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1 **Influence of suspended particles on the emission of**
2 **organophosphate flame retardants from insulation**
3 **boards**

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12

Abstract

The influence of the presence of so-called seed particles on the emission rate of Tris(2-chloroisopropyl) phosphate (TCIPP) from polyisocyanurate (PIR) insulation board was investigated in this study. Two Field and Laboratory Emission Test cells (FLEC) were placed on the surface of the same PIR board and respectively supplied with clean air (reference FLEC) and air containing laboratory generated soot particles (test FLEC). The behavior of the area specific emission rates (SE_{RA}) over a time period of 10 days was studied by measuring the total (gas + particles) concentrations of TCIPP at the exhaust of each FLEC. The estimated SE_{RA} of TCIPP from PIR board at the quasi-static equilibrium were found to be $0.82 \mu\text{g}\cdot\text{m}^{-2}\cdot\text{h}^{-1}$ in absence of seed particles, whilst the addition of soot particles led to SE_{RA} of $2.16 \mu\text{g}\cdot\text{m}^{-2}\cdot\text{h}^{-1}$. This indicates an increase of the SE_{RA} of TCIPP from PIR board with a factor of 3 in the presence of soot particles. The TCIPP partition coefficient to soot particles at the quasi-static equilibrium was $0.022 \pm 0.012 \text{ m}^3\cdot\mu\text{g}^{-1}$. In a next step, the influence of real-life particles on TCIPP emission rates was investigated by supplying the test FLEC with air from a professional kitchen where mainly frying and baking activities took place. Similar to the reference FLEC outcomes, SE_{RA} was also found to increase in this real-life experiment over a time period of 20 days by factor 3 in the presence of particles generated during cooking activities. The median value of estimated particle-gas coefficient for this test was $0.062 \pm 0.037 \text{ m}^3\cdot\mu\text{g}^{-1}$.

Keywords: Flame retardants, Gas-particle partition, Specific emission rates, Indoor environment

Introduction

Many semi-volatile organic compounds (SVOCs) are widely used as additive flame retardants (FRs) in consumer products and building materials to reduce their flammability. The additive (when not chemically bound to the material) use of FR chemicals results in an easy release to different environmental media during normal use of the treated product. Many studies have reported elevated concentrations of FRs in different indoor compartments, such as dust (Abdallah et al. 2008; Cequier et al. 2014; Van den Eede et al. 2011), air (Cequier et al. 2014; Newton et al. 2015) and also in humans (Cequier et al. 2015; Kucharska et al. 2015). The organophosphate flame retardants (PFRs) are a well-known suitable substitute of brominated FRs, because of their better technical characteristics (including ...). Tris(2-chloroisopropyl) phosphate (TCIPP) is a frequently used chemical, both as flame retardant and plasticizer, in various building materials and foam fillers. This chemical is one of the most frequently detected FRs in various indoor compartments (Wei et al. 2015). In spite

of its low acute toxicity, TCIPP is recognized as a human health concern because of its suspected carcinogenic impact and high bioaccumulative potential (van der Veen et al. 2012). In order to assess the human exposure to TCIPP, it is important to understand the mechanisms by which it is emitted and transported in indoor environments. As a chemical with low vapor pressure, it is expected that indoor surfaces, including airborne particles, play an important role in the transport of TCIPP in indoor environments (Weschler et al. 2008a; Xu et al. 2009).

Recent studies suggest that indoor airborne particles can influence emission rates of SVOCs from the products (Benning et al. 2013; Hu et al. 2013; Liu et al. 2012a; Liu et al. 2012b; Xu et al. 2006). For instance (Benning et al. 2013) found that the emission rate of other SVOC (di-2-ethylhexyl-phthalate (DEHP)) emitted from vinyl flooring significantly increased in a presence of ammonium sulfate particles. Moreover, the authors reported that the main mechanism explaining this finding was the rapid sorption of DEHP to the particles; the DEHP-particle sorption equilibration time was demonstrated to be less than 1 min. This resulted in a decrease of the gas phase concentration and, due to the gradient in concentrations, more DEHP was emitted from the source. Furthermore, the other studies demonstrated that the emission rates of SVOCs are influenced also by additional factors. For instance, Liang et al. (1997) demonstrated that the particle-air partition of SVOCs was stronger on organic versus inorganic particles (Liang et al. 1997). The model developed by Liu et al (2012) showed that the SVOC flux between the particles' surface and the bulk air depends on the size of the particles. Their model predicts that the flux will be significantly enhanced in the presence of particles with size less than 2 μm .

