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# Concentrations of selected chemicals in indoor air from Norwegian homes and schools

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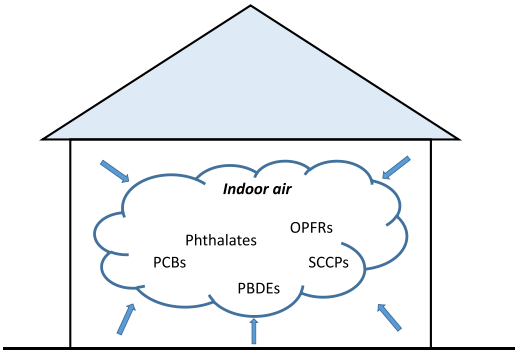
## HIGHLIGHTS

- Indoor air may contribute significantly to human exposure to different chemicals.
- Phthalates, PCBs, OPFRs, PBDEs, SCCPs were measured in total 54 indoor air samples.
- Levels of phthalates and SCCPs were higher in households compared to schools.
- Levels of phthalates, OPFRs and SCCPs up to 1000 times higher than PCBs and PBDEs.
- Estimated intakes of phthalates and SCCPs from air were equal to intakes from food.

## GRAPHICAL ABSTRACT

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## ABSTRACT

Both building materials and consumer products have been identified as possible sources for potentially hazardous substances like phthalates, polychlorinated biphenyls (PCBs), organophosphorous flame retardants (OPFRs), polybrominated diphenyl ethers (PBDEs) and short chain chlorinated paraffins (SCCPs) in indoor air. Thus, indoor air has been suggested to contribute significantly to human exposure to these chemicals. There is lack of data on the occurrence of several of the aforementioned chemicals in indoor air. Therefore, indoor air (gas and particulate phase) was collected from 48 households and 6 classrooms in two counties in Norway. In both the households and schools, median levels of low molecular weight phthalates (785 ng/m$^3$), OPFRs (55 ng/m$^3$) and SCCPs (128 ng/m$^3$) were up to 1000 times higher than the levels of PCBs (829 pg/m$^3$) and PBDEs (167 pg/m$^3$). Median concentrations of dimethyl phthalate (DMP), diethyl phthalate (DEP), di-isobutyl phthalate (DiBP) and SCCPs were 3–6 times higher in households compared to schools. The levels of OPFRs, PCBs and PBDEs were similar in households and schools. In univariate analysis, the indoor concentrations of different environmental chemicals were significantly affected by location of households (OPFRs), airing of living room (some PCBs and PBDEs), presence of upholstered chair/couch (OPFRs), pet animal hold (some PBDEs) and presence of electrical heaters (selected PCBs and PBDEs). Significant correlations were also detected for the total size of

*Abbreviations:* DMP, Dimethyl phthalate; DEP, diethyl phthalate; DiBP, di-isobutyl phthalate; DnBP, di-n-butyl phthalate; EFSA, European Food Safety Authority; PUF, polyurethane foam; PCBs, polychlorinated biphenyls; PBDEs, polybrominated diphenyl ethers; OPFRs, organophosphorous flame retardants; LCCP, long chain chlorinated paraffin; MCCP, medium chain chlorinated paraffin; SCCP, short chain chlorinated paraffin; PVC, polyvinyl chloride; TnBP, tri-n-butyl phosphate; TCEP, tris(2-chloroethyl) phosphate; TCPP, tris(1-chloro-2-propyl) phosphate; TPHP, triphenyl phosphate; TBOEP, tris(2-butoxyethyl) phosphate; TDCPP, tris(1,3-dichloro-2-propyl) phosphate.

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households with OPFRs, frequency of vacuuming the living room with selected PCBs and PBDEs, frequency of washing the living room with selected PCBs and the total number of TVs in the households with selected phthalates and SCCPs. Finally, intake estimates indicated that indoor air contributed more or equally to low molecular weight phthalates and SCCPs exposure compared to food consumption, whereas the contribution from indoor air was smaller than the dietary intake for the other groups of chemicals.



## 1. Introduction

Most individuals in western countries spend the majority of their time in indoor environments (offices, stores, homes, etc.) (Klepeis et al., 2001). Thus, if hazardous substances are present in the indoor environment a significant human exposure may occur by inhalation, dermal uptake and dust ingestion (Beko et al., 2013; Weschler and Nazaroff, 2012). Phthalates, polychlorinated biphenyls (PCBs), organophosphorous flame retardants (OPFRs), polybrominated diphenyl ethers (PBDEs) and short chain chlorinated paraffins (SCCPs) are among chemicals that have been widespread used in building materials and consumer products and have gained interest in the recent years due to both their widespread presence in indoor environments and their suggested potential for adverse health effects. Depending upon their molar mass, boiling point, saturation vapor pressure and octanol/air partitioning coefficient, these chemicals may be distributed between gas phase and particulate phase in indoor air as well as settled dust (Takeuchi et al., 2014; Wei et al., 2019; Weschler et al., 2008).

