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Review

Exposure to Atmospheric Particulate Matter-Bound Polycyclic Aromatic Hydrocarbons and Their Health Effects: A Review

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Abstract: Particulate matter (PM) is a major factor contributing to air quality deterioration that enters the atmosphere as a consequence of various natural and anthropogenic activities. In PM, polycyclic aromatic hydrocarbons (PAHs) represent a class of organic chemicals with at least two aromatic rings that are mainly directly emitted via the incomplete combustion of various organic materials. Numerous toxicological and epidemiological studies have proven adverse links between exposure to particulate matter-bound (PM-bound) PAHs and human health due to their carcinogenicity and mutagenicity. Among human exposure routes, inhalation is the main pathway regarding PM-bound PAHs in the atmosphere. Moreover, the concentrations of PM-bound PAHs differ among people, microenvironments and areas. Hence, understanding the behaviour of PM-bound PAHs in the atmosphere is crucial. However, because current techniques hardly monitor PAHs in real-time, timely feedback on PAHs including the characteristics of their concentration and composition, is not obtained via real-time analysis methods. Therefore, in this review, we summarize personal exposure, and indoor and outdoor PM-bound PAH concentrations for different participants, spaces, and cities worldwide in recent years. The main aims are to clarify the characteristics of PM-bound PAHs under different exposure conditions, in addition to the health effects and assessment methods of PAHs.

Keywords: particulate matter; polycyclic aromatic hydrocarbons; personal exposure; indoor; outdoor; health effects

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1. Introduction

Air pollution has become a mainstream global environmental pollution problem in recent decades [1,2]. Particulate matter (PM) is one of the major factors contributing to air quality deterioration, leading to adverse health effects on humans [3–6]. PM is an extremely complex mixture defined in many ways, including formation pathway, emission source, chemical composition, and PM size [5]. The formation pathway of PM involves the direct release by emission sources into the atmosphere or secondary formation via nucleation, vapour condensation, adsorption, and absorption of gaseous precursors, primary PM, or secondary PM [5]. In terms of the source, natural sources include dust, sea salt, living vegetation, volcanic activity, and forest fires, whereas anthropogenic sources mainly involve combustion processes, including stationary sources (domestic, industrial, and agricultural activities) and mobile sources (vehicles, aircraft, and shipping traffic exhaust) [7,8]. Regarding the chemical composition, PM contains many kinds of inorganic

and organic compounds, including water-soluble ions, trace elements, crustal material, elemental carbon, and organic carbon, many of which are harmful to human health [9–13]. According to the size, PM is generally divided into inhalable coarse particles (PM with an aerodynamic diameter smaller than 10 μm, PM_{10}) and inhalable fine particles (PM with an aerodynamic diameter smaller than 2.5 μm, $PM_{2.5}$) [14]. However, other sizes of PM also can be collected in the atmosphere by using different types of air sampler, such as PM_1, PM_2, $PM_{2.1}$, and PM_4, which aerodynamic diameters are smaller than 1, 2, 2.1, and 4 μm, respectively [15–18].

PM is of concern not only to researchers but also to the general public. Numerous toxicological and epidemiological studies have proven the adverse links between exposure to PM and health effects [19–26]. The cancer risk resulting from PM exposure has also been demonstrated by the International Agency for Research on Cancer (IARC) [27]. According to the report by the World Health Organization (WHO), PM in outdoor air is responsible for approximately 4.5 million premature deaths every year, or close to 10% of the total deaths on a global scale [28]. Among these deaths, approximately 2 million deaths, which represent approximately 5% of the global total deaths, are due to damage to the lungs and respiratory system directly attributable to PM [28]. Moreover, WHO also reported that almost 3 billion people worldwide still rely on solid fuels for cooking and heating, leading to approximately 4 million people premature deaths due to household indoor air pollution [29], which is almost equal to the deaths caused by outdoor PM pollution.

To protect public health, relatively strict indoor and outdoor air quality standards have been prescribed by WHO. For indoor household fuel combustion, WHO has strongly recommended that the emission rate target of $PM_{2.5}$ should not exceed 0.23 mg/min under unvented conditions, and 0.80 mg/min under vented condition [29]. For outdoor air, the annual average $PM_{2.5}$ and PM_{10} concentrations cannot exceed 10 and 20 $\mu g/m^3$, respectively, and 24-h average concentrations that cannot exceed 25 and 50 $\mu g/m^3$, respectively [30]. Currently, outdoor PM regulation standards also have been implemented in many cities worldwide. Table 1 lists various $PM_{2.5}$ and PM_{10} regulation standards set by the governments of several countries according to their national conditions.

Table 1. Regulation standards of $PM_{2.5}$ and PM_{10} set by several governments.

Continent	Country	$PM_{2.5}$ ($\mu g/m^3$)		PM_{10} ($\mu g/m^3$)		References
		24-h	Annual	24-h	Annual	
North America	USA	35	12	150	None	[3]
	Mexico	45	12	75	40	[31]
	Canada	27	8.8	None	None	[32]
South America	Brazil	25	10	50	20	[33]
	Chile	50	20	150	50	[34]
Australia	Australia	25	8	50	25	[35]
Africa	South Africa	None	None	75	40	[39]
Europe	EU	None	25	50	40	[37]
	Russia	35	25	60	40	[38]
Asia	China	75	35	150	70	[36]
	Japan	35	15	None	100	[36]
	South Korea	25	25	100	50	[36]
	Mongolia	50	25	150	50	[36]
	India	60	40	100	60	[36]

Specifically, in North America, the 24-h and annual average $PM_{2.5}$ concentration standards in the United States of America (USA) are 35 $\mu g/m^3$ and 12 $\mu g/m^3$, respectively [3]. Mexico (12 $\mu g/m^3$) and Canada (8.8 $\mu g/m^3$) have also defined relatively low standards for the annual average $PM_{2.5}$ concentration [31,32]. In South America, Brazil determined the final standards of $PM_{2.5}$ and PM_{10} in 2018, which 24-h and annual average are the same as those prescribed by WHO [33]. However, Chile had relatively higher standards of $PM_{2.5}$ (24-h: 50 $\mu g/m^3$; annual: 20 $\mu g/m^3$) than these above countries [34]. In Australia, the

24-h average $PM_{2.5}$ and PM_{10} concentrations are the same as ones prescribed by the WHO, while the annual average of $PM_{2.5}$ (8 μg/m^3) is lower and of PM_{10} (25 μg/m^3) is higher than the WHO-prescribed levels [35]. In the European Union (EU), Russia, and some Asian countries, the $PM_{2.5}$ and PM_{10} regulation standards are mostly higher than those in the above countries. The annual average $PM_{2.5}$ concentration standards are 15 μg/m^3 (Japan), 25 μg/m^3 (EU, Russia, South Korea, and Mongolia), 35 μg/m^3 (China), and 40 μg/m^3 (India), respectively [36–38]. In South Africa, only the PM_{10} regulation standard has been defined, which does not exceed 40 μg/m^3 for the annual average concentration and 75 μg/m^3 for the 24-h average concentration [39].

