Impacts of agricultural waste burning on the enhancement of $PM_{2.5}$-bound polycyclic aromatic hydrocarbons in northern Thailand

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Abstract

In Northern Thailand, wildland fires during the cold period release large amounts of smoke and fine particles into the atmosphere. The fine particles include several persistent organic compounds such as PAHs. In this study, $PM_{2.5}$-bound PAH concentrations in the air of nine administrative provinces, namely Chiang-Mai, Chiang-Rai, Nan, Phayao, Mae Hong Son, Phrae, Lampang, Lamphun, Uttaradit (northern Thailand), were determined during the wildland fire and non-wildland fire seasons. The monitoring strategy comprised two campaigns in each season. $PM_{2.5}$ was collected using MiniVolTM portable air samplers (Airmetrics) with quartz fibre filters. Both PAHs and their B[*a*]P equivalent concentrations of other urban cities around the world were significantly higher than those of northern provinces for both seasons. The average cancer risks observed at nine administrative provinces were $8.525\times10^{-4}\pm3.493\times10^{-3}$ and $2.558\times10^{-3}\pm6.986\times10^{-3}$ for ingestion rate of 50 and 100 mg day^{-1}, respectively. The excess cancer risks of world cities for ingestion rate of 50 and 100 mg day^{-1} were much higher than those of northern Thailand for 851 and 567 times in that order. Dust ingestion was exceedingly critical to non-dietary PAH exposure in comparison with $PM_{2.5}$ inhalation. These results are in good agreement with those of previous studies, underlining the significance of indoor air quality on long-term adverse respiratory diseases in Asian cities.
Keywords: wildland fire, PAHs, northern Thailand, PM2.5, incremental lifetime cancer risk.


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1 Introduction

The environmental fate of particle bounded polycyclic aromatic hydrocarbons (PAHs) has been comprehensively examined during the past few years [1–5]. PAHs are a group of aromatic rings that are formed during the imperfect combustion of agricultural waste, domestic garbage, biomass, lignite, and volcanic eruption [6–11]. Since PAHs responsible for many adverse health effects such as cancer, endocrine disruption, reproductive and developmental effects, several studies have been focusing on the fate and behavior of $PM_{2.5}$-bound PAH concentrations in many cities around the world [12–14]. Despite numerous researches focusing on the atmospheric concentrations of PAHs in many cities, the impacts of agricultural waste burnings on fluctuations of PAH contents are not yet well studied.

Previous reports seem to concentrate on the air pollution problems triggered by wildland fires, accidental biomass burnings, and agricultural waste combustions connected with transboundary haze pollution from Laos, Myanmar, and Thailand [15–18]. As a result, it is crucial to pay more attentions on human exposure risk to $PM_{2.5}$-bound PAHs, and thus to conduct ecological risk assessment. To the extent of our knowledge, there have been no studies focusing on neither chemical analysis nor ecological risk assessment of $PM_{2.5}$-bound PAHs in nine administrative northern provinces of Thailand. Overall, the major aims of this research are to evaluate the atmospheric contents of particulate PAHs before and after the haze episode and to conduct the ecological risk assessment of $PM_{2.5}$-bound PAHs at nine administrative provinces in northern part of Thailand.

2 Methodology

2.1 Sampling site descriptions and monitoring period

All monitoring stations are categorized as tropical wet and dry climate (Köppen climate classification: Aw). The topographical situations of individual air quality observatory sites were described as follows: Chiang-Mai Observatory Site (CMOS) (E: 498805, N: 2077713), Chiang-Rai Province Observatory Site (E: 593783, N: 2258302), Nan Province Observatory Site (NPOS) Thewarat Hotel (E: 687123, N: 2077209), Phayao Province Observatory Site (PYOS) Arunothai Coffee House Homestay (E: 594420, N: 2119215), Mae Hong Son Province Observatory Site (MHOS) Mae Hong Son Provincial Forestry Office (E: 391834, N: 2134869), Phrae Province Observatory Site (PHOS) Nana Charoenmuang Hotel (E: 620935, N: 2006155), Lampang Province Observatory Site (LMOS) Maemoh Training Center (E: 568200, N: 2020017), Lamphun Province Observatory Site (LPOS) Lamphun Provincial Administration Organization Stadium (E: 500441, N: 2052987), Uttaradit Province Observatory Site (UTOS) OUM Hotel (E: 615923, N: 1948269) (See Fig. 1).

