Since January 2020 Elsevier has created a COVID-19 resource centre with free information in English and Mandarin on the novel coronavirus COVID-19. The COVID-19 resource centre is hosted on Elsevier Connect, the company's public news and information website.

Elsevier hereby grants permission to make all its COVID-19-related research that is available on the COVID-19 resource centre - including this research content - immediately available in PubMed Central and other publicly funded repositories, such as the WHO COVID database with rights for unrestricted research re-use and analyses in any form or by any means with acknowledgement of the original source. These permissions are granted for free by Elsevier for as long as the COVID-19 resource centre remains active.
Profiling of phthalates, brominated, and organophosphate flame retardants in COVID-19 lockdown house dust; implication on the human health

Nadeem Ali a,⁎, Muhammad Imtiaz Rashid a, Nabil A. Alhakamy b, Sultan Hassan Alamri c, Syed Ali Musstjab Akber Shah Eqanid d

a Center of Excellence in Environmental Studies, King Abdulaziz University, Jeddah 21589, Saudi Arabia
b Pharmacists Department, Faculty of Pharmacy, King Abdulaziz University, Jeddah 21589, Saudi Arabia
c Department of Family Medicine, Medical College, King Abdulaziz University, Jeddah 21589, Saudi Arabia
d Public Health and Environment Division, Department of Biosciences, COMSATS Institute of Information Technology, Islamabad 45550, Pakistan

HIGHLIGHTS

- Increased levels of pollutants were found in indoor dust collected during COVID-19 lockdown.
- DEHP was the major pollutant in analyzed dust samples.
- Health risk from DEHP was above the set limits for exposed young population.
- Study highlights another aspect of COVID-19 on human lives.

GRAPHICAL ABSTRACT

ABSTRACT

In this study, brominated flame retardants (BFRs), phthalates, and organophosphate flame retardants (PFRs) were analyzed in indoor household dust collected during the COVID-19 related strict lockdown (April–July 2020) period. Floor dust samples were collected from 40 households in Jeddah, Saudi Arabia. The levels of most of the analyzed chemicals were visibly high and for certain chemicals multifold high in analyzed samples compared to earlier studies on indoor dust from Jeddah. Bis (2-ethylhexyl) phthalate (DEHP) was the primary chemical in these dust samples, with a median concentration of 769,500 ng/g of dust. Tris (2-butoxy ethyl) phosphate (TBEP) and Decabromodiphenyl ether (BDE 209) contributed the highest among PFRs and BFRs with median levels of 5990 and 940 ng/g of dust, respectively. The estimated daily exposure in the worst case scenario (23,700 ng/kg bw/day) for Saudi children was above the reference dose (20,000 ng/kg bw/day) for DEHP, and the hazardous index (HI) was also >1. The long-term carcinogenic risk was above the 1 × 10−5, indicating a risk to the health of Saudi young children from getting exposed to DEHP from indoor dust. This study draws attention to the increased indoor pollution during the lockdown period when all of the daily activities by adults and children were performed indoors, which negatively impacted human health, as suggested by the calculated risk. However, the current study has limitations and warrants more monitoring studies from different parts of the world to understand the phenomenon. At the same time, this study also highlights another side of COVID-19 related to our lives.

⁎ Corresponding author at: Center of Excellence in Environmental Studies, King Abdulaziz University, P.O Box: 80216, Jeddah 21589, Saudi Arabia.
E-mail address: nabahadar@kau.edu.sa (N. Ali).

http://dx.doi.org/10.1016/j.scitotenv.2022.158779
Received 1 June 2022; Received in revised form 5 September 2022; Accepted 11 September 2022
Available online 15 September 2022
1. Introduction

Phthalates, brominated flame retardants (BFRs), and organophosphate flame retardants (PFRs) are semi-volatile organic compounds (SVOCs) that are added to the consumer products to meet the regulation of fire hazards, provide longevity and elasticity [Blum et al., 2019; Benjamin et al., 2017; Zuidervene et al., 2020]. These SVOCs are added to various applications such as personal care products, electrical appliances, building materials, PVC flooring, electronics, children’s toys, medical devices, food containers, etc. [Blum et al., 2019; Benjamin et al., 2017; Zuidervene et al., 2020]. Several recent studies from different countries indicate that the use of PFRs has been increasing recently, especially after the regulations on the use of Penta-, Octa-, and Deca- brominated diphenyl ethers (BDEs) [Blum et al., 2019]. Instead of being chemically bound, most of these SVOCs are additives in the host polymer, which makes these chemicals prone to leach into the environment during production, use, and recycling [Blum et al., 2019; Benjamin et al., 2017; Covaci et al., 2011; Guo and kilomet, 2011; Stacey et al., 2009; Zuidervene et al., 2020]. Many studies have reported their presence in the environment and noted the link between exposure to those chemicals and several health conditions [Benjamin et al., 2017; Bastiaensen et al., 2021; Kamrin, 2009]. Humans get exposed to these chemicals via contaminated food intake, air inhalation, involuntary dust intake, and dermal uptake [Abdallah et al., 2016; Ali et al., 2021a, 2021b; Zhu et al., 2019]. High levels of these chemicals are reported in various countries’ indoor settings, and involuntary intake of indoor dust is considered one of the significant exposure pathways for these chemicals [Ali et al., 2021a, 2021b; Giorgonec et al., 2021; Santillo et al., 2003]. Monitoring SVOCs in indoor household settings is essential for specific age groups such as young children, older people, and homemakers who spend most of their time at home [Larsson et al., 2017; Lee et al., 2021; Santillo et al., 2003]. Exposure to young children with developing immune system and older people with weak immune system is of particular concern to these chemicals [Hwang et al., 2008; Larsson et al., 2017].

