a decade until 2011, when BCOs were finally eliminated.[7]

To evaluate the environmental and health benefits of the ban, a quantitative assessment was conducted previously, aiming to identify lung cancer risk attributable to carcinogenic benzo(a)pyrene (BaP).[8] By reconstructing temporally and spatially resolved historical BCO production, compiling BaP emissions, and modeling ambient concentrations and population exposure, lung cancer risk associated with BaP from BCOs was quantified.[8] It was shown that several thousand lung cancer cases were attributed to BCOs during a period from 1982 to 2011, and the number would have been almost tripled if there had been no ban.[8] On the other hand, if the legislation had been fully implemented immediately after the law came into force, the risk would have been reduced by approximately two-thirds, suggesting equal importance of the legislation and its implementation.[8]

Received: July 17, 2019
Revised: September 4, 2019
Accepted: September 5, 2019
Published: September 5, 2019

DOI: 10.1021/acs.est.9b04282

Among all air pollutants, $PM_{2.5}$ (particulate matter with aerodynamic size less than 2.5 μm) is the most harmful in terms of its health impacts and can lead to a range of diseases.[9] It has been estimated that approximately 80% of premature deaths caused by air pollution resulted from exposure to $PM_{2.5}$.[9] Because BaP is only one of many toxic components of $PM_{2.5}$,[10] the overall population health impact of $PM_{2.5}$ can be orders of magnitude higher than that of BaP.[9] Ambient $PM_{2.5}$ originating from BCOs results not only from direct emissions but also from secondary formation. Secondary $PM_{2.5}$ precursors, such as SO_2, NO_x, and semivolatile organic carbons,[11] can also be emitted in large quantities from BCOs.[12] The main objectives of this study were to evaluate the historical health impact of $PM_{2.5}$ originating from BCOs and the health benefits of the BCO ban, by quantitatively modeling the emissions, chemical transport in the atmosphere, population exposure, and health consequences.

METHODS

BCO Production. Historical BCO coke production data compiled in a previous study were adopted.[8] In brief, values of annual production from 1982 to 2014 in China were from statistical yearbooks.[13] Detailed spatial distribution and interannual variation were extracted from satellite images (Landsat) using composition (TM721 and TM751 bands) technology from 1987 to 2011.[14] The results agree well with recorded BCO coke production statistics in total quantities.[14] To cover a period from 1982 to 1986, which was regarded as the first stage of the economic reform in China, values from these years were extrapolated by assuming that the temporal and spatial patterns were the same as those in 1987. BCO coke production for the no-banning scenario was estimated previously.[8] In brief, a multivariate regression model was developed based on production data before 1996. The significant independent variables identified include fine coal production, total coke production, gross domestic production, and producer's price index for coke.[8]

Emissions. Gridded emissions of primary $PM_{2.5}$ and secondary $PM_{2.5}$ precursors from various sources in mainland China, including BCOs, with 0.1 × 0.1° spatial resolution and monthly temporal resolution were compiled from PKU emission inventories.[15] Chemical species or their sources missing from the PKU database were derived from other information sources. Specifically, ammonia (NH_3) from the agricultural sector, nonbiogenic volatile organic compounds (VOCs), and biogenic VOCs were obtained from EDGAR, HTAP, and MEGAN.[16−19]

Transport Modeling. The Model for Ozone and Related chemical Tracers, version 4 (MOZART-4) was used to model $PM_{2.5}$ concentrations in ambient air for a period from 1982 to 2014.[20] The horizontal resolution was 1.895° (latitude) × 1.875° (longitude), with 28 vertical layers and a 15 min time step. Meteorological inputs were NCEP/NCAR reanalysis products for all years.[21] Spatial resolution was downscaled to 0.1 × 0.1° using aerosol optical depths from MODIS as a surrogate.[22,23] The model was run twice based on total emissions from all sources and emissions with no BCOs, respectively. The differences in the calculated $PM_{2.5}$ between the two model runs were derived as BCO contributions. Because the base model (with all emissions) was previously run in another study using the same model and the same emission database, and was successfully validated against a large quantity of observations,[24] model validation was not repeated in this study.

Health Impact. Population exposure to $PM_{2.5}$ was derived based on the downscaled $PM_{2.5}$ concentrations. Numbers of premature deaths associated with $PM_{2.5}$ exposure were derived for five diseases of acute lower respiratory infections for children under five, lung cancer, ischemic heart disease (IHD), cerebrovascular disease, and chronic obstructive pulmonary disease (COPD), based on the latest hazard ratios in the Global Exposure Mortality model (GEMM) established by Burnett et al.[25] Background disease burdens were from the Global Burden of Disease study.[26]

Data Analysis. Statistical analysis with a significance level of 0.05 was conducted using SPSS 23.0 released by the International Business Machines Corporation.[27] Monte Carlo simulations were performed 1000 times to assess model uncertainty, using MATLAB R2016b released by Math-Works.[28] Coefficients of variation of model-estimated $PM_{2.5}$ concentrations were determined by the sensitivities of $PM_{2.5}$ concentrations to emissions of various species based on a series of sensitivity tests conducted in previous studies.[24,29] Grid cells were not treated individually. Distribution parameters of dose−response curves after GEMM are listed in Table S1.[25]

RESULTS AND DISCUSSION

EFs for BCOs. Among a variety of industrial processes using coal as fuel, coke production is the one with the highest emissions of a range of air pollutants, largely due to limited oxygen supply and high fugitive leakage, which are difficult to control.[30] The pollutants of greatest concern included incomplete combustion products (such as primary $PM_{2.5}$, black carbon (BC), organic carbon (OC), and carbon monoxide (CO)),[12,31] oxidized fuel components (such as SO_2), and high-temperature byproducts (such as NO_x). While emissions of these pollutants from ICOs can be well controlled, all pollutants from BCOs are released into the environment with no mitigation efforts.[2] High emission potential can be observed from the mean EFs of seven major air pollutants from BCOs, as shown in Figure S1 and Table S2 in comparison with the EFs for several other industrial processes and other sectors.[15] The EFs of most pollutants, including primary $PM_{2.5}$, CO, BC, OC, NH_3, and BaP, from BCOs are approximately one order of magnitude higher than those for other major industrial and nonindustrial sources. Based on the results of global atmospheric chemical transport modeling, the four most important pollutants contributing to ambient $PM_{2.5}$ are primary $PM_{2.5}$, SO_2, NO_x, and NH_3;[24] BCOs contribute significantly to all of these pollutants.

