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ORIGINAL PAPER

Profiling and potential cancer risk assessment on children exposed to PAHs in playground dust/soil: a comparative study on poured rubber surfaced and classical soil playgrounds in Seoul

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Abstract Children can get affected by polycyclic aromatic hydrocarbons (PAHs) while they interact with play area soil/rubber surfacing and exposed to PAHs by dermal contact, inhalation and hand-to-mouth activity. A comparative study has been conducted on PAHs profiling and probable cancer risk of children from PAHs present in uncovered playground surface soil and poured rubber surfaced playground dust. Surface soil and dust samples have been collected from 14 different children parks around the Korea University campus, Seoul, Republic of Korea. Concentrations of 16 PAHs in the soils/dust were found to be in a range of 2.82–57.93 μg g^{-1}. Profiling of the PAHs from the playground soils/dust reveals 3-ring PAHs are dominating with 79.9% of total PAHs content, on an average. The diagnostic ratio analysis confirms that vehicular exhaust and fossil fuel burning are likely the main sources of high molecular weight carcinogenic PAHs, whereas low molecular weight PAHs have pyrogenic origin. The probabilistic health risk assessment using Monte Carlo simulations for the estimation of the 95% cancer risk exposed to the PAHs from the surfaced playgrounds shows a little higher value than the USEPA safety standard (1.3×10^{-5}). Sensitivity analysis revealed exposure duration and relative skin adherence factor for soil as the most influential parameters of the assessment. Noticeably, cancer risk is approximately 10 times higher in poured rubber surfaced playgrounds than in uncovered soil playgrounds.

Keywords Poured rubber surfacing · Children playground · Monte Carlo simulation · Cancer risk assessment · Polycyclic aromatic hydrocarbons

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Introduction

With the recent expansion of childhood cancers, it is the second major driving reason for childhood death in South Korea. According to a recent report, a sum of 15,113 cancer patients of 0–14 years old were treated in the period of 1999–2011 in South Korea, and the age-normalized occurrence rate of all cancers was 144.0 and 124.9 per million for males and females, respectively (Park et al. 2016). The age-normalized incidence rate increased from 117.9 per million children in 1999 to 155.3 per million children in 2011, with an annual percentage increment of 2.4% (Ju et al. 2018). With these data in hand, it is evident that childhood cancer is a serious issue in South Korea.

As urban communities are winding up increasingly industrialized, polycyclic aromatic hydrocarbons (PAHs) contamination has turned into a major issue because of the expanding utilization of fossil fuel and industrial plants (Rajpara et al. 2017). PAHs present a critical hazard to the environment and human well-being as they are hydrophobic and sorbed to environmental matrices due to their low aqueous solvency (Tarafdar et al. 2017). The significant mechanisms of carcinogenesis initiated by PAHs are the association of PAHs with DNA to shape adducts and the formation of reactive oxygen species (epoxides). Both the actions result in detrimental effect on DNA and mutagenesis in imperative destinations of the genome. Various PAHs including chrysene, benzo[*a*]pyrene and fluoranthene offer ascent to metabolites that frame DNA adducts in animals (Munoz and Albores 2011). The International Agency for Research on Cancer (IARC) investigated some PAHs as potential human oncogenic agents, as short-term and long-term exposure to PAHs in laboratory tests caused hazardous/cancerous impacts on skin, lung, liver, urinary tract, hematopoietic, neurological, immune and reproductive systems of animals (IARC 2010). USEPA has considered sixteen PAHs as priority pollutants (USEPA 2014).

Soil is the most important reservoir of the human activity-generated PAHs. The high hydrophobicity and stable synthetic structure cause insolubility of PAHs, and subsequently, they can be sorbed to soil surfaces as well as soil organic matter (SOM) (Lasota and Błońska 2018). Again, many playgrounds in South Korea are poured rubber surfaced (PRS), which is basically consisted of shredded scrapped tires (rubber mulch) with a polyurethane cover layer. A few examinations about the hazard of the chemical substance of tire–rubber–reused items have been led previously (Ottesen et al. 2008; Kanematsu et al. 2009; Li et al. 2010). It is well reported that rubber tire materials contain aromatic extender oil and it has a PAHs content of 300–700 mg kg^{-1} (Aatmeeyata and Sharma 2010). And thus, the presence of PAHs in recycled rubber goods/playgrounds is being taken account of in several recent studies (Llompart et al. 2013; Celeiro et al. 2014; Marsili et al. 2014; Diekmann et al. 2018). Along this viewpoint, children could get affected by PAHs while they interact with play area surfacing and exposed to PAHs by dermal contact, inhalation and hand-to-mouth activity (soil ingestion) (Islam et al. 2018).

In the present study, we measured PAHs content in dust/surface soil from various children playgrounds in Seoul and arranged a statistical simulation on potential cancer risk of children exposed to them. Eventually, we compared the probable risk scenarios between the poured rubber surfaced and classical soil playgrounds.

Materials and methods

Sample collection

Surface soil and dust samples from 14 different children parks around the Korea University campus (37°35′18.2″N 127°01′43.4″E), Seoul, Republic of Korea, have been collected for the detection of PAHs content. The children park facilities for sampling were chosen in a manner that almost all of them reside beside prominent roads with heavy traffic loads (Fig. 1). Samples were collected during August–September 2018 using a brush (made of natural/organic material) and a stainless steel gimlet up to a depth of 1–5 cm for uncovered playgrounds and surface dusts for poured rubber surfaced playgrounds.

Poured rubber surfacing (PRS) is a consistent elastic/rubber surface made of two layers that is poured in place (PIP). The principal layer, or "wear layer," is normally 3/8" thick and made of EPDM (ethylene propylene diene monomer rubber), SBR (styrene-butadiene rubber) or TPV (thermoplastic vulcanizate) granules. The second layer, or "cushion layer," is 1–5 inches thick and made of shredded rubber or reused tires. The thickness of the surface relies on utilization and play gear with 15–20 mm for hard surfaces and 40 mm for packed stone. The crude rubber particles are bound together with a polyurethane cover and blended or made nearby. The surface arrives in an extensive variety of hues and even blends of colors. Many children playgrounds in Seoul have this kind of surfaced facility for physical safety.