In PM, polycyclic aromatic hydrocarbons (PAHs) are a class of persistent organic chemicals with at least two aromatic rings, mainly directly emitted as a result of the incomplete combustion of various organic materials, including both natural sources, such as forest fires and volcanic eruptions, and anthropogenic sources, such as the combustion of fossil fuels and biomass [40,41]. Several hundred PAHs have been detected worldwide, and the United States Environmental Protection Agency (US EPA) has classified 16 PAH species in a priority control pollutant list. Table 2 lists the information of these 16 PAHs and certain non-priority PAHs, which can be generally divided into low-molecular-weight PAHs (LMW PAHs, MW < 200 g/mol), medium-molecular-weight PAHs (MMW PAHs, 200 ≤ MW < 250 g/mol), and high-molecular-weight PAHs (HMW PAHs, MW ≥ 250 g/mol) these three categories. LMW PAHs exhibit a relatively high vapour pressure and easily occur in the gaseous phase, whereas HMW PAHs exhibit a much lower vapour pressure than that of LMW PAHs, and they mainly occur in the particle phase [42–47]. The vapour pressure of MMW PAHs is between those of LMW and HMW PAHs [47–50], suggesting that they may occur in both the gaseous and particle phases, and phase partitioning largely depends on factors such as meteorological conditions (their occurrence in the gaseous phase increases at a relatively high ambient temperature, and their occurrence in the particle phase increases at a relatively low ambient temperature) [48,51]. In contrast, a previous study has reported that the half-lives of PAHs range from a few hours to days, and the half-lives of MMW and HMW PAHs are longer than that of LMW PAHs [52], which indicates that particulate matter-bound (PM-bound) PAHs could be transported across long distances to other regions worldwide before attenuation [53–57].

Table 2. Information of US EPA 16 PAHs and non-priority PAHs.

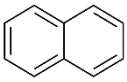
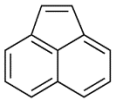
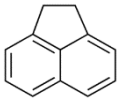
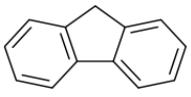
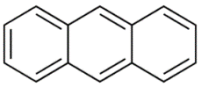
Species (Abbreviation)	CAS Number	MW [a] Category	Vapor Pressure [b]	Structure
US EPA 16 PAHs				
Naphthalene (Nap)	91-20-3	128.17 LMW	11.3	
Acenaphthylene (Acy)	208-96-8	152.19 LMW	0.64	
Acenaphthene (Ace)	83-32-9	154.21 LMW	0.29	
Fluorene (Flu)	86-73-7	166.22 LMW	0.08	
Anthracene (Ant)	120-12-7	178.23 LMW	0.08	

Table 2. *Cont.*

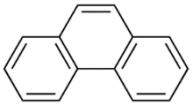
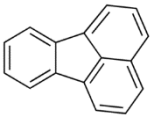
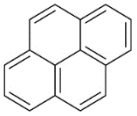
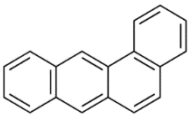
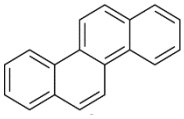
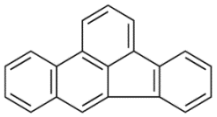
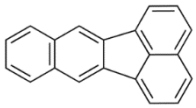
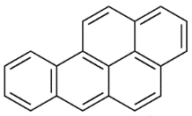
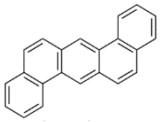
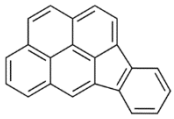
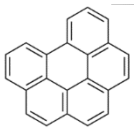
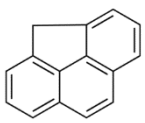
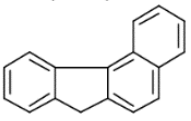
Species (Abbreviation)	CAS Number	MW [a] Category	Vapor Pressure [b]	Structure
Phenanthrene (Phe)	85-01-8	178.23 LMW	1.61×10^{-2}	
Fluoranthene (FR)	206-44-0	202.25 MMW	1.23×10^{-3}	
Pyrene (Pyr)	129-00-0	202.25 MMW	6.00×10^{-4}	
Benz[*a*]anthracene (BaA)	56-55-3	228.30 MMW	2.80×10^{-4}	
Chrysene (Chr)	218-01-9	228.30 MMW	8.31×10^{-4}	
Benzo[*b*]fluoranthene (BbF)	205-99-2	252.30 HMW	6.67×10^{-5}	
Benzo[*k*]fluoranthene (BkF)	207-08-9	252.30 HMW	1.29×10^{-7}	
Benzo[*a*]pyrene (BaP)	50-32-8	252.30 HMW	7.32×10^{-7}	
Dibenz[*a*,*h*]anthracene (DBA)	53-70-3	278.30 HMW	1.27×10^{-7}	
Indeno[1,2,3-*cd*]pyrene (IDP)	193-39-5	276.30 HMW	1.67×10^{-8}	
Benzo[*ghi*]perylene (BgPe)	191-24-2	276.30 HMW	1.33×10^{-8}	
Non-priority PAHs				
Cyclopenta[def]phenanthrene (CdefP)	203-64-5	190.24 LMW	- [c]	
Benzo[*c*]fluorene (BcF)	205-12-9	216.28 MMW	- [c]	

Table 2. *Cont.*

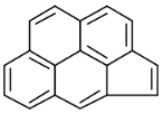
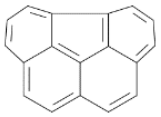
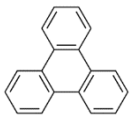
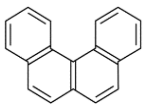
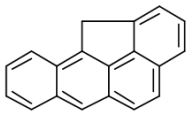
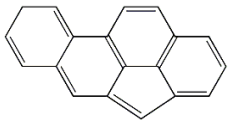
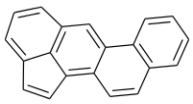
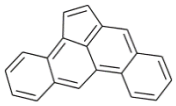
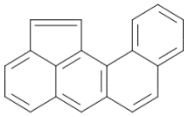
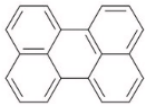
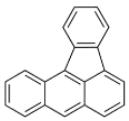
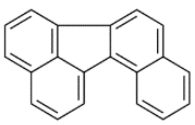
Species (Abbreviation)	CAS Number	MW [a] Category	Vapor Pressure [b]	Structure
Cyclopenta[*c*,*d*]pyrene (CcdP)	27208-37-3	226.30 MMW	- [c]	
Benzo[*ghi*]fluoranthene (BghiF)	203-12-3	226.30, MMW	2.56×10^{-5}	
Triphenylene (Tri)	217-59-4	228.30 MMW	2.8×10^{-6}	
Benzo[*c*]phenanthrene (BcP)	195-19-7	228.30 MMW	- [c]	
Retene (Ret)	483-65-8	234.30, MMW	- [c]	
11H-Benz[*bc*]aceanthrylene (11H-BbcA)	202-94-8	240.30 MMW	- [c]	
4H-Cyclopenta[def]chrysene (4H-CdefC)	202-98-2	240.30 MMW	- [c]	
Benz[*j*]aceanthrylene (BjA)	202-33-5	252.30 HMW	- [c]	
Benz[*e*]aceanthrylene (BeA)	199-54-2	252.30 HMW	- [c]	
Benz[*l*]aceanthrylene (BlA)	211-91-6	252.30 HMW	- [c]	
Perylene (Per)	198-55-0	252.30 HMW	6.67×10^{-8}	
Benzo[*a*]fluoranthene (BaF)	203-33-8	252.30 HMW	- [c]	
Benzo[*j*]fluoranthene (BjF)	205-82-3	252.30 HMW	3.60×10^{-6}	

