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1 **The Air that we Breathe: Neutral and volatile PFAS in Indoor Air**

2

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10

11 **Abstract**

12 Sources of exposure to per- and polyfluorinated alkyl substances (PFAS) include food,
13 water, and given that humans spend typically 90% of our time indoors, air and dust.
14 Quantifying PFAS prevalent indoors, such as neutral, volatile PFAS, and estimating their
15 exposure risk to humans is thus important. To accurately measure these compounds indoors,
16 polyethylene (PE) sheets were employed and validated as passive detection tools, and analyzed
17 by gas chromatography-mass spectrometry. Air concentrations were compared to dust and
18 carpet concentrations reported elsewhere. Partitioning between PE sheets of different
19 thicknesses suggested that interactions of the PEs with the compounds are occurring by
20 absorption. Volatile PFAS, specifically fluorotelomer alcohols (FTOHs), were ubiquitous in
21 indoor environments. For example, in carpeted Californian kindergarten classrooms, 6:2 FTOH
22 dominated with concentrations ranging from 9-600 ng m⁻³, followed by 8:2 FTOH.
23 Concentrations of volatile PFAS from air, carpet and dust were closely related to each other,
24 indicating that carpets and dust are major sources of FTOHs in air. Nonetheless, air posed the
25 largest exposure risk of FTOHs and biotransformed perfluorinated alkyl acids (PFAA) in

26 young children. This research highlights inhalation of indoor air as an important exposure
27 pathway and the need for further reduction of precursors to PFAA.

28

29 Passive sampling, Polyethylene sheets, PFAS precursors, gas-phase, Carpet, Dust, Risk
30 Assessment

31

32 Polyethylene (PE) sheets are effective passive samplers for PFAA precursors which are
33 ubiquitous in indoor air and dominate indoor exposure.

34

35 Introduction

36 Human exposure to fluorotelomer alcohols (FTOHs), perfluorooctane sulfonamides
37 (FOSAs) and perfluorooctane sulfonamidoethanols (FOSEs) and other precursors to
38 perfluoroalkyl acids (PFAA) comes primarily from consumer and industrial products readily
39 available in people's homes.¹⁻⁴ FTOHs were the dominant polyfluorinated compounds in
40 indoor air⁵ where ~60% of detected per- and polyfluorinated alkyl substances (PFAS) were
41 associated with the particle phase.² Since most people spend more than 90% of their time
42 indoors,² indoor air and dust are important uptake pathways for human PFAS exposure⁶ in
43 addition to the widely recognized exposure sources of diet and water.⁷⁻⁹ Indeed, correlations
44 between elevated indoor air exposure to precursors and increased PFAS serum concentrations
45 have been reported.^{10,11}

46 The use of passive sampling, which can measure the concentration of freely dissolved
47 or gas-phase trace organic contaminants, has been widely accepted as an effective detection
48 tool.^{12,13} Single-phase polymers, such as polyethylene (PE) sheets, have been able to detect a
49 wide range of non-polar and moderately polar contaminants in the gas phase or dissolved in

50 water.^{14,15} In addition, PE sheets are inexpensive, easy to handle, and can be easily transported
51 and deployed.¹³ Recently, neutral PFAS were successfully measured in outdoor air and water
52 using PE sheets.¹⁶ However, the partitioning of neutral PFAS into or onto the PE sheets indoors
53 is not yet fully understood.

54 To further assess the role that indoor environments play as an exposure source of
55 airborne PFAS in gas-phase and dust, the main objectives of this research were to (i) derive
56 indoor PE-air partitioning coefficients (K_{PE-air}); (ii) compare the volatile PFAS composition in
57 different indoor environments using PE sheets as passive samplers; (iii) evaluate the air-dust
58 partitioning of PFAS in carpeted kindergarten classrooms, and (iv) estimate daily intake (EDI)
59 in children 2-6 years of age.