The sampling procedure was based on the sampling guidance by Environmental Hazards Assessment

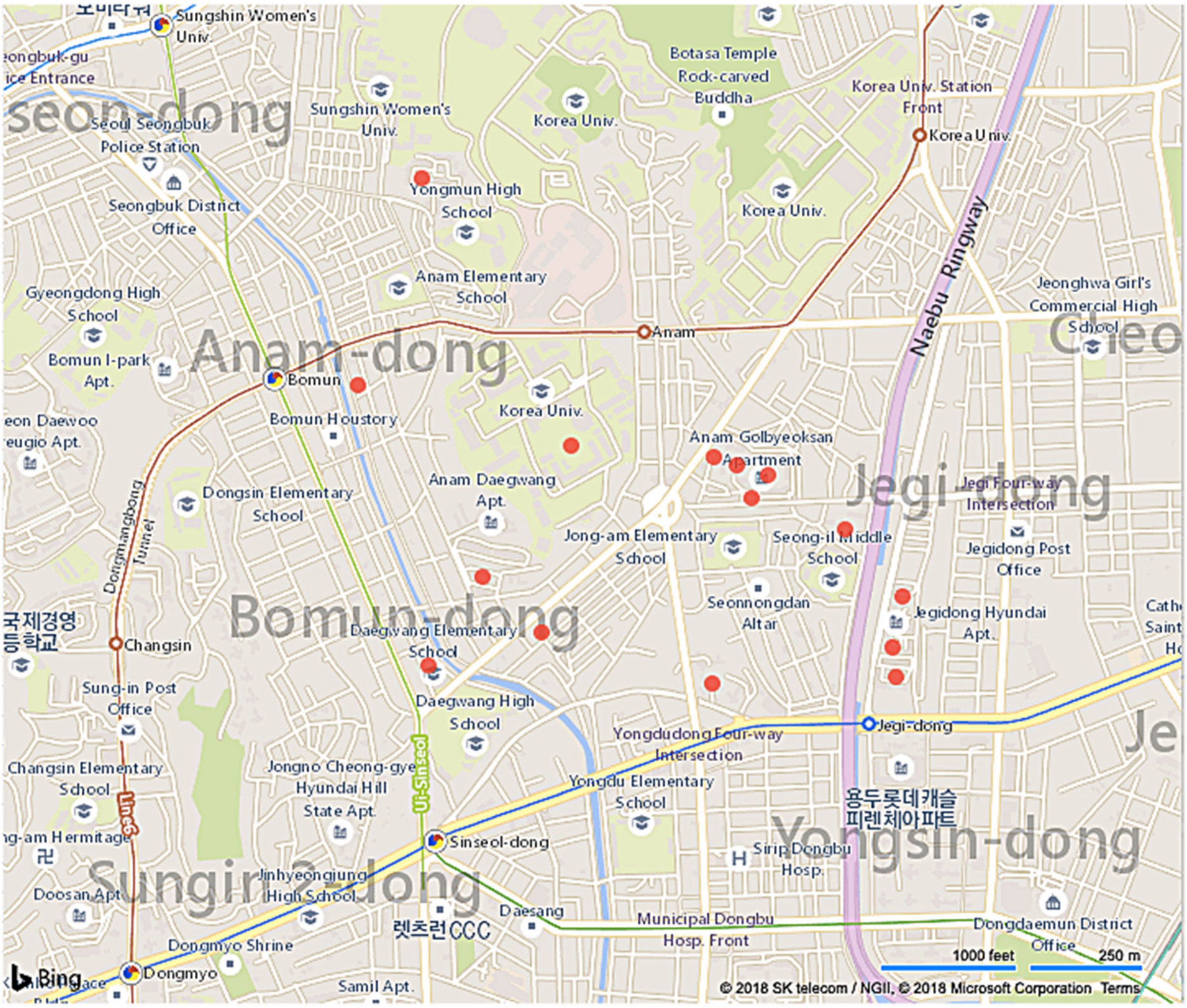


Fig. 1 Red dots represent dust/soil sampling children playground sites around Korea University, Seoul, South Korea

Program (EHAP), Department of Pesticide Regulation (DPR), California Environmental Protection Agency (Cal/EPA) (Sava 1994). From each of the sampling points/playgrounds, 8 surface soil/dust samples of approximately 10 g each (total 112 samples) were collected in amber glass vials. Samples were sieved through 2-mm sieve, pooled and homogenized to form a composite sample to represent each playground, and preserved in dark at 4 °C. Microscopic study revealed 658 μm of mean particle size of the samples. A surface soil was also collected from Hana Square of Korea University as a background control (Sample no. 15) because this particular area is surrounded by university buildings, which may resist the PAHs contamination via air, and this place is also well partitioned from the frequent vehicular tracks.

Chemicals

All organic solvents (acetone, *n*-hexane, acetonitrile, etc.) used for sample processing and analysis were of HPLC grade (J.T. Baker [Center Valley, PA, USA] and Daejung [Siheung, Republic of Korea]). Standard mixture (TCL PAH Mix) containing 16 PAHs was purchased from Supelco (product code CRM48905, Bellefonte, PA, USA). Milli-Q water was utilized to execute the diagnostic system.

Sample extraction

The dichloromethane/acetone (DCM/Ace) sonication technique (US EPA method 3550) was used for sample extraction. Extraction was encouraged by two sonications rehashed three times. This strategy has been utilized previously and shows high recuperation rates for PAHs including benzo[*a*]pyrene (BaP) (93–100%) (Contreras-Ramos et al. 2008). 1.5 g soil blended with 3 g anhydrous sodium sulfate and extricated three times utilizing 10 mL extractant (DCM/Ace at 1:1 ratio, *v/v*). Each extraction sonicated two times (40 kHz, 15 min) with vortex mixing before every sonication. Extractants were isolated by centrifugation at 3452 g for 20 min. The DCM/Ace concentrates were consolidated and dissipated under delicate nitrogen gas stream and re-dissolved in 2 mL acetonitrile pursued by filtration (0.45 µm PTFE non-sterile disposable membrane filter unit, Dismic-13HP, Advantec, Japan) into 2-mL HPLC vials (Duan et al. 2016).

The firmly significant connection between DCM-/Ace-extractable BaP and bioavailability studied by Duan et al. (2014), using a swine model, recommends that this extractant could be utilized for surveying BaP bioavailability for humans in sandy soils. The microscopic characterization of the collected samples matches the criteria of sandy type soil (Supplementary data 1).