Table 2. *Cont.*

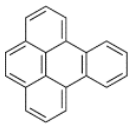
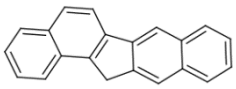
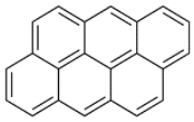
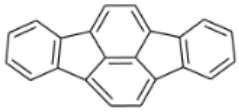
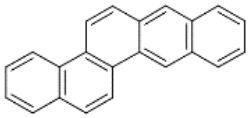
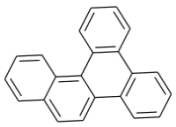
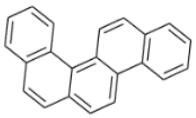
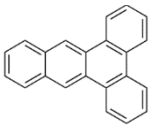
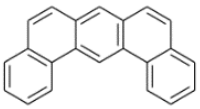
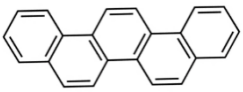
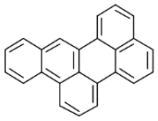
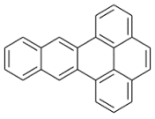
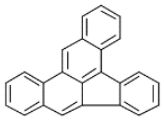
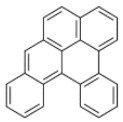
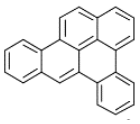
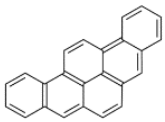
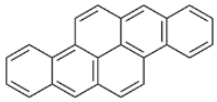
Species (Abbreviation)	CAS Number	MW [a] Category	Vapor Pressure [b]	Structure
Benzo[*e*]pyrene (BeP)	192-97-2	252.30 HMW	7.60×10^{-7}	
13H-Dibenzo[*a*,*h*]fluorene (13H-DahF)	239-85-0	266.3 HMW	- [c]	
Anthanthrene (Anth)	191-26-4	276.30 HMW	- [c]	
Indeno[1,2,3-*cd*]fluoranthene (IDF)	193-43-1	276.30 HMW	- [c]	
Benzo[*b*]chrysene (BbC)	214-17-5	278.30 HMW	- [c]	
Benzo[*g*]chrysene (BgC)	196-78-1	278.30 HMW	3.07×10^{-6}	
Benzo[*c*]chrysene (BcC)	194-69-4	278.30 HMW	1.20×10^{-7}	
Dibenz[*a*,*c*]anthracene (DBacA)	215-58-7	278.30 HMW	1.33×10^{-8}	
Dibenz[*a*,*j*]anthracene (DBajA)	224-41-9	278.30 HMW	- [c]	
Picene (Pic)	213-46-7	278.30 HMW	- [c]	
Coronene (Cor)	191-07-1	300.40 HMW	2.89×10^{-10}	
Benzo[*b*]perylene (BbPer)	197-70-6	302.40 HMW	- [c]	
Naphtho[2,3-e]pyrene (NeP)	193-09-9	302.40 HMW	- [c]	

Table 2. *Cont.*

Species (Abbreviation)	CAS Number	MW [a] Category	Vapor Pressure [b]	Structure
Dibenzo[*a*,*e*]fluoranthene (DBaeF)	5385-75-1	302.40 HMW	9.77×10^{-9}	
Dibenzo[*a*,*l*]pyrene (DBalF)	191-30-0	302.40 HMW	6.40×10^{-8}	
Dibenzo[*a*,*e*]pyrene (DBaeP)	192-65-4	302.40 HMW	6.93×10^{-9}	
Dibenzo[*a*,*i*]pyrene (DBaiP)	189-55-9	302.40 HMW	2.40×10^{-9}	
Dibenzo[*a*,*h*]pyrene (DBahP)	189-64-0	302.40 HMW	8.53×10^{-10}	

[a]: molecular weight (g/mol); [b]: Pa at 25 °C. Data referred from Ambrose et al. [42], Coover et al. [43], Goldfarb et al. [44], Lee et al. [45], Lei et al. [46], Yamazaki et al. [48], Hoyer et al. [49], Sonnefeld et al. [50], and PubChem (National Library of Medicine) [47]; [c]: No reference data.

PAHs are widely known for their carcinogenicity, mutagenicity and toxicity, and they pose a serious threat to human health [58,59]. A previous study has noted that with increasing MW, the carcinogenicity and acute toxicity of PAHs increase and decrease, respectively [60]. However, LMW and MMW PAHs can react with other gaseous air pollutants, such as ozone (O_3, which is a strong oxidizing agent that can damage human lung function, thus threatening human health [61]) and NO_x, to produce derivatives with a relatively low vapour pressure that more easily occur in the particle phase than their parent PAHs, and their mutagenicity and toxicity may be higher than those of the parent PAHs [62,63]. Table 3 summarizes the evaluation of PAHs (including heterocyclic PAHs) and their derivatives by the IARC [27,64,65]. In addition to benzo[*a*]pyrene (BaP), which is classified in Group 1 (carcinogenic to humans), seven species are classified in Group 2A (probably carcinogenic to humans), and twenty-five species are classified in Group 2B (possibly carcinogenic to humans). Moreover, several emission sources in outdoor air, including coal combustion, coal tar pitch, coke production, diesel engine exhaust, tobacco smoke, and wood dust, are classified in Group 1 by the IARC [27], which may release many PAHs and derivatives. Due to these harmful effects on human health, it is necessary to clarify the concentrations, compositions, and major contributors of PAHs in the atmosphere.

In contrast to PM observations, real-time observations of PAHs have not been commonly analysed. According to Li et al. [66] real-time observation of PM can offer a high temporal resolution, but low specificity of chemical characterization. Amador-Muñoz et al. [67] performed real-time PAHs sampling in parallel to non-real-time PM sampling. The non-real time concentrations of PAHs in PM were all lower than the results from real-time PAHs sampling, possibly because PAH derivatives and heterocyclic species were also detected as PAHs when using the real-time method, and the PAHs concentration in the atmosphere is overestimated [67]. On the other hand, the techniques with high temporal resolution and high specificity for the chemicals in PM are very expensive for routine air quality monitoring [68]. Therefore, the non-real-time determination method is still a well-

recognized method to determine the PAHs, including sampling, experimental treatment, and instrumental analysis. However, although the non-real-time observation method can measure different PAH species as needed, sample processing is a time-consuming task, the information of PAHs such as the atmospheric concentration cannot be promptly provided. To clarify the characteristics of the PAH species in PM, in our previous review, we summarized the size distributions of PM-bound PAHs freshly released from combustion sources and the distribution patterns of PM-bound PAHs in the atmosphere [69]. It was found that PAHs released from stationary sources are mainly bound to fine particles, but the sizes were slightly larger than that of PAHs released from mobile sources. In the atmosphere, PM-bound PAHs are more likely to be bound to large particles than to initial-mode particles, and the size decreases with increasing PAH MW [69]. Because the concentrations of PM-bound PAHs may differ according to people, the microenvironment, and area, in this review, we summarize the personal exposure concentrations of PM-bound PAHs and indoor and outdoor PM-bound PAHs in cities worldwide in recent years to further examine the exposure routes to atmospheric PM-bound PAHs and their health effects.

Table 3. Evaluation of PAHs (including heterocyclic PAHs) and their derivatives by IARC.

Evaluation	Species
Group 1 [a]	Benzo[*a*]pyrene
Group 2A [b]	Dibenz[*a*,*h*]anthracene, Cyclopenta[*cd*]pyrene, Dibenzo[*a*,*l*]pyrene, Dibenzo[*c*,*h*]acridine, 1-Nitropyrene, 6-Nitrochrysene, 2-Nitrotoluene,
Group 2B [c]	Naphthalene, Benz[*a*]anthracene, Chrysene, Benzo[*b*]fluoranthene, Benzo[*k*]fluoranthene, Benzo[*j*]fluoranthene, Indeno[1,2,3-*cd*]pyrene, Benzo[*c*]phenanthrene, Dibenzo[*a*,*e*]pyrene, Dibenzo[*a*,*h*]pyrene, Dibenzo[*a*,*i*]pyrene, Dibenzo[*a*,*h*]acridine, Dibenzo[*a*,*j*]acridine, Dibenzo[*c*,*g*]carbazole, 5-Methylchrysene, 2-Nitrofluorene, 4-Nitropyrene, 3,7-Dinitrofluoranthene, 3,9-Dinitrofluoranthene, 1,3-Dinitropyrene, 1,6-Dinitropyrene, 1,8-Dinitropyrene, 2,6-Dinitrotoluene, 3-Nitrobenzanthrone, 5-Nitroacenaphthene

[a]: Carcinogenic to humans; [b]: probably carcinogenic to humans; [c]: possibly carcinogenic to humans.

