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Reduction in polycyclic aromatic hydrocarbon exposure in Beijing following China's clean air actions

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ABSTRACT

Exposure to polycyclic aromatic hydrocarbons (PAHs) in the Chinese population was among the highest globally and associated with various adverse effects. This study examines the impact of China's two-phase clean air initiatives, namely the Air Pollution Prevention and Control Action Plan (APPCAP) in 2013–2017 and the Blue-Sky Defense War (BSDW) in 2018–2020, on PAH levels and human exposures in Beijing. To evaluate the effects of APPCAP, we measured 16 PAHs in 287 PM_{2.5} samples collected in Beijing and 9 PAH metabolites in 358 urine samples obtained from 54 individuals who traveled from Los Angeles to Beijing between 2014 and 2018. The concentration of PM_{2.5}-bound benzo[a]pyrene equivalents (BaPeq) decreased by 88.5% in 2014–2018 due to reduced traffic, coal, and biomass emissions. PAH metabolite concentrations in travelers' urine decreased by 52.3% in Beijing, correlated with changes in PM_{2.5} and NO₂ levels. In contrast, no significant changes were observed in Los Angeles. To evaluate BSDW's effects, we collected 123 additional PM_{2.5} samples for PAH measurements in 2019–2021. We observed sustained reductions in BaPeq concentrations attributable to reductions in coal and biomass emissions during the BSDW phase, but those from traffic sources remained unchanged. After accounting for meteorological factors, China's two-phase clean air initiatives jointly reduced Beijing's PM_{2.5}-bound BaPeq concentrations by 96.6% from 2014 to 2021. These findings provide compelling evidence for the effectiveness of China's clean air actions in mitigating population exposure to PAHs in Beijing.

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1. Introduction

Polycyclic aromatic hydrocarbons (PAHs) have long been known to cause cancers and many other adverse health outcomes [1]. Past decades witnessed a clear shift in the exposure burden of PAHs from developed to developing countries [2]. Although PAH emissions in developed countries drastically decreased from 1970 to 2008, global PAH emissions continued to rise due to rapid increases in PAH emissions in developing countries (e.g., China) along with their industrialization and economic growth [2]. Developing countries became the dominant PAH emitters and contributed to 90% of global PAH emission in 2000, with the highest release from China [2,3].

It was estimated that 30% of the Chinese population lived in areas where ambient benzo[a]pyrene equivalent (BaPeq) concen-

trations in 2003 [4]. A more recent analysis suggested that exposure to PAHs caused 15,198 excess lifetime cancer cases in 2013 [5]. We and others have also shown in multiple independent cohorts that long-term exposure to PAHs was associated with increased risk of cancer [6,7], cardiopulmonary diseases [8,9], and adverse birth outcomes [10,11], while short-term exposure to PAHs induced cardiopulmonary effects even among healthy individuals [12,13]. Improving regional (e.g., air pollution control for 2008 Summer Olympics [14]) or indoor (e.g., using air purifiers [15]) air quality has been shown to significantly reduced personal exposure to PAHs. However, the benefit reversed after the cessation of these short-term air pollution control measures.

In September 2013, the State Council of China issued the Air Pollution Prevention and Control Action Plan (APPCAP) targeted at reducing the ambient PM_{2.5} concentration to 15%–25% below the 2012 level by 2017, with ten specific measures including strengthen industrial and vehicle standards, phasing out outdated industrial manufacturing, and promoting clean energies

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in residential sectors [16,17]. As a result, the annual $\text{PM}_{2.5}$ concentrations from 496 stationary monitors in 74 cities of China have decreased by 33.3% (from 72.2 to 47.0 $\mu\text{g}/\text{m}^3$) between 2013 and 2017 [17]. In July 2018, the State Council of China further issued a three-year action plan for cleaner air (also named Blue Sky Defense War, BSDW), to continue reducing emissions of major air pollutants and greenhouse gases mainly through structural adjustments and clean energy promotion in four key sectors (i.e., industry, energy, transport, and land use). The BSDW further reduced, though at lower speeds compared to APPCAP, the national annual $\text{PM}_{2.5}$ concentrations to 33 $\mu\text{g}/\text{m}^3$ in 2020 [18]. Although these air pollution control measures did not target PAHs, they may influence PAH emission through controlling common anthropogenic sources of $\text{PM}_{2.5}$.

This study aims to determine the effectiveness of China's air pollution control actions in reducing ambient PAH levels and consequential human exposures in Beijing, by continuously monitoring ambient PAHs levels in Beijing from 2014 to 2021 and biomonitoring exposure to PAHs among 54 healthy adults who traveled from Los Angeles (LA) to Beijing in 2014–2018, a period witnessing substantial air quality improvement in Beijing but not in LA. From these participants, 358 urine samples were collected before, during, and after their trips to Beijing for the measurement of PAH metabolites. Samples collected while the participants were in LA served as controls in our temporal trend analysis of the biomarkers in relation to air quality change in Beijing.