Analytical procedure for HPLC

The Waters® ACQUITY UltraPerformance LC® (UPLC®) system outfitted with PDA eλ and FLR detectors (Waters, Milford, MA, USA) were utilized to examine all the samples. An ACQUITY UPLC column (2.1 × 100 mm, 1.7 micron BEH C18) was used for the separation of the PAHs. A water: acetonitrile gradient from 66 to 90% acetonitrile at a flow rate of 0.4 mL/min was used for 7 min of runtime, and the injection volume was 5 µL (Benvenuti 2007). Among the four-channel detection, Ex 275, Em 350 used for benzo[*a*]anthracene, chrysene, fluorene; Ex 330, Em 420 used for benzo[*ghi*]perylene, dibenz[*a,h*]anthracene, pyrene; Ex 375, Em 460 used for benzo[*a*]pyrene, benzo[*k*]fluoranthene, benzo[*b*]fluoranthene, fluoranthene and rest of the PAHs are detected using the PDA eλ detector at 254 nm. External standard calibration curves of five different concentrations were utilized for measurement of the concentrates. Peak area responses and calibration factors were utilized to process the concentration of each compound. The retention time of each detected peak and signal-to-noise ratio (USP s/n) calculated by Empower 3 software (Waters) are given in the Supplementary data 2.

Analytical strategies were inspected for the exactness and precision. Triplicate examinations of the samples gave an error between ± 10% and ± 15%. Intermittent benchmark standards were run to guarantee the exactness and precision of results. The recovery efficiency was inspected by soil tests spiked with known concentration of PAHs standard. The mean esteem of spiking recovery for 16 PAHs was 97.2%.

Risk assessment

The cancer-causing intensity of all the PAHs recognized can be assessed as the total of every individual BaP potency equivalent (BaP_{eq}) (Nisbet and LaGoy 1992; Tsai et al. 2004). A probabilistic risk assessment (PRA) model (exhibit 3–6, USEPA 2001) has been used in the current study, where a probability distribution of the parameter values (geometric means and geometric standard deviations) has been considered, rather than a single (mean) value. The chronic day-by-day admission of PAHs indicates the PAHs in soil particles received by the exposure end (i.e., respiratory organs and dermal layer) and is meant by chronic daily intake (CDI) (Yang et al. 2014). The CDI of PAHs in other medium was not considered as this investigation just spotlights on the PAHs conveyed by playground soil/dust particles. The CDI (mg kg^{-1} day^{-1}) of PAHs via soil dust was assessed using formulae as follows:

$$\mathrm{CDI_{ingestion}} = \frac{C_{\mathrm{soil}} \times \mathrm{IR_{soil}} \times \mathrm{CF} \times \mathrm{ED} \times \mathrm{EF}}{\mathrm{BW} \times \mathrm{AT}} \tag{1}$$

$$\mathrm{CDI_{inhalation}} = \frac{C_{\mathrm{soil}} \times \mathrm{HR} \times \mathrm{ED} \times \mathrm{EF} \times \mathrm{ET}}{\mathrm{PEF_{soil}} \times \mathrm{BW} \times \mathrm{AT}} \tag{2}$$

$$\mathrm{CDI_{dermalcontact}} = \frac{C_{\mathrm{Soil}} \times \mathrm{CF} \times \mathrm{SA} \times \mathrm{AF} \times \mathrm{ABS} \times \mathrm{ED} \times \mathrm{EF}}{\mathrm{BW} \times \mathrm{AT}} \tag{3}$$

where $CDI_{ingestion}$ is the chronic daily intake linked to soil particle ingestion (mg kg^{-1} day^{-1}), C_{soil} symbolizes the sum of transformed PAHs concentrations in soil based on toxic equivalents of BaP (mg kg^{-1}), IR_{soil} is the short form of soil ingestion rate (mg day^{-1}), EF stands for exposure frequency (day a^{-1}), ED is the abbreviation of exposure duration (a), ET is exposure time for receptor (= 1 h/day), BW denotes body weight of the exposed individual (kg), AT is an averaging time of 365 day $year^{-1}$ for 81.41 years lifetime exposure of cancer risk, CF implies the conversion factor (1×10^{-6} kg mg^{-1}); $CDI_{inhalation}$ symbolizes the ceaseless regular intake via inhalation of soil particles (mg kg^{-1} day^{-1}), HR is the expression for the rate of air inhalation (m^3 day^{-1}), PEF_{soil} stands for soil particle emission factor; $CDI_{dermal\ contact}$ signifies the chronic daily intake for dermal contact of soil (mg kg^{-1} day^{-1}), SA is the abbreviation of surface area of the skin that directly or indirectly contacts soil (cm^2 day^{-1}) (assumed that exposed skin surface is limited to head, hands and forearms), AF is respective adherence factor of soil for skin (mg cm^{-2}) and ABS denotes the fraction for dermal absorption.

The values of the variables are collected from the literature and are provided in Supplementary data 3. The risks of cancer (r) were determined by multiplying the corresponding chronic daily intakes with their cancer potency factor (USEPA 1989).

$$r_x = CDI_x \times CSF_x \quad (4)$$

x in subscripts stands for repeating the equation for ingestion, inhalation and dermal contact chronologically.

The total cancer risk of the children (R_{soil}) calculated is the aggregation of the risks of the different routes of PAHs exposure (Chiang et al. 2009):

$$R_{soil} = r_{ingestion} + r_{inhalation} + r_{dermal\ contact} \quad (5)$$

The sensitivity study positions the assumptions/parameters from the most crucial to the slightest essential in the model. On the off chance that an assumption and a forecast have a high correlation coefficient, it implies that the assumption significantly affects the estimate (through both, its vulnerability and its model sensitivity).

To measure and limit the vulnerabilities of the above hazard estimations, Monte Carlo simulation was applied to assess the cancer risk. The Monte Carlo simulation (MCS) is a statistical tool in which the input variables to a condition are fluctuated at the same time and the yield is probabilistically displayed (Liang et al. 2013). An aggregate of 50,000 iterations were considered in the wake of guaranteeing numerical stability. After a certain large number of iterations, the results do not show any change in risk esteems. The MCS assessment and sensitivity investigation were actualized utilizing Crystal Ball (11.1.2.4.600) software from Oracle (CA, USA). The upper 95% limits of probable risk evaluation were produced by the same software. As a matter of course, Crystal Ball attempts to fit a normal distribution to the forecast esteems. One can enter a significance level to indicate the threshold beneath which the presumption of normality is rejected. The default dimension of 0.05 converts into a 95% certainty that a dismissal of normality will be corrected. Crystal Ball also ascertains sensitivity by figuring rank connection coefficients between each assumption and each forecast while the simulation is running. Correlation coefficients give a significant proportion of how much assumptions and forecasts change together.