Since the late 1980s, when the Air Pollution Prevention and Control Law came into force,[4] a series of mitigation actions have been taken to battle pollution in China,[32] and emission control facilities have been extensively installed or upgraded in power generation, industrial, and transportation sectors.[33] Consequently, the EFs of major air pollutants from these sources have gradually decreased.[24] Taking primary $PM_{2.5}$ as an example, the temporal trends of EFs from major emission sources, including BCOs, coal-fired power plants, transportation, and residential coal stoves, from 1982 to 2014 in China are shown in Figure S2.[15] Because particulates are technically easy and less expensive to remove than other gas-phase pollutants, the average EFs of $PM_{2.5}$ from most activities showed substantial decreases during this period. In fact, similar decreasing trends can be observed for other pollutants,

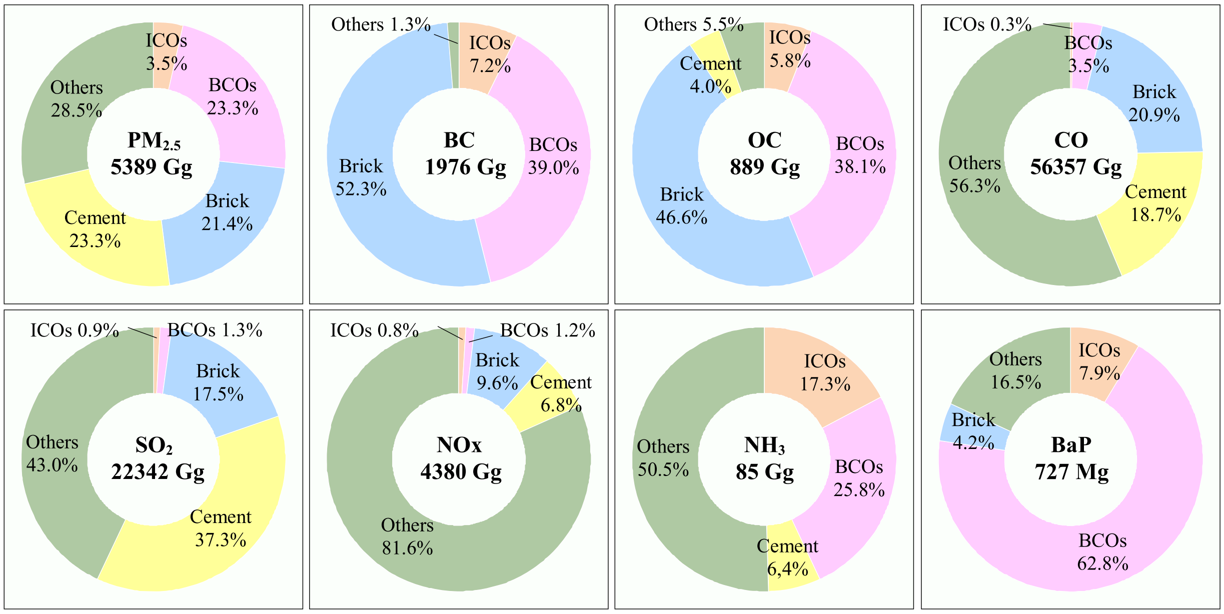


Figure 1. Relative contributions of BCOs to the total annual emissions of major air pollutants from industrial emissions. Contributions of ICOs, brick production, and cement production are also shown for comparison. All other industrial sources are lumped together as "others".

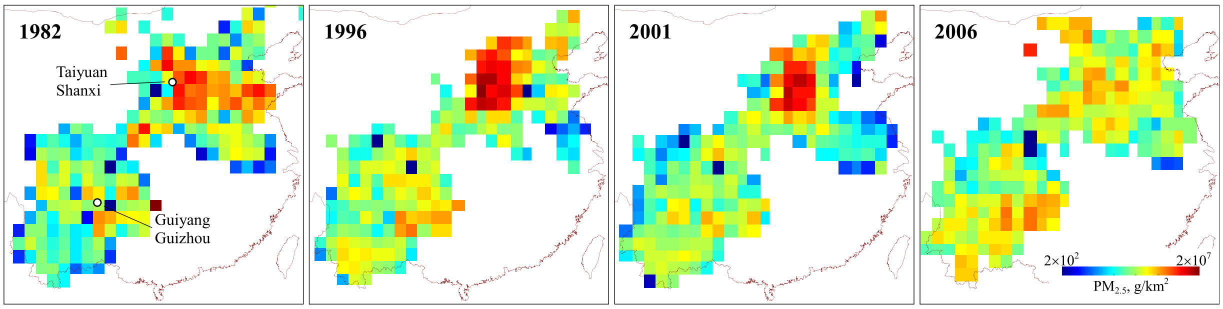


Figure 2. Spatial distribution of annual emissions of primary $PM_{2.5}$ from BCOs in eastern China in 1982, 1996, 2001, and 2006.

including SO_2 and NO_x, though at a slower pace.[4] Two exceptions were residential coal burning and BCOs, whose pollutant removal facilities were totally absent and for which EFs remained constant over the period.