2. Concentrations of Atmospheric PM-Bound PAHs

2.1. Personal Exposure to PM-Bound PAHs

Research data have indicated that several factors, including conditions of the body, exposure routes, and environmental conditions, can influence the mechanisms of PAHs that are absorbed or adsorbed by human bodies [70]. PAHs may affect human health via inhalation, ingestion, and dermal (skin) exposure, with inhalation being the main exposure pathway for PM-bound PAHs [70]. Exposure varies from person to person due to variations in various physical factors such as the breathing rate. Table 4 lists several studies of personal exposure to PM-bound PAHs in different cities, which have focused on different participants, including non-occupational and occupational exposure.

Regarding non-occupational exposure, residents living in rural areas [71,72] generally inhale higher PM-bound PAH concentrations (4.2–655 ng/m^3) than residents living in urban areas (0.4–11.9 ng/m^3) [73,74]. This is because solid fuels such as coal and wood are still mainly used by rural residents for cooking and heating an many PAHs are emitted from these sources due to their low combustion efficiency, while urban residents mostly use clean fuels such as liquefied petroleum gas and natural gas. Thus, children living in Italy exhibited very low exposure levels and slight seasonal variation (0.65 and 0.63 ng/m^3) [75], whereas children living in China exhibited relatively high exposure levels and a relatively large seasonal variation (27.31 and 58.18 ng/m^3) [76]. A possible reason is the different urban type, whereby Rome (Italy) is a typical commercial city and Tianjin is one of the largest industrial centers of China, leading to the background concentration of PM-bound PAHs in Tianjin being higher than that in Rome [75,76]. In addition, the heating systems in Tianjin can lead to more PM-bound PAHs being released into the atmosphere in the winter [76]. Harbin is a large industrial city in northeast China, and it has been reported that the annual average PM-bound PAH exposure concentration in patients with chronic obstructive pulmonary disease is 186.85 ng/m^3 [77], which is much higher than that seen

in residents living in Hong Kong, Zhuhai, and Wuhan, China [73,74]. In Harbin, the annual temperature range can be up to 60 °C and the heating period can account for half of the year, thus, coal combustion for heating is also a large factor, in addition to industrial coal combustion, leading to the annual average concentration of PM-bound PAHs being higher than other cities in South and Central China [77].

Table 4. Personal exposure concentration (average and/or range, ng/m^3) of PM-bound PAHs in several studies.

Participants	PM	PAHs	Period	Concentration	Country, City
Rural residents	PM	28	July	655 ± 250	China, Laiyang [71]
Rural residents	$PM_{2.5}$	10	9–12 March 2013	4.2–224	Thailand, Lampang [72]
Health residents	$PM_{2.5}$	26	2014–2016	1.7 (0.4–5.2)	China, Hongkong [73]
Residents	$PM_{2.5}$	16	2015–2018	8.27	China, Zhuhai [74]
Residents	$PM_{2.5}$	16	2014–2017	11.9	China, Wuhan [74]
Children	$PM_{2.5}$	8	11 April–9 May 2012 15 July–3 November 2012	0.65 0.63	Italy, Rome [75]
Children	$PM_{2.5}$	16	17 May–23 June 2010 8 November–13 December 2010	27.31 58.18	China, Tianjin [76]
COPD patients	$PM_{2.5}$	16	June 2017–October 2018	186.85	China, Harbin [77]
Drivers	PM_4	12	2015–2018	9.97	Greece, Thessaloniki [15]
Office workers	PM_2	13	2015	4.0 ± 2.3	Australia, Canberra [16]
Office workers	$PM_{2.5}$	8	6–13 March 2009 10–19 June 2009	15.19 ± 15.15 3.04 ± 1.38	Czech Republic, Ostrava [78]
Policemen	$PM_{2.5}$	8	8–20 February 2009 17–27 May 2009	4.27 ± 2.95 1.03 ± 0.61	Czech Republic, Prague [78]
Policemen	$PM_{2.5}$	8	2–6 March 2009 6–10 June 2009	39.08 ± 17.33 4.27 ± 1.99	Czech Republic, Karvina [78]
Housewife	$PM_{2.5}$	19	4–21 November 2016	310 ± 443	China, Xingping [79]
Housewife	$PM_{2.5}$	19	January 2018	116 (32–224)	China, Xi'an, [80]
Highway toll station workers	$PM_{2.5}$	16	March–May 2014	319.90	China, Tianjin [81]
Newsagent	PM	16	2013	5570	Iran, Tehran [82]
Seafarers	PM	32	July 2016	760–8400	Sweden [83]
Chinese kitchen worker	PM	16	4 September–1 November 2014	1794–12,108	China, Taiwan [84]

Regarding occupational exposure, drivers, office workers, and policemen, as indicated in Table 4, did not exhibit very high PM-bound PAH exposure concentrations (1.03–39.08 ng/m^3) but the values differed among cities and revealed seasonal variations [15,16,78]. In addition to the low background concentrations of PM-bound PAHs in cities, a possible reason is that few direct emission sources were impacting the above professionals in space. However, both housewives and highway toll station workers exhibited relatively high exposure levels [79–81], which were several times higher than those for the professionals described above. The high exposure levels for housewives (116 and 310 ng/m^3) mainly occurred due to the use of fuel for cooking, while those for highway toll station workers (319.90 ng/m^3) largely occurred due to the inhalation of traffic-related PAHs, such as vehicle exhaust [79–81]. Traffic emissions are also a major source for exposure for newsagents because their workplaces are usually located near roads. However, the exposure levels of PM-bound PAHs for newsagents were much higher in Tehran (Iran) (5570 ng/m^3) [82] than those measured for highway toll station workers in Tianjin (China) [81]. A possible reason could be that the pollution levels may be higher in Tehran than that in Tianjin, and the background PM-bound PAH concentrations may be very high. Moreover, studies have revealed very high exposure levels of PM-bound PAHs for seafarers and Chinese kitchen workers, ranging from approximately 760 to 12,108 ng/m^3 [83,84]. The high exposure levels for the former occur due to ship engine emissions and their long residence times on ships, while those for the latter occurred due to factors such as the use of various fuels, food type, cooking location and methods. Also, many PM-bound PAHs contained in food can be ingested by the human body through eating [85,86].

The results found that background concentrations in different cities could affect the exposure levels of residents, and the indoor microenvironment could also influence the exposure level because most people spend a lot of time indoors. In addition, the charac-

teristics of the PAH phase distribution determine that the PM-bound PAH concentration is higher during cold periods than that during warm periods. Moreover, people with various jobs exhibit different exposure levels, especially regarding occupational exposure, and workers are exposed to very high health risks in spaces with high PM-bound PAH concentrations. Overall, although the PM type and determined PAH number differed among the above studies, personal exposure to PM-bound PAH depends on many complex factors, including indoor microenvironments, outdoor environments, daily activities (such as jobs), and body conditions.

2.2. Indoor Concentrations of PM-Bound PAHs

Indoor air quality is crucial to human health because people spend more than 80% of their time indoors [73]. The indoor PM-bound PAHs in different microenvironments are emitted by many sources, including cooking, smoking, heating, stoves, chemical spraying, machines such as laser printers, and outdoor sources. Studies have noted that indoor air may even be worse than the outdoor air with the PM-bound PAHs [71,87]. Table 5 summarizes the indoor concentrations of PM-bound PAHs in different microenvironments in certain cities in recent years. Some studies have simultaneously examined personal exposure, as described in Section 2.1 [71,73,79,80,83,84].