Many recent studies have reported significantly improved outdoor environmental quality during the lockdowns to limit the spread of COVID-19 [Islam and Chowdhury, 2021; Lolli et al., 2020; Mousazadeh et al., 2021]. Lockdowns were considered the most effective tool to control the human-to-human transmission of COVID-19. These lockdowns reduced industrial activities and traffic, consequently improving the overall environmental quality of the outdoors [Islam and Chowdhury, 2021; Lolli et al., 2020; Mousazadeh et al., 2021]. Similar to other countries Kingdom of Saudi Arabia imposed a strict lockdown in the country for many months with restricted movement of the population. Young kids were not allowed to go out even during the open hours; therefore, they spent all their time indoors performing all their activities, including education and playing [Alamri et al., 2021]. With people spending more time at home, increased activities such as cooking, cleaning, indoor smoking, and children’s activities might have impacted the indoor air quality [Alamri et al., 2021]. However, studies have not focused on conducting the impact of lockdown on indoor environmental quality. Monitoring indoors is even more important in households with young children and people with compromised health. Even after >20 months into the COVID-19 pandemic, most young children were still spending most of their in indoors since their educational activities were online until January 2022. Therefore, it was significant to monitor and understand the dynamics of indoor chemical pollution and study its impact on public health.

In recent years indoor dust has gathered a lot of interest from the scientists to measure the indoor environmental quality by studying the occurrence of organic pollutants in it [Abdallah et al., 2016; Hwang et al., 2008; Lee et al., 2021]. Few studies reported organic pollutants in indoors during the lockdowns, and there is still insufficient data on this topic and no study have reported presence of PFRs, BFRs, and phthalates in indoor dust during COVID-19 lockdown [Alamri et al., 2021; Du and Wang, 2020]. Considering the knowledge gap, this study was designed to analyze these chemicals in household dust of Saudi Arabia collected during the COVID-19 related lockdown period. The study findings provide a different angle on COVID-19 impact on the environment and people.

2. Materials and methods

2.1. Chemicals, solvents, and sampling

The analytical standards for the selected BFRs, PFRs, and phthalates (Table 1) and labelled- internal standards were purchased from Restek, Wellington laboratories, and AccuStandard with >99 % purity. All solvents used, namely n-hexane (n-Hex), dichloromethane (DCM), isoctoane, toluene, and acetone, were acquired from Sigma Aldrich. Iso-octane and toluene were used to make the stock solutions for the selected chemicals. Silica BondElut (500 mg, 3 mL) cartridges for the sample clean-up were also obtained from Sigma Aldrich (Bellefonte, PA, USA). All glassware used during sample preparation, glass tubes, and glass Pasteur pipette were first baked at 400 °C overnight and then were kept at 100 °C until use.

Sampling details are provided in Alamri et al. [2021]. Briefly, indoor floor dust samples were obtained from different households in Jeddah (N = 40) from April–July 2020, when the COVID-19-related lockdown was in place. During the lockdown, the movement of people was restricted even in open hours to their residential areas only. At the same time, people were uncomfortable meeting others due to the fear inflicted by the COVID-19 pandemic; therefore, indoor household dust sampling was a strenuous task. We requested colleagues with a scientific background living in different city districts to collect samples from their homes and areas. A detailed written sampling methodology with a well-prepared questionnaire (with required information from their household) was sent to them (Table S1). Participant households were asked to collect a fresh floor dust sample using their vacuum cleaner. The sampled floor dust was wrapped in the aluminum foil, sealed, labelled, and kept in the freezer until it reached the lab for chemical analysis. The samples were pre-treated using 200 um sieving mesh for the homogenized dust samples and were kept at −20 °C until investigation.

2.2. Sample preparation for the instrument analysis

Sample preparation is discussed elsewhere [Alamri et al., 2021; Ali et al., 2021a, 2021b]. In short, typically ~70 mg was accurately measured and spiked with Is. Hexane/acetone (4/1, v/v) mixture was added to the test tubes with dust and was kept overnight in the dark to achieve equilibrium. The next day ultrasonication (20 min) was used for chemical extraction. Following ultrasonication, centrifugation was performed and the supernatants were transferred to a pre-cleaned labelled individual glass tubes for samples. The whole extraction procedure was repeated twice with leftover dust. The pooled extracts were evaporated using a gentle flow of nitrogen to incipient dryness and were then reconstituted in 1 mL of a solvent mixture of hexane/acetone (2:1 v/v). After re-solubilisation, samples were further cleaned on the pre-conditioned 3 mL silica (500 mg) BondElut cartridges with 8 mL of a solvent mixture of hexane/DCM (2:1 v/v) followed by 6 mL ethyl acetate.

The fraction was evaporated to incipient dryness under a gentle nitrogen flow and reconstituted in 100 µL of isoctoane: toluene (1/1; v/v). Gas chromatography-mass spectrometry (GCMS) was used for the quantitative analysis. Detailed instrumentation is described elsewhere [Ali et al., 2021a, 2021b]. In short, GCMS- QP2010 (Shimadzu) equipped with a fused silica capillary column (TR5 30 M × 0.25 mm × 0.25 µm) was used in electron ionization (EI) mode for the quantitative analysis of PFRs. For quantitative analysis, m/z 99 and 113 (TEHP), 155 and 167 (TEP), 155 and 211 (TBP), 249 and 251 (TCEP), 277 and 279 (TCPP), 397 and 381 (TDCPP), 325 and 326 (TPhP), 251 and 170 (DPEHP), 199 and 299 (TBB), 99 and 113 (TEHP), 367 and 369 (TCP) were used.