Emissions from BCOs, as reflected by the very high and unchanged EFs, have contributed significantly to the total emissions of major air pollutants over the past few decades. The relative importance of BCOs increased until the ban was enforced in 1996 because of slowly increasing or even declining emissions from most other sources. Figure 1 shows the relative contributions of BCOs to the total annual emissions of eight major air pollutants from all industrial sources in 1996, when BCO production was at its peak. If the emissions of major incomplete combustion products from coke production are separated into BCOs and ICOs, emissions from BCOs accounted for 23, 39, and 38% of the total emissions of primary $PM_{2.5}$, BC, and OC, respectively, from all industrial processes in 1996. Even when all anthropogenic sources were considered, the relative contributions to the total emissions of these air pollutants were still as high as 10, 23, and 8.3%, respectively. On the other hand, the contributions of BCOs to major precursors of secondarily formed $PM_{2.5}$ were less important. For example, although more than one-fourth of NH_3 from industrial sources stemmed from BCOs, NH_3 from the agricultural sector dominated the overall emissions.[34]

BCOs in China were not evenly distributed in space. Figure 2 shows the geospatial distributions of annual emissions of primary $PM_{2.5}$ from BCOs in eastern China in 1982, 1996, 2001, and 2006, which were derived based on the EFs and spatially resolved BCO production data. It appears that the strong emissions of $PM_{2.5}$ from BCOs were not localized. Instead, numerous BCOs were located extensively across a vast region across all of eastern China, stretching from Hebei and Shandong in the north to Guizhou and Yunnan in the southwest, with the highest emissions in Shanxi. The entire region was under the direct influence of BCO emissions, even without taking atmospheric dispersion and transportation into consideration. Furthermore, this region was densely populated with a total population of approximately 600 million, constituting almost half of the total population of China. These high-emission areas share common features of rich coal reserves and poor rural residents with low income.[7] In terms of BCO production, Shanxi was superior to other provinces.[7] Local farmers often had easy access to coal produced from many small coal mines,[35] and the booming iron-steel industry in northern China created a huge demand for coke.[36] In 1996, coal production in Shanxi reached 331 million tons, 12.7% of which was from below-scale coal mines.[14] Meanwhile, the province contributed 25.7% of the total coal production and 85.8% of BCO coke production in China.[7] Over more than 2

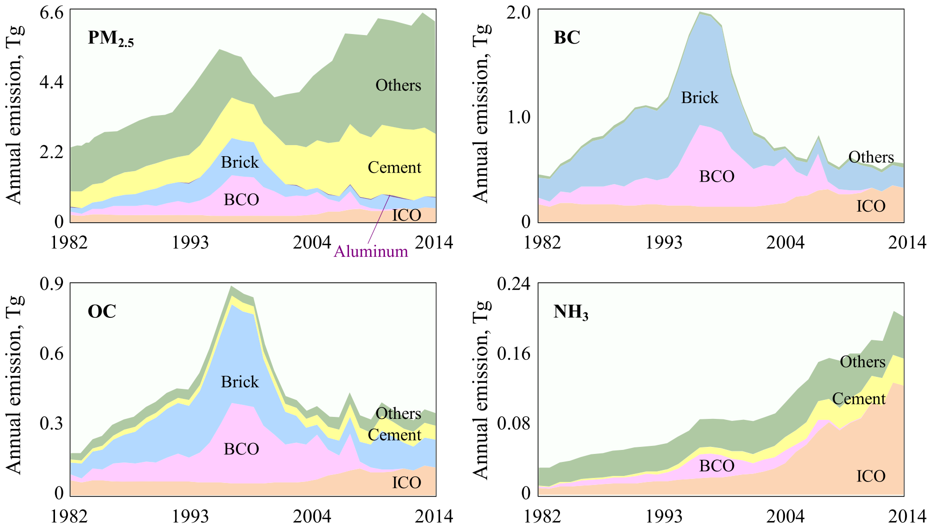


Figure 3. Temporal trends in annual emissions of major air pollutants from BCOs and other industrial sources from 1982 to 2014.

decades from 1982 to 2006, the spatial pattern of primary $PM_{2.5}$ emissions from BCOs did not change much. The exception was that there was a slight increase in emissions in southwestern and eastern China between 2001 and 2006, as well as a significant emission reduction in Shanxi and adjacent provinces. This is in line with provincial BCO productions.