2. Materials and methods

2.1. Measurements of $\text{PM}_{2.5}$ -bound PAHs and source tracers

$\text{PM}_{2.5}$ samples were collected routinely every six days at the Peking University Urban Atmosphere Environment Monitoring Station (PKUERS) as described previously [19]. From 2014 to 2021, we collected a total number of 410 $\text{PM}_{2.5}$ samples for subsequent chemical analysis. The sample was not collected between February 2020 and July 2020 due to COVID-19 lockdowns. To measure the concentrations of 16 PAHs in $\text{PM}_{2.5}$ samples, a punch of each sampling filter was spiked with deuterium-labeled recovery surrogates. The samples were then Soxhlet extracted, concentrated, purified with silica gel column, and then analyzed by an Agilent 8890-7010 GC-QQQ-MS with more details of analytical procedures and quality control measures described in the [Supplemental Materials](#) and [Table S1](#) (online). The tracers of coal combustion (picene), biomass burning (retene), and vehicle emissions (17(α),21(β)-hopane) were measured with an Agilent 7890B-7200 GC-ToF-MS as described previously [20].

2.2. Measurements of urinary PAHs metabolites

The biomonitoring study was built upon a joint research program between University of California Los Angeles (UCLA) and Peking University (PKU). In each year, this program supports ~15 UCLA students every year to visit PKU for ten weeks in the summer. Participants were recruited from UCLA students who participated in this summer program during 2014–2018 as described previously [13]. For each participant, multiple morning urine samples at more than one-week intervals were collected in LA, Beijing, and in LA again. The urine samples were not collected until one week after the arrival at the new city to exclude the exposures from the previous city. This wash-off period is long enough given short half-lives of PAHs in the human body [21,22]. Participants were asked to fast for >8 h before sample collection to diminish dietary effects. Each urine collection was coupled with a questionnaire focusing on activities related to PAHs, including cooking

behaviors, traffic-related activities, and passive smoking in the past three days. Human exposure to PAHs was assessed by measuring their metabolites in the urine with a previously established method [23]. The analytes include 9 hydroxylated PAHs (OH-PAHs), namely 2-hydroxylated dibenzofuran (2-OH-DBF), 2- and 3-hydroxylated fluorenes (OH-FLUs), 1-, 2-, 3-, 4- and 9-hydroxylated phenanthrenes (OH-PHEs), and 1-hydroxylated pyrene (1-OH-PYR). The concentrations of metabolites from the same PAHs were summed as an exposure indicator for their parent compound. Details of the analytical procedure and quality control measures are described in [Supplemental Materials](#) and [Table S2](#) (online). Urinary creatinine levels were measured based on the Jaffe reaction [24] and were used to normalize OH-PAHs levels. The study was performed in accordance with guidelines and approval of the Institutional Review Boards of both UCLA and PKU.

2.3. Measurements of ambient air quality

At PKUERS, we used a mid-volume sampler with four Teflon filters to gravimetrically determine the daily average concentration of $\text{PM}_{2.5}$ every six days. We measured daily NO_2 concentrations every six days using a chemiluminescence analyzer (Thermo Scientific 42i-TLE). In addition, we collected daily concentrations of $\text{PM}_{2.5}$ and NO_2 from national ambient air quality monitoring sites within 30 km of UCLA ($n = 4$) and PKU ($n = 10$). The measurements at PKUERS and the average concentrations of 10 nearby air monitoring sites were in good agreement ($R^2 = 0.66$ for $\text{PM}_{2.5}$ and $R^2 = 0.67$ for NO_2). In this study, we matched data of $\text{PM}_{2.5}$ -bound PAHs and urinary PAHs metabolites with the measurements at national air monitoring sites for subsequent data analysis, because these data are available in both LA and Beijing and have been foundations for evidence-based evaluation of air pollution control policies in previous studies [25,26].

2.4. Data analysis

In the air monitoring study, we used linear regressions to examine the temporal trend of $\text{PM}_{2.5}$ -bound PAHs concentrations as well as the difference in PAHs concentrations between the heating and nonheating seasons. In Beijing, central heating normally starts on November 15th and ends on March 15th. Nevertheless, it is common that the heating starts 1–2 weeks earlier than scheduled and ends 1–2 weeks later. Therefore, we considered heating seasons from November 1st to March 31st of the second year. We performed stratified analysis to examine the season-specific temporal trends of $\text{PM}_{2.5}$ -bound PAHs concentrations. Linear regression models were used to test whether the meteorological factors have significant temporal trends and if yes, we will adjust for meteorological factors when examining the temporal trend of PAHs. We used a receptor model (EPA PMF 5.0 version) for the source apportionment of ambient PAHs concentrations [27]. The input includes 16 PAHs, $\text{PM}_{2.5}$ mass concentrations, and tracers for traffic exhaust (hopane), biomass burning (retene), and coal combustion (picene). More details of the PMF analyses are provided in the [Supplemental Materials](#).

In the biomonitoring study, we used linear mixed effects models with random intercepts at participant levels to examine the difference in biomarkers between LA and Beijing. We used linear or logistic regression models to test whether participants' demographic characteristics or behaviors have significant temporal trends. To examine the temporal trends of urinary OH-PAHs levels, we used mixed effects models with random intercepts of participants in which biomarker concentrations were modeled as a function of city, year, and their interactions. To examine the city-specific associations between urinary OH-PAHs and air pollutant

concentrations up to 7 days prior to the urine collection, we used distributed lag models that include a flexible cross-basis function with a linear function for the dependent variable and a polynomial function of degree 3 for the lagged independent variable, adjusted for fixed effects of participants' exposure-related activity patterns and random intercepts of participants. Statistical significance was considered with a P -value < 0.05 . All analyses were performed in R (www.r-project.org).