Regarding residential indoor air, the PM-bound PAH concentrations in rural households (738 ± 321 ng/m^3) were much higher than those in urban households (0.186–276 ng/m^3), and the concentrations observed in northern Chinese cities (15–276 ng/m^3) were higher than those observed in southern Chinese cities (1.0–7.3 ng/m^3) [71,73,79,80,88], similar to those of personal exposure (Section 2.1). The PM-bound PAH concentrations in households in Jeddah (Saudi Arabia) (18.5 ng/m^3) [89] were comparable to those in Bursa (Turkey) (22 ng/m^3) [90], while in Madrid (Spain) [91] they were much lower (0.186 ng/m^3) than in the above cities. In both the Jeddah and Bursa studies air samplers were placed in the living room of residences, while in Madrid, Spain, it was placed on the first floor of a residential building [89–91]. The different locations of air samplers could lead to different PM-bound PAH concentration measurements. On the other hand, a high concentration of PM-bound PAHs (318 ± 314 ng/m^3) was observed in infants' rooms in Harbin (China) in winter, was not only suggested a high health risk for infants, but also for people who staying in the room [92].

Regarding school indoor air, Table 5 does not indicate a large concentration difference in Wuhan (China), among university dormitories (31.3 ng/m^3), laboratories (27.0 ng/m^3), and offices (32.4 ng/m^3) [93]. The annual average concentration differences observed in universities between dormitories and offices in Beijing (34.1 and 32.1 ng/m^3) were also not large [88], and comparable to those seen in Wuhan [93]. However, the concentrations observed in university offices in Jeddah (12.7 ± 5.1 ng/m^3) [89] were lower than those observed in university offices in Beijing, consistent with the household results. Moreover, the PAH concentrations observed at the laboratories and offices in Harbin (115 and 96.6 ng/m^3) in winter [94] were approximately three times higher than those observed in Beijing and Wuhan [88,93]. In contrast, the PM-bound PAH concentrations in primary or secondary classrooms ranged from 0.45 to 29.83 ng/m^3, and the concentrations were comparable across Beijing, Warsaw and Gliwice (Poland), and Porto (Portugal), but they were very low in São Paulo (Brazil) [17,95–97]. Different from other sampling cities, São Paulo is close to the Equator and its hot climate can greatly influence the gaseous/particles phase distribution of PAHs. Moreover, the shorter sampling period used in São Paulo than in the other cities also had an impact on the PM-bound PAHs results [96].

Regarding public indoor air, the lowest concentration level (2.39–7.4 ng/m^3) was observed in shopping malls in Islamabad (Pakistan), bakeries in Bari (Italy) and hotels in Jeddah [89,98,99]. This may occur because these public places contained few PAH sources. The second-highest concentration level (39.58–155.11 ng/m^3) was observed in hotels in Jinan (China) in public bars in Warri (Nigeria) and in office buildings in Changchun (China) [100–102]. The higher concentration of PM-bound PAHs in hotels in Jinan than

that in hotels in Jeddah possibly occurred due to the local background urban concentrations, and is consistent with the results for Beijing and Jeddah [88,89]. The relatively high concentration in public bars may be the result of factors such as tobacco smoking, while in office buildings, this may occur due to emissions from machines such as printers [103]. The seasonal differences observed in office buildings may occur due to the gaseous/particles phase distribution of PAHs at different temperatures and various outdoor pollution sources via window opening or ventilator operation [102]. The highest concentration level (550–39,000 ng/m^3) was observed in fire stations, ships, and Chinese kitchens [83,84,104]. Smoke originating from fires contains a large number of PM-bound PAHs, which could be adsorbed onto the helmets and clothes of firefighters and transported to fire stations [104]. The high concentrations observed in ships and Chinese kitchens determine the personal exposure levels of seafarers and Chinese kitchen workers (Section 2.1), respectively, because of engine fuel combustion and cooking, respectively [83,84]. However, the lower concentration in kitchens than the personal exposure concentration for kitchen workers possibly occurred because the air sampler was likely not located close to the cooking bench for safety reasons (high temperature) [84].

Table 5. Indoor concentrations (average and/or range, ng/m^3) of PM-bound PAHs in several studies.

Place	Country, City	PM	PAHs	Periods	Concentration
Rural households	China, Laiyang [71]	PM	28	July	738 ± 321
	China, Hongkong [73]	$PM_{2.5}$	26	2014–2016	3.0 (1.0–7.3)
	China, Xingping [79]	$PM_{2.5}$	19	4–21 November 2016	211 ± 120
	China, Xi'an [80]	$PM_{2.5}$	19	January 2018	92 (15–276)
Households	China, Beijing [88]	$PM_{2.5}$	16	December 2014–February 2016	39.8
	Saudi Arabia, Jeddah [89]	PM_{10}	13	-	18.5 ± 11.2
	Turkey, Bursa [90]	PM	16	July 2014– January 2015	22
	Spain, Madrid [91]	PM_{10}	14	May 2017–April 2018	0.186
Infant room	China, Harbin [92]	PM	16	December 2013–March 2014	318 ± 314
University (dormitory)	China, Beijing [88]	$PM_{2.5}$	16	December 2014–February 2016	34.1
	China, Wuhan [93]	$PM_{2.5}$	16	December 2014–June 2015	31.3
University (laboratory)	China, Wuhan [93]	$PM_{2.5}$	16	December 2014–June 2015	27.0
	China, Harbin [94]	$PM_{2.5}$	16	January 2015	115
	China, Beijing [88]	$PM_{2.5}$	16	December 2014–February 2016	32.1
University (office)	Saudi Arabia, Jeddah [89]	PM_{10}	13	-	12.7 ± 5.1
	China, Wuhan [93]	$PM_{2.5}$	16	December 2014–June 2015	32.4
	China, Harbin [94]	$PM_{2.5}$	16	January 2015	96.6
	China, Beijing [95]	$PM_{2.5}$	12	October 2016–March 2017	29.83
Classroom	Brazil, São Paulo [96]	PM	15	7–11 November 2016	0.45
	Poland, Warsaw [17]	PM_1	16	April–June 2015	10.9
	Poland, Gliwice [17]	PM_1	16	April–June 2015	21.6
	Portugal, Porto [97]	$PM_{2.5}$	18	March–May 2014	5.03–23.6
Shopping malls	Pakistan, Islamabad [98]	$PM_{2.5}$	16	February–April 2014	2.39 ± 1.45
Bakery	Italy, Bari [99]	$PM_{2.5}$	7	7–19 April 2013	7.4
Hotels	Saudi Arabia, Jeddah [89]	PM_{10}	13	-	6.3 ± 1.3
	China, Jinan [100]	$PM_{2.5}$	19	January 2016	39.58–115.63
Public bars	Nigeria, Warri [101]	PM	16	-	43.43–155.11
Office building	China, Changchun [102]	$PM_{2.5}$	16	April–October 2018	48.6
				December 2017–April 2018	67.9
Fire station	Poland, North Poland [104]	PM_4	15	September 2018	1882–5924
Ship	Sweden [83]	PM	32	July 2016	550–39,000
Chinese kitchen	China, Taiwan [84]	PM	16	4 September–1 November 2014	1648–5342

In contrast to personal exposure studies, air samplers remain fixed indoors rather than being portable. Hence, the indoor concentrations of PM-bound PAHs are related to the type of space and sampler location. Different space uses to determine whether direct sources of PAH emissions occur, and different air sampler locations may determine whether more or less PAHs are collected. However, the air is exchanged between indoor

and outdoor environments via opening windows or operating fans. Therefore, the air exchange speed and local outdoor concentration exert some considerable impacts on the indoor concentration. For example, if PM-bound PAHs are notably generated indoors and the air exchange speed is low, the indoor concentrations may be higher than the outdoor concentrations (such as in Chinese kitchens [84]). Although the PM type and determined PAH number differed among the above studies, the resulting impacts of PM-bound PAHs concentration levels were not large for the overall characteristics of different microenvironments, e.g., the concentrations were much higher in kitchens than those in classrooms, and higher in rural households than those in urban households.