Influence of the BCO Ban on Emissions. Figure 3 shows the temporal trends in annual emissions of major air pollutants, including primary $PM_{2.5}$, BC, OC, and NH_3, which influence $PM_{2.5}$ levels in the ambient air from BCOs and other industrial sources from 1982 to 2014. Coke production from BCOs and, consequently, emissions of major air pollutants from this source changed over the years. Consistent with emissions from many other industrial sources, variations were driven either economically by market forces or legislatively by regulations. There were continuous increases in emissions from all of these sources for 2 decades starting in the early 1980s, which resulted from rapid industrialization and extensive infrastructure expansion driven by the open-door policy and rapid economic development.[8] During that period, the total output of industry (purchasing power parity) in China increased more than 15 times from 218 billion RMB in 1982 to 3366 billion RMB in 1996, and the annual productions of coal and coke increased from 621.8 and 43.4 million tons to 1292.2 and 124.2 million tons, respectively.[13] Annual emissions of primary $PM_{2.5}$ from all industrial sources and from BCOs increased from 2168 and 168 Gg to 4779 and 1253 Gg, respectively. The industrial emissions reached the peak in approximately 1996 and began to decrease due both to the economic depression and to the nationwide campaign to control air pollution in most industrial sectors.[15] For example, the annual emission of PM from industrial sources decreased from 8.5 million tons at its peak in 1996 to 5.4 million ton in 2009.[37] The rebound of $PM_{2.5}$ emissions from cement production and other industrial sources was associated with a decade-long infrastructure boom, as the cement production increased at an average annual rate of 12% from 2000 to 2010.[15] For BCOs, 1996 happened to be the year when the Coal Law came into force. Although BCOs were legally banned by the end of that year, emissions decreased but did not end. The legal ban was difficult to implement because (1) BCOs were easily built at low cost, (2) they were scattered widely in rural regions, especially in mountainous areas, and (3) production benefited local economies.[2] In fact, BCOs were not completely eliminated until 2011, after a decade-long cat-and-mouse game between local governments and rural villagers. Meanwhile, after BCOs were banned, production from conventional coke ovens was intended to compensate. However, industrial-scale coke ovens took years to construct, and their capacity did not begin to catch up until the early 2000s,[8] leaving a gap between demand and supply, which was also a driving force for the continuation of BCO production.

To evaluate the impact of the BCO ban on emission of BaP from 1996 to 2014, a regression model was developed by Xu et al. to simulate BCO production for a business-as-usual scenario assuming that there was no BCO ban at all.[8] Based on several socioeconomic parameters, the annual BCO coke production driven by market demand was simulated for this particular scenario[8] and compared with the actual production to evaluate the effectiveness of the legislation. In this study, the same model was applied to estimate the emissions of major air pollutants, including primary $PM_{2.5}$ and secondary $PM_{2.5}$ precursors subject to the realistic and business-as-usual scenarios, respectively. Taking primary $PM_{2.5}$ as an example, the emissions for the two scenarios with and without the ban are shown in Figure S3. The difference between the two is the emission reduction due to the ban. For the realistic scenario, the emission did not terminate immediately after the ban came into force in 1996. Instead, BCO coke production and, consequently, emissions of various air pollutants continued until 2011, indicating poor implementation of the law. Without the ban, the annual emissions of primary $PM_{2.5}$ and other pollutants would have soared after 2000 and reached 2.1 Tg in 2014, nearly double the quantity in 1996 (1.2 Tg).

Ambient $PM_{2.5}$ Originating from BCOs. Based on the calculated emissions, ambient concentrations of both primary and secondary $PM_{2.5}$ were predicted by atmospheric transport modeling. Because BCOs were absent in the west, the northeast, and the southeast of China, emissions of various air pollutants from BCOs led to only a slight increase in the national average $PM_{2.5}$ concentration. In the peak year of 1996, the contribution of BCOs to the national average $PM_{2.5}$ level (26.1 $\mu g/m^3$) was merely 0.44 $\mu g/m^3$. In contrast, the population-weighted $PM_{2.5}$ concentration increased by 2.9

μg/m^3 due to BCO emissions in that year (58.9 μg/m^3 as the total from all sources), indicating the importance for health assessment of the spatial overlap of BCOs and population. The greatest contribution of BCOs to a local grid cell with approximately 100 km^2 resolution can be as high as 56 μg/m^3 (1996), which exceeded the national standard.

Although BCOs were scattered in an elongated region extending from Hebei in north China to Yunnan in the southeast, $PM_{2.5}$ concentrations over the entire region and even in neighboring areas were affected by BCO emissions due to atmospheric transport. The resulting change in ambient $PM_{2.5}$ over eastern China is mapped in Figure 4, showing both log-scaled absolute and relative BCO contributions to annual mean $PM_{2.5}$ in 1996. Consequently, not only the villages where BCOs were in operation but also the villages close to the production areas were severely affected. In fact, although a relatively small number of rural residents were involved in BCO coke production, the entire population of a vast region were exposed to extra $PM_{2.5}$ with levels exceeding 10 μg/m^3. As shown in Figure 4, the air quality in Shanxi, the province with the highest BCO coke production, was affected most significantly. As the most affected area in China, the contribution of BCOs in Jiexiu, Shanxi, to annual average $PM_{2.5}$ concentrations was approximately 50% in 1996. Although BCO production in Guizhou was higher than that in all other provinces except Shanxi, it was a distant second after Shanxi in terms of emissions and contribution of BCOs to ambient $PM_{2.5}$ concentrations. In 1996, the levels of total BCO coke production in Shanxi and Guizhou were 46.4 and 1.9 million tons, and the average BCO-originating additive $PM_{2.5}$ concentrations were 11.5 and 1.8 μg/m^3, respectively. The large differences in the absolute and relative contributions between the two provinces were due to much high emissions in Shanxi.[38] Distribution of population-weighted $PM_{2.5}$ from BCO, as an intermediate variable for population exposure calculation, is mapped in Figure S4, after being divided by the mean concentration. As shown, the overall risks are strengthened in the regions with high population density.

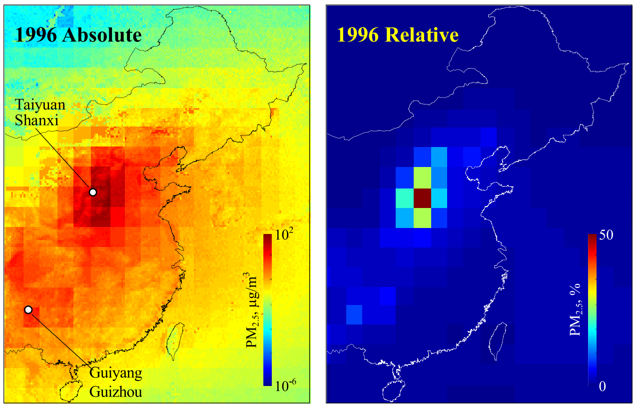


Figure 4. Contributions of BCO emissions to annual mean ambient $PM_{2.5}$ in eastern China in 1996. The results are shown in both absolute (A) and relative (B) terms.

