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Occurrence of liquid crystal monomers in indoor and outdoor air particle matters (PM_{10}): Implications for human exposure indoors

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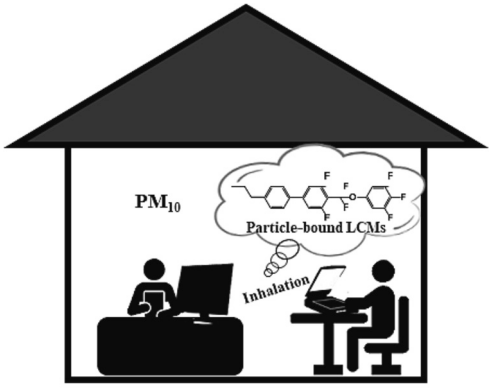
HIGHLIGHTS

- Indoor and outdoor PM_{10} LCMs in residential areas near and away from an e-waste area were analyzed.
- LCMs in indoor PM_{10} were significantly higher than those in outdoors.
- The release of various types of electronic products indoors is a non-negligible source of LCMs.
- Higher exposure doses of LCMs were observed indoors compared with outdoors.
- A low risk of exposure to LCMs via inhalation was found for adults and toddlers.

GRAPHICAL ABSTRACT



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ABSTRACT

Liquid crystal monomers (LCMs) are potentially persistent, bioaccumulating, and toxic substances. However, limited data are available on the occurrence of LCMs in indoor and outdoor air particle matter (PM_{10}) in residential areas. Herein, residential areas near an e-waste dismantling center (Guiyu Town, Shantou City), as well as areas away from the e-waste site (Jiedong District, Jieyang City) were selected as the sampling areas. PM_{10} was collected from the indoor environments of Guiyu (IGY) and Jieyang (IJY), as well as those from the outdoor environments (OGY and OJY) using the high-volume air samplers (TH-10000C). The levels of 57 LCMs in PM_{10} were analyzed, and the highest concentrations of LCMs were found in IGY (0.970–1080 pg/m^3), followed by IJY (2.853–455 pg/m^3), OGY (0.544–116 pg/m^3) and OJY (0.258–35.8 pg/m^3). No significant difference was

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observed for LCM levels in indoor PM_{10} between the two areas ($p > 0.05$), which were significantly higher than those in outdoors ($p < 0.05$), indicating that the release of electronic products in general indoor environments is a source of LCMs that cannot be ignored. The compositions of LCMs in outdoors were not consistent with those of indoors. The correlation analysis of individual LCMs suggested potential different sources to the LCMs in indoor and outdoor environments. The median daily intake values of Σ_{46}LCMs via inhalation were estimated as 0.440, 1.46×10^{-2}, 0.170 and 1.19×10^{-2} ng/kg BW/day for adults, and as 2.27, 2.60×10^{-2}, 0.880 and 2.10×10^{-2} ng/kg BW/day for toddlers, respectively, indicating much higher exposure doses of LCMs indoors compared with the outdoors, and much higher doses for toddlers compared with adults ($p < 0.05$). These results reveal the potentially adverse effects of LCMs on vulnerable populations, such as toddlers, in indoor environments.

1. Introduction

Liquid crystal displays (LCDs) are widely used for mobile phones, personal computer (PC), televisions, and solar panels (Liu et al., 2020; Liu et al., 2016). The total global shipments of large thin-film transistor liquid crystal displays (TFT-LCDs) reached 792.5 million units between July 2017 and July 2018, almost four times of those in 2012 (Li et al., 2007; Song et al., 2020). Liquid crystal monomers (LCMs), as the key materials of LCDs, are a class of synthetic organic chemicals with specific optical anisotropy embedded between layers of glass and electrical control elements (Geelhaar et al., 2013; Li et al., 2018). Generally, most LCMs consist of a diphenyl backbone and other functional groups, such as cyano-, fluorine-, or bromine- groups, and are usually classified into three categories according to their functional groups, i.e. biphenyls and analogs (BAs), cyanobiphenyls and analogs (CBAs), and fluorinated biphenyls and analogs (FBAs) (Li et al., 2018; Su et al., 2019). Only a few studies have investigated the toxicological effects of LCMs, which suggested that LCMs are emerging organic pollutants with potential persistence, bioaccumulation, and toxic properties (Li et al., 2021; Li et al., 2018; Su et al., 2019).

Since LCMs are not covalently bound to any of the base materials in LCDs, they are easily released into the environment during production, use, and recycling (Chen et al., 2017; Liang et al., 2021). As thus, LCMs have been frequently detected in a variety of environmental matrices associated with e-waste dismantling, including sediments (Su et al., 2021), dust (Cheng et al., 2022; Shen et al., 2022; Su et al., 2019; Yang et al., 2023), and air (Shen et al., 2022). In addition, LCMs volatilized from the use of various electronic products (e.g. mobile phones, computer and television displays) in the indoor environment may also be an important source of LCMs (Cheng et al., 2022; Feng et al., 2022; Liang et al., 2021; Liu and Abbatt, 2021; Shen et al., 2022; Zhu et al., 2021). LCMs are semi-volatile organic compounds (SVOCs), and air is the main medium for their release and transport from electronic devices to the surrounding environments (Shen et al., 2022). In air, due to the organics and the relatively large specific surface area of particulate matter, significant amounts of SVOCs will be adsorbed to the particulate matter. Hence, particulate matter (PM_{10}) could be an appropriate repository for SVOCs, which pose a high threat to human health.

Humans spend most of their time working or living indoors (Mercier et al., 2011), PM_{10} in the indoor environment is closely related to human health. Besides, the release of LCMs from outdoor sources, such as industrial emissions and human activities, is suspended in indoor and outdoor environments via atmospheric transport and other effects. Exposure to LCMs can occur when occupational workers and the general population carry LCMs from contaminated particulate matter into their bloodstream via inhalation (Chen and Hu, 2014; Chen et al., 2018; Zhang et al., 2021). LCMs have also been found in the serum of occupational workers and the general population (7.78–276 ng/mL) (Cheng et al., 2022; Li et al., 2022). However, there is still a lack of information concerning possible health effects of LCMs, and knowledge of the occurrence, fate and risk of human exposure to LCMs in PM_{10} remains generally limited. Therefore, there is an urgent need to investigate the pollution characteristics and human exposure to LCMs in indoor and outdoor PM_{10}.

Herein, indoor and outdoor ambient PM_{10} samples were collected from an e-waste dismantling area (Guiyu Town) and a reference residential area (Jieyang City). The major objectives were to: (1) investigate the levels and compositions of LCMs in PM_{10} in e-waste and reference residential areas, and (2) assess the risks of exposure to LCMs for adults and toddlers via respiratory inhalation. The results of this study provide basic data for assessing the occurrence of LCMs in indoor and outdoor environments and the health risks of human exposure to LCMs through respiration.

2. Materials and methods

2.1. Chemicals and reagents

A total of 57 LCMs (purity ≥97 %), including 9 BAs, 8 CBAs, and 40 FBAs, were selected as target chemicals, and were purchased from Maclean Biochemical Technology Co. (Shanghai, China), Aladdin Biochemical Technology Co., Ltd. (Shanghai, China), RHAWN (Shanghai, China), and Bidepharm (Shanghai, China). The details of the target analytes are presented in Table 1. As there are no isotopic-labelled internal standards (ISs) for LCMs, four surrogates—biphenyl-d_{10} (BPh-d_{10}), 2,2′,3,3′,4-Pentachlorobiphenyl (PCB-82), 2,2′,3,4,5,5′-Hexachlorobiphenyl (PCB-141), and 2,2′,3,3′,4,5,5′,6-Octachlorobiphenyl (PCB-198)—were chosen as the ISs for LCMs, and were purchased from Maclean Biochemical Technology Co.