3. Results

3.1. $PM_{2.5}$ -bound PAHs concentrations and sources during APPCAP phase

Ambient temperature, relative humidity, or wind speed were not significantly changed during the monitoring period in 2014–2018 (Table 1). The ambient concentrations of BaP exceeded the national air quality standard (i.e., 2.5 ng/m^3) in 72.4% of the sampling days in 2014 but only in 35.6% of those in 2018 (Fig. S1 online). From 2014 to 2018, the annual average BaPeq concentration in Beijing $PM_{2.5}$ samples decreased by 88.5% (119 ng/m^3 in 2014 and 10.2 ng/m^3 in 2018). The concurrently measured concentrations of $PM_{2.5}$ and NO_2 in Beijing decreased by 43.1% ($97.2 \text{ }\mu\text{g/m}^3$ in 2014 and $52.6 \text{ }\mu\text{g/m}^3$ in 2018) and 23.9% ($30.3 \text{ }\mu\text{g/m}^3$ in 2014 and $23.1 \text{ }\mu\text{g/m}^3$ in 2018), respectively (Fig. S2 online). Consequentially, the mass percentage of BaPeq in $PM_{2.5}$ samples also decreased from 0.21% in 2014 to 0.02% in 2018 (Fig. S3 online). The PAHs concentrations are substantially higher in the heating seasons as compared to the nonheating seasons (Fig. 1). Stratified analyses indicate that the average BaPeq concentration decreased by 87.7% (207 ng/m^3 in 2014 and 26.0 ng/m^3 in 2018) in the heating seasons (Fig. 1) and by 88.2% (16.8 ng/m^3 in 2014 and 2.36 ng/m^3 in 2018) in the nonheating seasons (Fig. 1) from 2014 to 2018. The temporal trend of individual PAHs was consistent with that of BaPeq, except for fluorene in the nonheating seasons when a large fraction of the fluorene existed in the gaseous phase (Fig. S4 online).

Assisted by source tracers, we identified four factors in PMF analysis that jointly explained 84.6%–96.2% of the variance for 16 individual PAHs. Among them, three factors – traffic exhaust, biomass burning, and coal combustion jointly contributed to 98% of the average BaPeq concentrations throughout the study period. The fourth factor, interpreted as other sources or processes, largely explained the variability of ambient $PM_{2.5}$ and fluorene levels, but merely contributed to other PAH species (Fig. S5 online). Consistent with previous studies in Beijing [19,28], the source intensities of traffic exhaust, biomass burning, and coal combustion were sub-

stantially higher in the heating seasons than in the nonheating seasons (Fig. S5 online), due to increased domestic heating activities and meteorological changes (e.g., lower temperature, lower boundary layer height, and more frequent stable weather conditions) in the heating seasons [19].

During the APPCAP phase, there were continuous decreases in the annual average BaPeq concentrations originating from traffic exhaust (–90.9%, from 40.8 ng/m^3 in 2014 to 3.7 ng/m^3 in 2018), coal combustion (–91.5%, from 42.2 ng/m^3 in 2014 to 3.6 ng/m^3 in 2018), and biomass burning (–80.2%, from 9.4 ng/m^3 in 2014 to 1.9 ng/m^3 in 2018) (Fig. 2). In the nonheating seasons, biomass burning made negligible contributions to BaPeq concentrations and the decrease in BaPeq concentrations from 2014 to 2018 was mainly driven by reductions in traffic exhaust and coal combustion. The contribution of coal combustion was fully eliminated during the nonheating season in 2018 (Fig. 2). In the heating seasons, we also observed rapid decreases in traffic exhaust (–95.0%, from 64.8 ng/m^3 in 2014 to 3.2 ng/m^3 in 2018). In contrast, the BaPeq concentrations originating from coal combustion and biomass burning were decreased to a less extent due to the rigid demand for domestic heating. In 2018, coal combustion became the predominant PAHs source in the heating season, accounting for 66.5% of the $PM_{2.5}$ -bound BaPeq concentrations (Fig. 2).

3.2. Biomarkers of PAHs exposure among travelers during the APPCAP phase

From 54 healthy travelers (26 males and 28 females, with an average age of 23.8 years), 358 urine samples were collected before, during, and after their trips from LA to Beijing, with the timing of urine collection and ambient $PM_{2.5}$ concentrations during study periods shown in Fig. 3. In 2014–2018, the air quality continuously improved in Beijing while remained unchanged in LA (Fig. 3), despite the consistently higher air pollution level in Beijing compared to LA. There was no significant temporal trend in travelers' demographic characteristics from 2014 to 2018 (Table 1).

Traveling from LA to Beijing led to significant increases in urinary OH-PAHs concentrations ($P < 0.05$), which recovered after returning to LA (Fig. 3). Nevertheless, the average concentrations of summed PAH metabolites in Beijing decreased by 52.3% from 2014 ($4.94 \text{ }\mu\text{g/g creatinine}$) to 2018 ($2.36 \text{ }\mu\text{g/g creatinine}$) and were consistently lower than those measured in another group of LA-Beijing travelers in 2012 [23] before China's clean air actions ($6.05 \text{ }\mu\text{g/g creatinine}$) (Table S3 online). The concentration of individual PAH metabolites was also significantly decreased in 2014–2018 in Beijing from the baseline but not in LA (Fig. 4). Noting that ambient $PM_{2.5}$ and NO_2 concentrations were highly correlated with

Table 1
Demographic information of study participants and sampling information of ambient $PM_{2.5}$ between 2014 and 2018.

