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ORIGINAL PAPER

Assessment of Dioxin-Like POP's Emissions and Human Exposure Risk from Open Burning of Municipal Solid Wastes in Streets and Dumpyard Fire Breakouts

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Abstract
India, a highly populated economy in transition generates huge quantity of municipal solid wastes and its management is posing great challenges. Infrastructural limitations force the urban local bodies to rely on traditional routes such as open dumpyards and landfills, where incidents of massive fire breakouts are often reported. In places where the collection system fails, the public seeks a much easier option of open burning of wastes in streets and households. The study reports a comparative assessment of 17 PCDD/Fs and 12 dl-PCBs emitted to air and residue during the repetitive incidents of massive fire breakouts at a municipal solid waste dumpyard and localized street waste burning in cities of India. The study also evaluated the direct exposure routes viz. inhalation as well as dermal and predicts the carcinogenic and non-carcinogenic health risks to the receiving population. The observed PCDD/F levels in the ambient air and burned residue samples ranged from 2.7 to 41.4 pgTEQ/m^3 and 79.8 to 860 ngTEQ/kg, while that of dl-PCB varied from 0.2 to 2.3 pgTEQ/m^3 and 6.0 to 46.2 ngTEQ/kg respectively. The dermal, as well as the inhalation daily exposure doses were estimated and the non-carcinogenic hazard indices of the children were found to be in levels of concern at two of the street burning sites while for adults the levels were found to be within the threshold limit. The cumulative Incremental Lifetime Cancer Risk (ILCR) values ranged from 2×10^{-6} to 2×10^{-4} suggesting moderate to low risk to cancer or cancer-linked illnesses to exposed individuals.

Graphical Abstract

Keywords Street littering · Dumpyard fires · Dioxins and PCBs · Ambient air · Burned residue · Cancer risk

Extended author information available on the last page of the article

Introduction

The domestic and commercial non-hazardous garbage generated from the community life activities poses great challenges in its management due to the variable compositions, high moisture content, fast putrescible nature and huge volume of generation (Nandy et al. 2015). Several factors such as outdated waste collection mechanisms, unskilled manpower, inadequate infrastructure, lack of scientific and professional approach in selecting suitable treatment processes and implementation of regulations are some of the major challenges for developing nations in the management of municipal solid wastes (MSW) (Ramaswami et al. 2016). India also faces significant difficulties in MSW management and out of the total generated solid waste quantity, only 20–30% gets treated while the remaining 70–80% ends up in smaller residential/street open dumps or larger MSW dumpyards (Sharma and Jain 2019). The indiscriminate dumping of wastes over the past few decades has led to the formation of several legacy dumpyards in India and the majority of them are devoid of any scientific or engineering measures to monitor or manage the formation of landfill gases (LFGs) (Waste Atlas, Sharma et al. 2019). Incidents of fires are very frequent in these landfill/dumpyard sites such as Ghazipur (Delhi), Deonar (Mumbai), Dhapa (Kolkata), Brahmapuram (Kochi) etc., which are triggered by combustible gases generated from heaps of putrescible wastes and burns out several hundred tons of waste every year (Annepu 2012; Project report on Indo-German initiatives: A case of waste management 2016).

Moreover, the non-hazardous nature of the MSW allows people to experiment and practice various options such as littering in streets, disposing of in running waters, open burning etc. and is observed commonly across India. The general public considers the open burning of MSW as a cheap and easy way to reduce waste volume and also to get rid of the associated smell, infectious vector breeding conditions, scavenging animals etc. (Kumar et al. 2015; Vreeland et al. 2016). Several studies reported that the uncontrolled combustion of MSW could lead to the emission of highly toxic fumes and respirable particulates into the breathing zones of the atmosphere. In addition, the dispersion of burned residue can contaminate the soil, surface and groundwater and sediments, all of which may finally reach the human food chain (Lemieux 2002). It was identified that open burning of MSW is a major source of unintentional persistent organic pollutants (U-POPs) such as polychlorinated dibenzo-*p*-dioxins (PCDDs), polychlorinated dibenzofurans (PCDFs) and polychlorinated biphenyls (PCBs) (commonly called dioxin-like POPs or dl-POPs) and hence has been notified as a source category in the 'standardized toolkit for identification and quantification of dioxin and furan releases' laid down by UNEP (Fiedler 2007). Considering its persistent, bioaccumulative and toxic properties, the dl-POPs have been classified as class-A carcinogens by International Agency for Research in Cancer (IARC) (McGregor et al. 1998). Hitherto, several studies reported high levels of dioxins and PCBs (4–4000 times) than that of control sites in the ambient air followed by biomass combustion, landfill or dumpyard fire breakouts (Shih et al. 2008; Fajkovic et al. 2018; Mazzucco et al. 2020; Weichenthal et al. 2015).

It can be noticed that massive fire breakout incidents in legacy dumpyards/landfill sites receive quick public attention as the spontaneous emission of thick smoke and possible allergies and breathing difficulties to people in the locality force the authorities to take necessary steps to extinguish the fire and to monitor the situation. Whereas the small-scale littering and burning of wastes in open places, roadsides, backyards etc. are often ignored by citizens as well as authorities since the discomfort caused by smoke/particulate emissions are relatively negligible owing to the lower volume of waste burned at any particular instance. Although the unit quantity of waste disposed of is less, such incidents occur routinely in several parts of a city and hence the annual cumulative emission will be much higher but are mostly ignored (Wiedinmyer et al. 2014; Cogut 2016). Often such waste heaps in streets will remain smoldering throughout the day, which are the most favourable conditions for the formation of products of incomplete combustions. Another danger associated with the episodes of street open burning is its close vicinity to residential, educational and commercial activities, where the probability of human exposure is high compared with relatively suburban located legacy dumpyard/landfill sites. Several studies reported the interdependence of adverse health effects and proximity of community life settlement to the dumpyard/landfill site, but investigations on the risks posed by street open burnings are very limited (Porta et al. 2009).

The present study has undertaken onsite ambient air and residual ash sampling followed by estimation and congener fingerprinting of the dl-POPs emitted during the massive fire breakout incidents at Brahmapuram MSW dumpyard and open burning of wastes in streets in urban city centres of Kerala, India. A detailed investigation to understand the daily exposure doses arising from the inhalation and dermal routes were carried out for two age groups (children and adults) and both carcinogenic and non-carcinogenic risks associated with the exposures were estimated. A quantitative assessment of the human health risk posed by the dl-POPs emission from the routine small-scale street burning practices and its comparison with that of massive dumpyard fires is the first such study reported to the best of our knowledge.

Methods

Sampling Sites

Street Waste Burning Sites

Three major roadside dumping sites in Thiruvananthapuram, Kerala—Pettah (8° 29′ 44″ N, 76° 55′ 49″ E), Thakarapparambu (8° 29′ 11″ N, 76° 56′ 45″ E) and Attakulangara (8° 28′ 46″ N, 76° 57′ 4″ E) were selected for the study (Fig. 1). The sites are situated in the Thiruvananthapuram corporation region (capital of the state of Kerala) where regular intentional/un-intentional waste open burning events are reported. The ambient air high volume PUF samplers were operated at approximately 2–5 m distant from the waste pile and are closer to the routine movement of the general public. The ambient air sampling at Pettah and Attakulangara was operated continuously for 22 and 25 h respectively in one stretch whereas at Thakarapparambu smoke ceased two times and correspondingly sampler was also switched off and restarted upon re-ignition and visible smoke generation to avoid dilution while ensuring collection of approximately 300 m^3 of the air sample.