All organic solvents were of high-performance liquid chromatography (HPLC) grade. Hexane (HEX), dichloromethane (DCM), toluene (TOL), acetone (ACE), ethyl acetate (ETAC), isooctane (ISO), and Florisil solid-phase extraction (SPE) cartridges (500 mg, 3 mL) were purchased from ANPEL Laboratory Technologies Inc. (Shanghai, China). The Milli-Q Integral system was supplied by Merck (Darmstadt, Germany).

2.2. Sampling area

Sampling campaigns were conducted in residential areas of Guiyu Town (Shantou City) and Jiedong District (Jieyang City) from October 17 to 24, 2021. Guiyu town in Guangdong Province, with a permanent population of 162,649, is one of the world's largest e-waste dismantling centers. Once there were thousands of small family-run workshops engage in recycling or dismantling e-wastes, including TVs, computers, solar panels, and cell phones, in a primitive manner (Chai et al., 2020). Jiedong District is located approximately 30 km from Guiyu Town, with a resident population of 1 million and there are no obvious sources of e-waste pollution around this area.

2.3. Sample collection

Sampling was carried out using high-volume air samplers (TH-10000C, Wuhan) operated at a constant flow rate (1.05 m^3/min) and equipped with a quartz fiber filter (QFF, 18.6 × 23.4 cm) to capture particle-bound SVOCs. The outdoor sampling sites of Guiyu (OGY) and Jieyang (OJY) were on the roofs of 4-storey buildings in residential areas, and approximately 12 m above the ground. For each particle sample, the sampling time was 24 h, and the sampling volume was

Table 1
Full names, abbreviations, chemical structures, CAS Nos, formulas, molecular weight (MW) and retention time (RT) of three classes of target LCMs, including 9 biphenyls/bicyclohexyls & analogues (BAs), 8 cyanobiphenyls & analogues (CBAs), and 40 fluorinated biphenyls & analogues (FBAs).