	Total	2014	2015	2016	2017	2018	P_{trend}^a
Ambient $PM_{2.5}$ samples in Beijing							
Number of samples	287	58	53	58	59	59	
Temperature (K, mean $\pm$ SD)	288 ± 11	289 ± 11	288 ± 10	287 ± 11	288 ± 11	288 ± 11	0.74
Relative humidity (% , mean $\pm$ SD)	41.1 ± 19.3	40.5 ± 17.0	41.5 ± 17.7	44.0 ± 22.0	45.3 ± 21.7	41.6 ± 21.3	0.46
Wind speed (m/s, mean $\pm$ SD)	2.15 ± 1.09	1.91 ± 0.69	2.11 ± 0.94	2.43 ± 0.95	2.28 ± 1.18	1.80 ± 1.14	0.98
Urine samples of LA-Beijing travelers							
Number of subjects	54	14	13	8	10	9	
Number of samples (LA/Beijing)	183/175	48/63	49/34	27/23	37/30	22/25	
Age (year, mean $\pm$ SD)	23.8 ± 7.6	23.3 ± 5.6	27.8 ± 13.6	22.6 ± 3.2	22.0 ± 2.7	21.8 ± 1.9	0.26
BMI (kg/m^2 , mean $\pm$ SD)	21.5 ± 2.7	21.7 ± 2.8	21.3 ± 2.1	20.1 ± 2.3	22.1 ± 2.4	22.0 ± 3.9	0.75
Race (Asian/Others)	39/15	8/6	12/1	6/2	7/3	6/3	0.94
Sex (M/F)	26/28	9/5	3/10	3/5	8/2	3/6	0.89
Smoking (yes/no)	2/52	0/14	0/13	2/6	0/10	0/9	0.81

^a Temporal trends were tested by linear regressions for temperature, relative humidity, wind speed, age and BMI, and logistic regressions for the others.

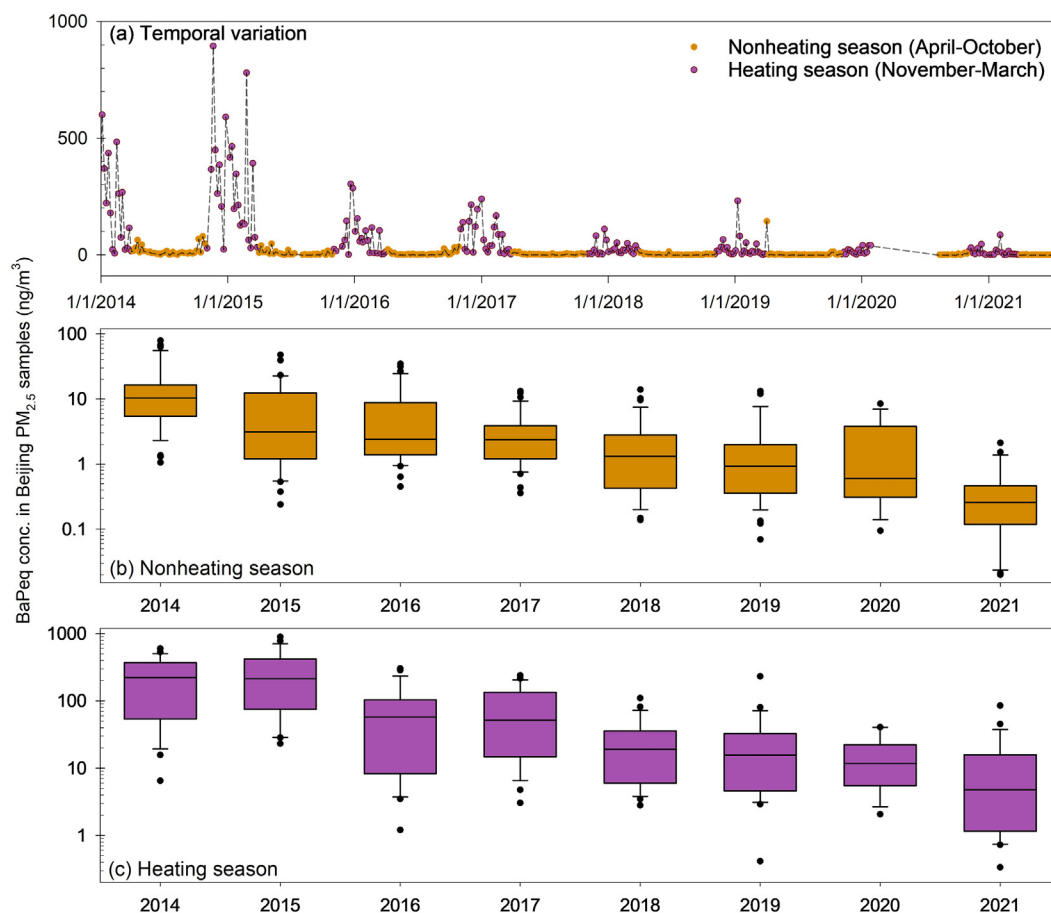


Fig. 1. Temporal trend of BaPeq concentrations in $PM_{2.5}$ samples in Beijing from 2014 to 2021 for (a) whole year, (b) nonheating seasons, and (c) heating seasons. Purple and orange symbols indicate data in the heating and nonheating seasons, respectively. The solid horizontal line represents the median. The box represents the 25th–75th percentiles, and the whiskers represent the 10th and 90th percentiles. The heating season of a year is defined as starting on November 1st of the preceding year and ending on March 31st of the current year (e.g., the heating season of 2015 spans from November 1st, 2014 to March 31st, 2015).

ambient PAHs concentrations in Beijing (Fig. S6 online), changes of urinary PAH metabolites in 2014–2018 were significantly associated with ambient $PM_{2.5}$ and NO_2 concentrations 0–7 days prior to the urine collection date in Beijing, but not in LA (Fig. 4 and Fig. S7 online). In addition to ambient air pollutant levels, longer time spent barbecuing and in public transportation was also associated with increased concentrations of urinary PAH metabolites in Beijing (Table S4 online).