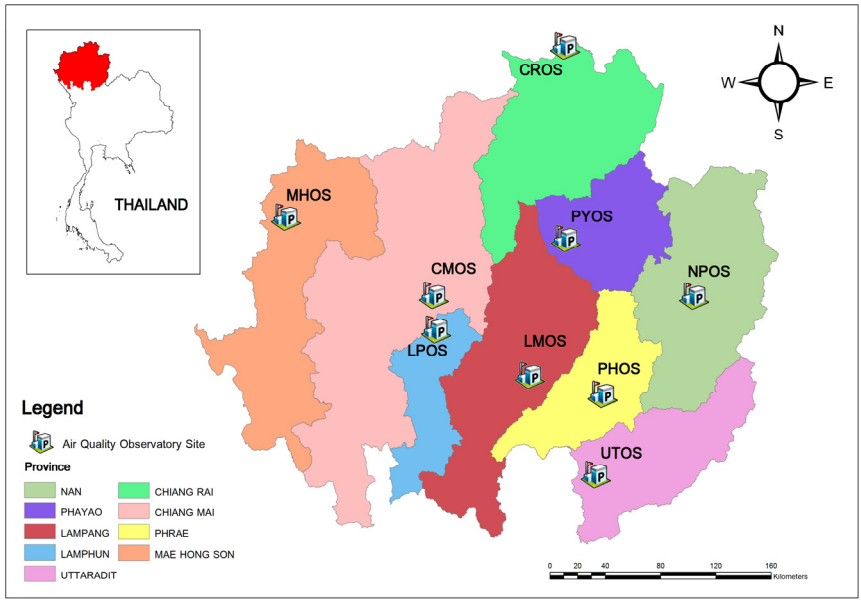

Figure 1: $PM_{2.5}$ monitoring stations in nine administrative provinces of Northern Thailand.

2.2 Sampling of $PM_{2.5}$

Samplings of $PM_{2.5}$ were conducted and collected every week on pre-baked (550°C for 12 h) quartz-fiber filters (QFFs; Whatman 47 mm; Article No. 28418542 (US reference)) with the assistance of MiniVolTM portable air samplers (Airmetrics) using the flow rate of 5 litres minute^{-1} through a particle size separator (impactor) and consequently through a 47 mm filter. More details of the $PM_{2.5}$ sampling method was provided in "EPA Quality Assurance Guidance Document: Method Compendium, Field Standard Operating Procedures for the $PM_{2.5}$ Performance Evaluation Program, United States Environmental Protection Agency Office of Air Quality Planning and Standards" (US-EPA, 2002). The weight measurement of $PM_{2.5}$ was strictly followed the instructions of US-EPA Quality Assurance Document: Method Compendium, $PM_{2.5}$ Mass Weighing Laboratory Standard Operating Procedures for the Performance Evaluation Program, United States Environmental Protection Agency Office of Air Quality Planning and Standards (US-EPA, 1998) with the employment of micro balances (Mettler Toledo, New Classic MF, MS205DU, Switzerland). The sampling campaigns can be separated into two groups according to the monitoring period during which they were conducted: *Campaign-I* was conducted before the "wildland fire episode" in the winter of 2012 (i.e. from the 7th to 22nd of December 2012), while air sampling during *Campaign-II* was done in March 2013 (i.e. from the 4th to 19th of March 2013).

Campaign-I (Nine Northern Provinces-I (NNP-I); sampling before the haze episode)
Observations of $PM_{2.5}$ at *cluster-I* sampling stations (CROS, PYOS and NPOS), *cluster-II* sampling stations (LMOS, PHOS and UTOS) and *cluster-III* sampling stations (MHOS, CMOS and LPOS) was conducted synchronously daily from the 28th of November to the 4th of December 2012, from the 7th to 13th of December 2012 and from the 16th to 22nd of December 2012, respectively.

Campaign-II (Nine Northern Provinces-II (NNP-II); sampling after the haze episode)
Air quality observations at *cluster-I* monitoring stations (CROS, PYOS and NPOS), *cluster-II* monitoring stations (LMOS, PHOS and UTOS) and *cluster-III* monitoring stations (MHOS, CMOS and LPOS) was performed synchronously daily from the 23th of February to the 2nd of March 2013, from the 4th to 11th of March 2013 and from the 13th to 20th of March 2013, respectively.