The objective of this study was to quantify the effect of generated soot particles on TCIPP emissions from polyisocyanurate (PIR) insulation boards and to estimate the TCIPP particle-gas partition coefficient to soot particles. Furthermore, the effect of particles generated during cooking activities on TCIPP emissions was assessed in a real-life experiment. The results of these experiments contribute to a better understanding of the emission and transportation processes of TCIPP in indoor environments and provide information on the validity of SVOC emission rates, assessed under controlled laboratory conditions, for health exposure modelling purposes.

Materials and methods

Chemicals

Analytical grade TCIPP (97.5 %, mixture of isomers) for calibration and identification was purchased from Sigma-Aldrich, USA. Triamyl phosphate (TAP) (98.9 %) was used as internal standard and it was purchased from Tokyo Chemical Industry, Japan. All standard solutions were prepared by dilutions in methanol. The solvents were GC grade with a purity >99.9% (Merck, Germany). Nitrogen gas of 99.999% purity was used for spiking thermal desorption sorbent tubes and 99.999% pure helium gas was used for chromatographic analysis.

Emission cells

The emissions of TCIPP were measured using Field and Laboratory Emission Test cells (FLEC) (CHEMATEC, Denmark) made from hand polished stainless steel with a volume of 35 cm³. Prior to the experiments, the internal surface of the FLECs was cleaned with acetone and methanol and finally dried under nitrogen flow in an oven at 120°C during 24h. Temperature and relative humidity were monitored and logged during the experiments with calibrated instruments. A Testo 175H1 (Testo, Germany) logger was used to measure and log the temperature and relative humidity, and a Gilian Gilibrator-2 (Sensidyne, USA) was used to measure the air flows.

Particle generation

Soot particles were generated with a miniCAST 6203 type A (Combustion Aerosol STandard) soot generator (Jing-CAST Technology, Switzerland) with a propane burner. The CAST burner enables generated soot particles to escape from the flame without contact with oxygen. Subsequently mixing of the particle stream with a quenching gas (N₂) prevents further combustion processes and stabilizes the soot particles. Moreover, the quenching gas inhibits the condensation in the particle stream at ambient air condition (E.Barthazy et al. 2006). To dilute the particle stream coming out of the soot generator, clean compressed air with a flow rate of 5 L min⁻¹ was added. Before entering the FLEC, the particle-loaded airstream was further diluted by mixing it with clean air in order to lower the total suspended particulate (TSP) mass concentration to about 100 µg m⁻³. According to previous studies, the size of the particles generated from the miniCAST soot generator is in the range of 10 – 160 nm (E.Barthazy et al. 2006). The TSP mass concentration was measured at the exhaust of the FLECs with DustTrak aerosol monitor TSI 8534 (TSI Inc., USA).

97 **Emission experiments**

98 Emission tests were designed to simulate two test scenarios: (1) controlled environment test when laboratory
99 generated soot particles are introduced into FLEC, and (2) real environment test when air containing particles
100 related to during cooking activities (mainly frying and baking) is introduced into FLEC. Whilst the first
101 scenario is characterized by a continuous and constant supply of particles, in the second experiment particles
102 are supplied in a discontinuous way. In both test scenarios, two FLECs (a reference FLEC and a test FLEC)
103 were placed on a PIR board of 0.50 x 0.50 x 0.09 m. The sample surfaces used in the two scenarios were
104 obtained from two (different for each experiment) PIR insulation boards (1.2 x 0.6 x 0.09 m) treated with
105 TCIPP, acquired from a Belgian do-it-yourself shop.