The uses of the chemicals under study cover a wide range of applications. Phthalates are widely used as plasticizers in polyvinyl chloride (PVC) plastics, building materials, paint and certain types of medical equipment, but are also present in consumer products such as cosmetics, toys and food packaging (Schettler, 2006). Phthalates are not chemically bound to the plastic polymers and may therefore easily leach out of the product. Chlorinated paraffin is a common term for a large group of chlorinated aliphatic substances. These can be divided into three groups based on the number of carbon atoms in the chain: short (C10–13), medium (C14–17) and long (C > 17) chain chlorinated paraffins (SCCPs, MCCPs and LCCPs, respectively) (van Mourik et al., 2016). Their common applications include additives in PVC, flame retardants (FRs) in plastics, rubbers and textiles, cooling fluids and lubricants in metal processing, and plasticizers in paints, rubbers and plastics (van Mourik et al., 2016). PCBs are a group of 209 congeners previously used in a wide range of industrial applications, mainly as insulators, sealants, and FRs in paints, and building materials (Diamond et al., 2010). Polybrominated diphenyl ethers (PBDEs) have been widely used as FRs in a variety of consumer products, electrical equipment and building materials for decades (Darnerud et al., 2001). They are commercially available in three different mixtures of congeners; penta-, octa- and deca-BDE. Organophosphorous (OP) triesters are a group of chemicals used mainly as FRs and also plasticizers in many consumer goods (Hou et al., 2016). PCBs, PBDEs and SCCPs are highly persistent whereas phthalates and OPFRs are non-persistent chemicals. The constant release of phthalates from different indoor sources results in their ubiquitous presence in indoor environment. Thus, they could be considered as pseudo-persistent chemicals.

These chemical groups have been associated with adverse health effects both in animal and human studies. Depending on the phthalate type, animal studies have suggested effects on reproduction, development, endocrine disruption, as well as cancer (Mathieu-Denoncourt et al., 2015; Ventrice et al., 2013), whereas epidemiological studies have associated phthalate exposure with asthma and allergy (Jurewicz and Hanke, 2011). The acute toxicity of SCCPs is low but they have been classified as potential carcinogens (van Mourik et al., 2016). PCBs are strongly accumulating in the food chain and human body. Their toxicity depends upon the chemical structure and includes adverse effects on immune system, hormone disruption, developmental and reproductive effects as well as cancer (Hens and Hens, 2017). Also PBDEs exhibit high fat solubility, bioaccumulation and are considered to be endocrine disruptors, and thereby affect development and reproduction (Linares et al., 2015). Human exposure to OPFRs has raised concern as indoor concentrations have been associated with asthma (Araki et al., 2014) and altered hormone levels and decreased semen quality in men (Meeker and Stapleton, 2010). Furthermore, in vivo and in vitro studies of OPFRs have shown relevant adverse health effects like carcinogenicity and neurotoxicity among others (van der Veen and de Boer, 2012).

Due to their potential adverse environmental and health effects, the use of several of these chemicals have been banned or restricted in many countries. For example, the use of the potentially most harmful phthalates is restricted in toys and products intended for infants and toddlers (EFSA, 2005a, 2005b, 2005c, 2005d, 2005e). There is a ban of SCCPs in Norway due to their classification as potential carcinogens and this chemical group has been included in Annex A (Elimination) of the Stockholm Convention on Persistent Organic Pollutants in 2017. The use of PCBs was banned in most countries during the late 1970s due to their persistence in the environment and documented adverse environmental and health effects (Hens and Hens, 2017). The production and use of penta- and octa-BDE were banned in 2004 both in EU and Norway (Directive 2003/11/EC of the European Parliament and of the Council, 2003). In addition, there has been worldwide restrictions on deca-BDE since 2006 (Cequier et al., 2014). No restrictions or regulations have so far been implemented on OPFRs. However, a screening assessment by the European Chemicals Agency in 2018 identified a risk for children from exposure to certain OPFRs in flexible polyurethane (PUR) foams in childcare articles and residential upholstered furniture (ECHA, 2018).

Widespread exposure of the general population to phthalates, PCBs, OPFRs and PBDEs has been documented through human biomonitoring studies (Haug et al., 2018; Saillenfait et al., 2018). However, there is a lack of data showing levels of many of these chemicals in indoor environments. The main objective of this study was to characterize and compare indoor air (gas and particulate phase) levels in Norwegian households and schools for semi-volatile phthalates, SCCPs, OPFRs, PCBs and PBDEs. In addition, the impact of building characteristics, indoor materials used and housekeeping activities of inhabitants on the levels of these chemicals were investigated. Finally, the human exposure was estimated based on the measured indoor air levels, and compared to dietary intake estimates from the literature.

## 2. Material and methods

### 2.1. Collection and analysis of air samples

A study group consisting of 48 households and 2 schools (6 classrooms in total) from two different neighboring counties in Norway (Oslo and Akershus, both urban places) was established in 2012 (Cequier et al., 2014). The participants included 48 mother-child pairs living in these households. Additionally, siblings of 8 children were also included in the study (in total 56 children). One school from each county was chosen. The school in Akershus County was larger compared to the one in Oslo, therefore indoor air samples from 4 classrooms were collected from the school in Akershus and from 2 classrooms in the Oslo County. The study was approved by the Regional Committee for Medical

and Health Research Ethics and informed consent was subsequently obtained from the participants. The indoor air samples and a detailed questionnaire about the building characteristics (type, age, county and building material used for house), indoor materials (type of floor, number and type of electronics, type of chair/couch etc.) and housekeeping activities (e.g., airing, vacuuming and washing) were collected for each participant. The parameters included in the analyses are listed in Table S1 for categorical variables and Table S2 for continuous variables.