60 Materials and Methods

61 Sampling of neutral PFAS was performed in carpeted kindergarten classrooms,
62 residences, an outdoor gear and apparel store in northern California; university offices,
63 classrooms, laboratories, and a carpet store in southern Rhode Island between 2018 and 2020.
64 A total of 90 PE sheets were deployed in the indoor locations, in addition to eight radiello
65 samplers (Sigma Aldrich) with precleaned XAD-4 as sorbent used for active sampling.

66 Two types of precleaned PE passive samplers differentiated by thickness (25 μm and
67 50 μm) were deployed for 14 days (validation study), 21 days (kinetic study), and 28 days
68 (measurements). Active sampling was performed on days 1, 7 and 14 where the radiello
69 samplers were attached to a QuickTake 30 SKC Pump at a constant flow of 5 L min^{-1} for 240
70 minutes. All samples were kept in a freezer at -20°C until extraction (for details, see SI).

71 **Instrumental analysis**

72 Samples were analyzed for nine neutral PFAS on an Agilent 7890B gas chromatograph
73 coupled to an Agilent 5977A mass selective detector (MSD) device operating in positive
74 chemical ionization mode using selected ion monitoring (for details, see SI).

75 **Data interpretation**

76 The partitioning constants of neutral PFAS between PE and air (K_{PE-air}) were derived
77 in the validation study as:

$$78 \quad K_{PE-air} = \frac{C_{PE}}{C_{air}} \quad (1),$$

79 where C_{PE} is the concentration in PE sheets (ng g^{-1}_{PE}), and

80 C_{air} is the gas-phase concentration (ng m^{-3}).

81 Active sampling was used in the K_{PE-air} validation study only. For all other campaigns, C_{air} was
82 calculated using equation (1). Partitioning within the PE sheets was derived as the ratio of the
83 25 μm passive sampler (C_{25}) to the 50 μm passive sampler (C_{50}) amounts at equilibrium (for
84 details, see SI).

85 **Daily intake**

86 The total estimated daily intake (EDI) of neutral PFAS via air and dust was calculated
87 from PFAS concentrations measured here, and dust concentrations reported elsewhere¹⁷ based
88 on established methods^{18,19} (for details, see SI).

89 **QA/QC**

90 Field blanks, matrix spikes, matrix blanks, and field duplicate samples were included
91 with each sample batch. Matrix spikes were prepared by spiking 50 μL of an 80 $\text{pg}/\mu\text{L}$ native
92 standard solution and 50 μL of an 80 $\text{pg}/\mu\text{L}$ mass labelled standard solution into a clean (unused
93 and never removed from the laboratory) sampler. Method detection limits (MDL) were
94 calculated as the blank average plus three times the standard deviation; however, when a
95 compound was not detected in the blanks, instrumental limits of detection (ILOD) were used.

96 Only values above limits of quantitation (LOQ) were reported (for details, see SI, Table S2).
97 Recoveries of the matrix spikes ranged between 81% (± 35) to 111% (± 19) for all compounds
98 (for details, see SI, Table S2).

99 Results and Discussion

100 PE-air Partitioning Constants

101 Results from the kinetic study showed that 6:2 FTOH and 8:2 FTOH reached
102 equilibrium after 14 days (see SI Figure S1). Log K_{PE-air} values were approximately 4 -5 for
103 the FTOHs, ~ 5 for 8:2 FTAc, and increased with molecular weight. Although 10:2 FTAc,
104 FOSAs and FOSEs were detected by PE sheets, none were detected by active sampling;
105 calculating their equilibrium partitioning constant was not possible (see SI Table S6). There
106 were only minor differences between the 25 μm and 50 μm K_{PE-air} results, indicating good
107 reproducibility of PE sheets as passive samplers.