106 In the controlled environment test (1), the reference FLEC was supplied with clean and dry air at a flow rate of
107 320 mL min⁻¹ (550 air changes per hour), whilst the test FLEC was continuously supplied with dry air
108 containing soot particles at a flow rate of 340 mL min⁻¹ (580 air changes per hour). The controlled
109 environment test (1) was performed at a temperature of 23 ± 1°C (controlled and continuously monitored). For
110 the real environment test (2), the test FLEC was supplied with air from a kitchen environment at a flow rate of
111 430 mL min⁻¹ (737 air changes per hour). Simultaneously, the reference FLEC was supplied with a clean and
112 dry air at flow rate of 255 mL min⁻¹ (437 air changes per hour). Cooking activities took place during week
113 days between 8 a.m. and 3 p.m. Samples from reference and test FLEC exhaust air were taken in parallel. For
114 the real environment test, sample collection was organized when cooking activities were most intensive
115 (between 10 a.m. and 2 p.m.). The temperature during the test was continuously monitored. The trend of the
116 temperature in the kitchen environment during the test is shown in figure 1. In each experiment, particle
117 concentrations in the exhaust air of both FLECs were continuously measured and logged. + duration of both
118 test scenarios

119

Figure 1. Temperature in the kitchen environment, measured during the real environment test

Sampling and analysis of TCIPP in FLEC air

To measure the concentration of TCIPP in the test cells, air samples were simultaneously taken on a regular base from the outlet on each of the cells. An active air sampling on mixed bed (polydimethylsiloxane (PDMS) / Tenax TA) sorption tubes with a constant sampling flow of 80 mL min⁻¹ for 4 hours (20 L sampling volume) were used for the collection of both (gas and particulate) phases. Programmable personal air pumps SG 350 (GSA Messgerätebau GmbH, Germany) were used to sample a defined air volume of 19.2 L. After sampling, the PDMS/Tenax TA sorbent tubes were sealed with end caps and stored under nitrogen atmosphere until analysis. All samples were taken in duplicate and analyzed within seven days after the collection.

Analysis of TCIPP was performed on a TD-GC-MS system, which consisted of a TD100™ automatic thermal desorber (Markes International Ltd., UK) coupled with a gas chromatograph Thermo Trace GC Ultra and mass selective detector Thermo DSQII (Thermo Fisher Scientific Inc., USA). The parameters, performance and validation of the system for analysis of TCIPP are described in (Lazarov et al. 2015).

To ensure that there was no TCIPP associated with the air and particles entering the cells, a blank test (FLECs placed on a precleaned glass plate) was done prior each experiment.

Area specific emission rates

In order to compare the results between the emission test cells regardless of the air change rates, the Area specific emission rates (SE_{RA}) of TCIPP were calculated from the concentration of TCIPP measured at exhaust of the FLECs. The SE_{RA} was calculated by followed equation:

$$SE_{RA} = \frac{c \times AC_F}{L_F} \quad (1)$$

where: SE_{RA} is the area specific emission rate of TCIPP [$\mu\text{g} \cdot \text{m}^{-2} \cdot \text{h}^{-1}$], c is the mass concentration of TCIPP sampled at the exhaust of the FLEC [$\mu\text{g} \cdot \text{m}^{-3}$], AC_F is the hourly air change rate of the FLEC [h^{-1}] and L_F is the loading factor inside the FLEC [$\text{m}^2 \cdot \text{m}^{-3}$] (CEN 2013). Because of the specific design of the FLEC, a maximal loading factor of $507 \text{ m}^2 \cdot \text{m}^{-3}$ was used in every test.

Partition coefficient

The TCIPP particle-gas partition coefficient, K_p , was calculated as:

$$K_p = \frac{C_{part}}{C_{gas} \cdot TSP} \quad (2)$$

where: K_p is the particle-gas partition coefficient [$\text{m}^3 \cdot \mu\text{g}^{-1}$], C_{part} is the particle phase concentration of the TCIPP [$\mu\text{g} \cdot \text{m}^{-3}$], C_{gas} is the gas phase concentration of the TCIPP [$\mu\text{g} \cdot \text{m}^{-3}$] and TSP is the total suspended particulate mass concentration [$\mu\text{g} \cdot \text{m}^{-3}$] (Pankow 1987; Yamasaki et al. 1982).