Accidental Fire Breakout Site at Brahmapuram MSW Dumpyard, Kerala

The Brahmapuram MSW treatment centre and dumping site (9° 59′ 28″ N, 76° 21′ 59″ E) is situated very close to the Smart city project, Kochi, the commercial capital of the state of Kerala and is approximately 7 km away from the city centre (Fig. 1). The plant receives approximately 350–400 tons of MSW per day. Presently the material recovery and composting units are non-functional/partially operated and hence a major share of waste received goes to open dumping and approximately 6 lakh tons of legacy wastes are dumped over 60 acres of land (Kochi Waste to Energy Project 2018). The dl-POPs emission during two major fire breakout incidents reported at the site: one in 2019 (22rd–25th February 2019) and the second in 2020 (18th–20th February 2020) were investigated in the present study. The air samples were collected at about 150–170 m away from the epicenter of the fire where the settlements of plant workers were located. The farther distance from the epicenter was chosen for operating the samplers to ensure the safety of the supporting staff and samplers from any possible escalation of fire. As the power supply to the area was disconnected due to fire hazards, a diesel-powered generator (Hitachi Corporation, Japan) was utilized for the uninterrupted operation of PUF samplers.

Sampling Procedure

The high-volume ambient air sampler (APM 460, Envirotech Instruments Pvt. Ltd, India) was used for the ambient air sampling consisted of a stainless-steel filter paper holder which can accommodate 25 × 20 cm filter paper, 15 cm long cylindrical glass-lined cartridge to hold polyurethane foam (PUF) media and a timer to set the sampling time. The glass made PUF cartridge was pre-cleaned by rinsing with acetone and the PUF plugs were Soxhlet extracted for 16 h with toluene and dried under high purity nitrogen gas to eliminate possibilities of cross-contamination before every sampling. Pre-weighed Whatman quartz micro-fibre filter (QFF) papers were used for the particulate phase collection and the PUF plugs were spiked with 100 pg of sampling standards (^{13}C-labelled congeners of 1234 TCDF, 1234 TCDD, PCB 79, PCB 60, PCB 127, PCB 159) to evaluate the sampling efficiency.

Two air samples each from two fire breakout incidents at Brahmapuram reported in 2019 & 2020 respectively and one sample each from street waste burnings at Pettah, Thakarapparambu and Attakulangara were collected for the present study. The air sampling was carried out in such a way that as far as possible a minimum of 300 m^3 of air at a sampling rate of 200 LPM was collected (USEPA method TO-9A 1999). It was not attained during the dumpyard fire incident at Brahmapuram due to delays in reporting of the incident and logistics from CSIR-NIIST located (350 km away) from the site of occurrence. At Brahmapuram dumpyard site, burned residue samples were collected from 4–5 points each from two of the fire ridden MSW heaps and were made into 2 Nos of composite samples representing each heap through coning and quartering method. One composite burned residue sample from each of the street waste burning sites at Pettah, Thakarapparambu and Attakulangara were also collected.

Sample Analysis

All the samples were extracted within 30 days from the date of sampling and analyzed within 45 days from the date of extraction. The QFF and PUF plugs were retrieved from the sampler and the final weight of the QFF was noted for calculating PM concentration. The mass-labelled (^{13}C) congeners of PCDD/Fs—17 nos (100 pg each) and MO&NO-PCBs—12 nos (500 pg each) were spiked onto the PUF plugs before extraction as internal standards (ISTD) to assess analytical recovery. The glass cartridge was rinsed with acetone followed by toluene for the complete transfer of residues. The rinsates were concentrated using a rotary evaporator (R-300, Buchi Corporation, Switzerland) and further added into the Soxhlet extractor solvent flask. The QFF and PUF were extracted together in a Soxhlet system with toluene as charging solvent for 16 h at 5 siphons per hour.

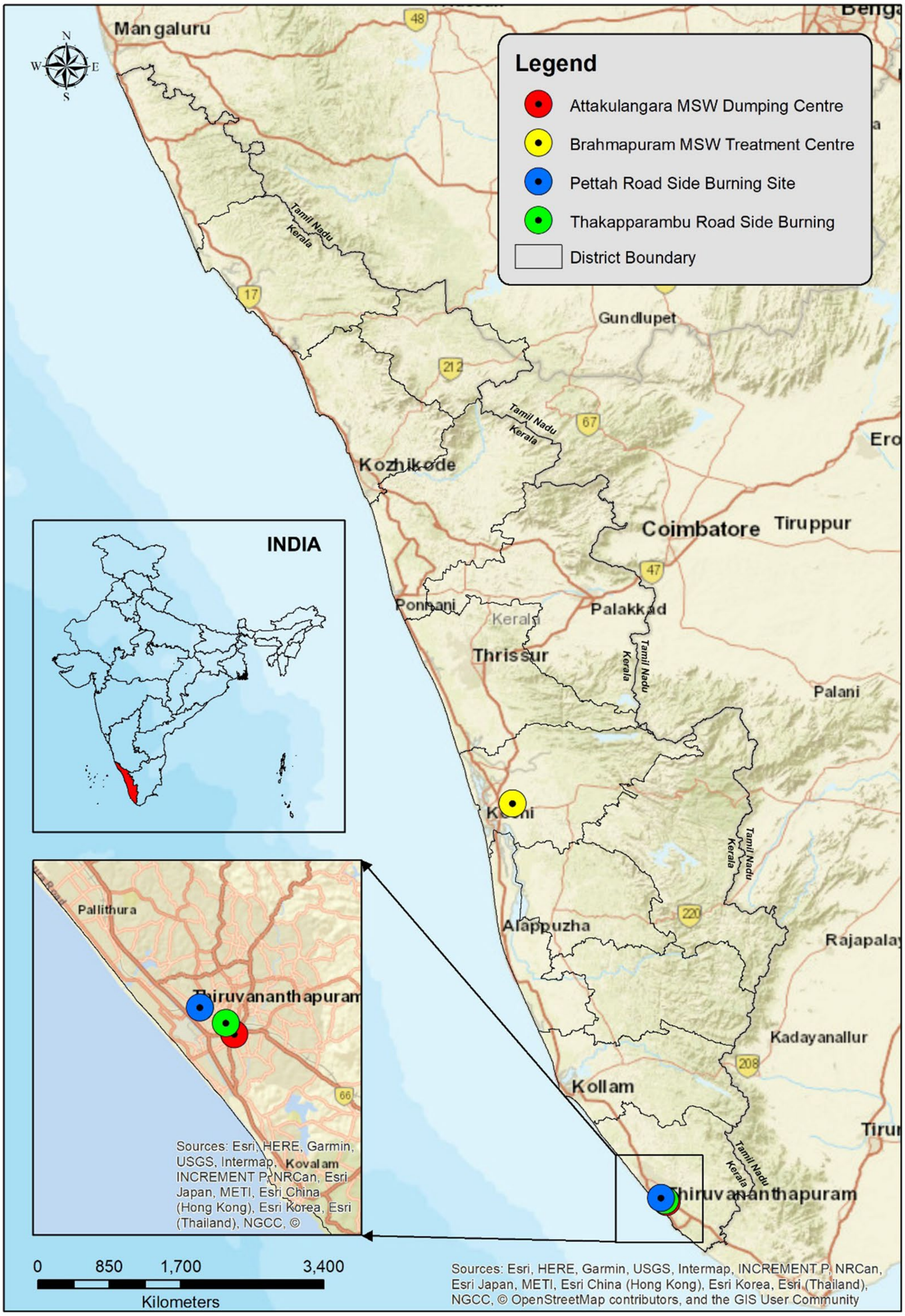

Fig. 1 Ambient air and burned residue sampling sites

The burned residue samples were dried in a hot air oven at 103 °C for 3 h to remove moisture content, weighed 5 g of dried sample, mixed with an equal amount of sodium sulfate to remove any residual moisture and were spiked with C^{13} labelled internal standards before Soxhlet extraction. The sample extracts were then concentrated using a rotary evaporator and were cleaned up and fractionated using three column based (Multilayer silica column, Alumina column and carbon column) automated system (DEXTech Pure, LCTech, Germany). The first fraction was obtained as 1:1 DCM: Hexane solution containing MO & NDL PCBs and the second fraction as toluene containing PCDD/F and NO-PCBs. A nitrogen evaporator (Supervap-6, FMS Inc, USA) was used for concentrating the sample fractions to dryness. Both the sample fractions were then spiked with 20 pg syringe standards (1278 TCDF, 123468 HxCDF, 1234689 HpCDF) and 100 pg (PCB 70, 111, 170) respectively, and finally reconstituted in 200 µL *n*-nonane.