Results and discussion

Profiling and diagnostic analysis of PAHs

Results of evaluated PAHs are shown as the arithmetic means of triplicated samples in Table 1. Total detected PAHs in the samples were in a range of 2.82–57.93 µg g^{-1} (arithmetic mean of 13.07 µg g^{-1}). The range of the detected total carcinogenic PAHs (CPAHs) was between 0.10 and 2.27 µg g^{-1} (arithmetic mean of 0.79 µg g^{-1}), and the ratio of CPAHs to total PAHs was noticeably lower, only 6.0% (on an average) of carcinogenic PAHs in the soil samples. Among all the sampling sites, no. 4, 6, 8, 10 and 14 were uncovered playgrounds. It is especially notable that the range of total PAHs in uncovered playground is 2.82–6.46 µg g^{-1} (mean 4.18 µg g^{-1}), whereas the range of total PAHs in the playgrounds covered with poured rubber surfacing granules is 4.91–57.93 µg g^{-1} (mean 18.01 µg g^{-1}), approximately 4–5 times higher than total PAHs content in covered playgrounds. The reason behind this high

Table 1 Concentration (mean) of each of the PAHs (μg g^{-1}) in surface soil/dust collected from corresponding sampling sites

	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15
Naphthalene	4.229	0.758	1.063	0.476	9.057	0.191	2.061	0.733	3.258	0.374	1.178	0.449	0.569	0.576	BDL
Acenaphthylene	5.095	0.741	2.647	1.165	19.700	0.170	BDL	0.425	3.790	0.647	1.286	0.717	2.844	0.675	0.162
Fluorene	0.100	0.122	0.044	0.065	2.674	0.037	0.884	0.127	1.159	0.024	0.041	0.101	0.901	0.098	0.060
Acenaphthene	2.377	0.529	0.478	0.249	5.567	0.588	1.191	1.379	1.656	0.945	1.750	0.667	3.649	0.982	0.728
Phenanthrene	0.358	0.445	0.208	0.161	0.732	0.108	0.498	0.356	2.070	0.387	0.356	0.393	1.116	0.155	0.162
Anthracene	4.793	5.890	2.771	0.496	19.321	2.029	8.817	0.742	10.828	1.619	0.769	1.122	8.030	3.646	0.432
Fluoranthene	0.041	0.039	0.003	BDL	0.026	BDL	0.032	0.009	0.182	0.006	0.006	0.007	0.026	0.027	0.004
Pyrene	0.004	0.009	BDL	BDL	BDL	BDL	0.014	0.003	0.039	BDL	0.004	0.012	0.006	0.003	BDL
Chrysene	0.294	0.353	0.065	0.090	0.623	0.032	0.914	0.437	1.208	0.050	0.087	0.965	0.423	0.149	0.040
Benzo[*a*]anthracene	0.007	0.181	0.003	0.013	0.068	0.010	0.243	0.025	0.150	0.034	0.051	0.056	0.123	0.024	0.003
Benzo[*b*]fluoranthene	0.039	0.043	0.007	0.004	0.035	0.026	0.217	0.034	0.117	0.009	0.111	0.121	0.035	0.014	BDL
Benzo[*k*]fluoranthene	0.013	0.027	0.007	0.003	0.035	0.008	0.178	0.008	0.039	BDL	0.049	0.031	0.008	0.004	BDL
Benzo[*a*]pyrene	0.011	0.026	0.003	0.003	0.014	0.016	0.171	0.010	0.039	BDL	0.048	0.048	0.008	BDL	BDL
Dibenzo[*a,h*]anthracene	0.009	0.023	0.003	0.004	0.006	BDL	0.244	0.004	0.034	BDL	0.064	0.046	0.026	0.004	BDL
Indeno[1,2,3-*cd*]pyrene	0.033	0.064	0.008	0.004	0.038	BDL	0.018	0.016	0.044	0.008	BDL	0.050	0.017	BDL	BDL
Benzo[*ghi*]perylene	0.392	0.092	BDL	0.092	0.039	0.014	0.286	BDL	0.351	BDL	0.111	0.124	0.374	0.103	0.069
Total PAHs	17.794	9.340	7.311	2.824	57.935	3.227	15.769	4.308	24.966	4.102	5.911	4.910	18.155	6.458	1.660
Total CPAH	0.798	0.808	0.096	0.212	0.857	0.105	2.272	0.534	1.982	0.101	0.520	1.442	1.014	0.298	0.112

BDL below detection limits

concentration of PAHs in polyurethane rubber surfaced playgrounds can be leaching of PAHs from recycled rubber tires (Llompart et al. 2013; Celeiro et al. 2014) and/or sorption of atmospheric PAHs to polyurethane that is frequently used for passive air sampler for occupational PAHs measurement (Strandberg et al. 2018).

As per our pre-assumption, the chosen background sampling site no. 15 (Hana Square, Korea university) showed the lowest concentration of total PAHs (1.66 μg g^{-1}). Ongoing road construction work between sampling locations no. 5 and 9 can be the additional reason for unusually high level of total PAHs content in the samples collected from there. The uncovered playground of sampling location no. 4 was well partitioned from high traffic road by apartment buildings, which can explain the lowest concentration of PAHs of all the sampling sites.

Here, we produced a diagram of spatial apportionment of the PAHs over the testing region (Fig. 2a) and the ternary plot of the composition profile of PAHs by ring number (Fig. 2b).

It is perceptible from the profiling data based on the ring number of PAHs that the sampling area is hugely dominated by 3-ring PAHs (79.9% of total PAHs content, on an average), followed by 2-ring (11.9%) and 4-ring PAHs (5.0%). We detected only 1.5% of 5 rings and 1.7% of 6 rings in the total PAHs content.