Full name	Abbreviation	CAS number	Formula	MW
Biphenyls/bicyclohexyls and Analogs, BAs				
1,1'-Bicyclohexyl,4-ethenyl-4′-propyl-, trans, trans	PVB (3VbcH)	116020-44-1	$C_{17}H_{30}$	234.42
trans, trans-4-(4-Methylphenyl)-4′-vinylbicyclohexyl	EBMB (MePVbcH)	155041-85-3	$C_{21}H_{30}$	282.463
trans-4-ethyl-4′-(4-propylcyclohexyl)-1,1′-biphenyl	EPB (3cH2B)	84540-37-4	$C_{23}H_{30}$	306.484
(trans,trans)-4-Butyl-4′-ethenyl-1,1′-bicyclohexyl	BVBC	153429-47-1	$C_{18}H_{32}$	248.447
1-[(trans, trans)-4′-(3-Buten-1-yl)[1,1′-bicyclohexyl]-4-yl]-4-methyl- benzene	MPhBB	129738-42-7	$C_{23}H_{34}$	310.516
(trans,trans)-4-(1*E*)-1-Propen-1-yl-4′-propyl-1,1′-bicyclohexyl	PPB (Pe3bcH)	279246-65-0	$C_{18}H_{32}$	248.447
4-(4-Methylphenyl)-4′-propyl-1,1′-bi (cyclohexyl)	MPCB	84656-75-7	$C_{22}H_{34}$	298.505
4-Methyl-4′-pentylbiphenyl	MPB (5MeB)	64835-63-8	$C_{18}H_{22}$	238.367
trans, trans-4-Propyl-4′-methoxybicyclohexyl	MOPB (MeO3bcH)	97398-80-6	$C_{16}H_{30}O$	238.409
Cyano biphenyls and Analogs, CBAs				
4-cyano-4′-ethoxybiphenyl	2OCB	58743-78-5	$C_{15}H_{13}NO$	223.27
4'-Propoxy-4-biphenylcarbonitrile	3OCB	52709-86-1	$C_{16}H_{15}NO$	237.296
trans-4-[4-[1-(E)-propenyl] cyclohexyl] benzonitrile	3eCHB	96184-40-6	$C_{16}H_{19}N$	225.329
4-Butyl-4′-cyanobiphenyl	4CB	52709-83-8	$C_{17}H_{17}N$	235.324
4-(4-Propylcyclohexyl) benzonitrile	3CHB	61203-99-4	$C_{16}H_{21}N$	227.345
4-(trans-4-Butylcyclohexyl) benzonitrile	4CHB	61204-00-0	$C_{17}H_{23}N$	241.371
4-(4-Ethylcyclohexyl) benzonitrile	EtCBN	73592-81-1	$C_{15}H_{19}N$	213.318
4′-(Octyloxy)-4-biphenylcarbonitrile	8OCB	52364-73-5	$C_{21}H_{25}NO$	307.429
Fluorinated biphenyls and analogs, FBAs				
4″-Ethyl-2′-fluoro-4-propyl-1,1′:4′,1″-terphenyl	EFPT (2F3T)	95759-44-7	$C_{23}H_{23}F$	318.427
1,2,3-Trifluoro-5-[(trans, trans)-4′-propyl[1,1′-bicyclohexyl]-4-yl] benzene	TPrCB	131819-23-3	$C_{21}H_{29}F_3$	338.45
4′-[(trans,trans)-4′-Ethyl[1,1′-bicyclohexyl]-4-yl]-3,4,5-trifluoro-1,1′- biphenyl	EBCTFB	137529-40-9 (web archive link,)	$C_{26}H_{31}F_3$	400.52
4-(3,4-Difluoro-phenyl)-4′-propyl-bicyclohexyl	DPrCB	82832-57-3	$C_{21}H_{30}F_2$	320.46
4-[2-(3,4-Difluorophenyl)ethyl]-4′-propyl-1,1′- bi(cyclohexyl)	DFEPBC	107215-66-7	$C_{23}H_{34}F_2$	348.513
trans, trans-4'-Propyl-4- (4-trifluoromethoxyphenyl) bicyclohexyl	PCTB (tFMeO-3bcHP)	133937-72-1	$C_{22}H_{31}F_3O$	368.476
3,4-Difluoro-4′-(trans-4-propylcyclohexyl) biphenyl	DPrB	85312-59-0	$C_{21}H_{24}F_2$	314.412
1-[4- (4-butylcyclohexyl) cyclohexyl]-4-ethoxy-2,3-difluoro -benzene	BCEDB (2OdFP4bcH)	473257-15-7	$C_{24}H_{36}F_2O$	378.539
trans-1-Ethoxy-2,3-difluoro-4-(4-propyl-cyclohexyl)-benzene	EDPrB (2O3cHdFP)	174350-05-1	$C_{17}H_{24}F_2O$	282.369
1,2,3-trifluoro-5-[2-fluoro-4-(4-propylphenyl) phenyl] benzene	TePT	205806-87-7	$C_{21}H_{16}F_4$	344.345
trans, trans-4-(3,4-Difluorophenyl)-4′-pentylbicyclohexyl	DPeCB	118164-51-5	$C_{23}H_{34}F_2$	348.513
trans, trans-4″-(4-ethoxy-2,3-difluoro-phenyl)-4-propyl-bicyclohexyl	EDPBB (2OdFP3bcH)	123560-48-5	$C_{23}H_{34}F_2O$	364.512
4′-(trans-4-Propylcyclohexyl)-2,3-difluoro-4-ethoxy-1,1′-biphenyl	EDPB (2O3cHdFB)	189750-98-9	$C_{23}H_{28}F_2O$	358.465
1,2,3-Trifluoro-5-[trans-4-[2-(trans-4-propylcyclohexyl) ethyl] cyclohexyl] benzene	TPrECB	131819-24-4	$C_{23}H_{33}F_3$	366.503
4′-(trans-4-Propylcyclohexyl)-3,4,5-trifluorobiphenyl	TrPrB	132123-39-8	$C_{21}H_{23}F_3$	332.403
3,4-Difluoro-4′- [(trans, trans)-4′-pentyl [1,1′- bicyclohexyl]-4-yl] biphenyl	DPeBB (5bcHdFB)	136609-96-6	$C_{29}H_{38}F_2$	424.609
trans,trans-4′-(4'-Pentyl-bicyclohexyl-4-yl)-3,4,5-trifluorobiphenyl	TPeBB	137529-43-2	$C_{29}H_{37}F_3$	442.599
trans, trans-3,4,5-Trifluoro-4′-(4′- propylbicyclohexyl-4-yl) biphenyl	TPrBB	137529-41-0	$C_{27}H_{33}F_3$	414.546
3,4-Difluoro-4′-[(trans,trans)-4′-propyl [1,1′-bicyclohexyl]-4-yl] biphenyl	DPrBB (3bcHdFB)	119990-81-7	$C_{27}H_{34}F_2$	396.556
trans, trans-4-(3,4-Difluorophenyl)-4′-butylbicyclohexyl	DBCB	82832-58-4	$C_{22}H_{32}F_2$	334.486
1,2-Difluoro-4-[trans-4-(trans-4-ethylcyclohexyl) cyclohexyl] benzene	DECB	118164-50-4	$C_{20}H_{28}F_2$	306.433
trans, trans-4-(4-Fluorophenyl)-4′-propylbicyclohexyl	FPCB	82832-27-7	$C_{21}H_{31}F$	302.469
trans, trans-4-(3,4-Difluorophenyl)-4′-vinylbicyclohexyl	DFPVBC	142400-92-8	$C_{20}H_{26}F_2$	304.417
trans-4-(4-Propylcyclohexyl)-4′-fluorobiphenyl	FPB	87260-24-0	$C_{21}H_{25}F$	296.422
1-[4-(4-ethylcyclohexyl) cyclohexyl]-4-(trifluoromethoxy) benzene	ECTB	135734-59-7	$C_{21}H_{29}F_3O$	354.45
2-Fluoro-4-(trans-4-pentylcyclohexyl)-4′-(trans-4-propylcyclohexyl) biphenyl	FPePrB	106349-49-9	$C_{32}H_{45}F$	448.698
1-ethoxy-2,3-difluoro-4-(4-pentylcyclohexyl) benzene	EDPeB	124729-02-8	$C_{19}H_{28}F_2O$	310.422
3,4-Difluoro-4′-(trans-4-pentylcyclohexyl) biphenyl	DPeB	134412-17-2	$C_{23}H_{28}F_2$	342.465
3,4-Difluoro-4′-(trans-4-ethylcyclohexyl) biphenyl	DFECB	134412-18-3	$C_{20}H_{22}F_2$	300.385
3,4,5-Trifluoro-1-[trans-4′-(trans-4″-pentylcyclohexyl)-cyclohexyl]-benzene	TPeCB	137644-54-3	$C_{23}H_{33}F_3$	366.503
4-[(trans, trans)-4′-(3-Buten-1-yl)[1,1′-bicyclohexyl]-4-yl]-1,2-difluoro- benzene	BBDB	155266-68-5	$C_{22}H_{30}F_2$	332.47
2′,3,4,5-Tetrafluoro-4′-(trans-4-propylcyclohexyl) biphenyl	TePrB	173837-35-9	$C_{21}H_{22}F_4$	350.393
[Trans(trans)]-1-(4′-ethyl[1,1′-bicyclohexyl]-4-yl)-2,3-difluoro-4-methylbenzene	EBDMB	174350-08-4	$C_{21}H_{30}F_2$	320.46
1-Butoxy-2,3-difluoro-4-(trans-4-propylcyclohexyl) benzene	BDPrB	208709-55-1	$C_{19}H_{28}F_2O$	310.422
2'-Fluoro-4-Pentyl-4"-Propyl-1,1′:4′,1"-Terphenyl	PFPT	95759-51-6	$C_{26}H_{29}F$	360.507
4-[Difluoro(3,4,5-trifluorophenoxy) methyl]-3,5-difluoro-4′-propyl-biphenyl	DTMDPB (tFPO-CF2-Df3b)	303186-20-1	$C_{22}H_{15}F_7O$	428.343
1-[4-(4-butylcyclohexyl) cyclohexyl]-4-ethoxy-2,3-difluoro-benzene	BCEDB (2OdFP4bcH)	473257-15-7	$C_{24}H_{36}F_2O$	378.539
4-[Difluoro(3,4,5-trifluorophenoxy) methyl]-3,5- difluoro-4′-ethyl-biphenyl	DTMDEB	303186-19-8	$C_{21}H_{13}F_7O$	414.316
2,3-Difluoro-1-methoxy-4-[(trans-4-propylcyclohexyl) methoxy] benzene	DMPMB	1373116-00-7	$C_{17}H_{24}F_2O_2$	298.368
3,4,5-Trifluoro-4′-(trans-4-pentylcyclohexyl)biphenyl)	TrPeB	137019-95-5	$C_{23}H_{27}F_3$	360.456
1,2-Difluoro-4-[trans-4-[2-(trans-4-propylcyclohexyl)ethyl] cyclohexyl]-benzene	DPrECB	117943-37-0	$C_{23}H_{34}F_2$	348.513
Internal standards, ISs				
D10-Biphenyl	BPh-d10	1486-01-7	$C_{12}D_{10}$	164.27
2,2′,3,3′,4-Pentachlorobiphenyl	PCB82	52663-62-4	$C_{12}H_5Cl_5$	326.433
2,2′,3,4,5,5'-Hexachlorobiphenyl	PCB141	52712-04-6	$C_{12}H_4Cl_6$	360.88
2,2′,3,3′,4,5,5′,6-Octachlorobiphenyl	PCB198	68194-17-2	$C_{12}H_2Cl_8$	429.77
Recovery standards, RSs				
2,3,6-Trichlorobiphenyl	PCB24	55702-45-9	$C_{12}H_7Cl_3$	257.543

1480.15 m^3. Indoor sampling sites of Guiyu (IGY) and Jieyang (IJY) were set up in the corresponding residential areas, and the sampling time ranged from 12 to 24 h. A total of 24 PM_{10} samples were collected from the e-waste dismantling area (16 indoors and 8 outdoors), and 23 PM_{10} samples were collected from the reference residential area (16 indoors and 7 outdoors). Blank samples were set during the sampling period to monitor background pollution.

Prior to the sampling campaigns, QFFs were baked at 450 °C for 6 h to remove any organic contaminant. The QFFs were wrapped in aluminum foil and sealed in polyethylene bags. After sampling, loaded QFFs were wrapped in aluminum foils and sealed in double-layer polyethylene bags, and stored at −20 °C until analysis. Concentrations of PM_{10} were determined by weighing QFFs before and after sampling at room temperature with a relative humidity (RH) of <30 %.