GC-triple quadruple mass spectrometer (Model: 7890B/7000C, Agilent Technologies, Germany) was used for the quantitative analysis. The MS/MS was operated in electron ionization mode (EI) at 70 eV coupled with the multiple reaction monitoring (MRM) method for quantification. The GC injection was made through solvent vent mode at 120 °C and the injection volume was 4 µL. 60 m DB-5MS UI (Agilent technologies, Germany) GC column was used (0.25 um film thickness, 0.25 mm internal diameter) for the analysis. The GC oven temperature program for the PCDD/F analysis was from 60 to 325 through three ramps with rates 30 °C/min, 2 °C/min and 10 °C/min. The final hold time was 5 min and the total run time was 35.5 min. The carrier gas (helium) flow rate was static 1 mL/min and MS source temperature was 330 °C.

Analytical Quality Control

Isotopic dilution mass spectrometric method was employed for the confirmatory analysis of PCDD/Fs and dl-PCBs (dl-POPs) and hence the obtained individual native congener concentrations were corrected with internal standard recoveries of the corresponding ^{13}C labelled congeners. The WHO-2005 TEF factors of PCDD/Fs and dl-PCBs were used for calculating the final TEQ (Van Den Berg et al. 2006). The ISTD recoveries for the corresponding congeners should be in the range of 60–120%. The deviations if any, was acceptable only when the contribution of the congener to total TEQ is less than 10%.

The limit of quantification (LOQ) for all the congeners under the study was calculated from the lowest acceptable calibration point complying with the validation criteria specified for GC–MS/MS as per EU regulations 644/2017 such as (i) the relative response factor (RRF) ≤ 30% (ii) relative standard deviation (RSD) ≤ 15% (iii) relative ion ratio tolerance ˂ 15% (EU (No.) 644/2017). The on-column concentration corresponding to the particular calibration point was considered as the LOQ (Law et al. 2018, L'Homme et al. 2015). For the congeners found below the limit of quantification (LOQ), the upper bound levels were taken as LOQ during the calculation of the final TEQ as specified in standard reporting protocols of dl-POPs. As the study was targeted to assess the human exposures to PCDD/F and dl-PCB emissions, upper bound levels obtained from the analysis were considered for all exposure dose calculations. To understand the background levels from any other sources such as vehicular exhausts in pristine and urban locations, a control and field blank site sampling were also carried out in the study. CSIR-NIIST institute campus was taken as the control site where no open littering and burning activity is practiced and field blank sample was collected from the Thakarapparambu site on a day when open burning activity was not occurring. The control and a field blank ambient air levels were used as reference values for comparison with open burning site emission data.

Statistical Analysis

The ratios between cumulative congener concentrations of PCDDs, PCDFs and dl-PCBs were analyzed for each sample. The concentration ratios can be used to understand the predominant formation mechanism underwent during open burning. Further congener specific contributions to total TEQ in samples were calculated and plotted using Microsoft Excel 2019. A correlation matrix was also generated to understand the interdependence between the detection frequencies of PCDD/Fs and dl-PCBs congeners in burned residue and air samples. Further, a ground-truthing attempt was also been conducted by estimating the correlations of congener fingerprint obtained from the present study with respect to the simulated open burning studies reported previously in the region.

Exposure Assessment and Risk Prediction

The emission levels observed in ambient air and residual ash samples were utilized for assessing the risk associated (non-carcinogenic and carcinogenic) with two direct routes of human exposure—inhalation and dermal pathways. The daily intake doses of PCDD/Fs and dl-PCBs through these two routes were calculated for two age categories—children (1–17 years of age) and adults (18–70 years of age). The Daily Exposure Dose through inhalation and dermal (DEDinh and DEDder—mg kg^{-1} day^{-1}) were calculated as per the Eqs. 1 and 2, following ATSDR Public health Assessment Guidance Manual (ATSDR Public Health Assessment Guidance Manual 2005).

$$\mathrm{DED_{inh}} = \frac{C_{\mathrm{air}} * IR * F * ED}{BW * AT} \tag{1}$$

$$\mathrm{DED_{der}} = \frac{C_{\mathrm{BR}} * A * AF * F * ED * CF}{BW * AT} \tag{2}$$

where, C_{air}—concentration observed in the air (mg TEQ/m^3), *IR*—inhalation rate (m^3/day), *F*—frequency of exposure (days per year), ED—exposure duration (years), C_{BR}—concentration observed in burned residues (mg TEQ/kg), A—total soil adhered (mg) AF—bioavailability factor (unitless), CF—conversion factor (10^{-6}), BW—average body weight (kg), AT—Average lifetime (days). Table 1 gives the values considered for the calculation of daily exposure dose (DED), hazard quotients and incremental lifetime cancer risk (ILCR) values. The average body weight and lifetime assigned to the Indian population was taken for calculations, whereas total soil adhered was considered as per the ATSDR document as no nation-specific data was available.

$$HQ_{\mathrm{inh}} = \frac{\mathrm{DED_{Inh}}}{\mathrm{RfD}} \tag{3}$$

$$HQ_{\mathrm{Der}} = \frac{\mathrm{DED_{Der}}}{\mathrm{RfD}} \tag{4}$$

$$HI_i = \sum_{i=1}^{n} HQ \tag{5}$$

Hazard Quotient (HQ), the ratio of daily exposure dose to reference dose is used for the non-carcinogenic risk assessment and is calculated as per Eqs. 3 and 4. The threshold value of HQ is 1, where $HQ < 1$ indicates lower exposure than no observed effect dose and is considered safe. For dioxin-like POPs, no reference doses are available to date in India and hence tolerable daily intake levels recommended by WHO for the ingestion route (TDI—1–4 pgTEQ/kg of body weight per day) was adopted (WHO 1998). HI—Hazard Index represents the cumulative effect of the HQs arising from various chemicals through different exposure pathways and in the present case it was calculated by summing the HQs emanating from dermal and inhalation route of PCDD/Fs and dl-PCBs (Eq. 5).

$$\mathrm{ILCR_{Der}} = \mathrm{DED_{der}} * SF \tag{6}$$

$$\mathrm{ILCR_{Inh}} = \frac{\mathrm{DED_{inh}} * \mathrm{IUR} * BW * 1000}{IR} \tag{7}$$

The cancer risk for the whole life exposure was determined through Incremental Lifetime Cancer Risk (ILCR) for both dermal and inhalation pathways as per the methodology specified by USEPA. An upper bound estimate of the response per unit chemical intake defined as the cancer slope factor (SF) was used for calculating ILCR (Eq. 6) through the dermal route. In the case of inhalation risk, IUR—inhalation unit risk factor was used to assess the cancer potency factor using the Eq. 7. ILCR was determined for both PCDD/Fs and dl-PCBs and cumulative risk was calculated by the summation of dermal and inhalation risks posed by PCDD/Fs and dl-PCBs, respectively (Eq. 8).

$$\mathrm{ILCR_{cum}} = \mathrm{ILCR_{Der}} + \mathrm{ILCR_{Inh}} \tag{8}$$

The carcinogenic benchmark described by USEPA was used for comparing the derived incremental lifetime cancer risk (ILCR) values. The ILCR values are classified as values $\le 1 \times 10^{-6}$ correspond to very low, 1×10^{-6} to 1×10^{-4} are low; 1×10^{-4} to 1×10^{-3} are moderate; 1×10^{-3} to 1×10^{-1} are high and values $> 1 \times 10^{-1}$ represent very high risk (ATSDR Public Health Assessment Guidance Manual 2005).