The procedure creating the PAHs in a source straightforwardly impacts the profile (Manoli et al. 2004). There are various diagnostic ratios of PAHs to track down the source of the PAHs whether it is petrogenic or pyrogenic. At any time, when source apportionment investigation of PAHs is done, it is accepted that proportions from source to receptors stay steady (Wang et al. 2010). As for the mass 178, anthracene-to-anthracene plus phenanthrene ratio [Ant/(Ant + Phe)] lower than 0.1 signifies a prediction of petroleum ancestry, whereas this ratio greater than 0.1 suggests a consequence of combustion. For mass 202, fluoranthene to fluoranthene plus pyrene [Fl/(Fl + Py)] proportion of 0.5 is edge limit for petroleum/combustion transition point. The proportions somewhere in the range of 0.4 and 0.5 were characterized as fluid petroleum derivative burning, though if the proportion > 0.5, it was described as that of biomass, wood or coal (Chawda et al. 2017). For benzo[*a*]anthracene to benzo[*a*]anthracene plus chrysene [BaA/(BaA + Chry)], proportion < 0.2 suggests petroleum combustion and the proportion > 0.2 shows pyrogenic or coal combustion of the source materials. Likewise, indeno[1,2,3-*cd*]pyrene to indeno[1,2,3-*cd*]pyrene plus benzo[*ghi*]perylene [IP/(IP + BghiP)] proportions < 0.2 likely imply petrogenic, somewhere in the range of 0.2 and 0.5 petroleum burning (vehicle and unrefined petroleum), and proportions > 0.5 demonstrate biomass, softwood and coal ignition (Yunker et al. 2002).

From Fig. 3 showing diagnostic ratio analysis of our samples, it is evident that around mass 178 (anthracene) and mass 202 (fluoranthene), PAHs are thought to be of pyrogenic/coal combustion/softwood biomass combustion origin. This signifies that lower and mid-ranged molecular weight PAHs (2–4 rings) have more prominent pyrogenic source. Noticeably, the measure of natural gas (NG) utilization in Seoul has been rapidly expanded since 1990 as the Ministry of Environment (MOE) in Korea has constrained the utilization of solid fuel (coals) for cooking in the Seoul metropolitan zones since 1985 and emphatically authorized the standard since 1995 (Lee and Kim 2007). Although it needs further investigations, there are chances that some of these PAHs can be of distanced origin.

It is well known that LMW-PAHs are highly volatile mixes staying as gases in media, while HMW-PAHs are low volatile compounds aggregated into or adsorbed onto particles (Mader and Pankow 2002). The hastier dispersion of LMW-PAHs voyaging all around effectively is thus more noticeable than HMW-PAHs. Kaya et al. (2012) suggest that air and soil PAHs densities are connected fundamentally hinting an active interaction of these compartments. Fugacity calculations in their study also advocate that atmospheric deposition is the dominant source for PAHs quantified in soils. It can be perceived from the above that the different origins of LMW–HMW-PAHs and higher concentrations of LMW-PAHs in playground dust might be because of the long-range transport of some of the PAHs (Lee and Kim 2007). However, relatively high density of 3-ring PAHs also suggests that their source can be the playground rubber surfacing (Llompart et al. 2013).

Again, for relatively heavier molecular weight PAHs, the values of the ratio for BaA/(BaA + Chry) are dispersed around 0.2 (mean 1.67), which confirms the petrogenic and pyrogenic both the origins of these PAHs. As for IP/(IP + B(ghi)P), all ratios are strictly