The TSP values used for individual estimates of K_p , were based on the average particle concentration measured at the exhaust of the FLEC for the corresponding day. The particle-phase TCIPP concentrations, C_{part} , were calculated as a difference between the TCIPP concentration measured at the exhaust of the FLEC supplied with particles and the concentration of TCIPP from the FLEC supplied with clean air for the same sampling period. The gas-phase TCIPP concentration, C_{gas} , used in the calculation was the concentration of TCIPP obtained from the exhaust of the FLEC supplied with clean air for the corresponding sampling period.

Results and discussion

Controlled environment test

The emission profiles of TCIPP from the test (supplied with soot particles) and the reference FLECs over a period of 10 days are shown in Figure 2. A continuous stream of soot particles was supplied into the test FLEC during the whole test period. The measured TSP mass concentration from the test FLEC's exhaust ranged between 74 and $140 \mu\text{g} \cdot \text{m}^{-3}$. Each data point in Figure 2 represents the mean value of the SE_{RA} estimated from the concentration measured at the exhaust of the FLECs in duplicate with error bars representing the standard

deviation. All measured TCIPP concentrations and calculated SER_A for both FLECs are summarized in Table S1.

Figure 2. SER_A of TCIPP from the PIR board with and without presence of soot particles. Lines drawn are not regression lines and inserted to illustrate the emissions trend.

The emission profile of TCIPP from PIR boards obtained from the reference FLEC shows a slow increase of the SER_A over the time to stable levels after 4 days (~56 000 air changes). The final value of SER_A for TCIPP was estimated from this quasi-static equilibrium at $0.82 \mu\text{g}\cdot\text{m}^{-2}\cdot\text{h}^{-1}$ (table S1). This estimated TCIPP SER_A without seed particles is found to agree well with results of previous emission studies of similar PIR boards (Kemmlin et al. 2003). In the FLEC supplied with a stream of soot particles, on the other hand, the SER_A was characterized by a different time profile. In fact with seed particles, SER_A shows the tendency to increase faster in the beginning of the experiment and then decreases to quasi-static equilibrium levels, passing some sort of a peak value of SER_A . The SER_A at the quasi-static equilibrium was estimated as $2.16 \mu\text{g}\cdot\text{m}^{-2}\cdot\text{h}^{-1}$ (table S1), which is 3 times higher than the SER_A assessed in the reference experiment.

Based on the measured concentrations of TCIPP in the exhaust air of both FLECs, K_p values were estimated. The time profile of the estimated K_p (Figure 3) showed a fast decrease over time, reaching a stable level after 4 days. This suggests that the main driving mechanism responsible for the increased emission rates of TCIPP in presence of soot particles is its partitioning to particles. In fact, the resulting increase of the material-air partitioning leads to augmented emission rates (Xu et al. 2006; Xu et al. 2009). The median value of the particle-gas partition coefficient for TCIPP onto soot particles was estimated after day 4 at $0.022 \pm 0.012 \text{ m}^3\cdot\mu\text{g}^{-1}$. This experimentally assessed K_p is compared with those estimated from the empirical relation of K_p

for SVOCs to the octanol-air partition coefficient described in literature (Finizio et al. 1997; Weschler et al. 2008a; Weschler et al. 2008b). To calculate the TCIPP K_p , an empirical relationship derived from a regression of $\log K_p$ vs. $\log K_{OA}$ for a series of PAHs have been used (Finizio et al. 1997):

$$K_p = 1.88 \times 10^{-12} \times K_{OA} \quad (3)$$

Figure 3. Profile of the gas-particles partition coefficient of TCIPP during the emission test. Line drawn is not regression line and inserted to illustrate the K_p trend.

The K_{OA} value for TCIPP was estimated from its octanol-water (K_{OW}) and air-water (K_{AW}) partition coefficients ($K_{OA} = K_{OW} / K_{AW}$) (Cousins et al. 2000). The values of K_{OW} and K_{AW} used to calculate K_p are marked in bold in Table 1. The results show that the measured value of K_p for TCIPP in presence of soot particles, $0.022 m^3 \cdot \mu g^{-1}$, is about a factor 3 lower than the estimated from the K_{OA} (Table 1). This difference is expectable and can be attributed to the condition that the empirical relationship between of K_p and of K_{OA} is estimated based on measurements of other groups of SVOCs (PAHs) than TCIPP.