For phthalates and BFRs, a TSG™ 8000 Evo triple quadrupole GC-MS (Thermo Fisher Scientific, Waltham, MA, USA) was installed with a fused silica capillary column (Rxi-5Sil MS 15 M × 0.25 mm × 0.10 µm). Phthalates were analyzed in electron ionization (EI) mode, while BFRs were analyzed in negative chemical ionization (NCI) mode. For BFRs m/z 79 and 81 (BDE 28, 47, 99, 100, 153, 154, 183, BTBPE, DPTE, PBBa, DDBDECH), 487 and 489 (BDE 209), 357 and 359 (TBB), 384 and...
Di-nonyl phthalate (DNP) 28 200 680 ± 1330 LOQ (LOQ - 5180)
Di-n-octyl phthalate (DNOP) 67 100 4650 ± 5580 1170 (LOQ - 26,980)
Bis(2-methoxyethyl) phthalate (BMEP) 69 50 2470 ± 8590 100 (LOQ - 48,300)
Bis(4-methyl-2-pentyl) phthalate (BMPP) 86 30 21,050 ± 101,040 40 (LOQ - 614,700)
Di-n-hexyl phthalate (DNHP) 56 10 16,720 ± 101,040 6 (LOQ - 2,020,040)
Benzyl butyl phthalate (BZBP) 100 10 29,330 ± 166,300 920 (100 - 470,900)
Bis(2-ethylhexyl) phthalate (DEP) 84 30 21,050 ± 1330 150 (100 - 1,050,000)
Bis(2-ethylhexyl) phthalate (DEHP) 94 15 740 ± 805 475 (LOQ - 4800)
Tricresyl phosphate (TCP) 81 10 260 ± 150 165 (LOQ - 7000)
Dimethyl phthalate (DMP) 94 10 41,040 ± 239,900 540 (LOQ - 1,460,700)
Diethyl phthalate (DEP) 100 15 6390 ± 9830 3800 (50-56,350)
Di-isobutyl phthalate (DIBP) 97 400 45,100 ± 34,010 33,600 (LOQ - 155,300)
D-i-butyl phthalate (DBP) 100 400 65,405 ± 67,390 47,700 (2000-396,950)
Bis(2-methoxyethyl) phthalate (DMEP) 69 50 2470 ± 8590 100 (LOQ - 48,300)
Bis(4-methyl-2-pentyl) phthalate (BMPP) 86 30 21,050 ± 36,360 810 (LOQ - 470,900)
Bis(2-ethylhexyl) phthalate (DEHP) 100 1800 841,140 ± 453,660 769,500 (8145-2,106,650)
D-n-octyl phthalate (DNOP) 67 100 3225 ± 5580 1170 (LOQ - 26,980)
Di-nonyl phthalate (DNP) 28 200 680 ± 1330 100 (LOQ - 1,460,700)
2,2′,4,4′-Tetrabromodiphenyl ether (BDE 47) 94 0.2 179 ± 468 30 (LOQ - 2372)
2,4,4′-Tribromodiphenyl ether (BDE 28) 100 0.2 3.2 ± 10.7 0.70 (LOQ - 64)
2,2′,4,4′,6-Pentabromodiphenyl ether (BDE 100) 80 0.2 60.4 ± 184 7.5 (LOQ - 955)
2,2′,4,4′,5-Pentabromodiphenyl ether (BDE 99) 87 0.2 235 ± 430 43.5 (LOQ - 2230)
2,2′,4,4′,6,6′-Hexabromodiphenyl ether (BDE 154) 75 0.2 32.6 ± 49.7 5.5 (LOQ - 475)
2,2′,4,4′,5,5′-Hexabromodiphenyl ether (BDE 153) 82 0.2 43.5 ± 120 7.2 (LOQ - 675)
2,2′,3,3′,4,5,6-Heptabromodiphenyl ether (BDE 183) 60 0.2 2.3 ± 3.3 2 (LOQ - 15)
Decabromodiphenyl ether (BDE 209) 97 10 1300 ± 1385 940 (LOQ - 6420)
2-ethylhexyl-2,3,4,5-tetrabromobenzolate (BDE 150) 80 2 64 ± 96 20.5 (LOQ - 415)
Bis(2-ethylhexyl) phthalate (DEHP) 100 0.2 8 ± 12 7 (LOQ - 50)
Bis(2-ethylhexyl)-2,3,4,5-tetrabromophthalate (TBPH) 80 5 245 ± 690 40 (LOQ - 3920)
1,2-Bis(2,4,6-trimethylphenoxyl)ethane (BTBPE) 60 0.2 1.5 ± 3.5 0.8 (LOQ - 25)
Pentabromobenzylacrylate (PBBA) 73 1 40 ± 65 6.5 (LOQ - 220)
1,2-Dibromo-4(1,2-dibromoethyl)cyclohexane (DBDBECH) 87 5 35 ± 30 29 (LOQ - 120)

CDI dermal contact = \( C_n \times \frac{SA \times SL \times ABSd \times EF \times ED}{BW \times AT} \times CF \) (3)

HQ = \( \frac{CDI \text{ (each exposure route)}}{RfD} \) (4)

HI = HQ (inhalation) + HQ (ingestion) + HQ (dermal contact) (5)

CDI represents the concentration (µg/g) (90th percentile) of chemicals in dust samples. R(inh) is the ingestion rate (mg/day), and a high dust intake by both adults (100 mg/day) and children (200 mg/day) was considered due to the dry and dusty conditions in the region [Alamri et al., 2021]. The households in this region use air conditioning to maintain the indoor temperature, which results in regular air circulation and accumulation of high dust particles indoors [Ali et al., 2021a, 2021b]. R(inh) indicates inhalation rate, i.e., 7.6 m³/day for children and 20 m³/day for adults [Ali et al., 2021a, 2021b]. Table S2 in the supplementary information provides the other exposure parameters in the above equations.