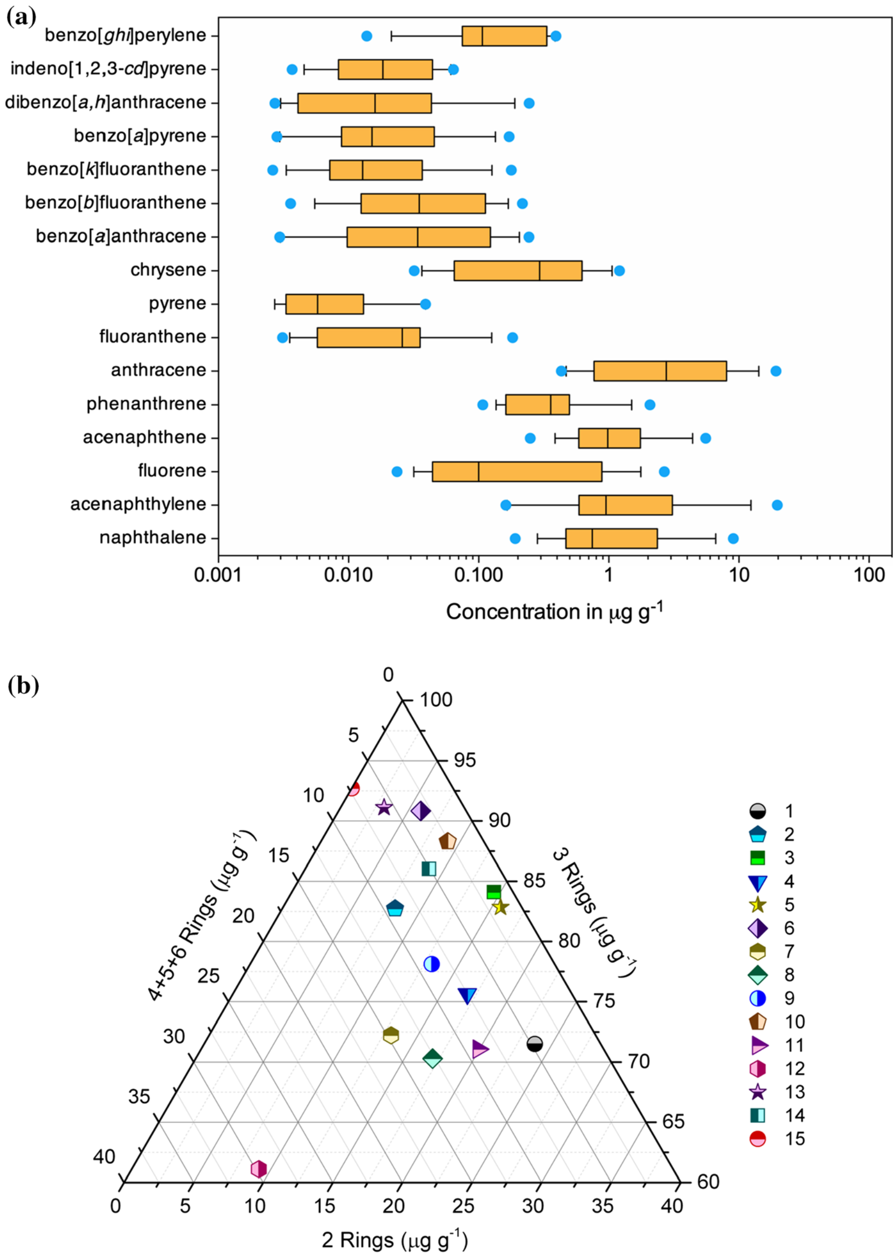


Fig. 2 **a** Distribution of individual PAHs in different sampling points depicted in box and whiskers plot in logarithmic scale base 10. Each box represents the lower and upper quartiles; the band within the box signifies the median value while whiskers denote the minimum and maximum values. **b** Ternary plot of PAHs composition based on ring numbers present at sampling playgrounds

below 0.5 (mean 0.20) with a significance of petrogenic or fuel combustion origin. The diagnostic ratio analysis proclaims that vehicular pollution and petroleum burning are the main source of high molecular weight CPAHs in the dust/soil samples from children playgrounds in Seoul. Increasing number of vehicles in South Korea can be an important contributor for CPAHs, as throughout the previous 20 years, vehicle increment was 137.5% while population increment was only 15.2% (Park 2015).

Cancer risk simulation

The total BaP_{eq} concentration of the dust/soil samples from playground was calculated and used for cancer risk assessment of children. The geometric mean of the BaP_{eq} in uncovered playgrounds was 0.04 μg g^{-1}

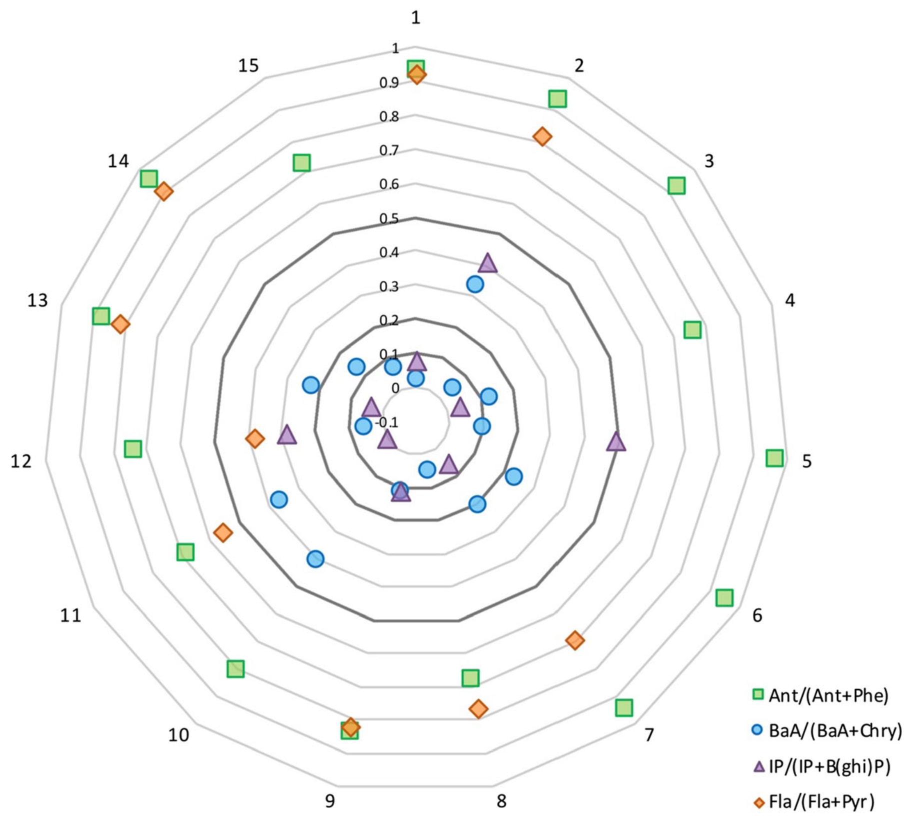


Fig. 3 Distribution of various diagnostic ratios: the radar plot. A value of > 0.1 for Ant/(Ant + Phe) and a value of > 0.5 for Fla/(Fla + Pyr) signify pyrolytic or coal combustion origin of relatively LMW-PAHs; a value of < 0.2 for BaA/(BaA + Chry) and a value of < 0.5 for IP/(IP + B(ghi)P) suggest petrogenic source of the HMW-PAHs