2.3. Outdoor Concentrations of PM-Bound PAHs

The atmospheric behaviours of outdoor PM-bound PAHs are more complex because they are not only dependent on various direct emission sources including industrial and traffic emission [105], wood and biomass burning [59], but also many complex atmospheric physical and chemical factors, including interactions with other pollutants, photochemical degradation, and dry and wet deposition [106,107]. Due to the different conditions in countries worldwide, the concentrations of PM-bound PAHs also vary among regions. In this review, relevant reports (in English) were retrieved on outdoor PM-bound PAHs worldwide in recent years, and the available data are summarized in Table 6.

As indicated in Table 6, Auckland (New Zealand) attained the lowest annual average concentration of outdoor PM-bound PAHs (0.31 ± 0.19 ng/m^3) [108] among all the cities/countries. Cities in the Americas also attained relatively low overall average concentrations levels of outdoor PM-bound PAHs, which ranged from 0.84 to 10.2 ng/m^3 [67,109–113]. In Europe, the observations in A Coruña (Spain) (7.56 ng/m^3) [114] exhibited a lower annual average concentration than that in cities in southern Spain (26.2 ng/m^3) [115]. A seasonal variation was observed in Milan (Italy) (0.40 and 72.8 ng/m^3) [116] and Wadowice (Poland) (10.5 and 80.6 ng/m^3) [117] which was larger during cold periods than that during warm periods. During cold periods, the concentration of PM-bound PAHs in Nicosia (Cyprus) (1.62 ng/m^3) [118] was the lowest among European cities. The concentrations observed in Brno (Czech Republic) (20.7 ng/m^3) [119] were comparable to those observed in Zagreb (Croatia) (25.4 ng/m^3) [120], whereas the concentrations in Sarajevo (Bosnia-Herzegovina) (64.8 ng/m^3) [120] were much higher than those in Zagreb, even though the samples were collected during the same period. During warm periods, slight differences in the PM-bound PAH concentration were observed among Moscow (1.32–7.68 ng/m^3), St. Petersburg (1.71–6.30 ng/m^3), and Kazan (2.95–9.61 ng/m^3) in Russia [121]. Although most countries in Europe as shown in Table 6 are developed countries, the overall concentration levels of outdoor PM-bound PAHs were relatively higher than those in the Americas. One reason was the different sampling periods that the concentrations were mostly annual average in the Americas, whereas were mostly seasonal average in Europe. In addition, different meteorological conditions in different periods can lead to the concentration differences of PM-bound PAHs. Moreover, the higher population density in Europe (~73/km^2) than in the Americas (~21/km^2), with more human activities including industrial and traffic emission also can increase PM-bound PAHs concentrations.

In Africa, the limited available studies involving short-term observations of outdoor PM-bound PAHs showed that the average concentrations in Algiers (Algeria) (7.47 ± 1.21 ng/m^3) [122] and Pretoria (South Africa) (4.11 ng/m^3) [123] were much lower than that in Kigali (Rwanda) (52.7 ng/m^3) [124]. On the other hand, Ofori et al. [125] summarized the studies on the PAHs from 2005 to 2019 in Africa. Only 14 reports among 121 papers focused on the indoor and outdoor PM-bound PAHs. Of those, only four papers related to outdoor PM-bound PAHs in the last five years, in which air samples were collected in Rwanda (this report), Tunisia (urban area, 2.8 ± 3.4 ng/m^3), Nigeria (industrial area, 73–143 ng/m^3), and Egypt (rural: 323 ng/m^3; suburban: 503 ng/m^3; urban: 417 ng/m^3) [125].

In Asia, China has the largest population globally. Since the severe haze event in 2013, researchers have paid special attention to the atmospheric environment. Yan et al. [126] re-

viewed 270 studies of PM-bound PAHs in 67 cities from 2001 to 2016, which covered seven typical regions, including North, Northwest, Northeast, East, Central, South, and Southwest China. It has been reported that the annual mean concentrations of PM-bound PAHs in these cities range from 3.35 to 910 ng/m^3, and those in the northern regions are higher than those in the southern regions [126]. In this paper, Table 6 lists the concentrations of PM-bound PAHs in several major Chinese cities collected within 5 years. The results for these cities indicate average concentrations ranging from 1.36 to 1056 ng/m^3 [18,73,127–136], and the seasonal and regional differences are consistent with the results reported by Yan et al. [126]. In other Asian cities, relatively low concentrations levels of PAHs have been observed in Japan [137–139], South Korea [140,141], Vietnam [142], Singapore [143], Malaysia [144], Thailand [145], Qatar [146], and Lebanon [147], ranging from 0.56 ng/m^3 (Doha, Qatar) to 29.5 ng/m^3 (Gwangju, South Korea). Relatively high PAH concentration levels have been observed in Mongolia [148], Pakistan [149], India [150–152], and Iran [153,154] ranging from 0.66 ng/m^3 (Bushehr, Iran) to 773 ng/m^3 (Ulaanbaatar, Mongolia), which countries are the most polluted areas in the world. In Mongolia, wood and biomass burning for cooking and heating were the largest emission sources of PM-bound PAHs in the winter [148], while in India and Iran, traffic emission was the main contributor for PM-bound PAHs through the years [151,153]. The air pollutants in Iran were also largely influenced by the air masses long-range transported from Iraq and other Middle East areas [155]. Moreover, due to the special location, frequent recirculation of air masses resulted in the increased residence time of PM in Iran and lead to high air pollutants levels [156].

Table 6. Outdoor PM-bound PAHs concentrations (average and/or range, ng/m^3) in some cities worldwide.

Country, City	PM	PAHs	Periods	Concentration
		Oceania		
New Zealand, Auckland [108]	$PM_{2.5}$	15	2016–2017	0.31 ± 0.19
		Americas		
Mexico, South Mexico [67]	$PM_{2.5}$	24	November 2016–March 2017	4.82 ± 1.97
Peru, Arequipa [109]	$PM_{2.5}$ PM_{10}	14	January–December 2018	7.4 ± 2.3 9.6 ± 3.9
Argentina, Cordoba [110]	PM_{10}	14	August 2011–August 2013	4.5 ± 4.34
Brazil, Belo Horizonte [111]	$PM_{2.5}$	16	May 2017–April 2018	1.68–6.24
Canada, Toronto [112]	PM_{10}	17	August 2016–August 2017	10.2 ± 2.5
US, Washington [113]	PM_{10}	19	April 2016–September 2018	0.84
		Europe		
Spain, Coruña [114]	PM_{10}	12	Januray–December 2017	7.56
Spain, South Spain [115]	$PM_{2.5}$ PM_{10}	16	July 2014–June 2015	23.0 26.2
Italy, Milan [116]	$PM_{2.5}$	-	December 2018–February 2019 May–July 2019	72.8 ± 16.6 0.40 ± 0.07
Poland, Wadowice [117]	PM_{10}	9	March 2017 August 2017	80.6 10.5
Cyprus, Nicosia [118]	$PM_{2.5}$	50	January–March 2018	1.62
Czech Republic, Brno [119]	PM_1	15	January–February 2017	20.7
Croatia, Zagreb [120]	PM_{10}	10	December 2017–February 2018	25.4
Bosnia and Herzegovina, Sarajevo [120]	PM_{10}	10	December 2017–February 2018	64.8
Russia, Moscow [121]	PM_{10}	9	June–July 2018	1.32–7.68
Russia, St. Petersburg [121]	PM_{10}	9	June–July 2018	1.71–6.30
Russia, Kazan [121]	PM_{10}	9	June–July 2018	2.95–9.61
		Africa		
Algeria, Algiers [122]	PM_{10}	22	June–September 2016	7.47 ± 1.21
South Africa, Pretoria [123]	$PM_{2.5}$	16	June–July 2016	4.11
Rwanda, Kigali [124]	$PM_{2.5}$	15	May–June 2017	52.7
		Asia		
China, Hongkong [73]	$PM_{2.5}$	26	2014–2016	3. 9 (1.5–9.6)