3.3. $PM_{2.5}$ -bound PAHs concentrations and sources after APPCAP

From 2018 to 2021, there is no significant temporal trend in ambient temperature or relative humidity during air sampling days. However, there is an increasing trend in the average wind speed which is driven by elevated levels in 2021 (2.79 m/s) as compared with 2018–2020 (1.80–2.15 m/s) (Table S5 online). Likewise, there is a decreasing trend in annual average BaPeq concentration from 2018 (10.2 ng/m^3) to 2021 (4.8 ng/m^3) which is driven by a sharp concentration drop in 2021 (Fig. 1). Consistently, the percentage of sampling days with BaP concentrations exceeding the national air quality standard was comparable between 2018 (35.6%) and 2020 (37.0%) and dropped to 14.3 % in 2021 (Fig. S1 online). In 2018–2020 (i.e., the BSDW phase), there were no significant changes in BaPeq concentrations (–2.5% per year, 95%CI: –34.2% to 44.3%). The trend of BaPeq concentrations in 2018–2020 was consistent in the heating and nonheating seasons. Nevertheless, several individual PAHs significantly decreased from

2018 to 2020 in the heating seasons but not in the nonheating seasons (Fig. S8 online). During the heating seasons in 2018–2020, there was a continuous decrease in BaPeq concentrations from coal combustion (17.1 ng/m^3 in 2018 and 6.6 ng/m^3 in 2020) and biomass burning (4.6 ng/m^3 in 2018 and 2.1 ng/m^3 in 2020) (Fig. 2), which explains the decrease in several individual PAHs in the heating seasons. In the nonheating seasons, however, coal combustion and biomass burning contributed little to BaPeq concentrations since 2018. The predominant sources of BaPeq during the nonheating seasons (i.e., traffic exhaust) did not show significant changes from 2018 to 2020, which explains the lack of a significant temporal trend in BaPeq concentrations during the BSDW phase.

From 2014 to 2021, the two-phase clean air actions (APPCAP and BSDW) have jointly decreased the annual average BaPeq concentration in Beijing $PM_{2.5}$ samples by 97.7% (119 ng/m^3 in 2014 and 4.8 ng/m^3 in 2021) without adjusting for meteorological factors and by 96.6% (95%CI: 94.4% to 97.9%) after adjusting for ambient temperature, humidity, and wind speed. In stratified analyses, the BaPeq concentration decreased by 96.5% (95.4% in adjusted models) in the heating seasons and by 97.9% (97.2% in adjusted models) in the nonheating seasons from 2014 to 2021.

4. Discussion

In this study, we present 8-year ambient air monitoring data (2014–2021) and 5-year biomonitoring data (2014–2018) to illustrate the impact of unprecedented changes in anthropogenic emis-