To calculate the daily dust intake (DI) via dust ingestion Eq. (6) was used.

\[
\text{Daily dust intake} \left( \frac{ng}{kg \ bw \ day} \right) = \left( C_n \times \frac{IR}{BW} \right) \times \text{Time} \tag{6}
\]

In this equation, Cn represents the concentration of pollutants found in the dust (ng/g). Median and 90th percentile levels were considered for typical and worst-case case scenarios. IR means the rate of dust ingestion;
for typical and worst-case scenarios, high and low dust intake (mg/day) for both adults (20 (low) and 100 (high)) and young children (50 (low) and 200 (high)) were considered. Time indicates the amount of time spent in their households, which was 24 h for both adults and children during lockdown periods [Alamri et al., 2021]. Bioavailability data is not available for many of these chemicals; therefore, 100 % bioavailability was assumed for these preliminary estimations. Body weight of 70 kg for adults, 25 kg for young children, and 12 kg for toddlers were considered.

2.4. Statistical analysis

For the descriptive statistical analysis and human risk assessment calculations Microsoft Excel 2013 was used. To study the correlation, Pearson’s correlation was performed among the levels of analyzed chemicals. An unpaired two-sample t-test was applied using GraphPad to study the significance of socio-economic indicators and difference between the levels of the current study and previous studies from the region with a significance level was p < 0.05.

3. Results and discussion

3.1. Levels and profile of analyzed chemicals

The concentrations of analyzed BFRs, PFRs, and phthalates are described in Table 1. All selected BFRs, PFRs, and phthalates were found in indoor dust samples collected during COVID-19 lockdown with varied detection frequencies (Table 1). The levels of phthalates were higher than both PFRs and BFRs, which is evident due to the broader application of phthalates in consumer products used indoors. Among phthalates, DEP, DnBP, BZBP, DCHP, and DEHP were present in all dust samples. While phthalates in consumer products used indoors. Among phthalates, DEP, both PFRs and BFRs, which is evident due to the broader application of samples with varying concentrations (Table 1, Fig. S2).

The concentrations of different BFRs in the present study dust samples ranged between <LOQ and 6420 ng/g (Table 1). Although the use of BDE 209 has been restricted, this chemical is still being reported in different environmental media at high levels (Covaci et al., 2011; EU-Commission, 2005). In this study, BDE 209 was the major congener at the median level of 940 ng/g of dust (Table 1). Among other PBDEs congeners BDE 47 and 99 were detected at the median concentrations of 30 and 43.5 ng/g, respectively, while all other PBDEs congeners were present at <10 ng/g of dust (Table 1). Other BFRs such as TBB, TBPH, and DDEPBEC were found in >80 % of dust samples with median levels of 20.5, 40, and 29 ng/g of dust, respectively (Table 1). The BFRs were present in >50 % of the dust samples, with wide variation in their levels and profile (Fig. S3). The majority of the households have PBDEs as major BFRs; however, there were few households where other BFRs were also present at high levels (Fig. S3). The skewed distribution of these chemicals indicates that various homes have a variety of stuff treated with different BFRs and released in the indoor environment. Levels of Penta-BDE (BDE 47 and 99) were higher than the components (TBB and TBPH) of FM 550 (Table 1). This indicates that even though regulated Penta-BDEs, these chemicals are still released from the previously treated consumer products and plausibly from the recycled stuff that might have used material treated with Penta-BDE. No significant correlation was found between the levels of TBB and TBPH (p > 0.05). This indicates other than FM 550 emission sources of these chemicals such as DF-45, which contains TBPH and is used in insulation cables, wires and coated fabrics, etc. [Great Lake Solutions, 2010; Sánchez and Villanueva, 2022]. This is the first study from the region reporting many of these analyzed BFRs, PFRs, and phthalates in indoor dust samples. Therefore this study highlights the need to include more chemicals for environmental monitoring in the region.