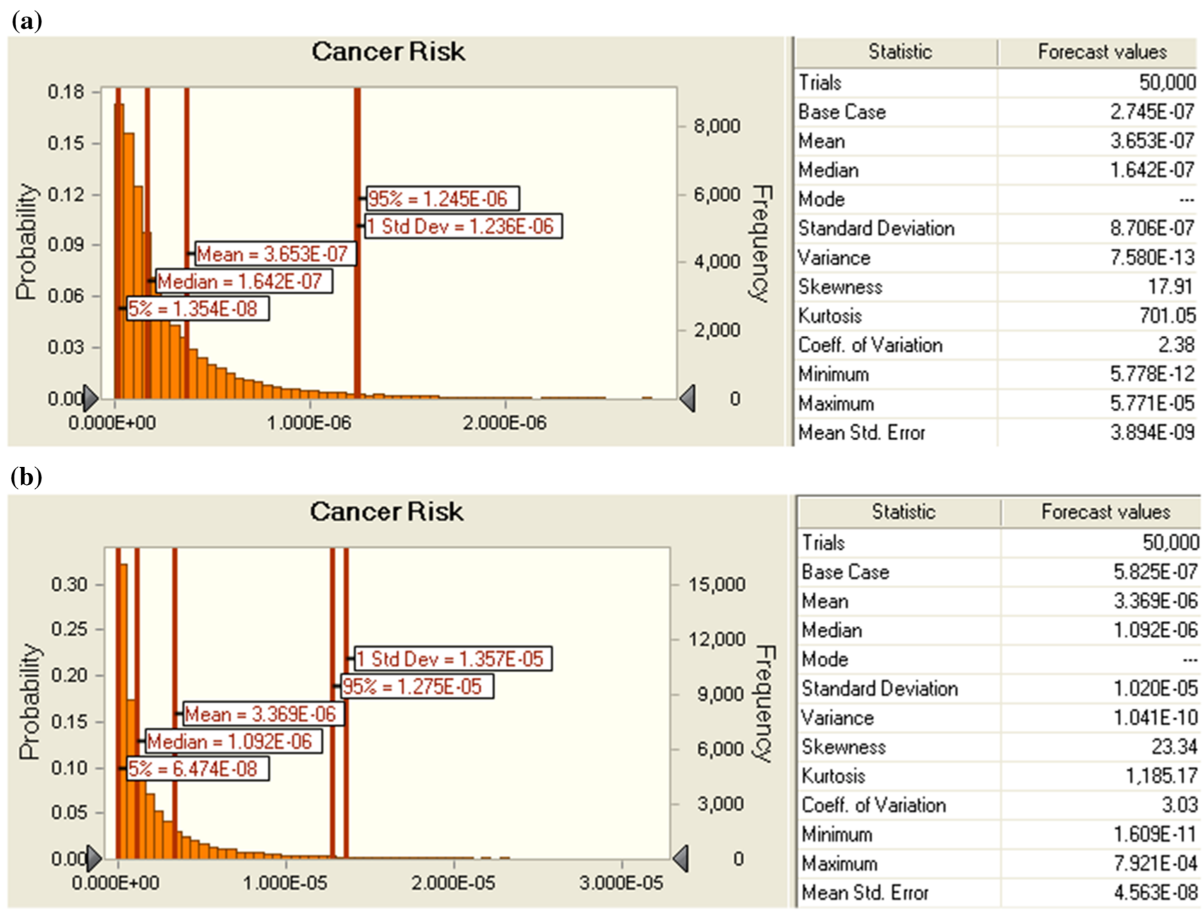
with the geometric standard deviation of 1.46. On the contrary, the covered playgrounds showed higher BaP_{eq} potential of 0.28 μg g^{-1} (geometric mean) with a geometric standard deviation of 2.49, which is 7 times higher than in uncovered playgrounds.

As shown in Fig. 4, the 95% probable cancer risk value for children from PAHs present in uncovered playground surface soil is 1.2×10^{-6} and the mean value is 3.7×10^{-7}. The 95% probable cancer risk due to PAHs present in the dust collected from poured rubber surfaced playgrounds is 1.3×10^{-5} with a mean value of 3.4×10^{-6}.

New York State Department of Health depicted quantitative portrayal of lifetime cancer risks as the esteem $\leq 10^{-6}$ signifies effective harmlessness, estimation of 10^{-6}–10^{-4} demonstrates a low plausible hazard, a risk appraise 10^{-4}–10^{-3} proposes a moderately potential threat, and 10^{-3}–10^{-1} is highly periled and very high when the value is $\geq 10^{-1}$ (NYS DOH 2012). Responding to the 1987 section 112 Clean Air Act decision (Natural Resources Defense Council v. U.S. Environmental Protection Agency 824 F. 2nd 1146 [1987]), EPA came up with the declaration that they would take the administrative steps on risk simulations utilizing the universal strategy that the lifetime additional cancer risk of the person with most exposure of one in ten thousand (1×10^{-4}) might comprise an adequate risk and that the edge of safety entailed by rule and fortified by the court ought to lessen the hazard for the best number of people to an additional lifetime hazard of close to one in million (1×10^{-6}) (Tarafdar and Sinha 2017a).

Although the simulated cancer risk from uncovered playgrounds is beyond (marginally) the crisis zone, 95% cancer risk from poured rubber surfaced playgrounds are a little above the tolerance safety margin according to USEPA. Noticeably, cancer risk is approximately 10 times higher in poured rubber surfaced playgrounds than in uncovered soil playgrounds.

Uncertainty investigations are required when there is no/less understanding from the earlier learning about uncertainty in the risk estimation and when quite possibly the inability to survey uncertainty may influence the choice of wrong options for risk reduction (Hammonds et al. 1994). Though Monte Carlo



(a)

Statistic	Forecast values
Trials	50,000
Base Case	2.745E-07
Mean	3.653E-07
Median	1.642E-07
Mode	---
Standard Deviation	8.706E-07
Variance	7.580E-13
Skewness	17.91
Kurtosis	701.05
Coeff. of Variation	2.38
Minimum	5.778E-12
Maximum	5.771E-05
Mean Std. Error	3.894E-09

(b)

Statistic	Forecast values
Trials	50,000
Base Case	5.825E-07
Mean	3.369E-06
Median	1.092E-06
Mode	---
Standard Deviation	1.020E-05
Variance	1.041E-10
Skewness	23.34
Kurtosis	1,185.17
Coeff. of Variation	3.03
Minimum	1.609E-11
Maximum	7.921E-04
Mean Std. Error	4.563E-08

Fig. 4 Probability density functions of cancer risk predicted for children from PAHs content of **a** uncovered and **b** poured rubber surfaced playgrounds

simulation has been used to lessen uncertainty, still some of it prevail in the present risk assessment process.

The BaP_{eq} values were received from analyses on rodents and not human, so they fluctuate after testing through various exposure courses. For instance, we can state that the BaP_{eq} value used in this study for environmental exposures of DBA is 5 (Nisbet and LaGoy 1992). This value may overestimate the true potency of this compound in the assessment, because another study claims a BaP_{eq} of 1.0 (Tsai et al. 2004). Once more, certain measure of uncertainty dependably connects with assessment of the toxicity of PAHs as a result of the endless number of conceivable PAHs blends and constrained dose–response information on cancer-causing nature. The probability distributions of exposure parameters like HR and SA were straightforwardly taken from USEPA prescribed qualities. These probably will not be an exact match to the Korean situation, making them uncertain parameters of the investigation.

Thus, our risk assessment contains both uncertainty and variability, and they must be treated individually. Separation of these concepts in the simulation leads us to more accurately detect the variation in the forecast due to lack of knowledge and the variation caused by natural variability. The 2D Simulation tool of Crystal Ball software runs an outer loop to simulate the uncertainty values and then freezes the uncertainty values while it runs an inner loop (of the whole model) to simulate the variability. We repeated this process for five hundred times of outer simulations, having a portrait of how the forecast distribution varies due to the uncertainty (Supplementary data 4).