108 Mean log K_{PE-air} values from the FTOHs of this study were approximately three log
109 units lower than those reported for outdoors (see SI Table S3)¹⁶ where break-through and
110 environmental factors could have affected the partitioning of the compounds. Missing K_{PE-air}
111 values were derived based on a correlation between previously reported¹⁶ and currently
112 measured PE-air partitioning constants. Further studies are needed to corroborate the
113 partitioning coefficients of the FOSAs and FOSEs.

114

115 **Table 1. Indoor log K_{PE-air} values from the validation study for 25 and 50 μm PE sheets.**

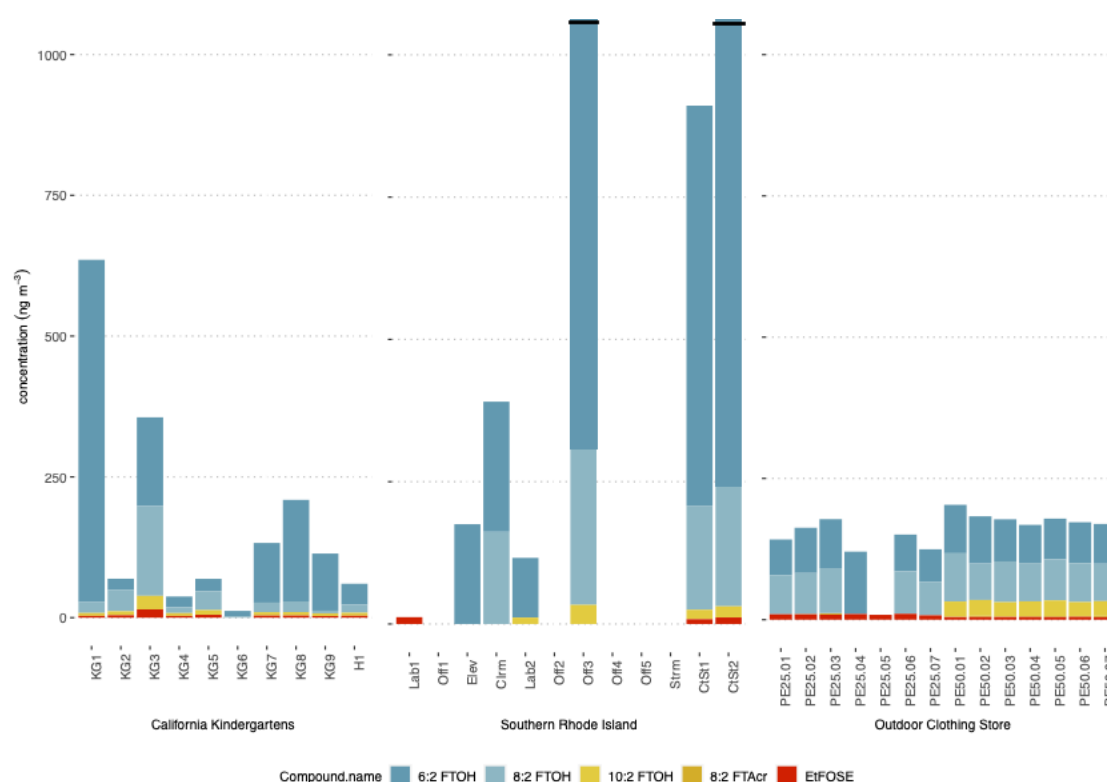
Compound	Molecular weight ($g\ mol^{-1}$)	Mean log $K_{PE-air\ 25}$ (this study)	Mean log $K_{PE-air\ 50}$ (this study)
6:2 FTOH	364.1	4.4 ± 0.1	4.3 ± 0.0
8:2 FTOH	464.1	4.3 ± 0.1	4.5 ± 0.0
10:2 FTOH	564.1	5.0	5.0 ± 0.0
8:2 FTAc	518.1	4.9 ± 0.4	5.0 ± 0.2