2.4. Sample analysis

The pretreatment procedures for the QFF samples were conducted according to those reported in previous studies (Shen et al., 2022; Tang et al., 2020), with minor modifications. Briefly, the collected QFF was cut into pieces, spiked with a mixture of IS solution (BPh-d_{10}, PCB-82, PCB-141, and PCB-198 each 20 ng) in a pre-clean glass tube, and ultrasonically extracted twice with 2.5 mL ACE/n-hex (3:1) and 0.5 mL toluene for 10 min. The extract was separated by centrifugation at 3000 rpm for 3 min, and the supernatant was transferred into a clean glass tube. The extracts were combined, concentrated to 1 mL, and purified using a Florisil SPE cartridge (500 mg, 3 mL) that preconditioned with 4 mL of ACE, 6 mL of ETAC, and 6 mL of HEX. After sample loading, the target LCMs were co-eluted with 8 mL of HEX/DCM (1/1). The eluates were then concentrated to near dryness under N_2 blowing, and the residues were reconstituted with 200 μL of ISO, transferred to a glass vial, and stored at −20 °C until instrumental analysis.

Quantification of the 57 target LCMs was performed using gas chromatography–tandem mass spectrometry in the electron impact (EI) ionization mode (GC–MS/MS, Agilent Technologies, USA), and analysis was performed in the multiple reaction monitoring (MRM) mode. A DB-5MS column (30 m × 0.25 mm × 0.25 μm, Agilent Technologies, Santa Clara, CA, USA) was used for separation. The injection volume was 1 μL, with high-purity helium as the carrier gas and a constant flow rate of 1 mL/min. The settings of the temperature program refer to those in a previous study (Shen et al., 2022): the temperature was maintained at 100 °C for 1 min, raised to 200 °C at 10 °C/min, to 240 °C at 3 °C/min for 1 min, and lastly to 300 °C for 16 min at 15 °C/min. The MS parameters of the LCMs are listed in Table S1 of the Supplementary Information (SI).

2.5. Estimation of exposure dose and risk

In this study, the estimated daily intake (EDI; ng/kg BW /day) of LCMs via inhalation for the occupational and general populations in Guiyu and Jieyang was calculated using Eq. (1) (Besis et al., 2016; Peng et al., 2011; Zhang et al., 2022).

$$EDI_{inh} = \frac{C \times R_{inh} \times EF}{BW} \quad (1)$$

where EDI_{inh} is the estimated chemical intake through inhalation (ng/kg BW /day); C is the total concentration(ng/m^3) of LCMs in the air; R_{inh} is the inhalation rate for adults (20 m^3/day) or toddlers (5 m^3/day) (Peng et al., 2011); BW is the average body weight for adults (70 kg) or toddlers (15 kg) (Peng et al., 2011); and EF is the exposure frequency in a day for adults (indoor = 19.3 h/day, outdoor = 4.68 h/day) or toddlers (indoor = 21.3 h/day, outdoor = 1.78 h/day) (Zhang et al., 2022).

The health risks of LCM exposure to adults and toddlers via inhalation were calculated using the Hazard Quotient (HQ). As there is no reference maximum intake dose of LCMs for adults and toddlers, the tolerable daily intake (TDI) was used to calculate the HQs (Eq. (2)) (Yang et al., 2023). The specific TDI values of the LCMs are listed in Table S2.

$$HQ = \frac{EDI}{TDI} \quad (2)$$

2.6. Quality assurance and quality control

To ensure the accuracy and precision of the analytical processes, three spiked blanks and three spiked matrix (pre-cleaned QFFs) samples were analyzed using the same methods as for sample pretreatment. The recoveries of target chemicals in spiked matrix were 69.8 %–141 % for BAs, 65.5 %–102 % for CBAs, and 80.1 %–143 % for FBA, with the relative standard deviations (RSDs) < 10 %. For the ISs, the recoveries were 61.6 ± 1.94 %, 82.0 ± 3.24 %, 82.0 ± 3.03 %, and 80.5 ± 3.42 % for BPh-d_{10}, PCB82, PCB141, and PCB198, respectively, with the RSD < 10 %. The specific recoveries and RSD of individual LCMs and ISs are listed in Table S3.

Moreover, one procedural blank sample (precleaned QFF) was analyzed every 12 samples to assess potential contamination and the average level of target chemicals detected in the QFF samples minus the procedural blank. The limit of quantitation (LOQ) was defined as the mean value in procedure blanks multiplied by three times the standard deviation for each analyte that was detected in the procedural blank samples; for analytes that were not detected (ND) in the procedural blank samples, the LOQ was defined as a signal/noise ratio of 10 (S/N = 10) (Tang et al., 2020). LOQs of BAs, CBAs and FBAs were 0.02–4.22, 0.01–1.39, and 0.01–1.93 pg/m^3, respectively.

2.7. Statistics analysis

Statistical analysis of chemicals with detection frequency (DF) > 50 % was conducted using SPSS 22 software for Windows (SPSS, Inc., USA). Chemicals with concentrations below the LOQ were assigned to values of half the LODs. Differences in concentration levels and exposure dose for LCMs in different regions were analyzed using the Kruskal-Wallis H test; correlations between chemicals were performed using Spearman correlation analysis.

3. Results and discussion

In this study, except for MOPB, BVBC, 4CB, DPrB, DPrECB, TPeCB, DFEPBC, DPeB, EBCTFB, DPrBB, and TPeBB, 46 LCMs were detected in the PM_{10} samples, including 7 BAs, 7 CBAs, and 32 FBAs (Table 2), suggesting the wide presence of LCMs in indoor and outdoor environments. The levels of FBAs and BAs detected in most samples were much higher than those of CBA, which is consistent with the results of previous studies (Liang et al., 2021; Shen et al., 2022).

3.1. Levels of LCMs

The total concentrations of Σ_{46}LCMs detected in indoor and outdoor PM_{10} in the two areas ranged from 0.258 to 1695 pg/m^3 (median 8.33 pg/m^3). Specifically, the levels of Σ_{46}LCMs were 0.970–1680 pg/m^3 (median 38.9 pg/m^3) in IGY, 2.85–455 pg/m^3 (median 26.4 pg/m^3) in IJY, 0.544–116 pg/m^3 (median 5.84 pg/m^3) in OGY, and 0.258–35.8 pg/m^3 (median 4.88 pg/m^3) in OJY (Table 2). The levels of LCMs in indoor PM_{10} samples from Guiyu and Jieyang were significantly higher than those in the corresponding outdoor environments (Kruskal-Wallis test, $p < 0.05$). There are three possible explanations for this finding. First, LCMs are mainly released by the displays of electrical and electronic products containing LCMs, such as computers, mobile phones, and TVs (Liu and Abbatt, 2021), and there are more potential sources of LCMs in indoor environments than in outdoor environments. Second, LCD products (e.g., TVs, computers) may be used more frequently in indoor environments than outdoors. Third, indoor environments have

Table 2
Descriptive statistics of concentrations (pg/m^3) of LCMs in indoor and outdoor from Guiyu (IGY, n = 16; OGY, n = 8) and Jieyang (IJY, *n* = 16; OJY, *n* = 7).