Results and Discussion

Dioxin Levels Observed During Street Waste Burning

Table 2 shows the PCDD/Fs and dl-PCBs levels in the ambient air and burned residue samples collected from street waste burning sites. The ambient air volume collected was in the range of 302–360 m^3 with sampling durations varying from 24 to 30 h. The observed levels of particulate matter ranged from 316 to 1311 µg/m^3 at these sites. Levels of PCDD/Fs observed in air and burned residue samples were found to be in the range of 13.0–41.4 pg TEQ/m^3 and 369–860 ng TEQ/kg, while that of dl-PCBs varied from 0.2 to 2.3 pg TEQ/m^3 and 11.9 to 46.2 ng TEQ/kg, respectively. The observed levels of PCDD/Fs at street waste burning sites

Table 1 Parameter values used for the exposure risk estimations

Sl. No.	Parameter	Value	References
1	IR	15.2 m^3/day	ATSDR Public Health Assessment Guidance Manual (2005)
2	BW	70 kg	World Population Prospects (2019)
3	AT	25,550 days	World Population Prospects (2019)
4	A	299 cm^2 (children), 326 cm^2 (adult)	ATSDR Public Health Assessment Guidance Manual (2005)
5	AF	0.1	ATSDR Public Health Assessment Guidance Manual (2005)
6	RfD	4 pgTEQ/kg BW	WHO (1998)
7	SF	1.56×10^5	Regional Screening Level-USEPA (2013)
8	IUR	38	Regional Screening Level-USEPA (2013)

Table 2 Particulate matter, PCDD/Fs and dl-PCBs levels in street waste burning site samples (ambient air and burned residue)

Sampling site	Sample	Observed levels		
		PM (particulate matter) (μg/m^3)	PCDD/Fs	dl-PCBs
Control site	Air	6	0.2 pgTEQ/m^3	0.2 pgTEQ/m^3
Field blank	Air	130	1.3 pgTEQ/m^3	0.2 pgTEQ/m^3
Pettah	Air	316	13.0 pgTEQ/m^3	0.9 pgTEQ/m^3
	Burned residue	–	860 ngTEQ/kg	46.2 ngTEQ/kg
Thakarapparambu	Air	1234	41.4 pgTEQ/m^3	2.3 pgTEQ/m^3
	Burned residue	–	369 ngTEQ/kg	11.9 ngTEQ/kg
Attakulangara	Air	1311	35.7 pgTEQ/m^3	0.2 pgTEQ/m^3
	Burned residue	–	414.0 ngTEQ/kg	12.6 ngTEQ/kg

were found to be 65–200 times higher than the control site and 10–32 times higher than the field blank values (Table 2). All the 17 PCDD/F congeners and 12 dl-PCB congeners were detected above the LOQs. The findings indicate that such kind of low intensity, high-frequency open burning incidents may contribute significantly to the total annual dioxin emission.

The congener wise contribution of PCDD/Fs and dl-PCBs towards total toxicity equivalence (TEQ) are shown in Fig. 2a and b and the congener profiles observed in the samples with standard error bars are shown in Figs. S1–S4. The predominant PCDD/F congeners found in the air and burned residue samples of Pettah and Thakarapparambu sites were 1234678-HpCDD and OCDD whereas in Attakulangara the pattern observed in the air indicates a higher proportion of 2378-TCDF and 12378-PeCDF and in residue samples, 1234678-HpCDF and OCDD were the major congeners. PCB-114 and PCB-118 were the major dl-PCB congeners in the air samples from Thakarapparambu and Attakulangara whereas PCB-114 and PCB-123 were the dominant congeners in ambient air sampled at Pettah site. In the case of burned residue samples also Thakarapparambu and Attakulangara showed similarity as PCB-77 and PCB-81 being the most predominant congeners and in samples from Pettah PCB-77 and PCB-105 were the prominent ones. The congener profiles observed in ambient air as well as in burned residue do not exhibit a uniform abundance pattern even at same sites. An important shortcoming associated with the onsite studies of dioxins emissions is the uncertainty associated with the waste composition, burned mass,

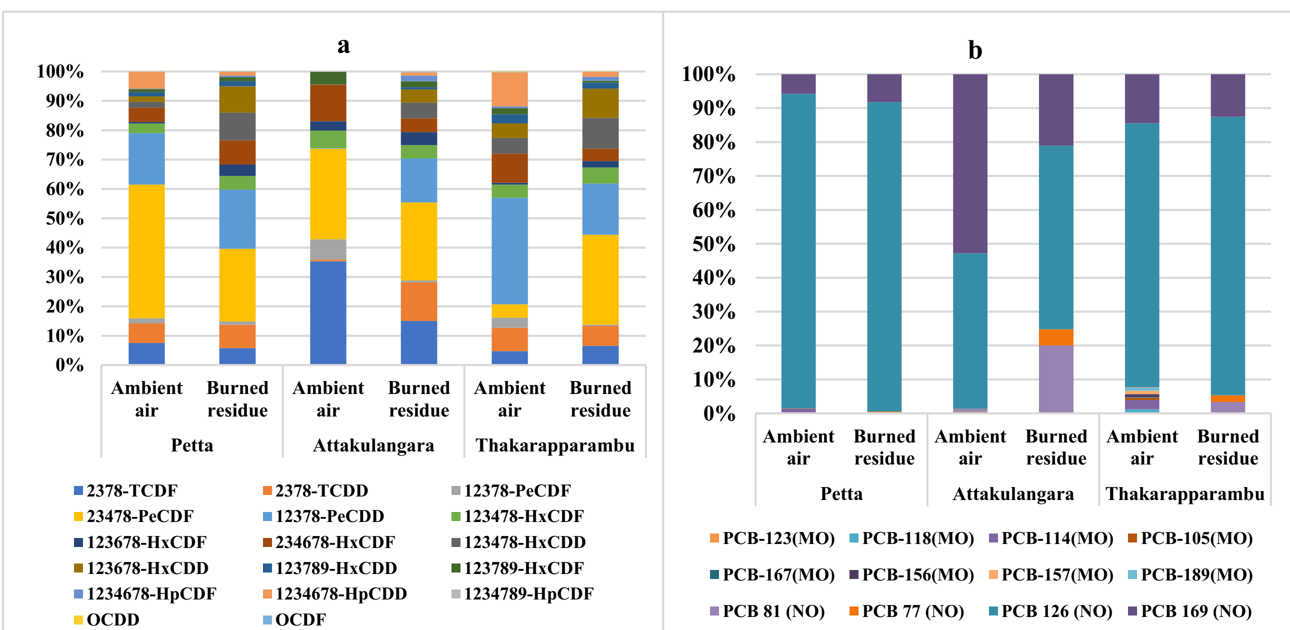

Fig. 2 **a**, **b** Plot showing the PCDD/F's and dl-PCB's congener wise contribution to respective total TEQ in street waste burning site samples (ambient air & burned residue)

combustion temperature and dilution factors (Lemieux et al. 2000). Dl-POPs emission from uncontrolled open combustion is highly dependent on these factors where even the emission factors generated from lab scale simulated studies varied over 3–4 orders of magnitude (Hedman et al. 2005; Wevers et al. 2004; Zhang et al. 2011, Ajay et al. 2021). This indicates that the variations in concentrations or congener profiles of dl-POPs with respect to different open burning sites monitored in the present study could be presumably due to variations in combustion conditions and waste compositions (Gullett et al. 2010).