Comparing the two-dimensional simulation to the previously done one-dimensional simulation (with both uncertainty and variability comingling together), we get that the 95th percentile risks in two-dimensional study (1.06×10^{-6} for uncovered and 1.15×10^{-5} for covered playgrounds) are negligibly lower than the 95th percentile risks of one-dimensional simulations (1.25×10^{-6} for uncovered and 1.28×10^{-5} for covered playgrounds—Fig. 4).

Contribution of various exposure routes

Hazard examination for the individual pathways of exposure (i.e., ingestion, inhalation and dermal contact) at 95% certainty level has been figured utilizing Monte Carlo simulation, and the near perspective of the outcome is portrayed in Fig. 5. As the contrast between dermal contact and inhalation course is more than enough high to plot in an immediate scale, we utilized logarithmic scale with base 10.

The cancer risk levels by means of soil ingestion were inside a similar request of magnitude (10^{-6}–10^{-7}) as through dermal contact exposure, showing that both ingestion and dermal contact extraordinarily added to the total risk. For both kinds of playgrounds (*i.e.,* covered and uncovered), dermal contact is the main mode of PAHs exposure with an exposure load of 72% for covered and 75% for uncovered playgrounds. Rest of the exposure occurs through the soil ingestion pathway (28% for covered and 25% for uncovered playgrounds). Risk estimation via dermal contact pathway is 9.4×10^{-6} for covered and 1.0×10^{-6} for uncovered playgrounds, while risk via soil ingestion is 3.7×10^{-6} for covered and 3.4×10^{-7} for uncovered playgrounds. Risk via inhalation exposure pathway is too small to be taken account for (9.2×10^{-11} for covered and 6.3×10^{-12} for uncovered playgrounds). The risk estimation of direct soil ingestion for kids is usually higher than that of the adults (Tarafdar and Sinha 2018) (mean 2.0×10^{-6}, for both kinds of playgrounds in the current study) which can be supported by their incessant hand-to-mouth movement.

Analysis of the parameter's sensitivity

A quantitative sensitivity investigation was led as contribution to variance, outlining the info parameters positioned by impact on probable yield risk (Fig. 6).

The tornado plots gave both the magnitudes (positive or negative) of the correlation. In this study, the exposure duration (ED) assumption has the greatest impact (48.7%) on the uncertainty of the overall risk outcome, followed by the respective adherence factor of soil for skin (AF) assumption (26.3%). Body weight of children (BW) is the third most important parameter of the assessment with − 15.2% of total load (negative value indicates the increase in the assumption is associated with a decrement in the forecast). ED and AF remained reliably critical patrons in some past investigations (Chen and Liao 2006; Tarafdar and Sinha

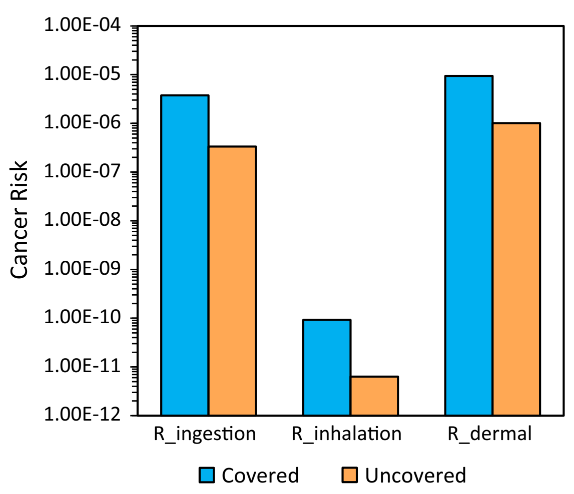


Fig. 5 Contributions of different exposure pathways to the total cancer risk (95% confidence values) represented in a logarithmic scale base 10

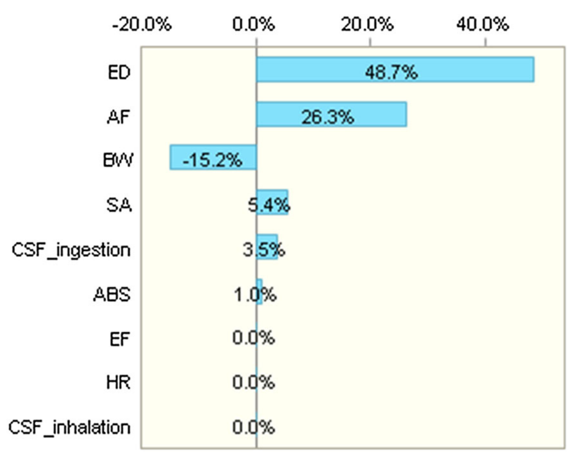


Fig. 6 Sensitivity analysis of the risk parameters. The sensitivity data portrayed in the contribution to variance view can be deciphered as the percent of forecast variance caused by each assumption

2017a, b, 2019). The sensitivity analysis uncovers that endeavors ought to be gone up against a superior meaning of probability distribution for two parameters (AF and ED) to enhance the unwavering quality of the risk evaluation.

Conclusion

Cancer risk assessment study using Monte Carlo simulation shows 10.2 times higher risk of cancer for children from poured rubber surfaced playgrounds than that of the uncovered ones. The 95% cancer risk from covered playgrounds (1.3×10^{-5}) is higher to the standards set by USEPA. Profiling of the PAHs from the playground dust/soil reveals 3-ring PAHs are dominating with a huge percentage (79.90% of total PAHs content, on an average). Leaching of PAHs from crapped tires used as the second layer of the poured rubber surface and adsorption of transported atmospheric PAHs can be the reasons of higher PAHs content in covered playgrounds. The diagnostic ratio analysis affirms that vehicular pollution and fossil fuel burning are the main source of high molecular weight CPAHs, whereas low molecular weight PAHs have pyrogenic origin. As LMW-PAHs have greater tendency to be transported, it can be concluded that LMW-PAHs can have a distanced source and HMW-PAHs may be of local vehicular exhaust origin. Among the three exposure routes, dermal exposure is proved to be the most important one and it suggests using most covered dresses for children in playgrounds to minimize cancer risk. The sensitivity analysis demonstrated that endeavors ought to be gone up against better comprehension of probability distribution for ED and AF to get an extended trustworthiness of the risk assessment. In addition, migration of PAHs from playground cover material as passive sampler to ingestible dusts/soils could be an important future research.

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