Compound	IGY(*n* = 16)			OGY(*n* = 8)			IJY(n = 16)			OJY(n = 7)		
	DF(%)	Median	Range	DF(%)	Median	Range	DF(%)	Median	Range	DF(%)	Median	Range
PVB	68.8	10.7	ND-184	ND	–	–	62.5	20.2	ND-121	0	–	–
PPB	6.25	ND	ND-3.29	ND	–	–	0	–	–	0	–	–
MPB	6.25	ND	ND-0.28	12.5	ND	ND-0.18	6.25	ND	ND-0.78	0	–	–
EBMB	25.0	ND	ND-361	62.5	2.01	ND-71.0	37.5	ND	ND-98.1	14.3	ND	ND-4.77
MPCB	0	–	–	ND	–	–	0	–	–	14.3	ND	ND-0.73
MPhBB	12.5	ND	ND-1.37	ND	–	–	6.25	ND	ND-4.68	0	–	–
EPB	6.25	ND	ND-0.09	ND	–	–	0	–	–	0	–	–
Σ_7BAs	**–**	**10.7**	**ND-550**	**–**	**2.01**	**ND-71.2**	**–**	**20.2**	**ND-225**	**–**	**–**	**ND-5.50**
EtCBN	25.0	–	ND-16.9	12.5	ND	ND-12.8	12.5	ND	ND-4.27	14.3	ND	ND-10.8
3CHB	0	–	–	25.0	ND	ND-1.46	0	–	–	0	–	–
3eCHB	56.3	1.49	ND-14.5	ND	–	–	18.8	ND	ND-7.08	0	–	–
4CHB	6.25	ND	ND-4.97	12.5	ND	ND-2.73	6.25	ND	ND-4.88	14.3	ND	ND-2.19
2OCB	68.8	0.24	ND-28.2	25.0	ND	ND-1.67	43.8	ND	ND-10.3	0	–	–
3OCB	25.0	ND	ND-2.12	50.0	0.35	ND-2.18	6.25	ND	ND-1.44	85.7	1.85	ND-2.30
8OCB	6.25	ND	ND-0.29	ND	–	–	6.25	ND	ND-0.97	28.6	ND	ND-0.23
Σ_7CBAs	**–**	**1.73**	**ND-67.0**	**–**	**0.35**	**ND-20.8**	**–**	**–**	**ND-28.9**	**–**	**1.85**	**ND-15.5**
EDPrB	18.8	ND	ND-0.31	ND	–	–	12.5	ND	ND-0.35	42.9	ND	ND-0.73
DMPMB	93.8	0.55	ND-2.54	87.5	0.74	ND-1.40	68.8	0.52	ND-3.35	14.3	ND	ND-0.50
BDPrB	12.6	ND	ND-0.75	12.5	ND	ND-0.05	12.5	ND	ND-0.14	14.3	ND	ND-0.19
EDPeB	12.6	ND	ND-4.54	12.5	ND	ND-0.43	6.25	ND	ND-0.37	0	–	–
ECTB	18.8	ND	ND-0.41	12.5	ND	ND-1.17	0	–	–	14.3	ND	ND-0.47
DFPVBC	81.3	18.70	ND-439	50.0	1.64	ND-12.4	31.3	ND	ND-16.3	57.1	2.38	ND-7.25
DECB	25.0	ND	ND-25.9	12.5	ND	ND-4.09	12.5	ND	ND-2.96	0	–	–
DTMDEB	25.0	ND	ND-0.48	37.5	ND	ND-0.06	6.25	ND	ND-0.02	0	–	–
DFECB	18.8	ND	ND-0.22	12.5	ND	ND-0.25	0	–	–	0	–	–
PCTB	18.8	ND	ND-0.71	37.5	ND	ND-1.00	25.0	ND	ND-1.83	28.6	ND	0.88
TPrCB	18.8	ND	ND-0.59	0	–	–	6.25	ND	ND-0.72	0	–	–
TePrB	12.5	ND	ND-0.09	0	–	–	0	–	–	0	–	–
FPCB	18.8	ND	ND-0.70	0	–	–	6.25	ND	ND-0.24	0	–	–
DPrCB	6.25	ND	ND-1.89	0	–	–	0	–	–	0	–	–
EBDMB	12.5	ND	ND-40.3	0	–	–	6.25	ND	ND-2.82	14.3	ND	2.50
TrPrB	18.8	ND	ND-0.29	25.0	ND	ND-0.14	0	–	–	14.3	ND	0.04
DTMDPB	81.3	0.15	ND-1.49	100	0.07	0.01–0.22	68.8	0.02	ND-0.61	71.4	0.02	0.03
FPB	0	ND	–	12.5	ND	ND-0.36	31.3	ND	ND-2.01	14.3	ND	0.13
TePT	43.8	ND	ND-0.19	37.5	ND	ND-0.04	25.0	ND	ND-0.05	28.6	ND	0.03
BBDB	18.8	ND	ND-1.37	12.5	ND	ND-0.57	12.5	ND	ND-1.19	0	–	–
DBCB	0	–	–	0	–	–	12.5	ND	ND-0.98	0	–	–
TPrECB	12.5	ND	ND-8.27	0	–	–	6.25	ND	ND-3.58	0	–	–
DPeCB	6.25	ND	ND-0.32	25.0	ND	ND-0.08	6.25	ND	ND-0.05	0	–	–
TrPeB	6.25	ND	ND-0.69	0	–	–	0	–	–	0	–	–
EDPBB	100	5.41	0.97–529	100	1.02	0.53–1.38	100	5.67	2.85–162	100	0.64	0.21–2.01
EFPT	25.0	ND	ND-0.13	25.0	ND	ND-0.06	12.5	ND	ND-0.05	14.3	ND	ND-0.01
EDPB	12.5	ND	ND-1.39	12.5	ND	ND-0.15	0	–	–	0	–	–
BCEDB	0	–	–	0	–	–	6.25	ND	ND-0.04	0	–	–
PFPT	6.25	ND	ND-0.02	0	–	–	18.8	ND	ND-0.10	14.3	ND	ND-0.01
TPrBB	43.8	ND	ND-1.44	37.5	ND	ND-0.04	12.5	ND	ND-0.04	14.3	ND	ND-0.01
DPeBB	37.5	ND	ND-0.76	25.0	ND	ND-0.14	6.25	ND	ND-0.17	0	–	–
FPePrB	0	–	–	0	–	–	6.25	ND	ND-0.30	0	–	–
Σ_{32}FBAs	**–**	**24.8**	**0.97–1060**	**–**	**3.48**	**0.54–24.0**	**–**	**6.21**	**2.85–201**	**–**	**3.03**	**0.26–14.8**
Σ_{46}LCMs	**–**	**38.9**	**0.97–1680**	**–**	**5.84**	**0.54–116**	**–**	**26.4**	**2.85–455**	**–**	**4.88**	**0.26–35.8**

ND: Not detected (> LOQs).

limited space and relatively low air exchange and circulation rates, which are not conducive to the diffusion of air pollutants (Tao et al., 2016; Zhao et al., 2020). The results of this study are consistent with those of a previous study (Shen et al., 2022), where the concentrations of LCMs in air samples from e-waste disassembly areas were found to be much higher in indoor environments (68800–385,000 pg/m^3) than in outdoor environments (15000–43,400 pg/m^3) (Shen et al., 2022). However, the concentrations of LCMs in PM_{10} indoors in Guiyu in the present study were much lower than those in a previous study (2696–14,200 pg/m^3) (Shen et al., 2022), as the air samples in that study were within an industrial park where e-waste is dismantled, whereas the sampling site in the present study was a residential area around the e-waste dismantling industrial park. The dispersion of LCMs from e-waste dismantling activities is subject to atmospheric dilution and other influences, resulting in concentrations of LCMs higher than those in surrounding areas. Although LCM levels in PM_{10} were low in both indoor and outdoor environments in this study, LCM pollution in indoor environments still needs to be taken seriously considering the potential health hazards of LCM and the fact that people spend most of their time indoors. Besides, the levels of LCMs varied in indoor PM_{10} in the present study, which could be related to the usage of electric equipment and the cleaning frequency in individual residencies, and require further investigation in the future.