As discussed previously, BCOs differed greatly from many other sources in composition profile of emissions. BCO emissions were dominated by primary $PM_{2.5}$, including OC and BC, whereas many other sources, including most industrial sources, were rich in secondary PM precursors, such as SO_2 and NO_x. As a result of these differences in emission composition, ambient $PM_{2.5}$ originating from BCOs played an important role in primary $PM_{2.5}$, with OC, BC, and unspecified $PM_{2.5}$ accounting for 66, 25, and 3% of the total, respectively (Figure 5). In comparison, the fraction was less than 30% for other sources combined. Therefore, the principal outcome of the BCO ban was the reduction of primary $PM_{2.5}$ in the ambient air.

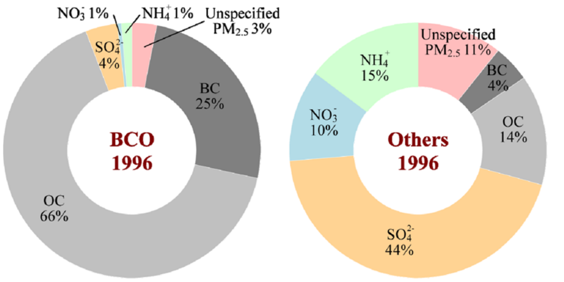


Figure 5. Major components of ambient $PM_{2.5}$ originating from BCOs (left) and from all other sources (right).

The temporal trends in BCO contributions to the national average and maximum grid cell $PM_{2.5}$ concentrations simulated for two scenarios, with and without BCO ban, are shown in Figure 6. The no-ban scenario assumed that BCO production was totally driven by market forces, and the temporal trend in BCO production was predicted using the regression model previously developed.[8] The general BCO production trends in these two scenarios were similar to those of emissions. In the realistic scenario, the contributions of BCOs to ambient $PM_{2.5}$ concentrations, as reflected in either the national average or maximum grid cell levels, had decreased rapidly after 1996, when the ban was formally introduced, although the influence did not immediately end as intended. Such a phenomenon was common in China during that period, when many environmental regulations and laws were issued but failed to be properly implemented in a timely manner.[39] The lag in implementation was particularly apparent for nonindustrial-scale enterprises such as BCOs, which were built and operated by rural farmers in poor and remote mountainous areas and could hardly be monitored or inspected. The banning process was practically a decade-long cat-and-mouse game. Governmental agencies had to send teams to demolish operating ovens in rural villages. However, more often than not, the ovens were soon rebuilt by local farmers, simply because the cost of building a new oven was inexpensive and the potential profit of producing and selling coke was very high, compared with the low income of the farmers.[2] As a result, similar regulations and notifications of the ban were issued again and again over these years.[40] By 2011, 15 years after the Coal Law was in place, all BCOs were finally terminated. Despite the reluctance in its implementation, the Coal Law showed positive influences on the phasing-out of BCOs and the decrease in BCO emissions.

As discussed above, if there had been no such regulation, the emissions of various pollutants from BCOs and, consequently, contributions to ambient $PM_{2.5}$ would have increased continuously, driven by the growing coke demand. The difference between the no-ban and the realistic scenarios can be seen in Figure 6. In the no-ban scenario, the annual average $PM_{2.5}$ concentration would have increased by approximately

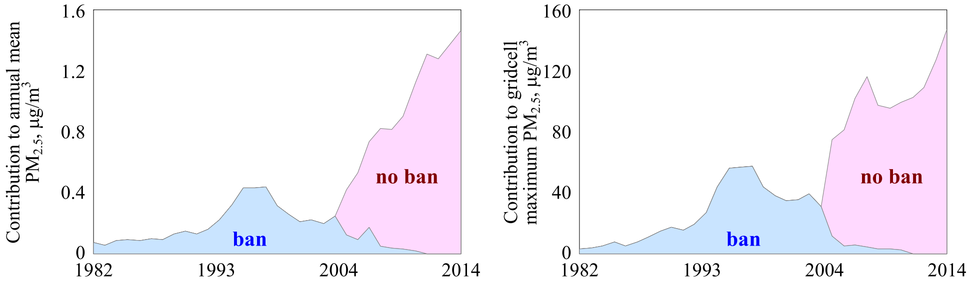


Figure 6. Temporal trends in ambient $PM_{2.5}$ originating from BCOs in eastern China from 1982 to 2014 for two scenarios, with and without the ban.

1.5 $\mu g/m^3$ in 2014 due to BCOs, which was three times that in the peak year of the realistic scenario (i.e., in 1996). The average population-weighted $PM_{2.5}$ concentration would have increased by 8.8 $\mu g/m^3$, with the concentration increases in single grid cells as high as 147 $\mu g/m^3$. As shown in Figure 7, the contribution in 2014 would have been more concentrated in a relatively small region around Shanxi, indicating severe adverse localized impacts without the ban. The distributions of primary and secondary $PM_{2.5}$ originally from BCO emissions are mapped in Figure S5, showing that the areas near the emission sources such as Shanxi are profoundly affected by primary $PM_{2.5}$ and the influence of secondary $PM_{2.5}$ reached a much larger region.