Table 6. *Cont.*

Country, City	PM	PAHs	Periods	Concentration
China, Xi'an [127]	$PM_{2.5}$ PM_{10}	16	December 2016–December 2017	63.1 (14.3–266) 66.8 (9.69–349)
China, Shanghai [18]	$PM_{2.1}$	9	July 2017 January 2018	1.36 ± 0.20 7.72 ± 3.33
China, Beijing [128]	PM_{10}	15	January 2017	98.1 ± 48.2
China, Zhengzhou [128]	PM_{10}	15	January 2017	77.9 ± 29.6
China, Guangzhou [129]	$PM_{2.5}$	16	June–July 2016 November–December 2016	5.49 10.5
China, Taiyuan [129]	$PM_{2.5}$	16	June–July 2016November–December 2016	29.5 197
China, Jinan [130]	$PM_{2.5}$	18	March–December 2016	39.8 (8.18–246)
China, Shanxi [131]	PM_{10} $PM_{2.1}$	17	January–February 2017	1056 ± 315 937 ± 294
China, Urumqi [132]	$PM_{2.5}$	16	September 2017–September 2018	448
China, Chengdu [133]	PM_{10}	16	March 2015–February 2016	82.0 ± 64.8
China, Changchun [134]	$PM_{2.5}$	15	October–November 2016	81.4 ± 46.0
China, Harbin [135]	$PM_{2.5}$	16	June 2017–May 2018	86.9
China, Lanzhou [136]	$PM_{2.5}$	9	July 2017–October 2018	9.86
Japan, Kanazawa [137]	$PM_{2.5}$	9	April 2017–February 2018	0.69
Japan, Chiba [138]	$PM_{2.5}$	21	June 2016–October 2017	2.9
Japan, Kirishima [139]	$PM_{2.5}$	9	November–December 2016	1.32 (0.36–2.90)
South Korea, Seoul [140]	$PM_{2.5}$	14	January–December 2018	5.6 ± 7.9
South Korea, Gwangju [141]	$PM_{2.5}$	17	October 2016–April 2017	1.04–29.5
Vietnam, Hanoi [142]	PM_{10}	9	2016–2018	8.51
Singapore, Singapore [143]	PM_{10}	16	May 2015–June 2016	0.68–5.97
Malaysia, Lumpur [144]	$PM_{2.5}$	16	June 2015–May 2016	2.04 ± 0.28
Thailand, Chiang Mai [145]	$PM_{2.5}$	8	February–April 2016	5.88 ± 1.97
Qatar, Doha [146]	$PM_{2.5}$ PM_{10}	36	May–December 2015	0.56 0.72
Lebanon, Beirut [147]	$PM_{2.5}$	15	December 2018–October 2019	0.95
Mongolia, Ulaanbaatar [148]	PM_{10}	15	January 2017 March 2017 September 2017	131–773 22.2–531 1.4–54.6
Pakistan, Islamabad [149]	$PM_{2.5}$ PM_{10}	16	January–September 2017	25.7 ± 12.0 40.1 ± 16.8
India, Jamshedpur [150]	$PM_{2.5}$	16	December 2016–February 2017 March–May 2017	109 ± 18.2 81.1 ± 13.3
India, Delhi [151]	$PM_{2.5}$	14	December 2016–December 2017	753 ± 252
India, Haryana [151]	$PM_{2.5}$	14	December 2016–December 2017	259 ± 64.6
India, Uttar Pradesh [151]	$PM_{2.5}$	14	December 2016–December 2017	535 ± 143
India, Pune [152]	$PM_{2.5}$ PM_{10}	16	March 2015–March 2016	342.4 ± 14.3 446.1 ± 25.6
Iran, Tehran [153]	PM_{10}	16	February–March 2018	213 ± 145
Iran, Bushehr [154]	$PM_{2.5}$	16	December 2016–September 2017	0.66–142.3

According to the simultaneously obtained $PM_{2.5}$ and PM_{10} data, as summarized in Table 6 [109,115,127,131,146,149,152], the concentrations of $PM_{2.5}$-bound PAHs accounted for approximately 65%~95% of the total PM-bound PAHs, proving that PAHs mostly occur in PM with a small size, which is consistent with previous studies reporting that the adsorption of PAHs depends on the PM type [69]. Although high outdoor concentrations of PM-bound PAHs were observed in certain cities, they were not as high as the personal exposure concentrations for seafarers and kitchen workers nor the indoor concentrations observed in ships and Chinese kitchens [83,84]. This occurs because expansive outdoor spaces facilitate the diffusion and dilution of air pollutants. Moreover, outdoor air pollutants easily degrade or transform in the atmosphere due to various meteorological conditions and other reasons. Moreover, although there have been a large number of environmental observation studies on PM-bound PAHs over the past few decades, different countries

and regions have focused on different PM-bound PAH research aspects. Some major cities have published many reports on various PAH species, while other cities have published no research reports on PAHs at all.

3. Health Effects and Assessments of PAHs

Over the last decades, many studies have been performed to better understand the health effects of PAHs. The health effects of PAH exposure can be divided into acute (short-term) effects and chronic (long-term) effects [59]. The acute effects mainly depend on the exposure time and PAH concentrations during exposure, while other factors, including pre-existing health conditions and age, may also influence health impacts [59]. Short-term exposure to high PAH levels may cause impaired lung function in patients with asthma and thrombotic effects in people suffering from coronary heart disease [157]. Acute occupational exposure to high PAH concentrations could cause eye irritation, nausea, vomiting, and diarrhea [158]. Repeated skin contacts with certain PAHs, such as anthracene (Ant) and naphthalene (NaP), are also known to cause skin irritation and inflammation [159]. However, PAHs to induce acute (short-term) human health effects at environmental concentrations are not fully understood.

In terms of chronic effects, long-term exposure to PAHs may induce DNA adduct formation in vitro and in vivo, in which the formation of DNA adducts is a key event regarding the mutagenicity and carcinogenicity of PAHs [160]. Rota et al. [161] summarized the studies of the respiratory and urinary tract cancers published between 1958 and 2014, that the high respiratory cancers (mainly lung cancer) were found in the workers who worked in iron and steel foundries for a long time. Petit et al. [162] investigated 93 exposure groups in nine industries and the results showed the highest lung cancer risk level was found in coke and silicon production, the lowest was in bitumen manufacture. Moreover, long-term exposure to PAHs can increase cardiovascular diseases (CVDs) risks and/or risk factors. Poursafa et al. [163] reviewed the related reports on exposure to PAHs and CVDs from 2000 to 2017. Most longitudinal with long-term follow-up studies indicated significant positive correlations of exposure to PAHs with CVDs increased risks. On the other hand, some studies reported that exposure to PAHs had negative effects on the development of children. For example, Kalantary et al. [164] summarized the studies of long-term exposure to PAHs with attention deficit hyperactivity disorder in children until 2018. Although overall studies did not show consistent results, the harm of exposure to PAHs on children is still worth noting and further research.