3.2. Evidence of increased phthalates and PFRs in lockdown indoor dust

At the start of the COVID-19 pandemic, a strict lockdown was introduced in KSA like the rest of the world to control the spread of infection. Studies from different parts of the world showed improved outdoor environmental quality, while some studies reported that indoor environmental quality was compromised during this period. Recently, a study from KSA showed increased PAHs in indoor dust collected during lockdown
Few studies have previously reported some of the analyzed various BFRs, PFRs, and phthalates in indoor household dust from KSA [Albar et al., 2017; Ali et al., 2021a, b; Ali et al., 2016a, b; Bannan et al., 2021]. Levels obtained in the current study were compared with earlier studies (Table S3) from the KSA by histogram graphs (Fig. 1) and using a two-sample t-test (Table S4). Most of the analyzed BFRs, PFRs, and phthalates were visibly higher in the current study than in previous studies (Fig. 1) [Albar et al., 2017; Ali et al., 2021a, b; Ali et al., 2016a, b; Bannan et al., 2021]. However, no statistically significant differences \( P > 0.05 \) were found except for the levels of BDE 209, TCEP, and TCPP between the current study and Ali et al. [2016a, b]. Levels of BDE 209, TCEP, and TCPP were significantly higher \( P < 0.05 \) in the present study than in Ali et al. [2016], this might indicate increased use of these compounds in products used indoors or their slow and gradual buildup in the indoor environment. However, BDE 209 was significantly higher in Bannan et al. [2021] than in the current study. Bannan et al. [2021] reported BFRs in indoor dust collected from the children's rooms, a small sampling area filled with many items/room compared to the complete household. Within compared chemicals, DiBP, BDE 44 & 99, TBPH, TCPP, and TDCPP were the major chemicals in KSA studies (Fig. 1). DEHP and BDE 209 were not included in Fig. 2 due to their overwhelming contribution to the phthalates and BFRs group; however, their levels were compared and are reported in Table S3. Levels of DEHP were higher than in the recent study by Ali et al. [2021a, b] but were lower than the earlier reported by Albar et al. [2017] (Table S4). This might indicate the decreased use of DEHP over the years but at the same buildup of DEHP in the indoor environment during the lockdown period. Comparing current results with previous studies is relevant since many households were also sampled and analyzed previously and are located in similar vicinity [Albar et al., 2017; Ali et al., 2021a, b; Ali et al., 2016a, b; Bannan et al., 2021]. Therefore, the increased levels of many chemicals in the present study indicate that the high indoor activities, such as office work, children's indoor activities (study and play), etc., might have impacted the indoor environmental quality. This is evident from the increased PAH levels in these samples than in previous studies [Alamri et al., 2021; Ali et al., 2016a, b & Ali, 2019]. To look at the various socio-economic factors (Table S1), such as number of cleaning per week, the number of people (young and adults) sharing the household, age of the building, and use of air purifiers, etc., were considered to study their influence on the chemical burden. However, no significant \( P > 0.05 \) influence of socio-economic indicators (Table S1) was found on the levels of analyzed chemicals. This was partly due to the small number of samples analyzed and the considerable variation of analyzed chemicals among different households. This also indicates diverse emission sources for these chemicals in separate homes. Pearson correlation was performed to study the common sources of these chemicals in the dust samples. For most of the chemicals, negative and non-significant correlations \( P > 0.05 \) were found among most of the chemicals. This might indicate that these chemicals have diverse emission sources within- and among different households.

### 3.3. Calculated human risk assessment

Human exposure to Phthalates, BFRs, and PFRs can occur through several routes such as contaminated food and water intake, involuntarily intake of contaminated dust, contaminated air inhalation, etc. Exposure to these chemicals has been linked with several health issues [Abdallah et al., 2016; Benjamin et al., 2017; Lee et al., 2021]. There is much evidence from the literature that exposure to these chemicals has adverse effects on human reproduction and development [Bastiaensen et al., 2021; Benjamin et al., 2017; Jurewicz and Hanke, 2011; Meeker et al., 2013]. DEHP, the primary chemical found in the present samples, is an endocrine disruptor with reported toxicity for different organs and has carcinogenic properties [Jurewicz and Hanke, 2011; Rowdhwal and Chen, 2018]. Studies have shown that phthalates exposure to young children impacts their social behavior similar to BPA [Benjamin et al., 2017]. Some epidemiological studies have highlighted that exposure to phthalates adversely affects thyroid function, anogenital distance, and reproductive hormones [Jurewicz and Hanke, 2011]. A recent study from California, USA, measured phthalates in dust and air and calculated that between 82 and 89 % of children had DBP exposure above the reproductive health benchmarks, and 8–11 % of children aged below two years were exposed to DEHP exceeding the cancer benchmarks [Lloyd et al., 2015].

Similarly, exposure to PFRs is linked with several health problems in lab animals, but limited epidemiological studies are available on indoor exposure to PFRs and its health implications [Araki et al., 2020; Araki et al., 2014; Meeker and Stapleton, 2010]. Meeker and Stapleton [42] found a negative correlation between levels of TDCIPP in house and free thyroxin, but its association was positive with prolactin. In contrast, a positive association between the levels of TnBP in floor dust and house-related mucous symptoms, asthma, and rhinitis is reported in the literature [Araki et al., 2020]. Likewise, a significant correlation was reported between TCIPP and TDCIPP in floor dust and the prevalence of atopic dermatitis [Araki et al., 2014]. Several studies have reported the impact on human health (neurobehavioral and reproductive disorder, thyroid hormone disruption, etc.) following exposure to PBDEs [ATSDR, 2017; Kim et al., 2014; Lyche et al., 2015; Sahlström et al., 2014]. However, no studies in the literature have reported that BFRs have carcinogenic potential in humans. Consequently, PBDEs are classified as Group 3 and Group D carcinogens by...
USEPA and the International Agency for Research on Cancer (IARC) [ATSDR, 2017]. This mean PBDEs are not classifiable as a carcinogen to humans. Similar to PBDEs, data is also limited on the impact of NBFRs on humans [Covaci et al., 2011; Xiong et al., 2019]. A recent study from China found a significant correlation between thyroid disruption and new BFRs in serum [Zhao et al., 2021]. Therefore to study the impact on the population from exposure to chemicals found in indoor dust during the lockdown period, daily and long-term exposure assessment was calculated using Eqs. (1)–(6).