2.3 Analysis of PAHs

A mix standard solution of 13 native PAHs (Norwegian Standard (NS 9815: S-4008-100-T): phenanthrene (Phe), anthracene (An), fluoranthene (Fluo), pyrene (Pyr), benz[*a*]anthracene (B[*a*]A), chrysene (Chry), benzo[*b*]fluoranthene (B[*b*]F), benzo[*k*]fluoranthene (B[*k*]F), benzo[*a*]pyrene (B[*a*]P), benzo[*e*]pyrene (B[*e*]P), indeno[1,2,3-*c,d*]pyrene (Ind), dibenz[*a,h*]anthracene (D[*a,h*]A), and benzo[*g,h,i*]perylene (B[*g,h,i*]P) and a mix of recovery internal standard (IS) PAHs [d_{12}-perylene (d_{12}-Per) and d_{10}-fluorene (d_{10}-Fl)) were ordered from Chiron AS (Stiklestadveine 1, N-7041 Trondheim, Norway) and used for this study. The exceptionally low concentrations of PAHs in $PM_{2.5}$ need their pre-concentration through Soxhlet extraction and fractionization prior to their quantitative analysis on a gas chromatograph coupled to a mass spectrometer (Shimadzu GCMS-QP2010 Ultra system with 60 m length × 0.25 mm i.d. capillary column of Agilent JW Scientific DB- 5 GC columns). The analytical procedure used in this study and relevant information has been explained in previous publications [19–21].

3 Results and discussion

3.1 $PM_{2.5}$-bounded PAHs

The average mass concentrations of particulate PAHs (pPAHs, reported in pg m^{-3}) of $PM_{2.5}$ samples collected at NNP-I (i.e. the average values of CROS, PYOS, NPOS, LMOS, PHOS, UTOS, MHOS, CMOS, and LPOS before the haze episode), at NNP-II (i.e. the average values of CROS, PYOS, NPOS, LMOS, PHOS, UTOS, MHOS, CMOS, and LPOS after the haze episode) and at world cities (WCs) are given in Table 1, in terms of their mean values with the corresponding standard deviations.

Table 1: Statistical descriptions of PAHs (pg m^{-3}) in $PM_{2.5}$ collected from nine administrative provinces before (NNP-I) and in the middle (NNP-II) of biomass burning episode coupled with the average of world cities.

PAH Congener	NNP-I (n = 9)	NNP-II (n = 9)	WCs (n = 39)
Ph	182 ± 355	185 ± 465	1,484 ± 3,477
An	57 ± 68	18 ± 15	396 ± 1,073
Fluo	84 ± 184	81 ± 191	1,948 ± 2,395
Pyr	182 ± 467	260 ± 696	2,867 ± 2,845
B[a]A	461 ± 1,372	337 ± 990	1,088 ± 1,220
Chry	333 ± 986	478 ± 1,387	2,140 ± 2,618
B[*b*+*k*]F	13 ± 25	83 ± 142	5,363 ± 8,476
B[*e*]P	1,767 ± 3,246	967 ± 1,855	686 ± 1,555
B[*a*]P	17 ± 27	7 ± 22	1,794 ± 2,389
Ind	5 ± 14	7 ± 21	2,975 ± 5,096
D[*a,h*]A	ND	ND	909 ± 1,292
B[*g,h,i*]P	7 ± 20	8 ± 24	3,057 ± 4,777
B[*a*]P-Equivalent			
$B[a]P_{Nisbet\text{-}Lagoy}$ [22]	70 ± 144	55 ± 112	7,345 ± 8,812
$B[a]P_{US\text{-}EPA}$ [23]	46 ± 80	34 ± 61	3,018 ± 3,759
$B[a]P_{Cecinato}$ [24]	69 ± 143	55 ± 111	3,667 ± 4,456