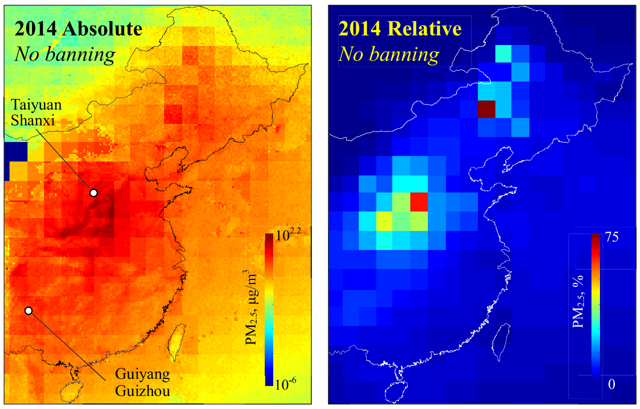


Figure 7. Spatial distributions of absolute and relative contributions of BCOs to ambient $PM_{2.5}$ in eastern China in 2014 assuming no ban.

Health Benefits of the Ban. Levels of BCO production were higher in northern and southwestern China, especially in Shanxi. Unfortunately, these provinces are among the regions with the highest population densities. With dispersion of the emitted air pollutants, a very large area was affected. For example, if 10 $\mu g/m^3$ additive ambient $PM_{2.5}$ concentration was selected as a threshold, the regions where air quality was significantly affected by BCOs in 1996 reached 200 000 km^2, covering a total population of 109 million; this population was almost 9% of the total population in China, suggesting that BCOs were an important source of air pollution at that time.

For the period from 1982 to 2014, the cumulative number of premature deaths caused by exposure to ambient $PM_{2.5}$ associated with BCOs reached 365 000 (95% confidence interval 259 000−402 000). Lung cancer, stroke, ischemic heart disease (IHD), acute lower respiratory infection, and chronic obstructive pulmonary disease (COPD) contributed 13, 27, 12, 10, and 37%, respectively, to premature mortality. A previous study revealed that approximately 3500 cases of cumulative lung cancer were attributable to exposure to polycyclic aromatic hydrocarbons (PAHs) originating from BCO production during the same period. Given that the majority of carcinogenic PAH compounds (i.e., high-molecular-weight PAHs) are bonded to fine particles, we estimated that PAHs contributed approximately 7% of the lung cancer incidence induced by exposure to $PM_{2.5}$ associated with BCOs. If the ban was not implemented, the estimated cumulative premature deaths from BCOs would have been 1 057 000 (769 000−1 16 0 000), which would have tripled the mortality observed in the ban case, suggesting a difference of 692 000 premature deaths.

In summary, operation of BCOs over the past few decades had caused severe health impact in almost entire eastern China. The BCO ban had led to a reduction of 692 000 premature deaths associated with $PM_{2.5}$ exposure, suggesting that legislation can lead to significant health benefits. On the other hand, if the ban had been implemented immediately after the law was in force, additional 183 000 lives would have been saved. In the past, many environmental laws and regulations have been established in China, covering a variety of areas of water, air, soil, ecosystem, and human health. However, in many cases, the effectiveness of the legislation was reduced by the slow implementation. A lesson that can be learned from this study is that legislation implementation is as important as the enactment. More attention needs to be paid by policymakers on the implementation of various existing environmental legislations.

ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.est.9b04282.

Results of EFs of major air pollutants for coke and other industrial activities, temporal trends of EFs of primary $PM_{2.5}$ for coke and other industrial activities, and annual emissions of primary $PM_{2.5}$ from BCOs (PDF)

AUTHOR INFORMATION

Corresponding Author

*E-mail: taos@pku.edu.cn. Phone/Fax: +86-10-62751938.

ORCID

Huizhong Shen: 0000-0003-1335-8477
Junfeng Liu: 0000-0002-7199-6357
Jianmin Ma: 0000-0002-6593-570X
Jianying Hu: 0000-0003-1965-7494
Yi Wan: 0000-0003-1312-6254

Xuejun Wang: 0000-0001-9990-1391
Chongguo Tian: 0000-0001-6058-9353
Shu Tao: 0000-0002-7374-7063

Notes

The authors declare no competing financial interest.

■ ACKNOWLEDGMENTS

This work was funded by the National Natural Science Foundation of China (Grants 41830641, 41629101, and 41821005).

■ REFERENCES

(1) The editorial board of China Steel Yearbook. *China Steel Yearbook*; Metallurgical Industry Press: Beijing, China, 2010.

(2) Ma, Y. Pollution and control of beehive coke ovens in China (in Chinese). *Environ. Sustainable Dev.* **1985**, *12*, 17−20.

(3) Shao, Y. Restriction and improvement beehive coke ovens (in Chinese). *Energy China* **1990**, *6*, 33−36.

(4) Standing Committee of the National People's Congress. *Law of the People's Republic of China on the Coal Industry*; Law Press: Beijing, China, 1996.

(5) Guo, Y. J. On the reasonable improvement of beehive coke ovens (in Chinese). *Resour. Econ. Compr. Util.* **1995**, *4*, 24−30.

(6) Huo, H.; Lei, Y.; Zhang, Q.; Zhao, L. J.; He, K. B. China's coke industry: Recent policies, technology shift, and implication for energy and the environment. *Energy Policy* **2012**, *51*, 397−404.

(7) The editorial board of China Coal Industry Yearbook. *Production of Comprehensive Utilization of Coal. China Coal Industry Yearbook*; Coal Industry Press: Beijing, China, 2012.

(8) Xu, Y.; Shen, H. Z.; Yun, X.; Gao, F.; Chen, Y. L.; Li, B. G.; Liu, J. F.; Ma, J. M.; Wang, X. L.; Liu, X. P.; Tian, C. G.; Xing, B. S.; Tao, S. Health effects of banning beehive coke ovens and implementation of the ban in China. *Proc. Natl. Acad. Sci. U.S.A.* **2018**, *115*, 2693−2698.