For all selected BFRs, PFRs, and phthalates, except for DEHP, DI was multifold below the reference dose (RfD) values for exposure to the indoor dust (Fig. 2; Table S5). The estimated DI for DEHP is above the RfD values for both toddlers and young children in worst-case scenarios (Fig. 2; Table S5). This indicates that indoor exposure to DEHP via dust ingestion during the lockdown period was a significant cause of concern, especially for toddlers and young children. In a previous study from Jeddah, children’s exposure to DEHP via dust was half the value of RfD [Ali et al., 2021a, b]. However, in the current study, the estimated DI for DEHP is above the RfD value, which indicates that during the lockdown, when children were spending their time at home, they were at higher risk of chemical exposure, especially DEHP. Incremental lifetime cancer risk (ILCR) was calculated for DEHP using (www.popstoolkit.com/tools/HHRA/Carcinogen.aspx) for toddlers, young children, and adults. For all groups, calculated exposure was more significant than $1 \times 10^{-5}$, indicating a potential cancer risk. To calculate the non-carcinogenic risk to both Saudi young and adult populations, CDI was calculated for each exposure route using Eqs. (1)–(3) (Table 2). Then HQ (Eq. (4)) was calculated for each exposure route using CDI values of each exposure route. Finally, HI was calculated (Eq. (5)) using values of HQs. The estimated HI and HQ for all chemicals except DEHP were <1, indicating low non-carcinogenic risk to the exposed population. However, for DEHP, both HQ- ingestion and HI were >1 (Table 2) for children, which is a cause of concern for long-term exposure. A recent study from Riyadh, Saudi Arabia, has reported a positive correlation between metabolites of DEHP in children’s urine and oxidative stress [Lee et al., 2019]. Indoor dust was suggested as a significant exposure pathway to exposed children [Lee et al., 2019]. There is no information available on the production and import volume of BFRs, phthalates, and PFRs within Saudi Arabia. Even though DEHP is a high production volume chemical and its broader application in consumer products is well documented, there is no information available on its production and use within Saudi Arabia. This lack of data on DEHP within Saudi Arabia is a concerning issue since its use is highly regulated now in western countries. According to EU REACH legislation, DEHP is under Category 1B reprotoxic [European Chemicals Agency, 2022].

The calculated health risk assessment showed that Saudi Arabian people were exposed to more chemicals during the COVID-19 lockdown. This was primarily because all the activities for both adults and children happened inside the home. Many households reported less cross ventilation during the lockdown period in fear of getting virus inside and increased use of cleaning chemicals to keep the house clean. This whole scenario made an ideal environment for the chemicals to buildup indoors. Even after 20 months, most young children in Saudi Arabia were still getting their education online. Consequently, they spend most of their time indoors, making them vulnerable to getting exposed to these chemicals. Although this study is based on a limited number of samples, it still highlighted another aspect of COVID-19 impact on our lives and health. Little interest and focus have been given to such research, and these findings warrant more studies on indoor environmental monitoring in similar situations.

4. Conclusion

This study analyzed phthalates, BFRs, and PFRs in household dust collected during COVID-19’s strict lockdown period in 2020. The analyzed PFRs and phthalates were higher than those previously reported from Jeddah, Saudi Arabia. This finding was in line with the hypothesis that indoor pollution might have increased during lockdown due to the increased indoor activities. The daily exposure and long-term non-carcinogenic risk were above the reference value of DEHP for young children, which signifies the risk to the young population. This study also reported some of these chemicals in the region’s indoor environment for the first time, highlighting the importance of analyzing more chemicals in the monitoring studies. Although calculated risk was below for most of the chemicals than the respective reference dose, little is known about the synergetic impact of these chemicals on the health of the exposed population. At the same time, this study throws light on another aspect of the COVID-19 in our lives. Limited studies are available on this aspect of COVID-19 lockdown from different parts of the world. More studies are warranted to understand this phenomenon from various regions.
CRediT authorship contribution statement

Conceptualization, N.A., M.I.R. and N.A.H.; methodology, N.A., M.I.R., S.A.M.A.S.E., and S.H.A.; formal analysis, N.A., M.I.R., and S.A.M.A.S.E.; investigation, N.A., M.I.R., and S.A.M.A.S.E.; writing—original draft preparation, N.A., M.I.R., and S.A.M.A.S.E.;—writing—review and editing, N.A., M.I.R., S.A.M.A.S.E., S.H.A., and N.A.H.; data curation, N.A., M.I.R., N.A.H.; visualization, N.A., S.H.A., and M.I.R.; resources, N.A., M.I.R., and S.A.M.A.S.E.; funding acquisition, N.A., M.I.R., S.A.M.A.S.E., and N.A.H.; supervision, N.A., M.I.R., and S.A.M.A.S.E.; project administration, N.A., M.I.R. and N.A.H.; funding acquisition, N.A., M.I.R., S.A.M.A.S.E., and N.A.H.; All authors have read and agreed to the published version of the manuscript.

Funding

This work was funded by Deanship of scientific studies, King Abdullah University (KAU), Jeddah, grant number GCV19-15-1441.

Data availability

All of the data is presented in the article and associated Supplementary Materials.

Declaration of competing interest

The authors declare no conflict of interest. The funders had no role in the design of the study; in the collection, analyses, or interpretation of data; in the writing of the manuscript, or in the decision to publish the results.

Acknowledgments

This work was funded by Deanship of Scientific Research, King Abdulaziz University (KAU), Jeddah, grant number GCV19-15-1441. We are grateful to all the volunteers who participated in the study. We are grateful to Mr. Sultan and Ms. Doha Hantoush for helping in the sample analysis.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2022.158779.