For SCCPs, OPFRs, PCBs and PBDEs, air samples (gas phase and particulate matter) were collected for 24 h in the living rooms of the participants and in the classrooms. A Leland Legacy pump (SKC Limited, UK) was set with a flow of 12 l/min with multiple sample holders. Each sample holder contained one filter and two polyurethane foam (PUF) cylinders. Filters and adsorbents were extracted together resulting in a total air concentration. For phthalates, air samples were simultaneously collected onto a porous polymer (Tenax TA) using a personal air sampling pump at a flow rate of 0.07 l/min for 24 h. Sample preparation and determination of the analytes and field blanks for each class of compound is described in the Supplementary Information.

### 2.2. Categorization of chemicals

The following phthalates were evaluated individually: dimethyl phthalate (DMP); diethyl phthalate (DEP); di-isobutyl phthalate (DiBP); di-n-butyl phthalate (DnBP). The concentrations of individual OPFRs (tri-n-butyl phosphate (TnBP), tris(2-chloroethyl) phosphate (TCEP), tris(1-chloro-2-propyl) phosphate (TCPP), triphenyl phosphate (TPHP), tris(2-butoxyethyl) phosphate (TBOEP) and tris(1,3-dichloro-2-propyl) phosphate (TDCPP) have been published previously by us (Cequier et al., 2014), and here only the sum of these six compounds are given. SCCPs (i.e., C10-C13) were evaluated as the sum calculated using technical mixtures as external standards. For PCBs and PBDEs, the congeners of interest were categorized according to the number of halogen atoms in their structure. Five different categories of PCBs were defined: TriCB (sum of PCBs 18, 28, 31, 33 and 37), TetraCB (sum of CB 47, 52, 66 and 74), PentaCB (sum of CB 99, 101, 105, 114, 118, 122 and 123), HexaCB (sum of CB128, 138, 141, 149, 153, 156, 157 and 167) and HeptaCB (sum of CB170, 180, 183, 187 and 189). Similarly, four PBDE categories were defined: TriBDE (BDE 28), TetraBDE (BDE 47), PentaBDE (BDE 85, 99 and 100) and HexaBDE (BDE 153 and 154). Individual air concentrations of OPFRs and PBDEs have been published elsewhere (Cequier et al., 2014).

### 2.3. Exposure estimates

The daily inhalation exposure was estimated for each group of chemicals by using the following formula: Daily estimated exposure from air (ng or pg/day/kg body weight) = Air concentration x Amount of air inhaled/body weight (kg). The volume of air inhaled by mothers and children were assumed to be 13.3 and 10.9 $m^3$/day, respectively (Cequier et al., 2014). The calculation assumes that the participants were at home for 24 h and that 100% of the inhaled chemicals were absorbed in the airways. This calculation is likely to represent an overestimation of the actual exposure since it is likely that only a fraction of the compounds in gas phase are absorbed in the airways. Additionally, for the chemicals adsorbed to particulate matter (PM) the inhaled and deposited fraction is known to be below 100% depending on the particle size.

The daily dietary intakes were taken from previous studies for SCCPs (Yuan et al., 2017), PCBs (Karjalainen et al., 2012; Kvalem et al., 2009), PBDEs (Karjalainen et al., 2012; Knutsen et al., 2008) and OPFRs (Poma et al., 2017) performed in the Nordic countries. The concentrations of phthalates were measured in 37 different Norwegian foods and beverages (Sakhi et al., 2014) and the food consumption of the participants in the present study was assessed through a 24-h recall conducted by a trained researcher as published elsewhere (Sakhi et al., 2018). Thus, the phthalate intake from food was estimated for both mothers and children by using the following formula:

Daily estimated exposure from food (ng or pg/day/kg body weight) = Food concentration x Amount of food consumed/body weight (kg). Exposures by other pathways like dust ingestion or dermal absorption were not estimated in the present study.

### 2.4. Statistical analysis

IBM SPSS version 23 was used to perform the statistical analyses. Our data was not normally distributed, therefore non-parametric tests were used to assess differences between the concentrations of environmental chemicals in households and schools (Mann-Whitney and Kruskal-Wallis), and to study correlations between indoor parameters and households (Spearman's rank correlation coefficient). With regards to the possible influence of building characteristics, indoor materials and housekeeping activities on the detected indoor chemicals, 11 categorical and 5 continuous variables were considered (Tables S1 and S2). For the multiple linear regression analysis, the data were log transformed prior to analysis. The statistical differences and correlations in Tables S1–S2 were used as a basis for the variables in the multiple linear regression analysis (Table 1). Significance was accepted for p-values $< 0.05$.

## 3. Results

### 3.1. Levels and determinants

The concentrations of phthalates, OPFRs and SCCPs were about 1000 times higher (ng/$m^3$) than those of PCBs and PBDEs (pg/$m^3$). The levels of PCBs and PBDEs decreased with the increasing number of halogen atoms in the molecules resulting in decreasing volatility (Fig. 1). The levels of the phthalates DMP, DEP and DiBP were 4–6 times higher in households compared to schools ($p \leq 0.004$)), with a slightly different pattern in households compared to classrooms. In households, we found highest median air concentrations for DiBP and DEP (304 and 284 ng/$m^3$, respectively), while in schools the highest air concentrations were found for DnBP followed by DiBP (91 and 69 ng/$m^3$, respectively) (Table S3). The levels of SCCPs were 3 times higher in the households compared to schools (128 and 43 ng/$m^3$, respectively) (Fig. 1 and Table S3). In contrast, the levels of the OPFRs, PCBs and PBDEs were similar for the two locations (Fig. 1, see Table S3 for numerical data).