(9) World Health Organization. WHO Global Ambient Air Quality Database (update 2018). https://www.who.int/airpollution/data/en/ (accessed 2018).

(10) Bai, X.; Liu, Y.; Wang, S.; Liu, C.; Liu, F.; Su, G.; Peng, X.; Yuan, C.; Jiang, Y.; Yan, B. Ultrafine particle libraries for exploring mechanisms of $PM_{2.5}$-induced toxicity in human cells. *Ecotoxicol. Environ. Saf.* **2018**, *157*, 380−387.

(11) Heo, J.; Adams, P. J.; Gao, H. O. Public Health Costs of Primary $PM_{2.5}$ and Inorganic $PM_{2.5}$ Precursor Emissions in the United States. *Environ. Sci. Technol.* **2016**, *50*, 6061−6070.

(12) Weitkamp, E. A.; Lipsky, E. M.; Pancras, P. J.; Ondov, J. M.; Polidori, A.; Turpin, B. J.; Robinson, A. L. Fine particle emission profile for a large coke production facility based on highly time-resolved fence line measurements. *Atmos. Environ.* **2005**, *39*, 6719−6733.

(13) National bureau of statistics of China. *China Statistical Yearbook*; China Statistics Press: Beijing, China, 2017.

(14) Kong, X. S.; Miao, F.; Liu, H. F.; Dong, Y. Y. Dynamic monitoring of ingigenous coke-production using multitemporal Landsat remote sensing images: a case study in south-east, Shanxi Province. *Remote Sens. Technol. Appl.* **2005**, *20*, 460−464.

(15) Peking University Inventory Dataset. version 2. http://inventory.pku.edu.cn/ (accessed, 2017).

(16) Sindelarova, K.; Granier, C.; Bouarar, I.; Guenther, A.; Tilmes, S.; Stavrakou, T.; Müller, J. F.; Kuhn, U.; Stefani, P.; Knorr, W. Global data set of biogenic VOC emissions calculated by the MEGAN model over the last 30 years. *Atmos. Chem. Phys.* **2014**, *14*, 9317−9341.

(17) Janssens-Maenhout, G.; Crippa, M.; Guizzardi, D.; Dentener, F.; Muntean, M.; Pouliot, G.; Keating, T.; Zhang, Q.; Kurokawa, J.; Wankmüller, R.; van der Gon, H. D.; Kuenen, J. J. P.; Klimont, Z.; Frost, G.; Darras, S.; Koffi, B.; Li, M. HTAP_v2.2: a mosaic of regional and global emission grid maps for 2008 and 2010 to study hemispheric transport of air pollution. *Atmos. Chem. Phys.* **2015**, *15*, 11411−11432.

(18) Crippa, M.; Guizzardi, D.; Muntean, M.; Schaaf, E.; Dentener, F.; van Aardenne, J. A.; Monni, S.; Doering, U.; Olivier, J. G. J.; Pagliari, V.; Janssens-Maenhout, G. Gridded Emissions of Air Pollutants for the period 1970−2012 within EDGAR v4.3.2. *Earth Syst. Sci. Data* **2018**, *10*, 1987−2013.

(19) van der Werf, G. R.; Randerson, J. T.; Giglio, L.; van Leeuwen, T. T.; Chen, Y.; Rogers, B. M.; Mu, M.; van Marle, M. J. E.; Morton, D. C.; Collatz, G. J.; Yokelson, R. J.; Kasibhatla, P. S. Global fire emissions estimates during 1997-2016. *Earth Syst. Sci. Data* **2017**, *9*, 697−720.

(20) Emmons, L. K.; Walters, S.; Hess, P. G.; Lamarque, J. F.; Pfister, G. G.; Fillmore, D.; Granier, C.; Guenther, A.; Kinnison, D.; Laepple, T.; Orlando, J.; Tie, X.; Tyndall, G.; Wiedinmyer, C.; Baughcum, S. L.; Kloster, S. Description and evaluation of the Model for Ozone and Related chemical Tracers, version 4 (MOZART-4). *Geosci. Model Dev.* **2010**, *3*, 43−67.

(21) Kalnay, E.; Kanamitsu, M.; Kistler, R.; Collins, W.; Deaven, D.; Gandin, L.; Iredell, M.; Saha, S.; White, G.; Woollen, J.; Zhu, Y.; Chelliah, M.; Ebisuzaki, W.; Higgins, W.; Janowiak, J.; Mo, K. C.; Ropelewski, C.; Wang, J.; Leetmaa, A.; Reynolds, R.; Jenne, R.; Joseph, D. The NCEP/NCAR 40-Year Reanalysis Project. *Bull. Am. Meteorol. Soc.* **1996**, *77*, 437−471.

(22) Levy, R. C.; Remer, L. A.; Kleidman, R. G.; Mattoo, S.; Ichoku, C.; Kahn, R.; Eck, T. F. Global evaluation of the Collection 5 MODIS dark-target aerosol products over land. *Atmos. Chem. Phys.* **2010**, *10*, 10399−10420.

(23) van Donkelaar, A.; Martin, R. V.; Brauer, M.; Kahn, R.; Levy, R.; Verduzco, C.; Villeneuve, P. J. Global estimates of ambient fine particulate matter concentrations from satellite-based aerosol optical depth: development and application. *Environ. Health Perspect.* **2010**, *118*, 847−855.