10:2 FTAcr	618.1	5.2*	5.3*
MeFOSA	527.2	5.1*	5.2*
EtFOSA	513.1	ND	ND
MeFOSE	571.2	5.2*	5.3*
EtFOSE	557.2	5.2*	5.2*

116 ND=not detected, *K_{PE-air} from this study was estimated based on a correlation between those measured here and those
 117 reported by Dixon-Anderson and Lohmann, (2018)¹⁶. Estimated K_{PE-air} = 0.44 x K_{PE-air, measured} (Dixon-Anderson and Lohmann,
 118 2018) + 1.30 (RSQ_{PE50}=0.67).
 119

120 PE-air Partitioning ratios

121 The partitioning ratios of the weight-normalized neutral PFAS between 25 and 50 μm
 122 thick PE sheets were ~1 (see SI Figure S2) implying absorption as the mechanism of
 123 partitioning. The greater mass of the 50 μm PE sheets for the same size resulted in easier
 124 detection and is thus preferable for future studies.



125 **Figure 1. Indoor air concentrations measured at California Kindergarten classrooms**
 126 **and an outdoor clothing store, and university classrooms, offices and laboratories, and a**
 127 **carpet store in southern Rhode Island.** H: home; KG: kindergarten classrooms; Lab: laboratory;
 128 Off: office; Elev: elevator; Clrm: classroom; Ctst: carpet store; Strm: storage room. Numbers (i.e.
 129

130 KG7) are indicative of separate/individual samples. Off3 and CtSt2 have concentrations above 1000
131 ng m⁻³.

132 **Neutral PFAS Indoor Air Concentrations**

133 Indoor air concentrations were derived from the PEs for neutral PFAS in (1) California
134 Kindergarten classrooms (SI Table S4); (2) offices, classrooms and laboratories at a university,
135 and a nearby carpet store in southern Rhode Island (SI Table S5); and (3) a storage room at an
136 outdoor clothing store in California (SI Table S6), see Figure 1. Neutral PFAS were present at
137 all locations, dominated by FTOHs, in-line with previous results.^{5,20,21} PFAS profiles and
138 concentrations varied between locations, though, likely driven by the different PFAS-
139 containing products present. These results indicated that PE-sheets can be used to determine
140 differences in PFAS profiles and concentrations in various indoor air settings.

141 In the California kindergarten classrooms, 6:2 FTOH dominated with concentrations
142 ranging from 10-600 ng m⁻³ (accounting for 29-96 % of sum of nine PFAS), followed by 8:2
143 FTOH (2-160 ng m⁻³, 3-54% of total PFAS) (Figure 1; SI Table S2). In three kindergarten
144 classrooms (KG2, KG3, and KG5), concentrations of 8:2 FTOH exceeded those of 6:2 FTOH.
145 In all kindergarten classrooms, EtFOSE was present at low concentrations, while MeFOSE was
146 below method detection limits (MDLs) (Figure 1; SI Table S2). EtFOSA, 8:2 FTAc, and 10:2
147 FTAc were not detected (SI Table 3).

148 When detected, 6:2 FTOH (with detection frequency of 83%, and ranging from < MDL
149 – 1900 ng m⁻³), and 8:2 FTOH (17%, < MDL-270 ng m⁻³) also dominated total PFAS in the
150 university rooms (Figure 1; SI Table S5). FTOHs were detected only in carpeted rooms and in
151 the analytical laboratory (SI Table S5). The detection of 10:2 FTOH was sporadic, with
152 concentrations up to 33 ng m⁻³ (Figure 1; SI Table S5). MeFOSA, EtFOSE, and MeFOSE were
153 at or < MDL at all sites, while EtFOSA and FTAc were rarely above MDLs (SI Table S5).