Dioxins Levels Observed During Fire Breakout Incidents at Brahmapuram

Table 3 shows levels of PCDD/Fs and dl-PCBs observed in ambient air and burned residue samples collected during the fire breakout incidents at Brahmapuram waste dumpyard site in consecutive years 2019 and 2020, respectively. The average levels of PCDD/Fs observed in the air and burned residue matrices during the 2019 incident were 10.3 pgTEQ/m^3 and 158.2 ngTEQ/kg whereas that of dl-PCB were found to be 0.3 pgTEQ/m^3 and 11.7 ngTEQ/kg, respectively. And in the 2020 incident, the average levels of PCDD/Fs observed in air and burned residue matrices were 3.2 pgTEQ/m^3 and 82.1 ngTEQ/kg respectively. In the case of dl-PCBs 0.3 pgTEQ/m^3 and 7.1 ngTEQ/kg were the average concentrations observed in the air and burned residue matrices. In 2019, the fire breakout incident lasted for 4 days and 24 h of active fumes were sampled whereas, during the fire breakout incident in 2020, the fire was brought under control on the 3rd day through watering of the waste piles and hence active smoke was absent for the majority of sampling duration. This could be the reason for lowered levels of PCDD/Fs and dl-PCBs in samples from the 2020 incident. The observed levels at dumpyard fire breakout sites were 10–20 folds lower than the street waste burning sites (Tables 2, 3) and this could be due to the fact that the PUF samplers were placed 150–170 m away from the epicenter of fire in the former case when compared to 2–5 m at the latter site. The observed levels of PCDD/Fs in ambient air during 2019 and 2020 incidents were 50 and 15 times higher than the control site levels and 8 and 2.5 times higher than the field blank levels.

The congener wise contribution to total PCDD/F and dl-PCB TEQ are shown in Fig. 3a and b and the congener profiles observed at the site with standard error bars are shown in Figs. S1–S4. 1,234,678-HpCDD and OCDD were the most prominent PCDD/F congeners in the air samples of 2019 and 2020 incidents and also in burned residue samples from 2019. The burned residue sample from 2020 showed higher levels of 1,234,678-HpCDD and 1,234,678-HpCDF. The major dl-PCB congeners in the air samples from 2019 and 2020 were PCB-118, PCB-114 and PCB-123 respectively. In the case of burned residue samples PCB-123, PCB-118, PCB-77 and PCB-123 were respectively the major congeners in 2019 and 2020.

Statistical Analysis and Correlation Studies

The trend of group-wise congener abundance observed in air samples was dl-PCBs > PCDFs > PCDDs, and in burned residue samples was dl-PCBs > PCDDs > PCDFs which were in agreement with the findings of simulated MSW combustion studies reported on Indian conditions (Ajay et al. 2021). Among PCDD/Fs the predominance of PCDDs over PCDFs was generally observed indicating the higher availability of oxygen due to open combustion conditions (Addink and Olie 1995). In addition, an abundance of PCDDs and higher chlorinated congeners in the majority of samples indicates that heterogeneous condensation of precursor molecules involving both gas and solid phase reactants was prevailing during open burning (Huang and Buekens 1995; Wikström et al. 2003).

Table 4 shows Spearman's correlation matrix to elucidate the interrelationship between dioxins, dl-PCBs and PM levels observed in ambient air and burned residue samples collected from open burning sites. The particulate matter

Table 3 Particulate matter, PCDD/Fs and dl-PCBs levels in ambient air and burned residue samples from Brahmapuram waste dumpyard fire breakout incidents

Site	Sample	Observed levels		
		PM (particulate matter) (μg/m^3)	PCDD/F	dl-PCBs
Brahmapuram Fire breakout incident in 2019	Air-1	338	9.5 pgTEQ/m^3	0.4 pgTEQ/m^3
	Air-2	326	11.1 pgTEQ/m^3	0.2 pgTEQ/m^3
	Burned residue-1	–	152.7 ngTEQ/kg	11.9 ngTEQ/kg
	Burned residue-2	–	163.8 ngTEQ/kg	11.4 ngTEQ/kg
Brahmapuram Fire breakout incident in 2020	Air-1	101	2.7 pgTEQ/m^3	0.4 pgTEQ/m^3
	Air-2	122	3.6 pgTEQ/m^3	0.2 pgTEQ/m^3
	Burned residue-1	–	84.3 ngTEQ/kg	6.0 ngTEQ/kg
	Burned residue-2	–	79.8 ngTEQ/kg	8.2 ngTEQ/kg

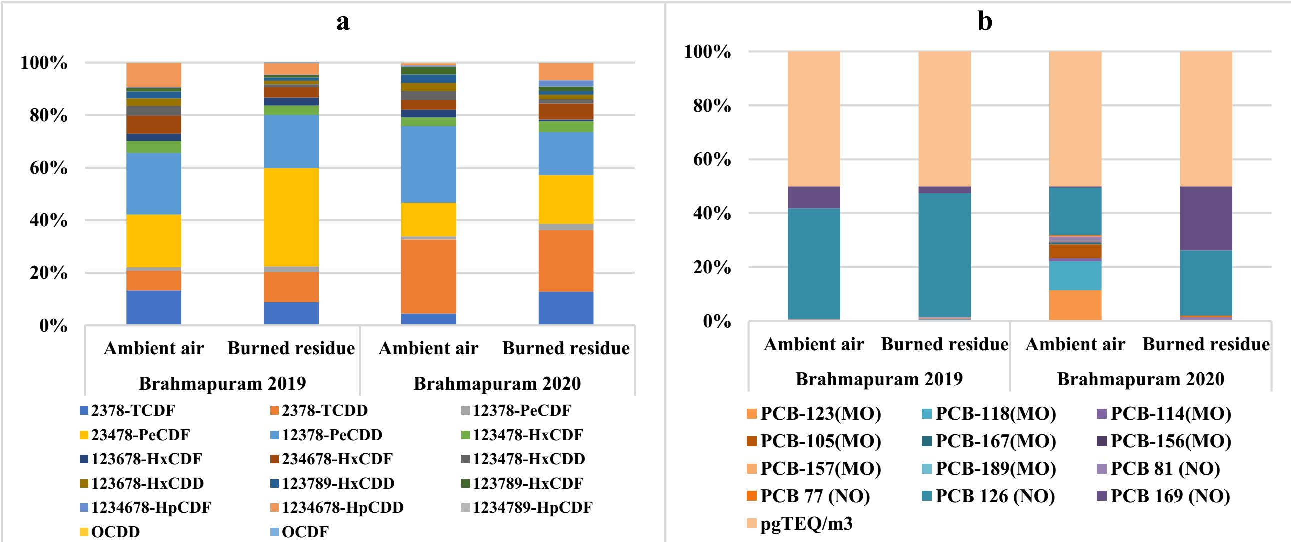

Fig. 3 **a**, **b** PCDD/F's and dl-PCB's congener wise distribution to respective total TEQ in ambient air and burned residue samples from Brahmapuram dumpyard site

Table 4 Spearman's correlation analysis between particulate matter, PCDD/Fs and dl-PCBs levels in ambient air and burned residue samples

	Particulate matter	PCDD/Fs air	PCBs air	PCDD/Fs residue	PCBs residue
Particulate matter	1				
PCDD/Fs air	1**	1			
PCBs air	0.14	0.14	1		
PCDD/Fs residue	0.82*	0.82*	0.14	1	
PCBs residue	0.93**	0.93**	0.39	0.82*	1

*Correlation is significant at the 0.05 level (2-tailed); **Correlation is significant at the 0.01 level (2-tailed)

concentration exhibited a very high correlation with PCDD/Fs air emission ($\rho = 1$), whereas poor correlations were observed with respect to dl-PCBs emissions ($\rho = 0.14$). This difference could be attributed to the antagonistic effect of two aspects i.e. high particulate bound nature of PCDD/F congeners and the relatively high volatility of dl-PCBs (Heo et al. 2014; Cindoruk and Tasdemir 2007). Further, the PCDD/Fs levels in air samples were highly correlated with burned residue levels, whereas a statistically significant correlation of dl-PCBs in air and residue samples was hardly observed. Although PCDD/Fs levels in air samples was not correlated with dl-PCB levels, both the groups were highly correlated in residue samples similar to those reported elsewhere (Cortés et al. 2014; Ba et al. 2009). The higher affinity of PCDD/Fs towards burned residues during open burning conditions has been previously reported which could be due to the planar structure of these compounds as it promotes strong binding towards carbonaceous residual matter. Some reports also suggest the role of shrinkage of the burning surface during combustion which could restrict the dioxins formed at the surfaces from emitting to air (Hazardous Chemicals from Open Burning of Waste in Developing Countries—Final Report 2010; Zhang et al. 2017). On the contrary, the commensurate rise of dl-PCBs levels in air and residue samples points to its lower particulate adsorption tendency which needs detailed investigation to elucidate the mechanism of congener distribution between phases depending on the degree of planarity of dl-PCBs (non-ortho & mono-ortho PCBs).