Regarding the assessment of PAHs, a large PAH database has been structured in various test systems including carcinogenicity, mutagenicity, and genotoxicity in the past decades [58,160]. Certain PAHs have been classified as carcinogenic to humans (Groups 1, 2A, and 2B, as indicated in Table 3) by IARC [27,64,65]. Several LMW PAHs, such as Ant and fluorene (Flu), have not been classified as carcinogenic. The carcinogenicities of some PAHs, such as acenaphthene (Ace), phenanthrene (Phe), and pyrene (Pyr), remain questionable [160]. On the other hand, because of the toxicity difference between different PAH species, their concentrations are insufficient to indicate the toxicity of PAHs. Hence, it is necessary to choose an appropriate index species for comparison. BaP is one of the most typical carcinogenic PAHs with the largest body of available data describing its exposure and health effects that BaP has commonly been adopted as the index species [160]. Table 7 summarizes the reference data of certain PAHs including toxicity equivalency factor (TEF) and relative potency factors (RPF) regarding their cancer risks based on BaP [160,165–173]. There is more available data for the 16 PAHs prioritized by the US EPA than for non-priority PAHs, and Table 7 indicates that among these 16 PAHs, the factor values are relatively low for most LMW PAHs and relatively high for most HMW PAHs. In addition to BaP, dibenz[*a*,*h*]anthracene (DBA) exhibits the highest factor value, which is 5 (TEF) and 10 (RPF) times higher than that reported by Nisbet and LaGoy [167] and the US EPA [160], respectively. Regarding the non-priority PAHs, certain PAHs exhibit relatively high factor values, such as benzo[*c*]fluorene (BcF), benz[*j*]aceanthrylene (BjA),

and dibenzo[*a*,*l*]pyrene (DBalP), whose RPF values are 20, 60, and 30 times higher than that of BaP [160], respectively, suggesting that even if the concentrations of these species in the atmosphere are very low, their health effects on humans are extremely notable.

Table 7. Reference data of certain PAHs regarding their cancer risks based on BaP.

Species [a]	1984 [b]	1988 [c]	1992 [d]	1993 [e]	1994 [f]	1997 [g]	1998 [h]	1998 [i]	2004 [j]	2010 [k]
US EPA 16 PAHs										
Acy			0.001		0.001					
Ace			0.001		0.001					
Flu			0.001		0.001					
Ant			0.01		0.01		0.0005			0
Phe			0.001			0.00064	0.005			0
FR			0.001		0.001		0.05			0.08
Pyr		0.081	0.001			0	0.001			0
BaA	0.013	0.145	0.1	0.1	0.1	0.014	0.005	0.1		0.2
Chr	0.001	0.0044	0.01	0.001	0.01	0.026	0.03	0.01		0.1
BbF	0.08	0.14	0.1	0.1	0.1	0.11	0.1	0.1	0.62	0.8
BkF	0.004	0.066	0.1	0.01	0.1	0.037	0.05	0.1	0.17	0.03
BaP	1	1	1	1	1	1	1	1	1	1
DBA	0.69	1.11	5	1	1	0.89	1.1			10
IDP	0.017	0.232	0.1	0.1	0.1	0.067	0.1	0.1		0.07
BgPe		0.022	0.01		0.01	0.012	0.02			0.009
Non-priority PAHs										
BcF										20
CcdP		0.023			0.1	0.012	0.02			0.4
BcP						0.023	0.023			
11H-BbcA										0.05
4H-CdefC										0.3
BjA										60
BeA										0.8
BlA										5
Per					0.001					
BjF		0.061			0.1	0.045	0.05	0.1	0.52	0.3
BeP		0.004			0.01	0	0.002			
Anth		0.32				0.28	0.3			0.4
DBacA					0.1					4
Cor					0.001					
NeP										0.3
DBaeF										0.9
DBalP							1	10		30
DBaeP							0.2	1		0.4
DBaiP						1.1	0.1	10	12	0.6
DBahP						1.2	1	10	11	0.9

[a]: Full name were shown in Table 2. [b]: Chu, 1984 [165]; [c]: Clement Associates, 1988 [166]; [d]: Nisbet and Lagoy, 1992 [167]; [e]: US EPA, 1993 [168]; [f]: Malcolm and Dobson, 1994 [169]; [g]: Muller et al., 1997 [170]; [h]: Larsen and Larsen, 1998 [171]; [i]: Collins et al., 1998 [172]; [j]: Cal EPA, 2004 [173]; [k]: US EPA, 2010 [160].

Currently, it is difficult to accurately assess the health effects of PAHs. The inhalation lifetime cancer risk (ILCR) model has been widely applied to estimate the health risk in people induced by inhalation exposure to PAHs [72], which is usually calculated according to two methods. One method is the occupational exposure assessment method, and the ILCR is calculated with Equation (1):

$$ILCR = BaP_{eq} \times UR_{BaP} \quad (1)$$

where BaP_{eq} is the BaP-equivalent concentration (ng/m^3), calculated by multiplying the PAH concentration by TEF. UR_{BaP} is the unit cancer risk resulting from BaP, which is estimated as 8.7×10^{-5} per ng/m^3, which is based on epidemiological data retrieved from studies on coke oven workers [58]. The other method is the non-occupational exposure assessment method, and the ILCR is calculated with Equation (2):

$$ILCR = BaP_{eq} \times SF \times IR \times EF \times ED \times CF/(BW \times AT) \quad (2)$$

where SF is the cancer slope factor ($mg \cdot kg^{-1} \cdot day^{-1}$) for BaP inhalation exposure, IR is the inhalation rate ($m^3 \cdot day^{-1}$), EF is the exposure frequency (350 $days \cdot year^{-1}$), ED is the exposure duration (year), CF is the conversion factor with a value of 10^{-6}, BW is the body weight (kg), and AT is the average lifespan for carcinogens (25,550 days). The population can be divided into males and females based on gender and subdivided into children, adolescents, adults, and senior adults based on age.

4. Conclusions

PM is one of the major factors contributing to air quality deterioration. PM regulation standards have been formulated in many countries, and the air quality index (AQI) of PM is monitored in real-time in most areas worldwide. Although PM-bound PAHs account for a very low percentage of PM, the behaviour of PAHs in the atmosphere is notable due to their carcinogenicity and mutagenicity. This review summarized several studies of personal exposure and indoor and outdoor PM-bound PAHs in recent years. The reviewed results indicated that personal exposure to PM-bound PAHs largely differed by the region, season, and job and depended on many complex factors. The indoor concentrations of PM-bound PAHs were related to the type of space use. The outdoor PM-bound PAH concentrations exhibited regional and temporal differences, and they were higher during cold periods than those during warm periods, while Asia was the region with the most serious PAH pollution. On the other hand, a novel coronavirus (COVID-19) was first discovered in China at the end of 2019, and after it broke out it then quickly spread all over the globe. To contain the COVID-19 epidemic, the outdoor activities of people (traffic, industry, recreation, etc.) were largely limited in many countries. Although the harm caused by COVID-19 is very large, several studies found that these control measures could greatly affect air pollutants emissions and the air quality, and the premature deaths due to improved air quality declined in many countries and regions [174–177], suggesting the control of air pollution should be more strengthened.

It has been more than four decades since the US EPA released a list of 16 critical PAHs. This list is known as the US EPA 16 PAHs, which has played a vital role in environmental and analytical research. However, there are several hundred PAH species globally, and some studies summarized in this review have analysed other PAH species, while certain non-priority PAHs exhibit higher toxicity than that of BaP. Furthermore, several PAH derivatives, such as oxygenated PAHs, nitrated PAHs, chlorinated PAHs (ClPAHs), and brominated PAHs (BrPAHs), are not included in the list, and ClPAHs and BrPAHs have only recently attracted attention. Toxicological studies have suggested that certain PAH derivatives exhibit even higher carcinogenicity and mutagenicity than those of their parent PAHs. For instance, 1-, 4-nitropyrene (1-, 4-NPs), and 6-nitrochrysene (6-NC) have been classified as Group 2A and 2B by IARC, which had higher TEF values (1-, 4-NPs: 0.1; 6-NC: 10) than Pyr and Chr [178]. These results indicate that the total health effects of PAHs are typically underestimated. A more effective measure of global health risk assessment of the exposure to PM-bound PAHs should be developed in the future.

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