No significant difference was observed in the levels of LCMs in indoor and outdoor environments, respectively, between the two sites in this study, despite the fact that IGY (0.970–1060 pg/m^3) had a greater range of LCM concentrations (Kruskal-Wallis test, $p > 0.05$). This indicates that in addition to e-waste dismantling activities, the release of LCMs in residential indoor environments during use should not be neglected. This further confirms that the release of LCM-containing products in indoor environments is an important source of LCMs (Liu and Abbatt, 2021).

As shown in Table 2 and Fig. 1, the relative proportions of BAs and FBAs were much higher than those of CBAs, overall, the contribution of

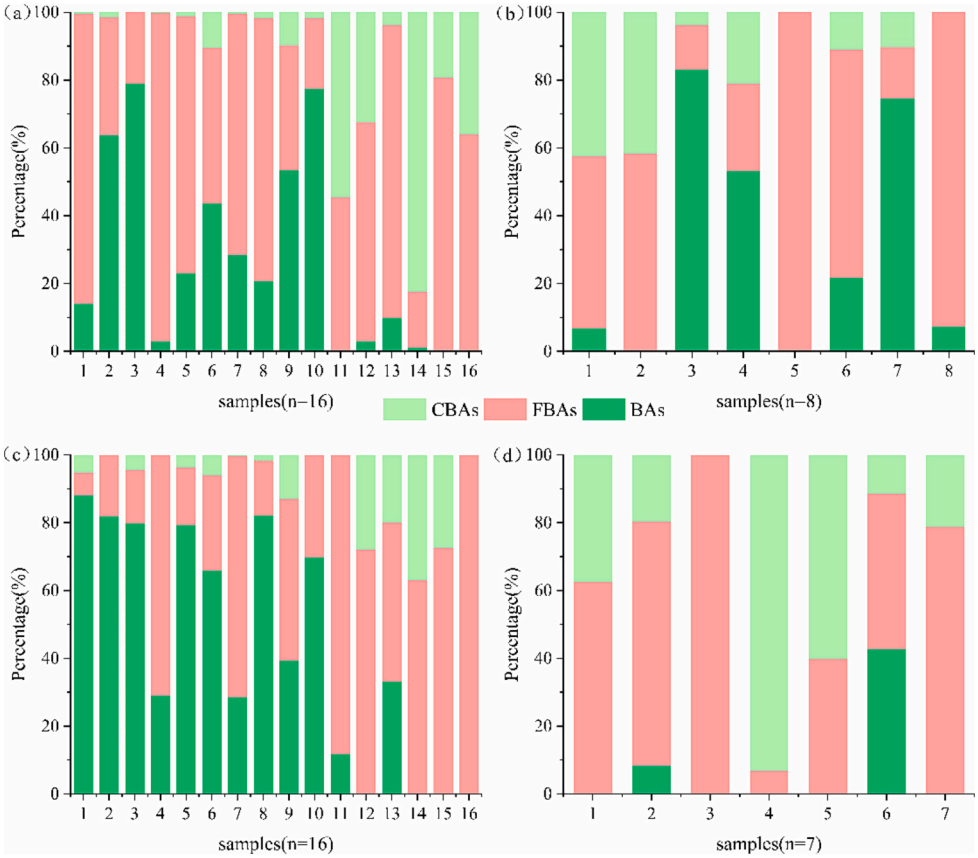


Fig. 1. Relative contributions of the three types of LCMs in indoor and outdoor PM_{10} from residential areas in e-waste recycling area (indoor: IGY, outdoor: OGY) and general residential areas area (indoor: IJY, outdoor: OJY), (a) IGY, (b) OGY, (c) IJY, and (d) OJY.

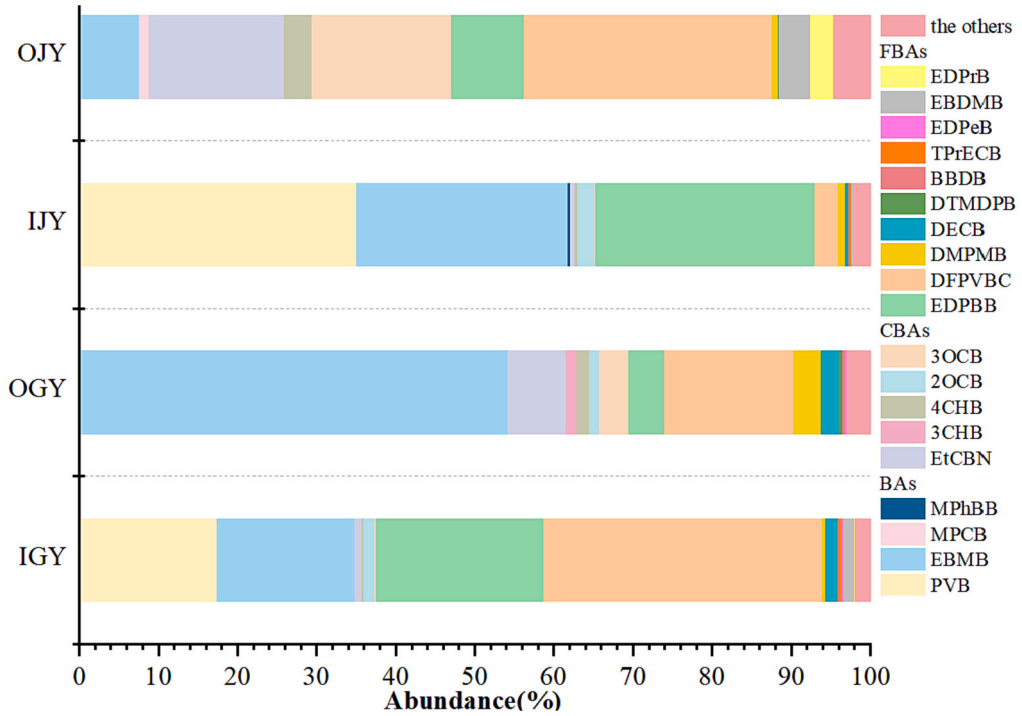


Fig. 2. Compositions of LCMs in indoor and outdoor PM_{10}samples from residential areas in e-waste recycling area (indoor: IGY, outdoor: OGY) and general residential areas area (indoor: IJY, outdoor: OJY).