154 Volatile PFAS were present in all replicates from the outdoor clothing store (Figure 1).
155 FTOHs were the most abundant and dominant group; consistent with previous studies on the

156 composition of PFAS in various indoor environments.^{5,20,21} The most abundant compound was
157 8:2 FTOH, with an average concentration of $\sim 200 \text{ ng m}^{-3}$, followed by 6:2 FTOH and 10:2
158 FTOH with average concentrations of 70 ng m^{-3} and 30 ng m^{-3} respectively (Figure 1). The
159 dominance of 8:2 FTOH is concerning since this and other longer-chain PFAS have been
160 phased out by PFAS producers in the United States, European Union, and Japan²². These
161 results show that these compounds are still being used for textiles, and possibly point to textile
162 imports from other countries where PFAS are poorly regulated.²³

163 Fraser et al. (2011)¹¹ reported concentrations of FTOHs ranging from <MDL to 11 ng
164 m^{-3} (6:2 FTOH), $0.3 - 70 \text{ ng m}^{-3}$ (8:2 FTOH), and $0.14 - 12 \text{ ng m}^{-3}$ (10:2 FTOH) in multiple
165 office spaces in Boston, Massachusetts, similar to results report here and in other studies.^{5,21} A
166 study in Ottawa, Canada, in 2005 reported concentrations of MeFOSE, EtFOSE and EtFOSA
167 in indoor air of $\sim 7 \text{ ng m}^{-3}$, 2 ng m^{-3} and 0.1 ng m^{-3} , respectively², and even lower in Vancouver,
168 Canada in 2011, at 0.4 ng m^{-3} , 0.06 ng m^{-3} , 0.03 ng m^{-3} , and 0.02 ng m^{-3} respectively.²¹ In the
169 present study, although present in many locations, FOSEs rarely exceeded concentrations of
170 0.001 ng m^{-3} . FOSAs were detected even fewer times. The difference in concentrations of the
171 FOSAs and FOSEs in different locations across North America could reflect geographic
172 differences of indoor sources. Additionally, the difference between older and newer data could
173 point to the phase out of PFOS-based chemicals, including FOSAs and FOSEs since 2002,
174 whereas the use of replacement FTOHs in North America has increased since 2000.^{21,24,25}

175

176 **Air-dust-carpet partitioning**

177 Concentrations of neutral PFAS in dust and carpet of the same kindergarten classrooms
178 were measured by Wu *et al.* (2020)¹⁷ (see SI Table S7). Strong correlations ($\text{RSQ} > 0.7$, $\text{P} \leq 0.05$)
179 were observed between different FTOHs in air-dust, and air-carpet (and dust-carpet from Wu

180 *et al.* (2020)¹⁷, see SI Table S8), except for 6:2 FTOH in air-carpet. On the other hand, FOSEs
181 were not strongly correlated in air, dust or carpet.

182 Distribution of PFAS between indoor air and floor dust were reported to be controlled
183 by partitioning between the gas phase and PFASs sorbed to the organic phases in the dust.²⁶
184 Our results corroborated that neutral PFAS were present in air and partitioned to dust. Given
185 that the origin of volatile PFAS in air in the (carpet-free) outdoor clothing storage room was
186 likely to be textiles, it is possible that multiple products in the kindergarten classrooms were in
187 fact the source of these compounds that also partitioned into carpet and dust. Previous work
188 demonstrated that FTOHs, FOSAs, and FOSEs degrade in the atmosphere into more stable
189 PFAA.^{27,28} Significant associations between precursors in air and PFCAs in dust have been
190 reported (e.g., 6:2 FTOH and PFHxA).²⁹ Similarly, significant associations were observed
191 between FOSAs/FOSEs in air and PFOS and PFDS in house dust.²⁸

192 In contrast to FTOHs, there were no significant correlations between the
193 FOSAs/FOSEs in air, dust and carpet from this study, suggesting that the sources of
194 FOSAs/FOSEs were different and likely not linked to carpets or textiles. A previous study did
195 not find significant correlations between the FOSEs in kindergarten classrooms either, but did
196 however find strong associations in offices³⁰, implying that there were common sources of
197 these sulfonamidoethanols in items associated with office spaces that perhaps were not usually
198 found in kindergarten classrooms. Additionally, as previously mentioned, the production of
199 FOSAs/FOSEs has been largely phased-out of production since 2002²⁴, and thus their low
200 concentration or absence is expected³¹.