The mean values of Fluo, Pyr, B[*b*+*k*]F, B[*a*]P, B[*g,h,i*]P, Σ3,4-rings PAHs and $B[a]P_{Equivalent}$ concentrations (i.e. $B[a]P_{Nisbet\text{-}Lagoy}$, $B[a]P_{US\text{-}EPA}$, $B[a]P_{Cecinato}$) at WCs (Fluo: 1,948 ± 2,395 pg m^{-3}, Pyr: 2,867 ± 2,845 pg m^{-3}, B[*b*+*k*]F: 5,363 ± 8,476 pg m^{-3}, B[*a*]P: 1,794 ± 2,389 pg m^{-3}, B[*g,h,i*]P: 3,057 ± 4,777 pg m^{-3}, Σ3,4-rings PAHs: 9,923 ± 13,628 pg m^{-3}, $B[a]P_{Nisbet\text{-}Lagoy}$: 7,345 ± 8,812 pg m^{-3}, $B[a]P_{US\text{-}EPA}$: 3,018 ± 3,759 pg m^{-3}, $B[a]P_{Cecinato}$: 3,667 ± 4,456 pg m^{-3}) were significantly ($p < 0.05$) much higher than those of samples collected at NNP-I and NNP-II (Table 1).

It is obvious that the measured PAH congeners in the present study fell within relatively low range, in comparison with other studies around the world (Table 2). Several factors such as unique tropical wet and dry climate (Köppen Aw) characterized by hot and rainy weather coupled with low industrial emission sources of northern provinces might have influenced the comparatively low PAH contents observed in NNP-I and NNP-II. While using the Σ3,4-rings PAHs level at NNP-I as the background level, the magnitude of Σ3,4-rings PAHs at the WCs increased by factors of 7.6, and that of $B[a]P_{Nisbet\text{-}Lagoy}$, $B[a]P_{US\text{-}EPA}$, and $B[a]P_{Cecinato}$ were roughly enhanced by factors of 105, 66, and 53, respectively. Previous studies have indicated that vehicular traffic was the main sources of $PM_{2.5}$ bounded PAHs in urban cities (Martellini *et al.* [25], Slezakova *et al.* [26, 27]). These findings underline the prominence of traffic emissions as the main contributors of $PM_{2.5}$ bounded PAHs, and it might have been exceeded numerous emission sources involving wild land fire and biomass burnings. As there were no significant differences detected at NNP-I and NNP-II (Table 1), it seems plausible to mention that the wild land fire episode plays a minor role on variations of $PM_{2.5}$ bounded PAHs in nine administrative provinces of Northern Thailand.

3.2 Ecological health risk assessment

In this study, non-dietary exposure is defined as human exposure to $PM_{2.5}$ bounded PAHs via both household air and dust. Table 2 displays the excess cancer risks associated with house dust exposure in NNP-I, NNP-II, and WCs, ranged from $3.732\times10^{-4}\pm7.929\times10^{-4}$ (NNP-II-$TEQ_{\text{US-EPA}}$ with ingestion rate of 50 mg day^{-1}) to 1.451 ± 1.741 (WCs-$TEQ_{\text{Nisbet-Lagoy}}$ with ingestion rate of 100 mg day^{-1}), depending on *TEQ* equations and ingestion rate (*IR*). The mean cancer risks of NNP were $8.525\times10^{-4}\pm3.493\times10^{-3}$ and $2.558\times10^{-3}\pm6.986\times10^{-3}$ for ingestion rate of 50 and 100 mg day^{-1}, respectively. The cancer risks of WCs for ingestion rate of 50 and 100 mg day^{-1} were greatly higher than those of NNP for 851 and 567 times in that order. Although the cancer risks of NNP were much lower than those of WCs, its average values ranged from 10^{-4} to 10^{-3} (i.e. one cancer incidence case per million), which can be considered as unacceptable cancer risk. These values are comparable to those of foundry workers (9.06×10^{-4} and 1.09×10^{-3}) in Taiwan [28], but higher than those predicted as occupational exposure for sinter metal workers (3.18×10^{-5} and 4.98×10^{-5}) and the Canadian maximum acceptable level of risk (1×10^{-5}). The current study indicates there is some concerns on adverse health impact related with $PM_{2.5}$ bounded PAHs exposure for preschool children via non-dietary exposure in home environment.