Different building characteristics, indoor materials and housekeeping activities affect the air concentrations of different environmental chemicals as shown in Tables S1–S2. Statistically significant differences were detected for some chemical groups regarding location of household (OPFRs), airing of living room (selected PCBs and PBDEs), presence of upholstered couch (OPFRs), pet animal hold (selected PBDEs) and presence of electrical heaters (selected PCBs and PBDEs). The mean levels of the 5 continuous building and indoor variables for the households, as well as the correlation between these variables and the detected chemical levels are given in Table S2. Statistically significant correlations were detected for some chemical groups and the total size of household (OPFRs), frequency of vacuuming the living room (selected PCBs and PBDEs), frequency of washing the living room floor (selected PCBs) and the total number of TVs in the household (selected phthalates and SCCPs).

When performing multivariate analysis with these variables (building characteristics, indoor materials and housekeeping activities), frequency of washing the living room floor reduced the levels of many PCBs by two fold (Table 1). However, vacuuming the living rooms significantly increased the air concentrations of OPFRs and PBDEs. Among indoor materials, the use of electric heaters was negatively associated to some phthalates and PBDEs.

**Table 1**
Multiple linear regression analysis of environmental chemicals measured in indoor air with respect to indoor parameters (building characteristics, indoor materials and housekeeping activities).

| Environmental chemicals[a] | | Variables in the model | Unstandardized beta[b] | 95% confidence interval | | p-value | $R^2$ (%) |
|---|---|---|---|---|---|---|---|
| | | | | Lower bound | Upper bound | | |
| Phthalates | DMP | Total number of TVs | 1.34 | 1.05 | 1.70 | 0.018 | 13 |
| | DiBP | Single carpet in the living room (Yes/no) | 1.97 | 1.08 | 3.58 | 0.028 | 17 |
| | | Electrical heaters (Yes/no) | 0.50 | 0.26 | 0.99 | 0.048 | |
| | DnBP | Electrical heaters (Yes/no) | 0.43 | 0.21 | 0.89 | 0.024 | 17 |
| | | Fire place/wood burning (Yes/no) | 2.03 | 1.08 | 3.84 | 0.029 | |
| Chlorinated paraffins | SCCPs | Age of the house (years) | 0.99 | 0.99 | 1.00 | 0.035 | 14 |
| | | Renovated living room (Yes/no) | 0.76 | 0.55 | 1.05 | 0.096 | |
| Organophosphorous flame retardants | OPFRs | Vacuuming living room (times per week)[c] | 1.32 | 1.08 | 1.62 | 0.009 | 22 |
| | | Upholstered chair/couch (Yes/no) | 2.46 | 1.12 | 5.39 | 0.026 | |
| PCBs | TriCB | PVC chair/couch (Yes/no) | 2.45 | 1.34 | 4.49 | 0.005 | 18 |
| | TetraCB | County (Oslo/Akershus) | 1.63 | 1.09 | 2.45 | 0.019 | 29 |
| | | Type of material used in the building (Wood/Bricks and concrete) | 2.03 | 1.35 | 3.04 | 0.001 | |
| | | Vacuuming living room (times per week)[c] | 0.88 | 0.77 | 1.00 | 0.049 | |
| | PentaCB | Washing living room (times per week)[d] | 0.59 | 0.40 | 0.87 | 0.010 | 15 |
| | HexaCB | Washing living room (times per week)[d] | 0.61 | 0.41 | 0.92 | 0.018 | 13 |
| | HeptaCB | Washing living room (times per week)[d] | 0.51 | 0.27 | 0.98 | 0.043 | 9 |
| PBDEs | TriBDE | Electrical heaters (Yes/no) | 0.48 | 0.28 | 0.82 | 0.008 | 23 |
| | | Vacuuming living room (times per week)[c] | 1.21 | 1.01 | 1.44 | 0.037 | |
| | TetraBDE | Electrical heaters (Yes/no) | 0.47 | 0.26 | 0.84 | 0.012 | 22 |
| | | Vacuuming living room (times per week)[c] | 1.25 | 1.03 | 1.51 | 0.026 | |
| | PentaBDE | Electrical heaters (Yes/no) | 0.41 | 0.23 | 0.72 | 0.002 | 25 |
| | | Vacuuming living room (times per week)[c] | 1.19 | 0.99 | 1.43 | 0.065 | |

[a] Only those environmental chemicals that had significant predictors in the model are presented.
[b] This shows change in concentration of the investigated environmental chemical for every one-unit increase in variable in the model.
[c] Vacuuming of the floor.
[d] Using water/soap solution to wash the floor.