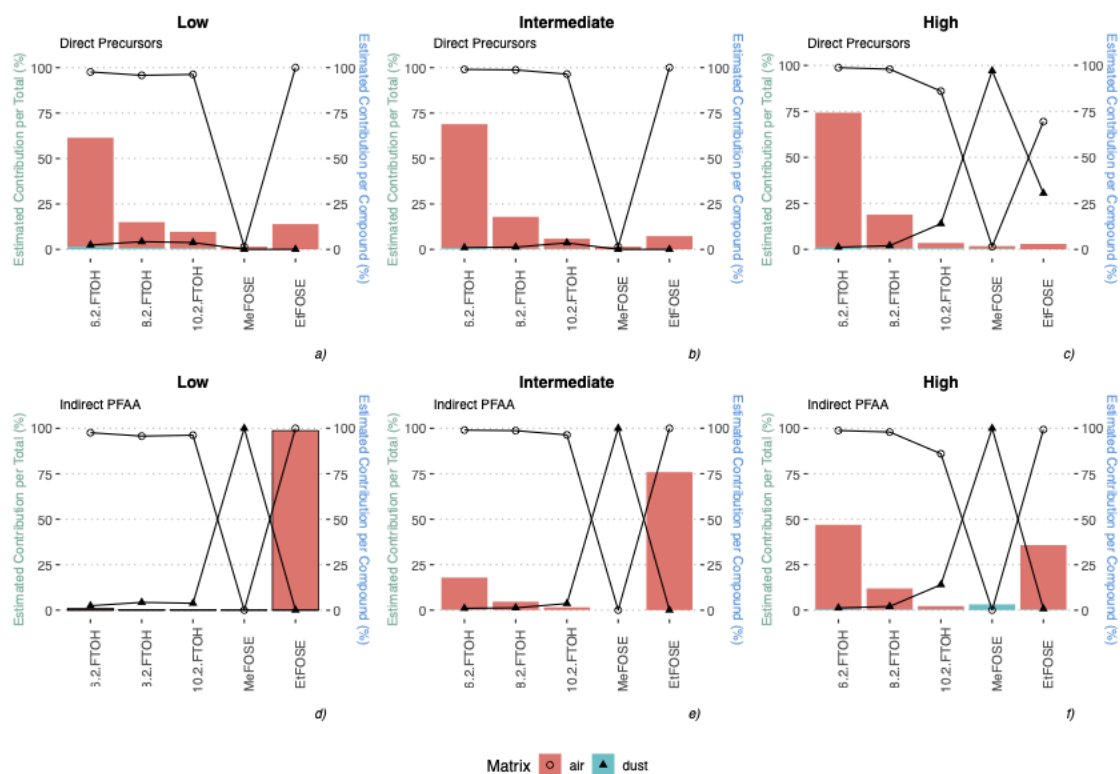
201

202 **Estimated daily intake of volatile PFAS through air and dust**

203 To assess the relevance of volatile and neutral PFAS in indoor air for children aged two
204 to six years old, the estimated daily intake (EDI) was calculated (SI Table S8) for three

205 exposure estimates (low, medium, high, see SI). Biotransformation constants for each
206 compound were used to calculate their contribution to the \sum PF_{AA} intake (SI Table S8).