The highest average values of *TEQ* daily exposure doses via house dust ($M_{\text{PDI-ingestion}}$; ng day^{-1}) for adults and children was $7.345\times10^{4}\pm8.813\times10^{4}$ and $1.469\times10^{5}\pm1.763\times10^{5}$ ng day^{-1} under high exposure condition (with ingestion rate of 100 and 200 mg day^{-1} for adults and children) with the application of WCs-$TEQ_{\text{Nisbet-Lagoy}}$, while 4.715 ± 12.08 and 62.33 ± 159.8 ng day^{-1} under low exposure condition (4.16 and 55 mg day^{-1} for adults and children) with the employment of NNP-II-$TEQ_{\text{US-EPA}}$, respectively (Table 3). It is also important to note that $M_{\text{PDI-ingestion}}$ values of NNP-II-$TEQ_{\text{US-EPA}}$ were almost similar to those of Guangzhou for adults and children with the values of 4.31 and 57.0 ng day^{-1}, respectively. For *TEQs* daily exposure via inhalation ($M_{\text{PDI-inhalation}}$; ng day^{-1}), WCs-$M_{\text{-PDI-inhalation-Nisbet-Lagoy}}$ shows the highest values for both adults and children with the values of 147 ± 176 and 735 ± 881 ng day^{-1}, in that order (Table 4). Since the values of $M_{\text{PDI-ingestion}}$ were two to three orders of magnitude exceed those of $M_{\text{PDI-inhalation}}$, it would be safe to assume that dust ingestion was extremely crucial to non-dietary PAH exposure in comparison with $PM_{2.5}$ inhalation. These findings in NNP are consistent with those of Guangzhou, highlighting the importance of indoor air quality on long-term adverse respiratory diseases in Asian cities.

4 Conclusions

In this study, PAHs analysis in $PM_{2.5}$ collected in nine administrative provinces of Northern Thailand from December 2012 to March 2013 did not show any significant differences between wildland fire and non-wildland fire seasons. These findings underline the influences of traffic emissions as regular contributors of $PM_{2.5}$-bound PAHs. While the excess cancer risks of $PM_{2.5}$-bound PAHs in

Table 2: The cancer risks associated with non-dietary ingestion of PAHs in house dust for preschool children.

	NNP-I (n = 9)		**NNP-I (n = 9)**		**NNP-II (n = 9)**		**NNP-II (n = 9)**		**WCs (n = 39)**		**WCs (n = 39)**	
Ingestion rate (mg day^{-1})	**50**	**50**	**100**	**100**	**50**	**50**	**100**	**100**	**50**	**50**	**100**	**100**
	Aver	**Stdev**	**Aver**	**Stdev**	**Aver**	**Stdev**	**Aver**	**Stdev**	**Aver**	**Stdev**	**Aver**	**Stdev**
Cancer Risk -Nisbet-Lagoy	2.305 E-03	6.774 E-03	4.610 E-03	1.355 E-02	6.037 E-04	1.456 E-03	1.207 E-03	2.912 E-03	7.256 E-01	8.707 E-01	1.451 E+00	1.741 E+00
Cancer Risk -US-EPA	1.515 E-03	3.764 E-03	3.030 E-03	7.527 E-03	3.732 E-04	7.929 E-04	7.464 E-04	1.586 E-03	2.982 E-01	3.715 E-01	5.963 E-01	7.429 E-01
Cancer Risk -Cecinato	2.272 E-03	6.727 E-03	4.544 E-03	1.345 E-02	6.037 E-04	1.443 E-03	1.207 E-03	2.886 E-03	3.623 E-01	4.403 E-01	7.246 E-01	8.806 E-01

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Table 3: The calculation of TEQs daily exposure via ingestion ($M_{PDI\text{-}ingestion}$; ng day^{-1}).