Table 1: Particle-gas partition coefficient, K_p , of TCIPP estimated from the octanol – air partition coefficient (K_{oa}), as well as the values of octanol –water partition coefficient (K_{ow}) and octanol-air partition coefficient (K_{aw}) of TCIPP.

K_{ow}	5.75×10^3 ^a	5.50×10^3 ^a			
K_{aw}	1.698×10^{-7} ^b	1.549×10^{-7} ^c	4.677×10^{-6} ^d	2.45×10^{-6} ^e	2.95×10^{-2} ^e
K_{oa}	3.390×10^{10}	3.236×10^{10}	3.715×10^{10}	3.548×10^{10}	
K_p	0.064	0.061	0.070	0.067	

The values of K_{ow} and K_{aw} used for estimation of K_{oa} in present work are given in bold.

^a (MacKay 2006); ^b (European Chemicals Agency (ECHA) 2014); ^c (European Communities 2008); ^d (Brooke D N 2009); ^e (Liagkouridis et al. 2015)

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200 *Real environment test*

201 In order to investigate the influence of a different particle composition on the emission of TCIPP from PIR
202 boards, and to explore the experiment's repeatability in real-life, the test FLEC was supplied with air from a
203 kitchen environment during cooking activities. Figure 4 shows the emission profiles of TCIPP obtained from a
204 FLEC supplied with kitchen air as well as the profile obtained from a reference FLEC (supplied with clean air)
205 over a simultaneous test period of 20 days. Each data point in figure 4 represents the mean SER_A estimated
206 from the concentration quantified at the exhaust air of the FLECs in duplicate, with error bars representing the
207 standard deviation. The measured TSP mass concentrations in exhaust air of the test FLEC are illustrated in
208 Figure 4. All measurements of the concentration of TCIPP and calculated SER_A for both FLECs are
209 summarized in Table S2.

210

Figure 4. SE_{Ra} profile of TCIPP from the PIR board with clean and kitchen air. Lines drawn are not regression lines and inserted to illustrate the emissions trend.

211

212 Overall, the estimated SE_{Ra} of TCIPP during this experiment shows higher values compared to those obtained
 213 from the controlled environment test. It should be noted that these elevated SE_{Ra} may be caused by a higher
 214 TCIPP content of studied PIR board (no information was available on the time elapsed between the board's
 215 manufacturing and the start of the experiments, nor about the boards' conservation in the shop). However, the
 216 emission profile of TCIPP obtained in FLEC supplied with clean air indicates a quasi-static equilibrium state
 217 after about 6 days (~60 000 air changes), which is comparable to those observed in the previous experiment.
 218 The final value of SE_{Ra} for TCIPP from this second board at the quasi-static equilibrium was estimated to be
 219 $2.33 \mu\text{g}\cdot\text{m}^{-2}\cdot\text{h}^{-1}$ (table S2).

220 When supplied with air from a real kitchen environment, the SE_{Ra} profile of TCIPP (figure 4) was found to be
 221 characterized by a similar trend to those obtained in presence of soot particles. SE_{Ra} based on concentrations
 222 of?? at the quasi-static equilibrium was $5.21 \mu\text{g}\cdot\text{m}^{-2}\cdot\text{h}^{-1}$ (Table S2). The larger variation of SE_{Ra} values in the
 223 quasi-static equilibrium of this real-life experiment compared to the first experiment (with continuous supply
 224 of soot particles), is most probably the consequence of the larger variation in particle concentrations over time
 225 in kitchen air during the experiment. The particle-gas partition coefficient for TCIPP onto particles during
 226 cooking activities, estimated as the median of the partition coefficients at the quasi-static equilibrium, is found
 227 to be $0.062 \pm 0.037 \text{ m}^3\cdot\mu\text{g}^{-1}$. This higher TCIPP partition coefficient in the kitchen experiment can result from
 228 the typically more abundant grease particles that are expected in kitchen air (mainly generated from frying

activities) (Abdullahi et al. 2013). The stronger tendency of SVOC to absorb to more grease organic matter is well known and confirmed by other studies (Liang et al. 1997). The results of this real environment test confirm an enhancement of TCIPP emission rates from PIR boards in a real-life, with a discontinuous generation of particles.