Further a comparative evaluation between the congener profiles observed in the present study with that of the general congener patterns reported from the open burning scenarios in the region was conducted to ensure that the emissions are exclusively from the MSW open burning incidents at the sites (Ajay et al. 2021). It could also lead to the derivation of dl-POPs congener fingerprint representative of MSW open burning sector in the region. The Spearman's correlation analysis to deduce the above hypothesis are shown in supplementary information, Tables S1–S4. It shows that the congener profiles of PCDD/Fs in burned residues samples from open burning sites reported in the present study showed a very high correlation with that observed in the simulated study. In the case of ambient air, the congener profile of all samples except the one from the Thakarapparambu site exhibited a very high correlation with simulated combustion study profiles. An interesting finding is that even though the ambient air levels observed at different sites varied over two orders in magnitude, the congener profiles were having statistically significant correlations between them. Therefore, the congener profiles can be considered as a source fingerprint of open burning activities in the region. The dl-PCBs have not shown any notable correlations in the case of air samples, whereas a significant correlation was observed for residue samples in the present field samples vis-à-vis the reported congener profile of the simulated study.

Comparative Evaluation of Emission Levels with Previous Studies

There are no threshold levels established for dioxins in the ambient air or the burned residues from open burning in Indian conditions or internationally. Moreover, it is inappropriate to compare the permissible levels specified for stack emission vis-à-vis the non-point/area sources of emissions and hence an assessment of the extent of contamination at the site cannot be derived from the observed air and residual level concentrations. From the literature survey, it was noted that only very few case studies based on real-time sampling during fire breakout incidents at waste dumpyards were reported previously and a comparative evaluation of available studies is given in Table 5. A rapid decline in the levels of PCDD/Fs and dl-PCBs in ambient air with the increase in distance from the fire focal point was a notable observation, which was inferred in some of the previous studies as well (Ruokojärvi et al. 1995; Mazzucco et al. 2020; Bergström and Björner 1992; Rada et al. 2018; Fajkovic et al. 2018; Weichenthal et al. 2015; Nadal et al. 2016a, b). Ruokojärvi's and Bergström's groups conducted sampling at 2–10 m away from the fire focal point and the levels observed are comparable with the present study. The predicted concentrations of 54.6–76.9 pgTEQ/m^3 through modelling study by Rada et al. is very much similar to the current findings considering the possible dilution effects that may incur at the sites. On the contrary, Mazzucco et al. reported a high level of dioxin deposition in the soil at 1–3 km apart from the Bellolampo fire accident site in Italy whereas Nadal et al. reported much lower emission levels from tyre landfill fires in Spain at a similar distance. This indicates that the comparative evaluation needs to address different aspects such as waste quantity, type of waste, duration of the incident, micro-meteorological factors and the applied sampling and analytical methods to understand the trends of emission.

Table 5 Comparison of the levels of PCDD/Fs and dl-PCBs in the present study vis-à-vis existing studies on landfill fires and open burning incidents

Sl No.	Landfill fire/street burning	PCDD/F in air	PCDD/F in burned residue/soils	dl-PCB in burned residue/soils	dl-PCB in air	Sampling point distance form fire focal point	References
1	Landfill fire (simulated and spontaneous incident)	51–427 pgTEQ/m^3	106–290	–	–	2–5 m	Ruokojärvi et al. (1995)
2	Landfill fire (spontaneous incident)	–	13–900 μg/kg	100–880 μg/kg	–	1–3 km	Mazzucco et al. (2020)
3	Landfill fire (simulated study)	66–518 ngTEQ (NORDIC)/m^3	–	–	–	5–10 m	Bergström and Björner (1992)
4	Landfill fire (modelled study)	54.6–76.9 pgTEQ/m^3	–	–	–	–	Rada et al. (2018)
5	Landfill fire (spontaneous incident)	25.7 fgTEQ/m^3	48.11 ngTEQ/kg	–	–	–	Fajkovic et al. (2018)
6	Landfill fire (spontaneous incident)	0.4 pgTEQ/m^3	–	–	–	1 km	Weichenthal et al. (2015)
7	Tyre landfill fire (spontaneous incident)	13.3–15.4 fgTEQ/m^3	0.1–1.3 ngTEQ/kg	0.02–0.3 ngTEQ/kg	1.3–1.5 fgTEQ/m^3	1–3 km	Nadal et al. (2016a, b)
8	Dumpyard and street fires	2.7–41.4 pgTEQ/m^3	79.8–860 ngTEQ/kg	6.0–46.2 ngTEQ/kg	0.2–2.3 pgTEQ/m^3	2–150 m	Present study

Health Risk Assessment

The non-carcinogenic, as well as carcinogenic risks associated with the dermal and inhalation exposures for the exposed community were estimated for all the sites. Approximately 1–5 fire breakout incidents occur annually at Brahmapuram waste dumpyard site and considering the average atmospheric lifetime of dioxins as 26–130 h (approximately 5–6 days), 30 days per year was considered as the exposure frequency (Atkinson 1991). The plant was commissioned in 2008 and the first fire breakout incident in the plant was reported in April 2010 and hence 10 years was considered as the exposure duration. In the case of open burning of street waste, the major centralized waste treatment facility in Thiruvananthapuram city was closed down in 2013. Since then, the frequency of road dumps and street waste burning incidents increased quite evidently and hence the subsequent 8 years was considered as the exposure duration. It was noted from surveillance studies that open burning of wastes is practiced at least once in every 2–3 days in the street sites of Thiruvananthapuram city. As per the ATSDR guidelines, the upper bound frequency of incidents needs to be considered for risk calculation. Thus 180 days per annum was taken as the exposure frequency for calculating the health risk assessment.

Daily Exposure Dose

Table 6 shows the estimated (daily exposure doses) DEDs through dermal and inhalation routes at the study sites. The PCDD/Fs dermal DEDs for children varied from 7.6×10^{-12} to 1.3×10^{-10} mgTEQ kg^{-1} bw day^{-1} and for adults, it varied from 1.8×10^{-12} to 3×10^{-11} mgTEQ kg^{-1} bw day^{-1}. The DEDs of PCDD/Fs from inhalation ranged from 4.8×10^{-11} to 4.5×10^{-9} mgTEQ kg^{-1} bw day^{-1} for children and 1.1×10^{-11} to 1.0×10^{-9} mgTEQ kg^{-1} bw day^{-1} for adults. In both routes, children were found to be more susceptible to the exposures of dioxins than adults. From Table 6 it can be noted that the cumulative exposure doses were up to 70 times higher at street burning sites than the fire breakout incident site. A similar trend was found in the case of dl-PCB daily exposure doses. The dermal doses of dl-PCBs varied from 6.6×10^{-13} to 7.2×10^{-12} mgTEQ kg^{-1} bw day^{-1} for children and 1.6×10^{-13} to 1.7×10^{-12} mgTEQ kg^{-1} bw day^{-1} for adults. Whereas the inhalation doses varied from 4.6×10^{-12} to 1.7×10^{-10} mgTEQ kg^{-1} bw day^{-1} for children and from 1.0×10^{-12} to 3.8×10^{-11} mgTEQ kg^{-1} bw day^{-1} for adults. When compared between the exposure pathways, inhalation exposure doses were higher for both the age groups whereas comparison between the congener groups revealed that PCDD/Fs dose levels were approximately 30 times higher than that of dl-PCBs (Table 6).