	NNP-I (*n* = 9) Children		**NNP-I (*n* = 9) Children**		**NNP-I (*n* = 9) Adult**		**NNP-I (*n* = 9) Adult**	
M-DID (mg day^{-1})	**55 Aver**	**55 Stdev**	**200 Aver**	**200 Stdev**	**4.16 Aver**	**4.16 Stdev**	**100 Aver**	**100 Stdev**
M-PDI- ingestion-Nisbet-Lagoy	1.283E+02	3.771E+02	4.667E+02	1.371E+03	9.707E+00	2.853E+01	2.333E+02	6.857E+02
M-PDI- ingestion-US-EPA	8.433E+01	2.095E+02	3.067E+02	7.619E+02	6.379E+00	1.585E+01	1.533E+02	3.810E+02
M-PDI-ingestion-Cecinato	1.265E+02	3.745E+02	4.600E+02	1.362E+03	9.568E+00	2.833E+01	2.300E+02	6.810E+02
	NNP-II (*n* = 9) Children		**NNP-II (*n* = 9) Children**		**NNP-II (*n* = 9) Adult**		**NNP-II (*n* = 9) Adult**	
M-DID (mg day^{-1})	**55 Aver**	**55 Stdev**	**200 Aver**	**200 Stdev**	**4.16 Aver**	**4.16 Stdev**	**100 Aver**	**100 Stdev**
M-PDI- ingestion-Nisbet-Lagoy	1.008E+02	2.933E+02	3.667E+02	1.067E+03	7.627E+00	2.219E+01	1.833E+02	5.333E+02
M-PDI- ingestion-US-EPA	6.233E+01	1.598E+02	2.267E+02	5.810E+02	4.715E+00	1.208E+01	1.133E+02	2.905E+02
M-PDI-ingestion-Cecinato	1.008E+02	2.907E+02	3.667E+02	1.057E+03	7.627E+00	2.199E+01	1.833E+02	5.286E+02
	WCs (*n* = 39) Children		**WCs (*n* = 39) Children**		**WCs (*n* = 39) Adult**		**WCs (*n* = 39) Adult**	
M-DID (mg day^{-1})	**55 Aver**	**55 Stdev**	**200 Aver**	**200 Stdev**	**4.16 Aver**	**4.16 Stdev**	**100 Aver**	**100 Stdev**
M-PDI- ingestion-Nisbet-Lagoy	4.040E+04	4.847E+04	1.469E+05	1.763E+05	3.056E+03	3.666E+03	7.345E+04	8.813E+04
M-PDI- ingestion-US-EPA	1.660E+04	2.068E+04	6.036E+04	7.520E+04	1.255E+03	1.564E+03	3.018E+04	3.760E+04
M-PDI-ingestion-Cecinato	2.017E+04	2.451E+04	7.334E+04	8.914E+04	1.525E+03	1.854E+03	3.667E+04	4.457E+04

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Table 4: Calculation of *TEQs* daily exposure via inhalation ($M_{PDI\text{-}inhalation}$; ng day^{-1}).

	NNP-I (n = 9) Adult		**NNP-I (n = 9) Children**		**NNP-II (n = 9) Adult**		**NNP-II (n = 9) Children**		**WCs (n = 19) Adult**		**WCs (n = 19) Children**	
Inhalation rate (m^3)	**20**	**20**	**10**	**10**	**20**	**20**	**10**	**10**	**20**	**20**	**10**	**10**
	Aver	**Stdev**	**Aver**	**Stdev**	**Aver**	**Stdev**	**Aver**	**Stdev**	**Aver**	**Stdev**	**Aver**	**Stdev**
M-PDI-inhalation-Nisbet-Lagoy	1.400 E+00	2.880 E+00	7.000 E-01	1.440 E+00	1.100 E+00	2.240 E+00	5.500 E-01	1.120 E+00	1.469 E+02	1.763 E+02	7.345 E+01	8.813 E+01
M-PDI-inhalation-US-EPA	9.200 E-01	1.600 E+00	4.600 E-01	8.000 E-01	6.800 E-01	1.220 E+00	3.400 E-01	6.100 E-01	6.036 E+01	7.520 E+01	3.018 E+01	3.760 E+01
M-PDI-inhalation-Cecinato	1.380 E+00	2.860 E+00	6.900 E-01	1.430 E+00	1.100 E+00	2.220 E+00	5.500 E-01	1.110 E+00	7.334 E+01	8.914 E+01	3.667 E+01	4.457 E+01

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Northern Thailand were greatly lesser than those of other world cities, its average values ranged from 10^{-4} to 10^{-3}, which was higher than the Canadian maximum acceptable level of risk (1×10^{-5}) and other previous occupation exposure studies. There were also some concerns associated with $PM_{2.5}$ bounded PAHs exposure for preschool children via non-dietary exposure in home environment. The cancer risk exposure through inhalation appeared to play a minor role, whilst the direct ingestion contributed greatly on children as a consequence of their hand-to-mouth activity, indicating this route of exposure should not be underestimated.

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