207 Total EDI (SI Table S9) was 1.5 ng kg⁻¹ bw day⁻¹ for low exposure, 14 ng kg⁻¹ bw day⁻¹
208 ¹ for intermediate exposure, and 150 ng kg⁻¹ bw day⁻¹ for high exposure. Compounds that were
209 regularly detected in both air and dust were 6:2 FTOH, 8:2 FTOH, and, to a lesser extent, 10:2
210 FTOH; while MeFOSE and EtFOSE appeared to have significantly larger contributions in dust
211 than air (Figure 2, SI Table S9). Volatile and neutral PFAS measured in air contributed 4.9-62
212 % to \sum PF_{AA} intake, while ionic PFAS measured in dust contributed 34-95 % (SI Table S9).
213 These results are similar to other studies that found precursors contributing 41–68 % to \sum PFOS
214 uptake via all investigated exposure pathways,³² and precursors responsible for 90 % to the
215 \sum PFOS intake in air (Figure 2).^{26,33} Our results imply that air inhalation was a major exposure
216 pathway for FTOHs, while dust ingestion was dominant for FOSEs in children, similar to prior
217 results.³⁰



219

220 **Figure 2. Percent of volatile and neutral PFAS (top panel) and indirect or**
 221 **biotransformed PFAA (bottom panel) intake via air inhalation (pink) and dust ingestion**
 222 **(blue) for children at ages 2 through 6.** Bars represent the relative contribution of
 223 individual precursors to total PFAS (left axis); bars are differentiated by color for both
 224 matrices. Lines represent the percent estimated contribution for each compound in air and
 225 dust (right axis). MeFOSE was detected at low concentrations in dust and <MDL in air.

226

227

228 Given the potential for precursors to be biotransformed into more stable PFAA,
 229 estimations of PFAA indirect exposure were also calculated as $1.2 \text{ ng kg}^{-1} \text{ bw day}^{-1}$, 75 ng kg^{-1}
 230 bw day^{-1} , $2800 \text{ ng kg}^{-1} \text{ bw day}^{-1}$ for the low, intermediate, and high exposure scenario
 231 respectively (SI Table S9). The major contributors to indirect PFAA exposure were 6:2 FTOH
 232 and 8:2 FTOH in air, and MeFOSE in dust (Figure 2). This study demonstrated that volatile
 neutral PFAS, such as FTOHs, are major contributors to exposure in air.

233 Supporting Information

234 The Supporting Information contains additional details on the analysis, data interpretation
235 and EDI calculation, and is available free of charge at <https://pubs.acs.org/doi/XXX>.

236

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242

243 Conflict of interest

244 The authors declare no competing financial interests.

245

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247

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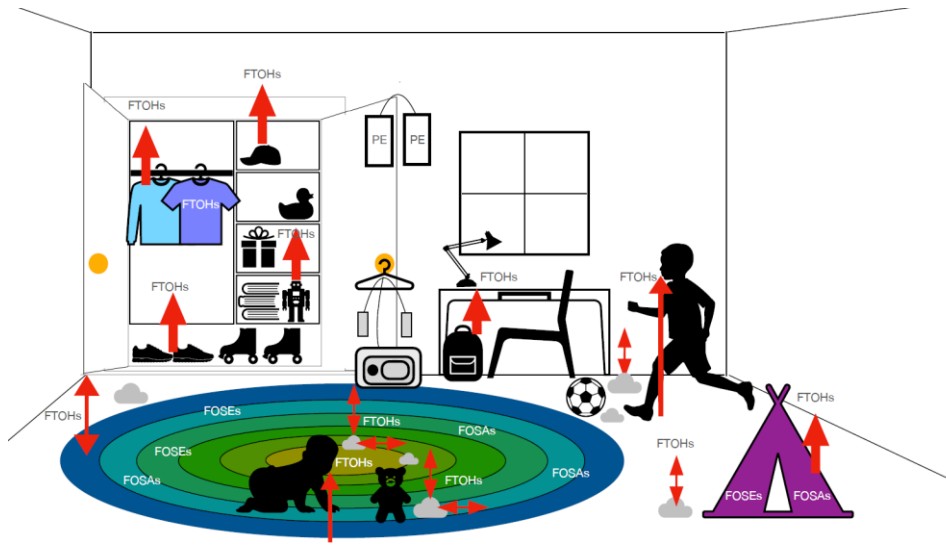
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369 **Graphical TOC**

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