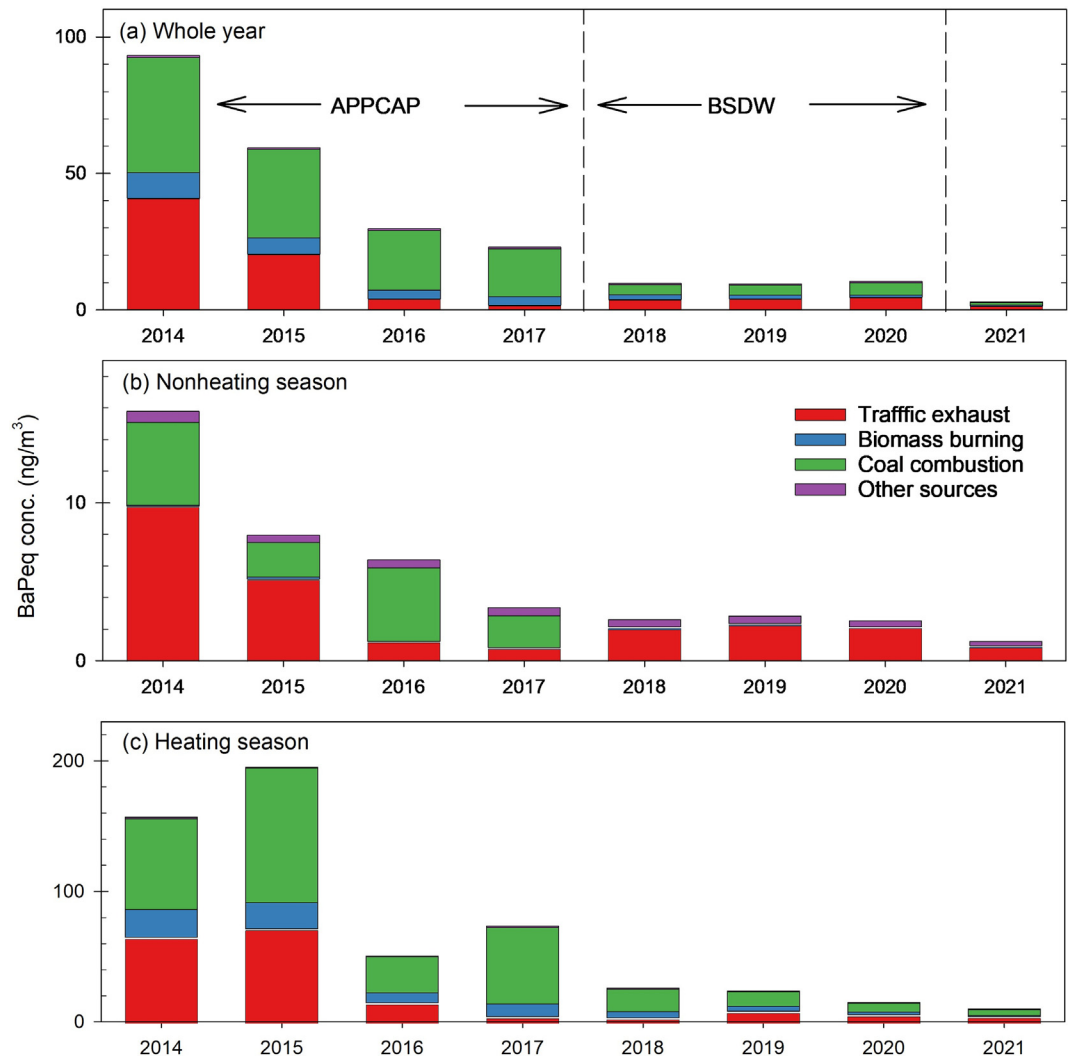


Fig. 2. Changes of source contributions to average concentrations PM_{2.5}-bound BaPeq in Beijing from 2014 to 2021 for (a) whole year, (b) nonheating seasons, and (c) heating seasons.

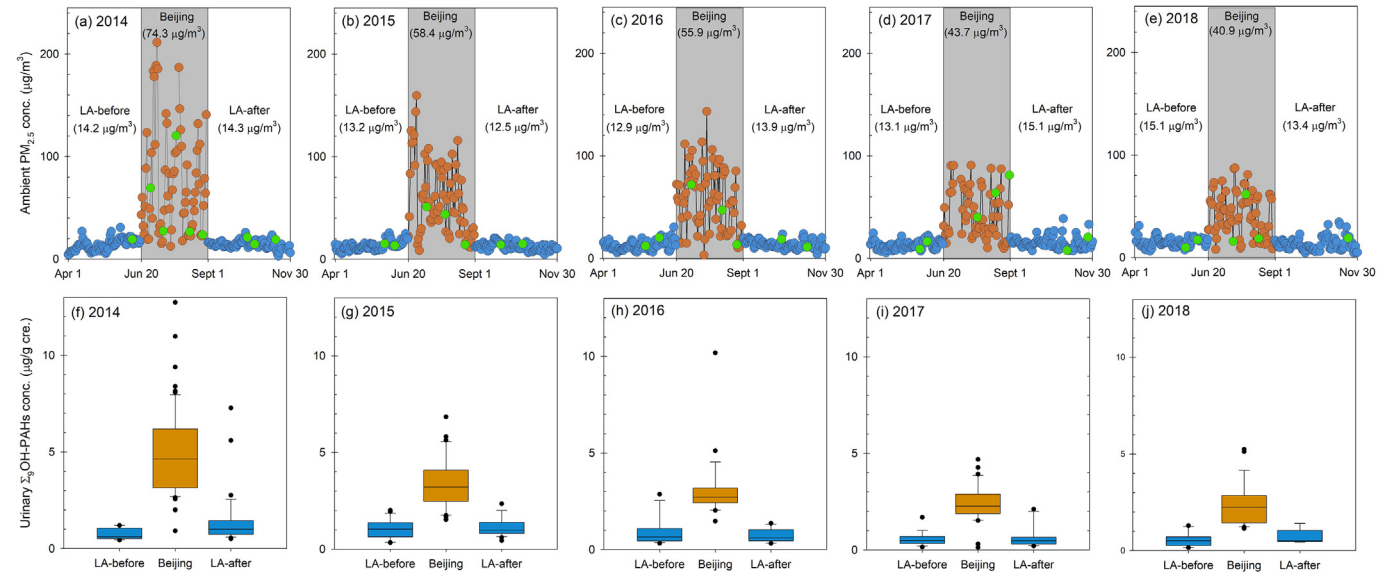


Fig. 3. Concentrations of ambient PM_{2.5} (a–e) and urinary PAHs metabolites (f–j) in the natural experiment among travelers between LA and Beijing in the summertime from 2014 to 2018. Urine samples were collected within $\pm$ four days of the date indicated by green circles. Blue and orange circles indicate data in Los Angeles and Beijing, respectively. Numbers in brackets indicate average PM_{2.5} concentrations in LA-before, Beijing, or LA-after. Daily PM_{2.5} concentrations in Los Angeles and Beijing were obtained from national air quality monitors within 30 km of UCLA (n = 4) and Peking University (n = 10), respectively.