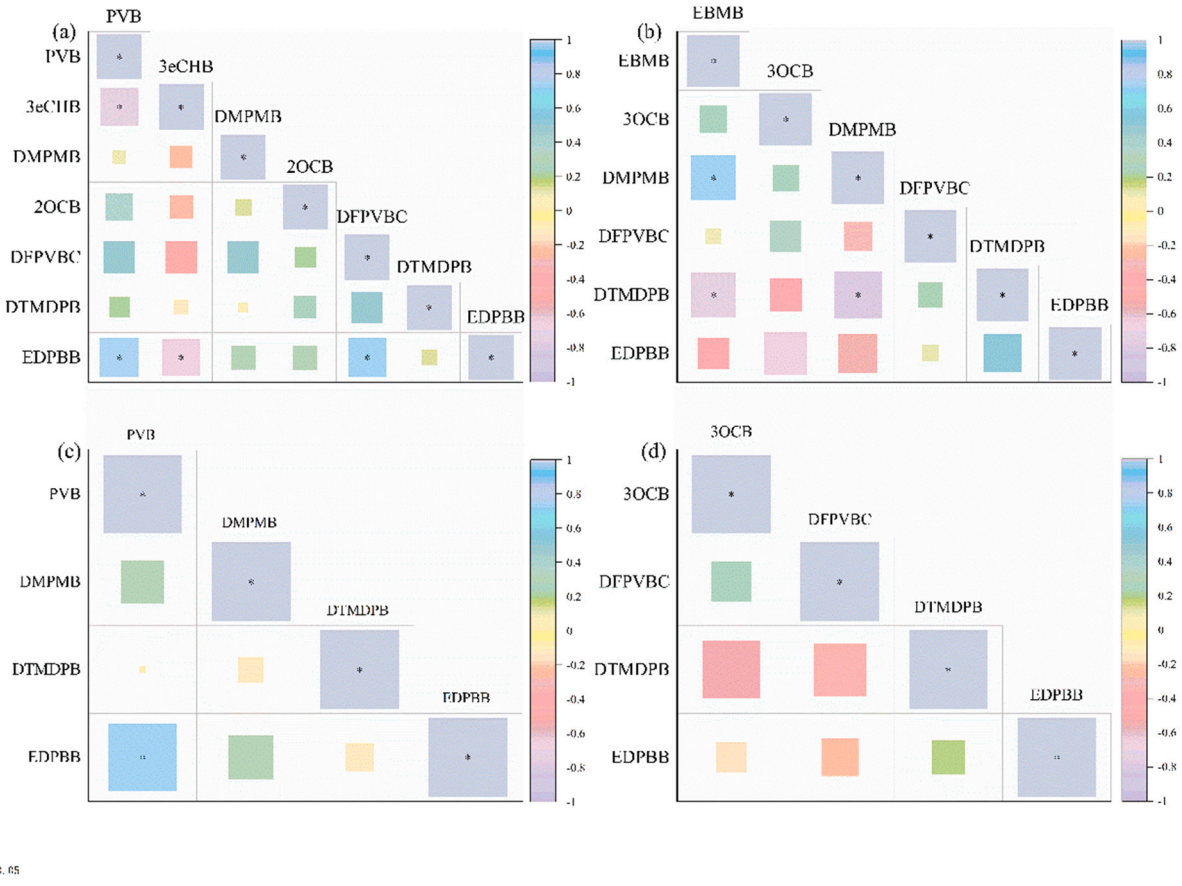
$\sum_7$CBAs to $\sum_{46}$LCMs was negligible in both IGY and IJY samples; similar results have been found in previous studies (Liang et al., 2021; Shen et al., 2022). In products from approximately 15 years ago, CBAs were found only in certain brands of computer/television panels (Liang et al., 2021). However, the contribution of $\sum_7$CBAs to $\sum_{46}$LCMs was 15.3 % and 38.8 % in the OGY and OJY samples, respectively, indicates that the application of CBAs in next-generation LCM products may be gradually expanding. In addition, different study areas, sampling seasons(Tian et al., 2011) and particle sizes may have an impact on the results of these studies (Allen et al., 2008; Cao et al., 2012; Ruby and Lowney, 2012; Tian et al., 2011).

3.2. Compositions and correlation of LCMs

The composition profiles of the individual LCMs in IGY, OGY, IJY and OJY are shown in Fig. 2. DFPVBC (35.3 %) was the most abundant LCM observed in IGY samples, followed by EDPBB (21.1 %), EBMB (17.4 %), and PVB 17.3 %. These four dominant chemicals collectively contributed to 91 % of the LCMs in IGY samples. The most abundant LCM in the OGY samples was EBMB, accounting for >50 %, followed by DFPVBC (16.3 %), EtCBN (7.33 %), EDPBB (4.52 %), 3OCB (3.76 %), and DMPMB (3.45 %). Major LCMs in the OGY samples were generally similar to those in the IGY samples, whereas the outdoor samples contained higher proportions of CBAs than the indoor samples (Fig. 2).

In addition, the results of the correlation analysis showed a significant positive correlation between PVB and EDPBB, and between PVB and EDPBB in the IGY samples ($p < 0.05$) (Fig. 3), suggesting that these LCMs have similar release sources. However, significant negative correlations between 3eCHB and PVB ($R = -0.787$, $p < 0.01$), and between 3eCHB and EDPBB ($R = -0.666$, $p < 0.05$) were found for IGY samples, suggesting that 3eCHB and the other two LCMs may not be co-added to the same LCD products. Besides, no significant correlation was found between the above LCMs in the OGY samples, implying they may have different sources. However, there was a significant positive correlation between EBMB and DMPMB ($R = 0.732$, $p < 0.05$), and a significant negative correlation between DTMDPB and DMPMB ($R = -0.810$, $p < 0.05$), and between DTMDPB and EBMB ($R = -0.732$, $p < 0.05$). It is hypothesized that LCMs in outdoor and indoor PM_{10} in Guiyu may have a similar source, that is, e-waste dismantling. However, because of the more complex environmental conditions (light, rain, and wind) in the outdoor environment, atmospheric transport and dilution, and the different lifetimes and persistence of individual LCMs in the atmosphere (Su et al., 2019), the environmental fates of LCMs in PM_{10} samples from indoor and outdoor environments differ (Su et al., 2016). This could be one of the main reasons why the major LCMs in outdoor environments are not necessarily dominant in indoor environments.

The main LCMs in the IJY samples were PVB (35.0 %), followed by EDPBB (27.7 %), EBMB (26.8 %) and DFPVBC (2.93 %). The major LCMs in the OJY samples were DFPVPC (31.3 %), 3OCB (17.6 %), EtCBN (17.1 %), EDPBB (9.23 %), EBMB (7.57 %) and EBDMB (3.96 %), which were significantly distinct from the composition of LCMs in the OJY samples. The results of correlation analysis of the LCMs for the IJY and OJY samples are shown in Fig. 3. In the IJY samples, there was a significant positive correlation between PVB and EDPBB ($R = 0.750$, $p < 0.01$), which is consistent with the findings of a previous study (Liang et al., 2021), in which PVB and EDPBB were present together in several different brands of TV-LCD panels. However, there were no significant correlations between the LCMs in the OJY samples. These results indicate that the LCMs in the OJY samples did not originate indoors but were the result of a combination of contamination from multiple sources in outdoor environments. The primary LCMs observed in this study were PVB, EBMB, EDPBB, EBDMB and DFPVBC. In previous studies, PVB,



* p< 0.05

Fig. 3. Correlation between the concentrations of LCMs in indoor and outdoor PM_{10}samples from Guiyu and Jieyang. (a) IGY, (b) OGY, (c) IJY, and (d) OJY.

EBMB, EDPBB, and EBDMB showed high contributions to various environmental matrices, such as indoor dust (Cheng et al., 2022; Su et al., 2019), air (Shen et al., 2022), and sediments (Su et al., 2021). However, DFPVBC, which had high concentrations and DF in this study, studies on DFPVBC are scarce, and its presence of has not been reported in any environmental matrices, and yet DFPVBC was found in the serum of occupational population engaged in e-waste dismantling and in the general population (Li et al., 2022). The different target LCM analyzed in these studies may also be one of the reasons for the lack of detection of DFPVBC in the environmental matrix in previous studies. The results of this study indicating that they are commonly used in LCMs products as important LCMs. Therefore, attention should be paid to widespread contamination and the risk of human exposure to these LCMs in the general environment.