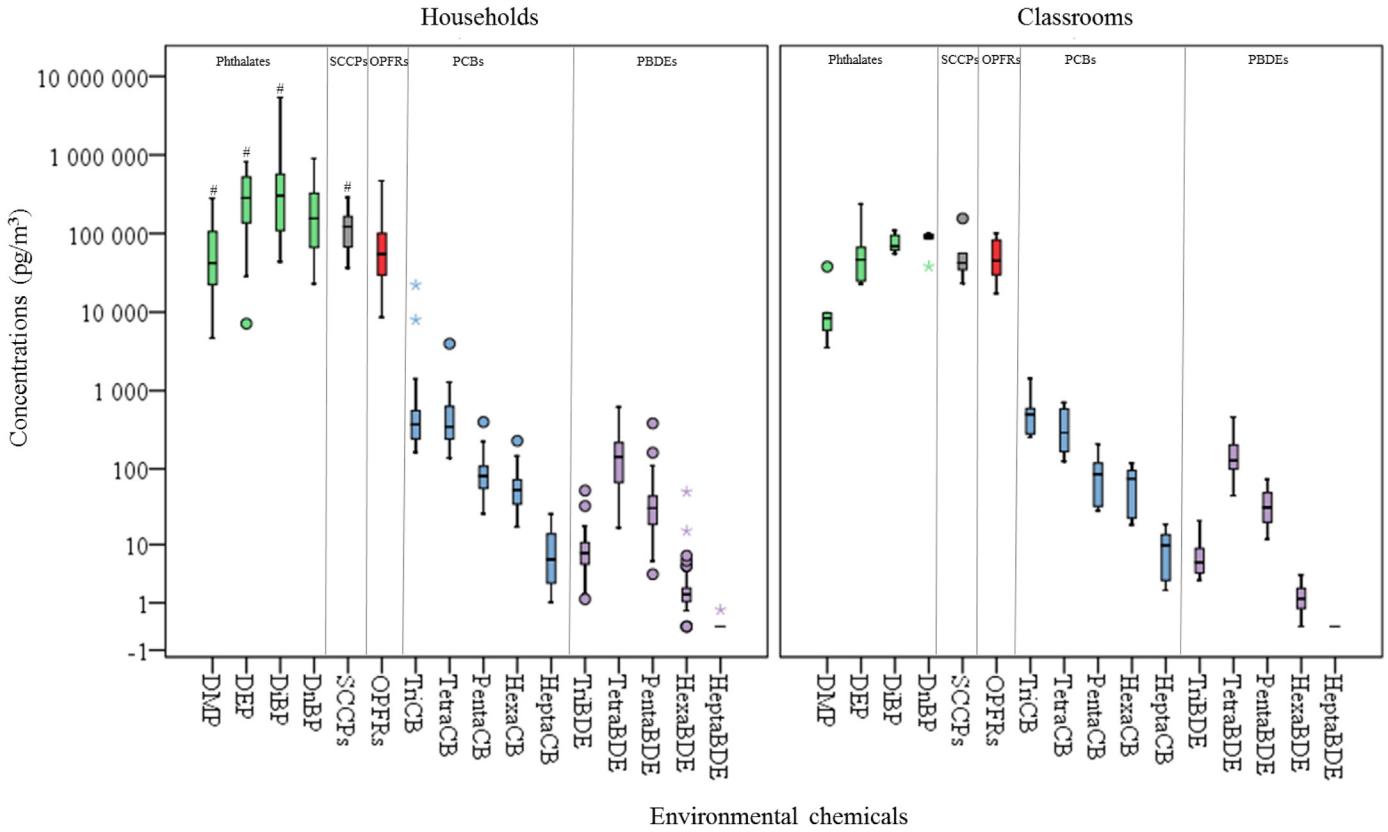


**Fig. 1.** Indoor air concentration of 4 different phthalates (DMP: dimethyl phthalate; DEP: diethyl phthalate; DiBP: di-isobutyl phthalate and DnBP: di-n-butyl phthalate), short chain chlorinated paraffins (SCCPs), organophosphorous flame retardants (OPFRs), PCBs (TriCB: sum of PCBs 18, 28, 31, 33 and 37; TetraCB: sum of PCBs 47, 52, 66 and 74; PentaCB: sum of PCBs 99, 101, 105, 114, 118, 122 and 123; HexaCB: sum of PCBs 128, 138, 141, 149, 153, 156, 157 and 167; HeptaCB: sum of PCBs170, 180, 183, 187 and 189) and PBDEs (TriBDE: PBDE 28; TetraBDE: PBDE 47; PentaBDE: PBDE 85, 99 and 100; HexaBDE: PBDE 153 and 154) in indoor air from houses and classrooms. Circles and stars represent moderate and extreme outliers, respectively. #air concentrations in houses are significantly different from classrooms, p-value < 0.05.

### 3.2. Estimated intakes

Table 2 shows the estimated daily intakes from air for both mothers and children. Among the chemicals, the highest median intakes were estimated for two phthalates (DEP and DiBP) in both groups. Also, the sum of phthalates represents the highest median intakes from all the chemical groups investigated (mothers: 150 and children: 226 ng/day/kg bw). The estimated median intakes were up to 1.7 times higher in children compared to mothers. For adults, the estimated intakes of DEP and DiBP from air were approximately 5 and 2 times higher compared to estimated intakes from food, respectively. For children, the estimated intake of DEP from air was approximately 3 times higher compared to estimated intakes from food, whereas for DiBP, the intakes were similar for both exposure routes. For both adults and children, estimated intakes for DnBP were similar for both exposure through food and air. For adults, the estimated intakes of SCCPs from air were similar to the intake from food when comparing with the Swedish dietary study (Yuan et al., 2017) (18 ng/day/kg bw) and 2–4 times lower than the two Japanese studies (Harada et al., 2011; Iino et al., 2005) (duplicate diet study: 55 ng/day/kg bw and market based study: 110 ng/day/kg bw) (not shown in Table 2). For OPFRs, the estimated intakes from air in adults were 8 times lower than the intakes from food (Table 2).