Conclusions

This paper is the first one in its kind to study the influence of seed particles in air on flame retardant emissions (TCIPP) from building materials, in a controlled laboratory environment, as well as in a real-life setting.

The study has investigated the influence of continuously laboratory generated soot particles on TCIPP emission rates from PIR boards. The experiment indicated that SER_A augmented up to 3 times in presence of carbonaceous seed particles. This seed particle-induced increase of the TCIPP SER_A from a PIR board was confirmed in a real-life experiment, where particles discontinuously generated during cooking activities were inserted into the test cell. The particle-gas partition coefficient of TCIPP onto these two different types of particles indicated a stronger affection of TCIPP to particles with higher organic (grease) content.

Therefore, in real-life a variable fate of TCIPP material emissions in indoor environments is expected, amongst other factors, depending on particles' origins and compositions. Because of the potential underestimated emission rates assessed under traditional emission testing conditions (without seed particles), outcomes of this study underline the need for a cautious use of SVOC emission rates for indoor air and exposure modelling for health impact assessments. A more detailed study on the impact of concentration variability and residence time of the seed particles inside the test cell is needed to further understand the fate and transport of these chemicals in indoor environment. Outcomes of this study may contribute to improve current SVOC emission test protocols, which will lead to a more accurate and precise source quantification and human exposure control.

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339 **Supporting information**

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Table S1. Concentrations of TCIPP measured from the exhaust of FLECs during the controlled environment test

Day ^a	Reference FLEC			Test FLEC		
	Air changes	C _{TCIPP} , µg.m ⁻³	SER _A , µg.m ⁻² .h ⁻¹	Air changes	C _{TCIPP} , µg.m ⁻³	SER _A , µg.m ⁻² .h ⁻¹
1	9757	0.05±0.01	0.05	10372	1.12±0.51	1.29
2	23017	0.10±0.05	0.11	24433	1.66±0.31	1.91
3	36184	0.30±0.11	0.33	38423	4.45±0.05	5.12
4	50284	1.20±1.36	1.30	53408	3.33±0.67	3.82
7	88931	1.11±0.64	1.20	94463	3.91±2.97	4.49
8	102166	0.55±0.01	0.60	108531	2.54±0.09	2.92
10	130240	0.89±0.39	0.97	138361	1.45±0.30	1.67

341 ^a time elapsed after the start of the test in days.

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Table S2. Concentrations of TCIPP measured from the exhaust of FLECs during the real environment test

Day ^a	Reference FLEC			Test FLEC		
	Air changes	C _{TCIPP} , µg.m ⁻³	SER _A , µg.m ⁻² .h ⁻¹	Air changes	C _{TCIPP} , µg.m ⁻³	SER _A , µg.m ⁻² .h ⁻¹
1	11211	5.30±1.67	4.57	19069	3.94±1.21	5.78
2	21617	5.02±0.51	4.33	36831	5.65	8.27
3	32155	4.63	4.00	54656	6.58±0.53	9.64
6	64058	4.71±0.47	4.06	108546	6.09	8.92
7	74333	3.30±1.89	2.85	125983	4.45±1.04	6.52
8	84782	3.09±1.19	2.66	143877	4.08±1.06	5.97
9	95237	2.76±1.06	2.38	161587	4.51±1.46	6.60
10	105774	3.40±0.82	2.93	179437	4.96±1.80	7.26
13	137355	2.06±0.97	1.78	233095	3.30±2.17	4.83
14	147792	2.83±1.00	2.44	250732	2.42±0.18	3.55
15	158432	2.20±0.50	1.90	268762	2.66±0.92	3.90
16	168956	3.62±3.26	3.12	286636	2.60±1.05	3.81
17	179300	2.25±0.23	1.94	304181	3.91±1.26	5.73
20	211013	1.48±0.06	1.27	357987	2.70±0.63	3.95

^a time elapsed after the start of the test in days.343
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