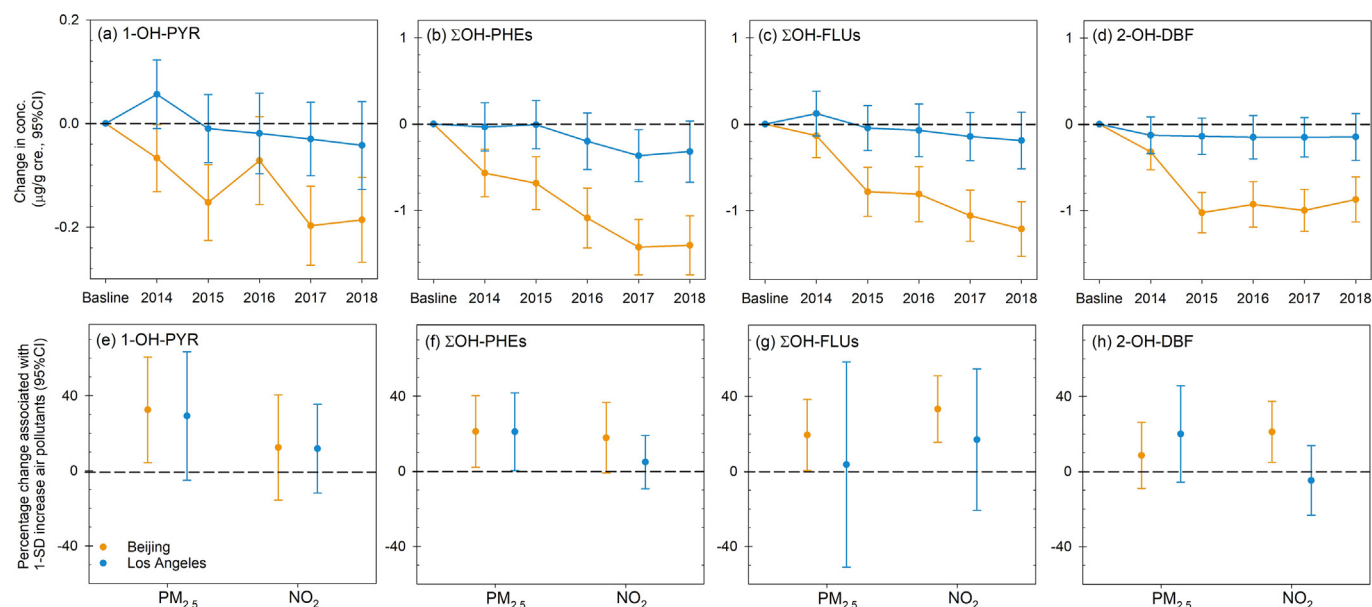


Fig. 4. Temporal trend of urinary PAH metabolites in LA and Beijing (a–d) and the city-specific associations of urinary PAH metabolites with ambient PM_{2.5} and NO₂ concentrations in 2014–2018 (e–h). Data from travelers in 2012 (before the implementation of APPCAP) are considered as baseline and were obtained from our previous study [23]. Percentage changes in PAH metabolites are associated with one standard deviation change in PM_{2.5} (4.43 μg/m³ in LA and 30.1 μg/m³ in Beijing) or NO₂ (7.23 μg/m³ in LA and 6.14 μg/m³ in Beijing) concentrations 0–7 days prior to the urine collection date. ΣOH-PHEs: sum of 1-, 2-, 3-, 4-, and 9-OH-PHEs; ΣOH-FLUs: sum of 2- and 3-OH-FLUs.

sions, following China's two-phase clean air actions, on human exposure to PAHs. Our results indicated a remarkable decline of 96.6% in ambient BaPeq concentrations from 2014 to 2021 and a 52.3% decline in the concentration of PAH metabolites in travelers' urine samples collected in Beijing from 2014 to 2018. These findings demonstrate the effectiveness of long-term air pollution control in reducing human exposure to PAHs.

It has been well-documented that APPCAP reduced PM_{2.5} mass concentrations [17,29,30] and improved population health [31,32]. This study provides the first biomonitoring evidence demonstrating that air pollution control measures reduced human exposure to airborne toxins such as PAHs. We used a natural experiment among a small group of well-characterized international travelers to study the effectiveness of national policies on human exposure to environmental pollutants, which allowed us to use a country without such policies as control. This approach has been previously used to demonstrate the effectiveness of government and manufacturer efforts to phase out bisphenol A in reducing population exposure in LA from 2012 to 2017 [33]. Herein, we reported that exposure to PAHs significantly decreased in Beijing but not in LA from 2012 to 2018. Although we also observed significant decrease in self-reported passive smoking in Beijing ($P < 0.05$) from 2012 to 2018 (Table S6 online) likely due to the smoking ban policy in Beijing since 2015 [34], changes in urinary PAHs metabolites were significantly associated with PM_{2.5} and NO₂ concentrations but not with self-reported passive smoking (Table S4 online and Fig. 4). In addition, our recent analysis of a subgroup of participants in 2014 and 2015 found that the drastic decrease in urinary levels of PAHs metabolites from 2014 to 2015 (Fig. 4) was accompanied with no significant changes in urinary cotinine levels [34], indicating that improved air quality is the likely cause of decreased PAHs exposure in Beijing.

We observed a greater reduction in BaPeq concentrations from 2014 to 2018 (88.5%) as compared with the concurrently measured PM_{2.5} mass concentrations (43.1%), implying that APPCAP not only reduce PM_{2.5} concentrations but also alter its chemical compositions. Our results support that APPCAP reduced the mass percentage of PAHs (and likely other combustion-originated chemicals) in PM_{2.5}, an index directly related to the PM_{2.5} toxicity [35,36], which

may also contribute to the frequently documented health benefits of APPCAP [31,32]. On the other hand, the decrease in urinary PAH metabolites from 2014 to 2018 (52.3%) was smaller as compared with PM_{2.5}-bound BaPeq concentrations during the same period (88.5%). Previous studies have suggested that non-inhalation routes (e.g., diets), that were less influenced by APPCAP, made considerable contributions to exposure to low molecular-weight PAHs [37,38] that can be measured in the urine. These studies also suggested that inhalation to particulate-phase PAHs was the primary exposure route of high molecular-weight PAHs and was the major contributor to carcinogenic risks caused by PAH exposure [37,38].