For PCBs, the intakes from food were taken from Kvalem et al. (Kvalem et al., 2009) and were not available for all the congeners. In Table 2, we have presented air intakes only for those congeners for which the available food intake estimates exist. The estimated intakes from food were higher compared to estimated intakes from air for adults. As expected, the contribution of intake from air decreased from 15% for the most volatile PCB (PCB 28) to 0.1% for the least volatile PCB (PCB 180). For children, no estimated intakes from food were available for single PCB congeners, but a total estimated intake from food of 8740 pg/day/kg body weight was available. The total intake of PCBs from air contributed approximately 2% to the children's total PCBs food intake. As for PCBs, the estimated inhalation intakes of PBDEs in adults from air were about 10–170 times lower than the dietary intake (Table 2). The estimated inhalation intakes of PBDEs in children from air contributed approximately 5% to the children's total PBDEs food intake (Table 2).

The estimated inhalation intakes were well below tolerable intakes (10,000 ng/day/kg body weight) allocated by the European Food Safety Authority (EFSA) for the phthalates (DiBP and DnBP) (EFSA, 2005c).

**Table 2**
Estimated daily intakes from air for different environmental chemicals in mothers and children compared to estimated dietary intakes. For PCBs and some PBDEs, the estimated intakes from air are shown only for congeners with available food intake data.

| Environmental chemicals | | Estimated intake from air | | | | | Estimated dietary intake |
|---|---|---|---|---|---|---|---|
| | | Number of participants | Percentiles | | Min | Max | |
| | | | 50th | 95th | | | |
| Mothers | | | | | | | |
| Phthalates | DMP | 42 | 8.4 | 38 | 0.92 | 59 | 9.4[a] |
| (ng/day/kg | DEP | 42 | 57 | 172 | 1.0 | 794 | 11[a] |
| body weight) | DiBP | 42 | 59 | 179 | 8.6 | 1034 | 29[a] |
| | DnBP | 42 | 26 | 175 | 3.8 | 188 | 29[a] |
| SCCPs (ng/day/kg body weight) | | 44 | 26 | 54 | 6.9 | 71 | 18[b] |
| OPFRs (ng/day/kg body weight) | | 48 | 11 | 89 | 1.7 | 115 | 85[c] |
| PCBs (pg/day/kg | PCB 28 | 44 | 21 | 440 | 8 | 1234 | 140[d] |
| body weight) | PCB 52 | 44 | 27 | 114 | 10 | 275 | 330[d] |
| | PCB 101 | 44 | 11 | 30 | 3.3 | 68 | 560[d] |
| | PCB 138 + 153 | 44 | 5 | 15 | 1.3 | 28 | 2760[d] |
| | PCB 180 | 44 | 0.3 | 1.9 | 0.0 | 2.3 | 370[d] |
| PBDEs | TriBDE | 47 | 1.5 | 8.2 | 0.2 | 11 | 20[e] |
| (pg/day/kg | TetraBDE | 47 | 24 | 127 | 2.8 | 148 | 440[e] |
| body weight) | BDE 99 + 100 | 47 | 5.7 | 55 | 0.70 | 102 | 210[e] |
| | HexaBDE | 47 | 0.29 | 6.9 | 0.0 | 67 | 50[e] |
| Children | | | | | | | |
| Phthalates | DMP | 50 | 10 | 78 | 1.9 | 96 | 27[a] |
| (ng/day/kg | DEP | 50 | 87 | 295 | 1.7 | 1846 | 26[a] |
| body weight) | DiBP | 50 | 85 | 886 | 15 | 1936 | 82[a] |
| | DnBP | 50 | 44 | 282 | 6.4 | 329 | 56[a] |
| SCCPs (ng/day/kg body weight) | | 52 | 39 | 96 | 11 | 114 | [g] |
| OPFRs (ng/day/kg body weight) | | 56 | 18 | 154 | 3.5 | 183 | [g] |
| PCBs (pg/day/kg | PCB 28 + 31 | 52 | 71 | 673 | 22 | 4476 | 8740[f] |
| body weight) | PCB 52 | 52 | 44 | 151 | 18 | 505 | |
| | PCB 101 + 118 | 52 | 20 | 61 | 6.7 | 110 | |
| | PCB 138 + 153 | 52 | 7.6 | 25 | 1.7 | 38 | |
| | PCB 180 | 52 | 0.5 | 3.6 | 0.0 | 3.8 | |
| PBDEs | TriBDE | 55 | 2.4 | 15 | 0.24 | 21 | 1125[f] |
| (pg/day/kg | TetraBDE | 55 | 42 | 245 | 4.1 | 348 | |
| body weight) | PentaBDE | 55 | 8.6 | 116 | 1.2 | 248 | |
| | HexaBDE | 55 | 0.49 | 15 | 0.0 | 156 | |

[a] Sakhi et al. (Sakhi et al., 2014; Sakhi et al., 2018).
[b] Yuan et al. (Yuan et al., 2017).
[c] Poma et al. (Poma et al., 2017).
[d] Kvalem et al. (Kvalem et al., 2009).
[e] Knutsen et al. (Knutsen et al., 2008).
[f] Karjalainen et al. (Karjalainen et al., 2012).
[g] Dietary intakes not available for children.