References

Abdallah, M.A.E., Pawan, G., Harrad, S., 2016. Human dermal absorption of chlorinated organophosphate flame retardants; implications for human exposure. Toxicol. Appl. Pharmacol. 291, 28–37.
Agency for Toxic Substances and Disease Registry, 2019. Toxicological Profile for Di-(2-ethylhexyl) Phthalate (DEHP). Environmental Toxicology Branch 1600 Clifton Road, N.E., Mail Stop 6102-1 Atlanta, Georgia, pp. 30329–42.
Alamri, S.H., Ali, N., Ali Albar, H.M.S., Rashid, M.I., Rajeh, N., Ali Qutub, M.M., Malavarannan, G., 2021. Polycyclic aromatic hydrocarbons in indoor dust collected during the COVID-19 pandemic lockdown in Saudi Arabia: status, sources and human health risks. Int. J. Environ. Res. Public Health 18 (5), 2743.
Albar, H.M.S.A., Ali, N., Shahzad, K., Ismail, I.M.I., Rashid, M.I., Wang, W., Ali, L.N., Eqani, S.A.M.A.S., 2017. Phthalate esters in settled dust of different indoor microenvironments; source of non-dietary human exposure. Microchem. J. 132, 227–232.
Ali, N., 2019. Polycyclic aromatic hydrocarbons (PAHs) in indoor air and dust samples of different saudi microenvironments: health and carcinogenic risk assessment for the general population. Sci. Total Environ. 696, 133995.
Ali, N., Eqani, S.A.M.A.S., Ismail, I.M.I., Malavarannan, G., Kadi, M.W., Albar, H.M.S., Rehan, M., Covaci, A., 2016. Brominated and organophosphate flame retardants in indoor dust of Jeddah, Kingdom of Saudi Arabia: implications for human exposure. Sci. Total Environ. 569, 269–277.
Ali, N., Ismail, I.M.I., Khoder, M., Shamy, M., Alghamdi, M., Costa, M., Ali, L.N., Wang, W., Eqani, S.A.M.A.S., 2016. Polycyclic aromatic hydrocarbons (PAHs) in indoor dust samples from cities of Jeddah and Kuwait: levels, sources and non-dietary human exposure. Sci. Total Environ. 573, 1607–1614.
Ali, N., Alhakamy, N.A., Ismail, I.M., Nazar, E., Summan, A.S., Shah Eqani, S.A.M.A.S., Malavarannan, G., 2021. Exposure to phthalate and organophosphate esters via indoor dust and PM10 is a cause of concern for the exposed Saudi population. Int. J. Environ. Res. Public Health 18 (4), 2125.
Ali, N., Kadi, M.W., Albar, H.M.S., Rehan, M., Ismail, I.M.I., Khoder, M., Shamy, M., Alghamdi, M., Costa, M., Ali, L.N., Wang, W., Eqani, S.A.M.A.S., 2016. Polycyclic aromatic hydrocarbons (PAHs) in indoor dust samples from cities of Jeddah and Kuwait: levels, sources and non-dietary human exposure. Sci. Total Environ. 573, 1607–1614.
Araki, A., Salto, I., Kanazawa, A., Morimoto, K., Nakayama, K., Shibata, E., Tanaka, M., Takahashi, T., Yoshimura, T., Ichiki, T., Saioj, Y., 2014. Phosphorus flame retardants in indoor dust and their relation to asthma and allergies of inhabitants. Indoor Air 24 (1), 3–15.
Araki, A., Bannai, Y.A., Bastianemos, M., Van den Ende, N., Kawai, T., Tsuboi, T., Miyahita, C., Itoh, S., Goudarzi, H., Konno, S., Covaci, A., 2016. Brominated and organophosphate flame retardants in indoor dust of Jeddah, Kingdom of Saudi Arabia: implications for human exposure. Toxicol. Appl. Pharmacol. 291, 28–37.
and phosphate flame retardants and plasticizers and their associations with wheeze and allergy symptoms among school children. Environ. Res. 183, 109212.

ATSDR, 2017. Public health statement Polybrominated diphenyl ethers (PBDEs). https://www.atsdr.cdc.gov/ToxProfiles/t0207-c1-h.pdf.

Bannan, D., Ali, N., Alhakamy, N.A., Alifafeh, M.A., Alharbi, W.S., Rashid, M.I., Rajeh, N., Malarvunan, G., 2021. Brominated flame retardants in Children’s room: concentration, composition, and health risk assessment. Int. J. Environ. Res. Public Health 18 (12), 6421.

Bastiaensen, M., Gys, C., Colles, A., Verheyen, V., Koppen, G., Govarts, E., Bruckers, L., Morrens, B., Loos, I., De Decker, A., Nelen, V., 2021. Exposure levels, determinants and risk assessment of organophosphate flame retardants and plasticizers in adolescents (14–15 years) from the Flemish environment and health study. Environ. Int. 147, 106368.

Benjamin, S., Masai, E., Kamimura, N., Takahashi, K., Anderson, R.C., Faisal, P.A., 2017. Sources and human exposure implications of concentrations of diethylhexyl phthalate, dipentyl phthalate, and diisononyl phthalate. Regul. Toxicol. Pharmacol. 52 (2), 90–101.

Bergh, C., Luongo, G., Wise, S., Östman, C., 2012. Organophosphate and phthalate esters in standard reference material 2585 organic contaminants in house dust. Anal. Bioanal. Chem. 402 (1), 51–59.

Blum, A., Behl, M., Birnbaum, L.S., Diamond, M.I., Phillips, A., Singla, V., Sipes, N.S., Stapleton, H.M., Venier, M., 2019. Organophosphate ester flame retardants: are they a re-grettable substitution for polybrominated diphenyl ethers? Environ. Sci. Technol. Lett. 6 (11), 638–649.

Brommer, S., Harrad, S., 2015. Sources and human exposure implications of concentrations of organophosphate flame retardants in dust from UK cars, classrooms, living rooms, and offices. Environ. Int. 83, 202–207.

Covaci, A., Harrad, S., Abdallah, M.A.E., Ali, N., Law, R.J., Herzke, D., de Wit, C.A., 2011. Novel brominated flame retardants: a review of their analysis, environmental fate and behavior. Environ. Int. 37 (2), 532–556.

Dirtu, A.C., Ali, N., Van den Ende, N., Noels, H., Covaci, A., 2012. Country-specific comparison for profile of chlorinated, brominated and phosphorus organic contaminants in indoor dust. Case study for eastern Romania, 2010. Environ. Int. 49, 1–8.