3.3. Exposure dose and risk assessment of LCMs via inhalation

Exposure to the target LCMs via inhalation was assessed for adults and toddlers using Eq. (1). The EDI under P_{50} exposure scenarios (EDI-P_{50}) for adults were 0.441 ng/kg BW/d in IGY,0.0147 ng/kg BW/d in OGY, 0.171 ng/kg BW/d in IJY, and 0.0118 ng/kg BW/d in OJY. The EDI-P_{50} values for toddlers were 2.27 ng/kg BW/d in IGY, 0.0260 ng/kg BW/d in OGY, 0.879 ng/kg BW/d in IJY, and 0.0210 ng/kg BW/d in OJY. There is no previous report on exposure assessment of LCMs in PM_{10}; however, EDI through dust ingestion for adults from e-waste dismantling sites (48.3 ng/kg BW/d) and reference sites (16.5 ng/kg BW/d) were reported in a previous study (Cheng et al., 2022). The EDI levels of adults and toddlers exposed to dust in general indoor (adults 0.0285 ng/kg BW /d, toddlers 0.230 ng/kg BW/d) and outdoor (adults 0.120 ng/kg BW /d, toddlers 0.0430 ng/kg BW/d) environments have also been reported (Zhang et al., 2022). The EDI-P_{50} in this study was not comparable to the EDI levels in the two previous studies, and the different amounts of LCM analytes and media of exposure may have contributed to this result.

As shown in Fig. 4, the median EDI for adults indoors were much higher than that outdoors in Guiyu and Jieyang ($p < 0.05$), which was associated with a greater enrichment of organic pollutants in indoor environments (Butte and Heinzow, 2002). In addition, the EDI levels in the IGY were not significantly different from those in the IJY ($p > 0.05$), indicating that the risk of exposure to LCMs in the general population deserves as much attention as that of occupational workers. EDI-P_{50} was much higher for toddlers than adults in IGY ($p < 0.05$), which was expected because toddlers spend a longer time indoors, and LCMs may have specific effects on toddlers (Zhang et al., 2022). However, no significant differences were found between the median EDI-P_{50} values of adults and toddlers in OGY, IJY and OJY ($p > 0.05$). These results suggest that further attention should be paid to the potential health effects of LCM on toddlers in e-waste dismantling areas.

As limited parameters were available for the risk assessment of LCMs, the HQs for EDPBB, EDPrB, BCEDB, FPCB, BDPrB, EDPeB, TePrB, DTMDPB, PCTB, BBDB, and EFPT were calculated (Yang et al., 2023). The HQ values of the individual LCM and their sums were < 1 (Table S3), indicating a low risk of LCMs ingestion from PM_{10} via inhalation in adults and toddlers. However, owing to the limitation of risk assessment parameters, only 11 LCMs were assessed in this study, which may have underestimated the true health risks for adults and toddlers. In addition, pathways other than inhalation may be important routes of LCM exposure, and further comprehensive analyses of LCMs in the environment should be conducted.

4. Conclusions

The occurrence of LCMs in PM_{10} and the associated human exposure were investigated in residential areas near and distant to an e-waste dismantling area. LCMs were frequently detected in PM_{10} from indoor and outdoor environments, with the highest concentrations observed in IGY samples, followed by IJY, OGY, and OJY. Although LCMs in indoor PM_{10} were significantly higher than those in outdoor PM_{10}, no significant difference was found in LCM levels in indoor and outdoor environments, between the e-waste dismantling area and reference residential area. These results indicate that LCM released from various types of electronic products in general indoor environments is a non-

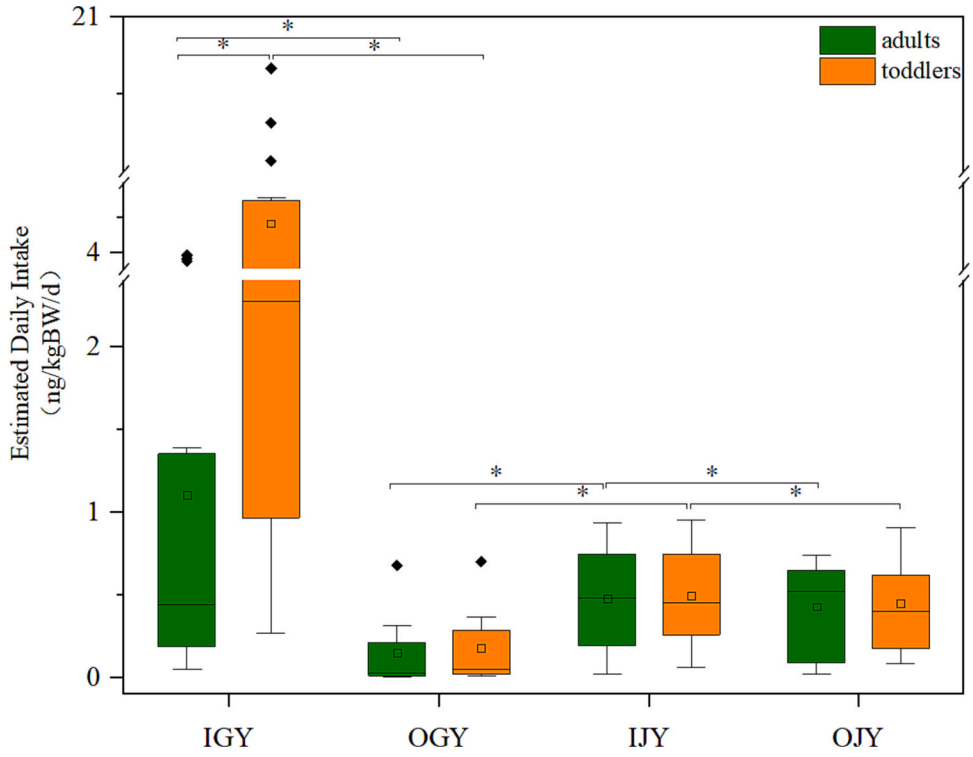


Fig. 4. Estimated daily intake (ng/kg BW/day) of LCMs for adults and toddlers (Asterisks (*) represent significance levels with $p < 0.05$).

negligible source of LCMs. PVB, EBMB, EDPBB and DFPVBC were the major contaminants in indoor samples; and the compositions of LCMs in outdoor samples were not completely consistent with those in indoor samples, suggesting multiple sources of LCMs outdoors. The EDI-P_{50} of LCMs was much higher for toddlers than for adults in indoor environments in Guiyu, suggesting that more attention needs to be paid to the health risks of LCMs for toddlers. The results of this study reveal the occurrence of LCMs in indoor and outdoor PM_{10} in e-waste dismantling and general residential areas, and also provide further insight into the assessment of the risk of human exposure to LCMs through inhalation.

However, it should be noted that the sample size in the present study was small, which is a major limitation of the present study. Meanwhile, the source apportionment of LCMs was not available in this study, which limited the identification of specific source or individual LCMs. Besides, the human exposure dose to LCMs was assessed solely through inhalation pathway, and the risk assessment was not available to most LCMs owing to the limitation of risk assessment parameters. For a more representative assessment of human exposure risk to LCMs, a larger sampling size, more risk assessment parameters, and source identification of LCMs are required in the future studies.

CRediT authorship contribution statement

Li—Li Yao: Methodology, Investigation, Writing- Original Draft Preparation

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Rong-Fa Xu: Investigation, Writing - Review & Editing

Ming Zhu: Investigation, Writing - Review & Editing

Yan Ma: Data curation, Visualization, Writing - Review & Editing

Bin Tang: Conceptualization, Formal analysis, Writing - Review & Editing,

Qi-Yuan Lu: Investigation, Writing - Review & Editing

Feng-Shan Cai: Formal analysis, Data curation, Writing - Review & Editing

Xiao Yan: Data curation, Writing - Review & Editing

Jing Zheng: Writing- Review & Editing, Supervision, Resources, Funding acquisition

Yun-Jiang Yu: Writing- Review & Editing, Supervision

Declaration of competing interest

The authors declare no competing financial interest.

Data availability

Data will be made available on request.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2023.166964.

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