## 4. Discussion

The indoor environment may be polluted by a wide variety of potentially toxic chemicals. These chemicals are either in common use and found in a range of consumer products and building materials, or have previously been used and are still present in the indoor environment. These chemicals leak from the various products into the indoor environment and lead to exposure and risk of adverse health effects among residents.

### 4.1. Levels

Only two studies (Giovanoulis et al., 2018; Rakkestad et al., 2007) have previously documented presence of phthalates in Norwegian indoor environments. The levels and patterns of phthalates in the present study were similar to those observed in other Norwegian households in the recent study by Giovanoulis et al. (Giovanoulis et al., 2018). Rakkestad et al. (Rakkestad et al., 2007) has earlier characterized particles with respect to adsorbed organic compounds including phthalates in different indoor environments in Norway and found up to 20 times variation in the total amount of phthalates between the different locations. A review by Bølling et al. (Kocbach Bolling et al., 2013) showed that the overall range in the median levels in various countries is wider for DEP (50–2700 ng/m$^3$, highest concentrations were found in New York, USA) followed by DiBP (50–800 ng/m$^3$) and DnBP (200–2300 ng/m$^3$), for both DiBP and DnBP highest concentrations were found in Poland. The air levels of phthalates in Norwegian households were comparable and nearer to the lower limit in the ranges presented in Bølling et al. (Kocbach Bolling et al., 2013). To our knowledge, the present study is the first to report the concentrations of SCCPs in indoor air in Norwegian households and classrooms. Since 2001, Norway has prohibited production and use of substances or preparations containing SCCPs ≥1%. Additionally, articles containing SCCPs ≥0.15% were also prohibited. Furthermore, all SCCPs use is prohibited in EU since 2015. However, monitoring of outdoor air at Zeppelin Observatory

(Svalbard, Norway) in 2013 showed that the levels of SCCPs (360 pg/m$^3$) at this Arctic location were almost 3 times higher than other legacy POPs like PCBs, PBDEs and perfluorinated compounds (PFAS). When comparing the SCCPs levels with other studies worldwide, we found that the levels in Norway were comparable to indoor air concentrations in Sweden (Friden et al., 2011) (total chlorinated paraffins (short and medium) <5–210 ng/m$^3$) and China (Gao et al., 2016) (SCCPs: 60–1350 ng/m$^3$). Although PCBs use was prohibited some decades ago, several studies have measured the concentrations of these chemicals in different environmental samples including indoor air as shown in Table S4. Not surprisingly, the levels were higher in buildings where PCBs were reported to be used in sealant (Table S4). The levels in our Norwegian households were even lower than those in low-contaminated buildings in other countries. Based on existing literature and the current data, it seems that PCB-congeners 18, 28, 52, 101, 118, 138, 153 and 180 are among the most commonly occurring PCBs in indoor air. We observed that the PCBs in classrooms were slightly higher compared to the levels in households, although the differences were not statistically significant. The total median indoor air PBDEs concentrations in the current study (170 pg/m$^3$) was comparable to households in UK (128 pg/m$^3$), Denmark (275 pg/m$^3$) and Sweden (58 and 330 pg/m$^3$), lower than in Americans and Chinese households (760 and 628 pg/m$^3$, respectively) and higher than in Greek (11 pg/m$^3$), German (38 pg/m$^3$) and Australian (19 pg/m$^3$) households (Besis and Samara, 2012). OPFRs are used as presumably "safer replacements" for PBDEs and their use in different consumer products is increasing (van der Veen and de Boer, 2012). The levels of OPFRs in the current study (55 ng/m$^3$) was comparable to results from a Swedish investigation (40 ng/m$^3$) (Bergh et al., 2011), but was approximately 4 times lower compared to a Japanese study (217 ng/m$^3$) (Kanazawa et al., 2010).

### 4.2. Determinants

In general, there is lack of data showing possible determinants that could affect the indoor air levels of the investigated chemicals. Thus, it is difficult to find data for comparison of the current results. In the present study, we observed that the range of phthalate levels in Norwegian households was highest for DEP followed by DiBP, DMP and DnBP. The reason for the observed wide range in phthalate levels could be the presence of different consumer products as well as use of different personal care products especially with regard to DEP. For some phthalates, the number of TVs and use of carpet were positively associated with their air concentrations. This is in line other results showing that use of carpets and plastic products might affect the indoor air and dust levels of selected phthalates (Sukiene et al., 2016).

The negative association of SCCPs with the age of the houses and renovation of the living room is in accordance with a gradually decreased use of SCCPs in different products due to the regulatory restrictions mentioned above. Thus, the sources of SCCPs are most likely old products and building materials. For PCBs, use of PVC, type of building material and localization had an impact on their air levels. Among different housekeeping activities, washing of the living room was negatively associated to the air levels of less volatile PCBs. This could be explained by the fact that frequent washing reduces the amount of floor dust and its possibility to resuspend into the indoor air.

The levels of PBDEs in air were negatively associated with use of electric heaters, while they were positively associated with frequency of vacuuming the living room. The reason for the latter could be the resuspension of dust containing PBDEs during vacuuming. The households that had more foam furniture had significantly higher air levels of OPFRs indicating that OPFRs might be used as flame retardants in this type of furniture. Vacuuming the living room also increased the air levels of OPFRs, probably due to the same cause as for the PBDEs.