Non-carcinogenic Risk: Hazard Quotient and Hazard Index

The non-carcinogenic risk estimates of PCDD/F/dl-PCBs from dermal and inhalation routes for children and adults are shown in Fig. 4a and b. The *HQ*s for dermal risk was found to be very low in the range of 4×10^{-3} to 1×10^{-2} for children and 5×10^{-4} to 3×10^{-3} for adults. Whereas the inhalation risk *HQ* was found to be 10–100 times higher than the dermal risk i.e. 1.0–1.1×10^{-2} for children and 3×10^{-3} to 3×10^{-1} for adults. Children were having higher *HQ* values in both the routes and can be considered as the higher risk community. In the case of dl-PCBs also a similar trend was observed where *HQ*s calculated for inhalation route was higher than that of dermal by a factor of 10–100. Inhalation risk *HQ*s for children and adults were in the range of 1×10^{-3} to 4×10^{-2} and 3×10^{-4} to 1×10^{-2} respectively. Whereas dermal risk *HQ*s ranged from 2×10^{-4} to 2×10^{-3} for children and 4×10^{-5} to 4×10^{-4} for adults. A cumulative non-carcinogenic risk index (*HI*) from dermal and inhalation exposure to PCDD/Fs and dl-PCBs was also calculated (Fig. 4a, b). Hazard quotients from dermal exposures at all the sites were having a very low contribution to *HI* ranging from 2 to 13% for children and 2–14% for adults. Hazard indices at street burning sites were approximately up to 80 times higher than that at dumpyard fire site and the highest *HI* for children (1.2) was at Thakarapparambu which crossed the threshold value of 1. At Attakulangara where the *HI* was

Table 6 Daily exposure doses for PCDD/Fs and dl-PCBs through dermal and inhalation routes at the study sites

Site	PCDD/Fs exposure doses (mgTEQ kg^{-1} bw day^{-1})				dl-PCBs exposure doses (mgTEQ kg^{-1} bw day^{-1})			
	DED_{Derm}		DED_{Inh}		DED_{Derm}		DED_{Inh}	
	Children	Adult	Children	Adult	Children	Adult	Children	Adult
Brahmapuram 2019	1.5×10^{-11}	3.5×10^{-12}	1.6×10^{-10}	3.5×10^{-11}	1.1×10^{-12}	2.6×10^{-13}	4.6×10^{-12}	1.0×10^{-12}
Brahmapuram 2020	7.6×10^{-12}	1.8×10^{-12}	4.8×10^{-11}	1.1×10^{-11}	6.6×10^{-13}	1.6×10^{-13}	5.4×10^{-12}	1.2×10^{-12}
Pettah	1.3×10^{-10}	3.0×10^{-11}	9.6×10^{-10}	2.1×10^{-10}	6.8×10^{-12}	1.6×10^{-12}	6.6×10^{-11}	1.5×10^{-11}
Attakulangara	6.1×10^{-11}	1.5×10^{-11}	2.6×10^{-09}	5.9×10^{-10}	7.2×10^{-12}	1.7×10^{-12}	1.5×10^{-11}	3.4×10^{-12}
Thakarapparambu	5.4×10^{-11}	1.3×10^{-11}	4.5×10^{-09}	1.0×10^{-09}	1.9×10^{-12}	4.5×10^{-13}	1.7×10^{-10}	3.8×10^{-11}

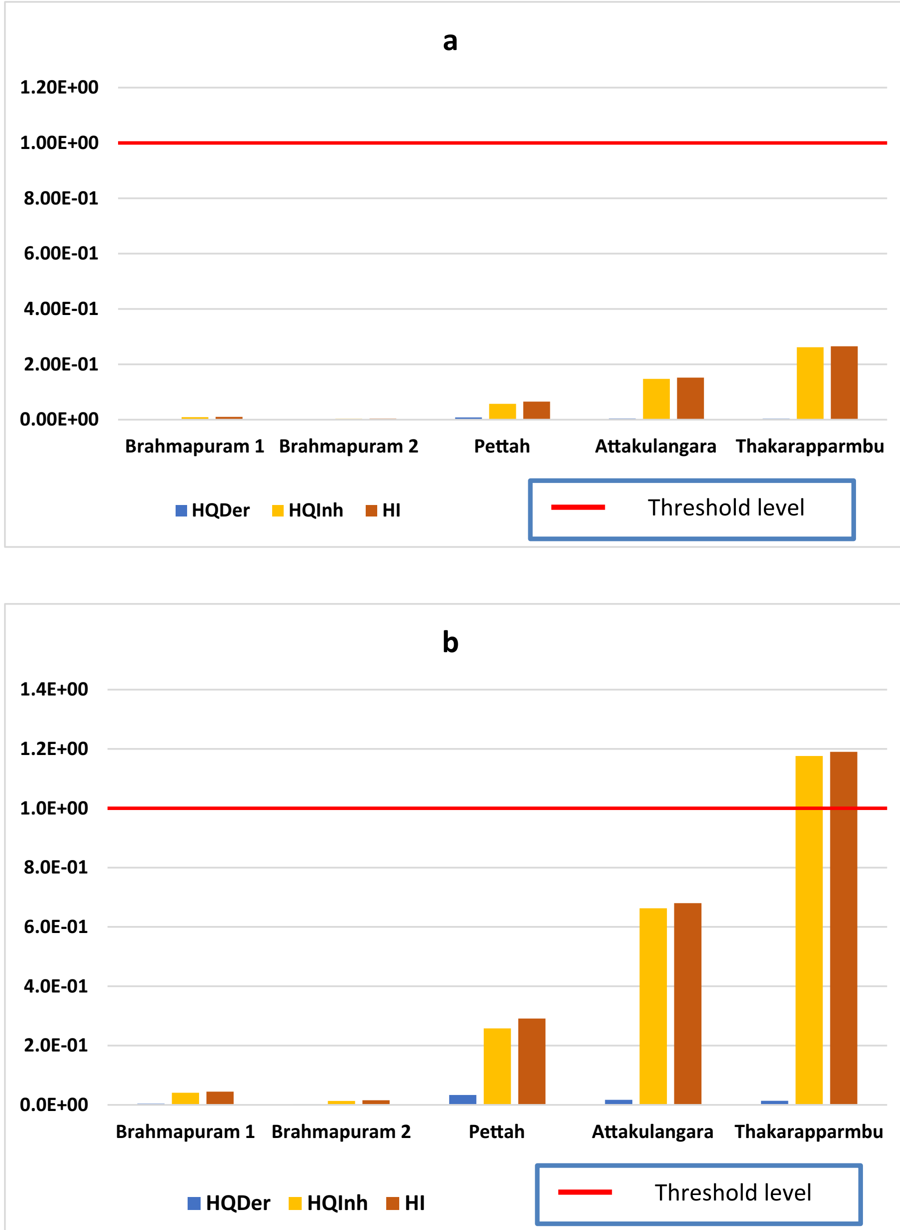

Fig. 4 **a**, **b** Hazard quotients (from dermal and inhalation routes) and hazard index from PCDD/F/dl-PCBs for adults and children at the study sites

found to be 0.7 also point towards significant non-carcinogenic effects to children underexposure. The highest *HI* for adults (0.3) was at Thakarapparambu which was much lower than the reference limit value indicating very low non-carcinogenic risk. The probability of non-carcinogenic risk arising from PCDD/Fs exposure during open burning is much higher than the dl-PCBs, as 90–99% of the cumulative hazard indices were contributed by PCDD/Fs.