(24) Zhong, Q. R.; Ma, J. M.; Shen, G. F.; Shen, H. Z.; Zhu, X.; Yun, X.; Meng, W. J.; Cheng, H. F.; Liu, J. F.; Li, B. G.; Wang, X. L.; Zeng, E. Y.; Guan, D. B.; Tao, S. Distinguishing Emission-Associated Ambient Air $PM_{2.5}$ Concentrations and Meteorological Factor-Induced Fluctuations. *Environ. Sci. Technol.* **2018**, *52*, 10416−10425.

(25) Burnett, R.; Chen, H.; Szyszkowicz, M.; Fann, N.; Hubbell, B.; Pope, C. A.; Apte, J. S.; Brauer, M.; Cohen, A.; Weichenthal, S.; Coggins, J.; Di, Q.; Brunekreef, B.; Frostad, J.; Lim, S. S.; Kan, H. D.; Walker, K. D.; Thurston, G. D.; Hayes, R. B.; Lim, C. C.; Turner, M. C.; Jerrett, M.; Krewski, D.; Gapstur, S. M.; Diver, W. R.; Ostro, B.; Goldberg, D.; Crouse, D. L.; Martin, R. V.; Peters, P.; Pinault, L.; Tjepkema, M.; Donkelaar, A.; Villeneuve, P. J.; Miller, A. B.; Yin, P.; Zhou, M. G.; Wang, L. J.; Janssen, N. A. H.; Marra, M.; Atkinson, R. W.; Tsang, H.; Thach, Q.; Cannon, J. B.; Allen, R. T.; Hart, J. E.; Laden, F.; Cesaroni, G.; Forastiere, F.; Weinmayr, G.; Jaensch, A.; Nagel, G.; Concin, H.; Spadaro, J. V. Global estimates of mortality associated with long-term exposure to outdoor fine particulate matter. *Proc. Natl. Acad. Sci. U.S.A.* **2018**, *115*, 9592−9597.

(26) Fischer, S. L.; Koshland, C. P. Daily and peak 1 h indoor air pollution and driving factors in a rural Chinese village. *Environ. Sci. Technol.* **2007**, *41*, 3121−3126.

(27) IBM SPSS Statistics Manuals. https://www-01.ibm.com/support/docview.wss?uid=swg27038407#en (accessed 2015).

(28) Math Works Documentation for MATLAB. https://www.mathworks.com/help/pdf_doc/matlab/index.html (accessed 2016).

(29) Shen, H. Z.; Tao, S.; Chen, Y. L.; Ciais, P.; Guneralp, B.; Ru, M. Y.; Zhong, Q. R.; Yun, X.; Zhu, X.; Huang, T. B.; Tao, W.; Chen, Y. C.; Li, B. G.; Wang, X. L.; Liu, W. X.; Liu, J. F.; Zhao, S. Q. Urbanization-induced population migration has reduced ambient PM2.5 concentrations in China. *Sci. Adv.* **2017**, *3*, No. e1700300.

(30) Pilarczyk, E.; Sowa, F.; Kaiser, M.; Kern, W. Emissions at Coke Plants: European Environmental Regulations and Measures for Emission Control. *Trans. Indian Inst. Met.* **2013**, *66*, 723−730.

(31) Shen, H. Z.; Tao, S.; Liu, J. F.; Huang, Y.; Chen, H.; Li, W.; Zhang, Y. Y.; Chen, Y. C.; Su, S.; Lin, N.; Xu, Y. Y.; Li, B. G.; Wang, X. L.; Liu, W. X. Global lung cancer risk from PAH exposure highly

depends on emission sources and individual susceptibility. *Sci. Rep.* **2014**, 4.

(32) Chen, H. Y.; Hao, Y.; Li, J. W.; Song, X. J. The impact of environmental regulation, shadow economy, and corruption on environmental quality: Theory and empirical evidence from China. *J. Cleaner Prod.* **2018**, *195*, 200−214.

(33) Zhang, H. F.; Wang, S. X.; Hao, J. M.; Wang, X. M.; Wang, S. L.; Chai, F. H.; Li, M. Air pollution and control action in Beijing. *J. Cleaner Prod.* **2016**, *112*, 1519−1527.

(34) Meng, W. J.; Zhong, Q. R.; Yun, X.; Zhu, X.; Huang, T. B.; Shen, H. Z.; Chen, Y. L.; Chen, H.; Zhou, F.; Liu, J. F.; Wang, X. M.; Zeng, E. Y.; Tao, S. Improvementin of a Global High-Resolution Ammonia Emission Inventory for Combustion and Industrial Sources with New Data from the Residential and Transportation Sectors. *Environ. Sci. Technol.* **2017**, *51*, 2821−2829.

(35) Hu, Y. Z. Development of cokin g industry in Shanxi Province since the reform and opening up of China for 30 years (in Chinese). *Coal Chem. Ind.* **2008**, *6*, 4−7.

(36) Dong, X. D. What to think about the soaring price of coke. *China Steel Focus* **2003**, *4*, 42−43.

(37) Liu, R. J.; Zhang, Z. H. Research on condition of China's industrial smoke emission (in Chinese). *Ecol. Environ. Sci.* **2012**, *21*, 694−699.

(38) Zhang, J. J.; Fu, M. C.; Geng, Y. H.; Tao, J. Energy saving and emission reduction: A project of coal-resource integration in Shanxi Province, China. *Energy Policy* **2011**, *39*, 3029−3032.

(39) Chang, M. M. Guangxi is going to strengthen the legislation about environment and law enforcement inspection to increase the implementation of laws and regulations. China Environment News. http://www.cqyuyang.cn/changzhi/407604.html.

(40) Shanxi Environmental Protection Board. Notice on further strengthening the remediation of the banned small and primitive industry. Shanxi EPB Official Documents 2008, *80*.