### 4.3. Exposure estimates

Ideally, comparison of estimated intakes from different sources (gas phase, particulate phase, settled dust and food) should done for measurements performed in the same study. In the present study, we measured gas and particulate phase only. Since our estimates for inhalation exposure assume 100% absorption of the chemicals from both the gas and particle phase of the air, they are likely to overestimate the actual burden from inhalation exposure. More specifically, only a fraction of the compounds in gas phase are absorbed in the airways and for the PM adsorbed chemicals, the inhaled and deposited fraction is known to be below 100%. An exception may be for volatile compounds such as the low molecular weight phthalates DEP and DnBP, which showed an inhalation uptake close to 100% in a recent human exposure study (Weschler et al., 2015). Thus, the estimates for these low molecular weight phthalates are likely to be a more correct approximation of the actual burden from inhalation exposure compared to estimates for less volatile compounds. Regarding chemicals adsorbed to PM, their lung absorption depends on the deposited fraction for the PM and their physicochemical properties. Since the distribution between particle and gas phase is not determined here, it is not possible to predict the error these factors will add to the estimates.

The estimated inhalation exposure for phthalates was either higher (DEP or DiBP) or of the same magnitude (DMP and DnBP) as the estimated food intake. This is in line with Dewalaque et al. (Dewalque et al., 2014) showing that dietary intake contributes <20% for DEP, DiBP and DnBP, but in contrast to Giovanoulis et al. (Giovanoulis et al., 2018) showing >60% contribution of the dietary intake to the total intake for these phthalates. Since the air concentrations in the present study were in the same range as those reported by Giovanoulis et al. (Giovanoulis et al., 2018), the observed differences are most likely due to differing dietary intake assessment.

For adults, inhalation can be an important source of exposure for SCCPs, and inhalation was recently reported to account for 76–98% of the exposure depending on the dust intake rate (Friden et al., 2011). Few studies are available showing SCCPs intakes from food. The intakes from food were comparable between the Swedish study (Yuan et al., 2017) (18 ng/day/kg bw) and two Japanese studies (55–110 ng/day/kg bw) (Harada et al., 2011; Iino et al., 2005). However, a Chinese study showed 10 fold higher concentrations in food compared to these studies and consequently SCCPs intakes from food were around 760 ng/day/kg bw (Harada et al., 2011). It is more appropriate to compare the intakes from air in the present study to food intakes from Sweden since the Norwegian food habits are more similar to those from Sweden rather than from Japan and China. The estimated intakes from air in the present study (26 ng/day/kg bw) are comparable to the food intakes (18 ng/day/kg bw) and are, thus, not negligible. This is in accordance with the observations made by Fridén et al. (Friden et al., 2011).

Our data showed that the exposure to PCBs from air (0.1–15%) contributes less than the exposure from food. This is in line with Harrad et al. (Harrad et al., 2009) showing that indoor air contributed 15% to the overall PCBs exposure in England. Nevertheless, the importance of inhalation exposure to PCBs is supported by data showing that indoor air levels are positively associated with an increased body burden of PCBs (Fitzgerald et al., 2011). Higher contributions of indoor air to overall PCBs exposures (up to 46%) were observed in countries outside Europe (US, Canada and New Zealand). This could indicate varying use of PCBs-containing building materials before their ban in different countries, thereby affecting the contribution that different exposure pathways have on the total exposure.

For PBDEs, inhalation contributed <10% to exposure compared to the dietary intake. This is in accordance with a previous study showing that food and dust ingestion are among the major exposure sources (Frederiksen et al., 2009). The estimated intake from air has been reported to vary extensively depending upon the type of OPFR studied

(Xu et al., 2016). A recent study from Sweden by Poma et al. (Poma et al., 2017) measured 8 different OPFRs in 53 composite food samples and estimated a total OPFRs intake of 85 ng/day/kg bw from food. Although the total OPFR intake from air in the present study is 8 times lower than intake from food, indoor air may still be an important source of exposure as the urinary metabolites of different OPFRs were shown to be more strongly correlated with air and dust exposure compared to diet (Cequier et al., 2015).

Overall, inhalation exposure is likely to result in higher doses of the more volatile of the investigated chemicals in the respiratory tract than in other organs. At least for phthalates, exposure from indoor air could be of relevance for respiratory conditions such as asthma and airway hyper reactivity as indicated by Bornehag et al. (Bornehag and Nanberg, 2010). In addition, exposure by inhalation could circumvent important detoxification mechanisms in the liver implying that such exposures may have a potential impact on the toxicity of these chemicals.

## 5. Conclusion

The present study showed that the indoor air levels of low molecular weight phthalates (DMP, DEP, DiBP and DnBP), OPFRs and SCCPs were up to 1000 times higher than the levels of PCBs and PBDEs in both households and schools. A number of parameters related to building characteristics, applied indoor materials and housekeeping activities of the participants had a significant impact on the levels of the targeted chemicals. Finally, indoor air contributed less to the total exposure compared to the diet for most of the chemicals investigated, except for low molecular weight phthalates (DMP, DEP, DiBP and DnBP) and SCCPs where the contributions from diet and air were of similar magnitude. Despite of the higher contribution from diet than air for most of the assessed chemicals, inhalation could still be an important exposure pathway when assessing the causes of adverse respiratory health outcomes.

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## Appendix A. Supplementary data

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