Carcinogenic Risk: Incremental Life Cancer Risk (ILCR)

Probabilistic cancer risk was estimated which is a depiction of the number of people in a million equally exposed persons to develop cancer or cancer-linked illnesses over a lifetime (Fig. 5). The cancer risk from the dermal exposure of PCDD/Fs and dl-PCBs were found in the range of 3×10^{-7} to 5×10^{-6} whereas the inhalation exposure ranged from 2×10^{-6} to 2×10^{-4}. Hence, dermal exposure is accountable for only low to very low cancer risks, while that associated with inhalation was found to be in the moderate to low range. The highest ILCR values through inhalation and dermal exposure were found at Thakarapparambu and Pettah respectively. The trends of carcinogenic risks arising from exposure to PCDDs were found to be 10–100 times higher than dl-PCBs. The ILCRs values of both PCDD/Fs and dl-PCBs through dermal and inhalation pathways were summed together to

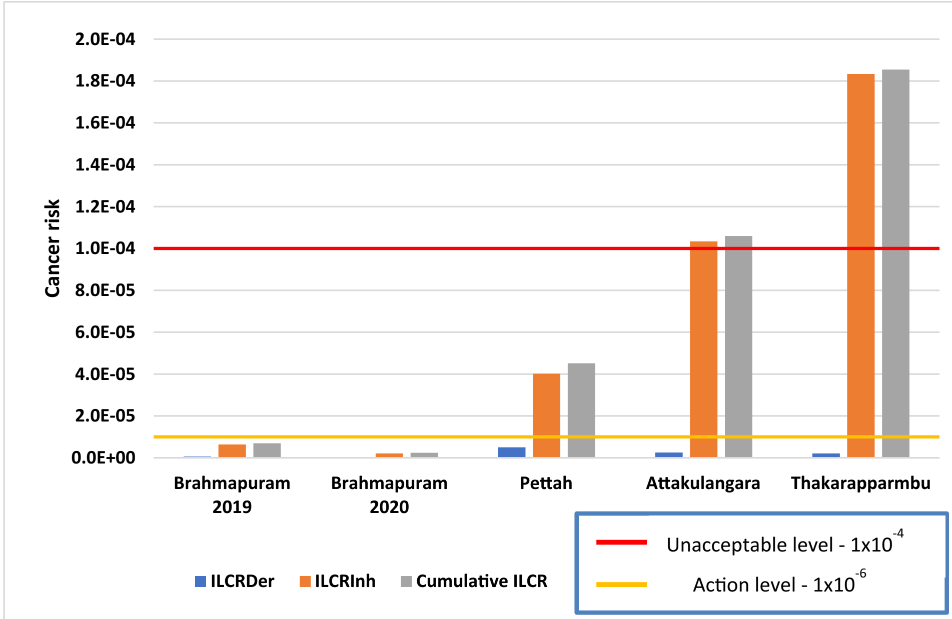

Fig. 5 Dermal, inhalation and cumulative incremental lifetime cancer risk (ILCRs) factors associated with PCDD/Fs and dl-PCBs exposures at the study sites

get the cumulative incremental lifetime cancer risk. The cumulative risk values ranged from 2×10^{-6} to 2×10^{-4} which indicates moderate to low risk to exposed individuals. As per USEPA, ILCR values lower than 1×10^{-6} indicates the probability of no additional cancer risk to the exposed community whereas ILCRs in the range of 1×10^{-6} to 1×10^{-4} indicate a moderate threat. The ILCR values higher than 1×10^{-4} suggests 'unacceptable' exposure conditions as per the ATSDR guidance manual and is likely to cause excess cancer risk to the community. At two of the street waste burning sites viz. Attakulangara and Thakarapparambu the ILCRs were higher than 1×10^{-4} and in all the other sites values were in the range of 1×10^{-6} to 1×10^{-4}. This indicates a moderate to unacceptable level of cancer risk exists at all the sites and requires in-depth studies such as long-term sampling programs and bio-monitoring studies for the further evaluation of the scenario.

Comparison of Carcinogenic Risk Assessment Studies

Assessment of cancer risk posed by dl-POPs from street waste burning based on real-time sampling studies was not available in the literature and hence, the findings of the present investigation were compared with a few studies which reported risk factors associated with MSW landfills (Table 7). The risk factors estimated in the present study was higher than most of the reported ones, as it reported the exposure risk based on the real-time sampling studies conducted during fire breakout incidents or street open burning activities whereas the former ones estimated risk in the vicinity of waste landfills from random sampling. A study conducted in Kenya on the risk assessment from open burning of MSW reported higher carcinogenic risk values and comparable non-carcinogenic risk with the present study (Shih et al. 2016). Another study on the risk factors associated with PCDD/Fs emission in a 3 km buffer zone of a non-hazardous waste landfill area reported the possibility

Table 7 Comparison chart showing estimated ILCR values of present study vis-à-vis elsewhere reported studies

Site	Study description	Non-carcinogenic risk	Carcinogenic risk	References
Montallegro, Italy	Solid waste landfill site—ambient air and soil within 3 km radius	1.4×10^{-9} to 2.5×10^{-7}	2.4×10^{-10} to 5.5×10^{-12}	Davoli et al. (2010)
Nairobi, Kenya	Open burning	0.02 to 0.54	2.1×10^{-4} to 5.8×10^{-6}	Shih et al. (2016)
Central Italy	Impact of MSW landfill site on local population	9.1×10^{-5} to 6.1×10^{-6}	1.4×10^{-8} to 2.2×10^{-9}	Palmiotto et al. (2014)
Catalonia, Spain	Impact of MSW landfill site on local population	<0.001	4×10^{-6} to 1×10^{-7}	Nadal et al. (2016a, b)
Kerala, India	Landfill fires	0.003 to 0.04	2×10^{-6} to 7×10^{-6}	Present study
Kerala, India	Street waste burnings	0.06 to 1.2	2×10^{-6} to 2×10^{-4}	Present study

of very low risk to the exposed community (Davoli et al. 2010). Similarly, few other studies on the impact of hazardous waste landfill sites to the nearby population reported lower ranges of risk factors (Palmiotto et al. 2014; Nadal et al. 2016a, b). ILCR reported at the street waste burning sites in the present study is the highest as per the comparison table (although the ingestion route is not included in the present study) and this is essentially due to the greater frequency of incidents and the possibility of lower radial dispersion of emitted dl-POPs.

Conclusions

The present study evaluated dl-POPs emissions from uncontrolled open burning of MSW and associated carcinogenic and non-carcinogenic risks through two pathways at selected streets and dumpyard sites in Kerala, India. The correlation studies elucidated the possibility of deriving congener 'fingerprints' for MSW open burning and points towards developing smart tools for source identification. The children were found to be susceptible to the non-carcinogenic effects at one site whereas adults were found to be in safe limits with comparatively lower hazard indices. The cumulative ILCR factors at the sites were in the range of 2×10^{-6} to 2×10^{-4} which is classified as low to moderate risk as per ATSDR guidelines. The major pathway of exposure was through inhalation (more than 90%) and PCDD/Fs accounted for 90% of the cumulative risk. The street waste littering and burning were found to pose more threats to human health than dumpyard fires due to their episodic nature, higher exposure probability (closeness to settlements and public places) and ground-level emissions resulting in minimum dispersive dilutions.

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Data Availability All authors herewith confirm that all data and materials as well as software application or custom code support the published claims and comply with field standards.

Code Availability Not applicable.

Declarations

Conflict of interest No potential conflicts of interest (financial or non-financial) is involved in the study.

Consent to Participate Not applicable.

Consent for Publication Not applicable.

Ethical Approval Not applicable.

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