Dodson, R.E., Perovich, L.J., Covaci, A., Van den Ende, N., Jonas, A.C., Dirtu, A.C., Brody, J.G., Rudel, R.A., 2012. After the PBDE phase-out: a broad suite of flame retardants in repeat house dust samples from California. Environ. Sci. Technol. 46 (24), 13056–13066.

Du, W., Wang, G., 2020. Indoor air pollution was non-negligible during COVID-19 lockdown. Indoor Air 31 (4), 1121–1123.

Guo, Y., Kannan, K., 2011. Comparative assessment of human exposure to phthalate esters from house dust in China and the United States. Environmental science & technology 45 (8), 3788–3794.

Hwang, H.M., Pak, E.K., Young, T.M., Harth, V., Lorber, M., Brüning, T.H., 2012. Di-n-butyl phthalate (DnBP) and diisobutyl phthalate (DiBP) metabolism in a human volunteer after single oral doses. Arch. Toxicol. 86 (12), 1829–1839.

Jia, J., Zhang, K., Zhang, H., Liao, C., Jiang, G., 2019. Phthalate esters in indoor dust: a review of the importance in children. Environ. Res. 183, 109212.

Kada, M.W., Ali, N., Albar, H.M.S.A., 2018. Phthalates and polycyclic aromatic hydrocarbons (PAHs) in indoor settled carpet dust of mosques, health risk assessment for public. Sci. Total Environ. 535 (23), 15531–15539.

Koch, H.M., Christensen, K.L.Y., Harth, V., Lorber, M., Brüning, T.H., 2012. Di-n-butyl phthalate (DnBP) and diisobutyl phthalate (DiBP) metabolism in a human volunteer after single oral doses. Arch. Toxicol. 86 (12), 1829–1839.

Lee, I., Alakeel, R., Kim, S., Al-Sheikh, Y.A., Al-Maneed, H., Alyousef, A.A., Kho, Y., Choi, K., 2019. Urinary phthalate metabolites among children in Saudi Arabian: occurrence, risks, and their association with oxidative stress markers. Sci. Total Environ. 654, 1350–1357.

Lee, I., Pülimäe, C., Ringbeck, B., Ihn, Y., Gotthardt, A., Lee, G., Alakeel, R., Alfrashd, M., Tosep, R., Jayalalipraja, E.A., Tantrakarnapak, K., 2021. Urinary concentrations of major phthalate and alternative plasticizer metabolites in children of Thailand, Indonesia, and Saudi Arabia, and associated risks. Environmental Sci. Technol. 55 (24), 16562–16567.

Li, X., Hauser, R., Drummer, O.H., Zock, P.L., Zhou, J., 2015. Assessment of phthalates/phthalate alternatives in children’s toys and childcare articles: review of the report including conclusions and recommendation of the chronic Hazard advisory panel of the consumer product safety commission. J. Expos. Sci. Environ. Epidemiol. 25 (4), 343–353.

Loli, S., Chen, Y.C., Wang, S.H., Vivone, G., 2020. Impact of meteorological conditions and air pollution on COVID-19 pandemic transmission in Italy. Sci. Rep. 10 (1), 1–15.

Lyche, J.L., Roselland, C., Berge, G., Polder, A., 2015. Human health risk associated with brominated flame retardants (BFRs). Environ. Int. 74, 170–180.

Meeker, J.D., Stapleton, H.M., 2010. House dust concentrations of organophosphate flame retardants in relation to hormone levels and semen quality parameters. Environ. Health Perspect. 118 (3), 318–323.

Meeker, J.D., Cooper, E.M., Stapleton, H.M., Hauser, R., 2013. Exploratory analysis of urinary metabolites of phthalates-containing flame retardants in relation to markers of male reproductive health. Endocr. Disruptors 1 (1), e2635.

Mousazadeh, M., Paital, B., Naghdali, Z., Mortezania, Z., Hashemi, N., Naziragh, E.K., Aghababaei, M., Ghorbakhiami, M., Lichthouse, E., Sillanpää, M., Hashim, K.S., 2021. Positive environmental effects of the coronavirus 2020 episode: a review. Environ. Dev. Sus. 1, 1–23.

Oteef, M.D., Elhassan, M.S., 2020. Plastic toys and child care articles as a source of child exposure to phthalates and other plasticisers in Saudi Arabia. Int. J. Environ. Anal. Chem. 1–15.

Rowe0whel, S.S.S., Chen, J., 2018. Toxic effects of di-2-ethylhexyl phthalate: an overview. BioMed Research Int. 2018.

Sahlström, L.M., Sellström, U., de Wit, C.A., Lignell, S., Därnerud, P.O., 2014. Brominated flame retardants in matched serum samples from Swedish first-time mothers and their toddlers. Environ. Sci. Technol. 48 (13), 7584–7592.

Santillo, D., Labunska, I., Davidson, H., Johnston, P., Strutt, M., Knowles, O., 2003. Consuming Chemicals: Hazardous Chemicals in House Dust as an Indicator of Chemical Exposure in the Home. Greenpeace Research Laboratories, Exeter, UK.

Stapleton, H.M., Klosterhaus, S., Eagle, S., Fahl, J., Meeker, J.D., Blum, A., Webster, T.F., 2009. Exposure to phthalates and other plasticisers in Saudi Arabia. Int. J. Environ. Anal. Chem. 1–15.

Zhu, Q., Jia, J., Zhang, K., Zhang, H., Liao, C., Jiang, G., 2019. Phthalate esters in indoor dust: a review of the importance in children. Environ. Res. 183, 109212.