We utilized a consistent protocol to continuously monitor ambient PAH concentrations for over eight years, spanning two phases of China's clean air actions (APPCAP and BSDW). A greater reduction in ambient BaPeq concentrations has been observed in the APPCAP phase compared to the BSDW phase. While this can be partially explained by the fact that the potential of emission reduction narrowed in BSDW, the source apportionment results show that BSDW was not as effective as APPCAP in reducing traffic emissions. As the most stringent-ever clean air policy in China, APPCAP phased out 20 million old vehicles and enacted two new vehicle emission standards [16,39], which is reflected by the observed 90.9% reduction traffic-originated BaPeq concentrations from 2014 to 2018. In contrast, the traffic-originated BaPeq concentration did not change from 2018 to 2020. It is possible that the rising number of vehicles in Beijing [40] may also offset the traffic pollution control measures of BSDW (e.g., optimizing the structure of freight transportation and further tightening vehicle emission standard [41]). Since the vehicle-use intensity is projected to further increase by 2050 [42], sustained efforts are needed to control vehicular emission and prevent a reversal in ambient PAHs concentrations.

During the heating seasons, the primary source of BaPeq concentration is coal combustion, which is consistent with the predominant use of large coal boilers for centralized heating in urban areas and scattered coal consumption in rural households for domestic heating in North China [43]. In the early phase of APPCAP (i.e., 2013–2015), efforts to reduce emissions from coal combustion were mainly through promoting the use of clean stoves

and washed coals [16]. Since 2017, greater efforts have been made to substitute coal with natural gas and electricity [16,44]. Our observations during the heating seasons indicate a declining trend in coal-originated BaPeq concentrations from 2014 to 2021, notably with a significant decrease evident between 2017 and 2018 (Fig. 2). These results supported the effectiveness of the clean heating renovation, such as the transition from coal to gas and electricity, in mitigating PAHs pollution.

Among the general population in China, exposure to PAHs has been associated with an increased risk of cancer and cardiopulmonary diseases [4,9]. Emerging evidence has also suggested that PAHs accelerate aging [45] and interfere with male and female fertility [46,47] among Chinese population. Most of these studies were conducted before the implementation of clean air actions in 2013. In our previous studies, traveling from LA to Beijing induced significant changes in a panel of circulating biomarkers indicative of increased cardiovascular risks, in associations with PAH exposure [13]. Greater adverse effects induced by the travel were observed in 2014 as compared with 2015 when the PAH exposure was much lower, suggesting potential health benefits of reducing population exposure to PAHs [13]. In the current study, we found that the China's clean air actions substantially reduced human exposure to PAHs; and the PM_{2.5}-bound PAHs levels in 2021 were only ~3% of the level in 2014. The drastic reduction in inhalable PAHs levels in Beijing since 2013 necessitates further studies to evaluate whether populations in other regions of China experienced similar exposure reduction as well as whether the contemporarily low levels of PAHs in the air remain a threat to population health.

Our study has several limitations. First, we did not measure gaseous PAHs. Although we did not observe significant temporal trends in ambient temperature from 2013 to 2021, there is a continuous decline in ambient PM_{2.5} concentrations, which may lead to significant changes in the gas-particle partitioning of PAHs during the study period. Second, we did not conduct biomonitoring studies in 2019 and 2020 or in the heating seasons. Therefore, we cannot evaluate the effects of BSDW on human exposure to PAHs, nor did we know whether the exposure was also reduced in the heating seasons when people's activity pattern (e.g., time spent in indoor environments) might be substantially different from the nonheating seasons. Third, we did not quantify external exposure to PAHs in the biomonitoring study. Nevertheless, we used ambient levels of PM_{2.5} and NO₂ as a proxy to PAHs concentrations because of the strong correlations of BaPeq with PM_{2.5} and NO₂ (Fig. S6 online). Additionally, we have previously shown that traffic emission was the predominant source of 3- and 4-ring PAHs during the nonheating seasons of Beijing [19], which further justify the use of ambient NO₂ levels as a proxy to PAHs in the air. Last, although urinary PAH metabolites quantified personal exposure to PAHs from all exposure routes, we collected fasting urine samples and therefore the results were less influenced by dietary PAH sources. Previous studies have suggested that diet contributes to a marked proportion of total daily PAHs intakes in Beijing [37,38] owing to the uptake of airborne PAHs in vegetables [48] and cooking of food at high temperatures [37]. To what extent clean air actions would influence PAHs levels in food remains unclear.

Conflict of interest

All authors declare that they have no conflict of interest.

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Author contributions

Yifang Zhu and Xinghua Qiu designed the study. Yan Lin and Xiaodi Shi performed the sample collection. Yan Lin, Xiaodi Shi, Xing Jiang, Jinming Liu, and Peiwen Zhong conducted the laboratory analysis. Yan Lin, Yihui Ge, and Chi-Hong Tseng conducted the data analysis. Yan Lin, Xinghua Qiu, Junfeng (Jim) Zhang, Tong Zhu, Jesus A Araujo, and Yifang Zhu wrote the manuscript.

Data availability

All study data are included in the article and/or Supplementary Appendix.

Appendix A. Supplementary materials

Supplementary materials to this article can be found online at <https://doi.org/10.1016/j.scib.2024.08.015>.

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