Indoor dust contamination by biocides in French dwellings

Pierre Martinache, Bertille Bonnaud, Régis Moilleron, Adèle Bressy

Indoor dust contamination by biocides in French dwellings

Pierre Martinache [1,*], Bertille Bonnaud [1], Régis Moilleron [1], Adèle Bressy [1]

[1] Leesu, Ecole des Ponts, Univ Paris Est Creteil, Marne-la-Vallee, France

[*]*Corresponding email: pierre.martinache@enpc.fr*

SUMMARY

Biocides are widely used in our daily life as disinfectants or protection products in building materials. The scientific literature on these substances mainly focuses on outdoor exposure despite the potential threat to people exposed indoors and the lack of information on indoor contamination. The objective of this study was to develop a protocol for biocide monitoring in indoor dust. 18 biocides were selected based on their consumption and their toxicity. The dusts from 12 dwellings were collected during 2021 using a vacuum cleaner equipped with a bag. After sieving at 100 µm, a microwave-assisted extraction was performed twice, followed by analysis by liquid chromatography coupled to a tandem mass spectrometer with internal standards. For the first time, it was shown that 16 out of 18 biocides were observed at least once in household dust. The highest contaminations were found for BAC C16 with 28,000 ng/g and BIT with 3,500 ng/g.

KEYWORDS

Biocides – Contamination – Dwellings – LC-MS/MS – Settled dust

1 INTRODUCTION

Biocidal products are regularly used in dwellings as antimicrobial and disinfectant products (Jones and Joshi, 2021). However, there is still little data on exposure to biocides indoor and on the concentrations of biocides in settled indoor dusts despite their potential toxic effects (Hwang *et al.*, 2021). The few studies comparing indoor and outdoor dusts have shown much higher concentrations for indoor dusts (Mahler *et al.*, 2009). The objective of this study is therefore to establish a first database of biocide contamination in the indoor environment.

2 MATERIALS/METHODS

Sampling campaigns

Indoor dust was collected using a bag hoover in 12 French dwellings between March and July 2021. Two parallel sampling campaigns were conducted: one with spot samples that collected indoor dust deposited on the floors in a single aspiration (n = 7, 25-76 m²) and a second with the collection of bags that had been used (n = 5, 38-180 m², 1-4 months). After sampling, the bags were sealed and kept in the dark. Except for a bag that includes car cleaning in addition to indoor dust, all of the vacuums were for the interior.

Biocides analysis

Biocides were selected (see Table 1) based on their massive use in households and also on their toxicity (Paijens *et al.*, 2020). Dust samples were sieved to 100 µm and stored at -20°C in amber vials. 100 mg of each sample was microwave-extracted twice. The extract was then evaporated to dryness under nitrogen flow then taken up in 1 mL of mobile phase and analysed by high performance liquid chromatography with tandem mass spectrometer (Paijens *et al.*, 2020). A QAQC procedure was followed and no significant contamination was detected.

3 RESULTS AND DISCUSSION

Table 1 presents the biocide contents in indoor dust (n = 12) and the maximum exposure calculated with an ingested mass of dust of 50 mg/day for a 70 kg adult according to the standard equation (Darney *et al.*, 2018). The detection frequency proves that the choice of biocides is relevant. 16 out of 18 molecules were quantified in 17 to 100% of the samples with three levels of median concentrations: under 100 ng/g (DCOIT, CMIT, DIU, MCPP, CBZ, TBZ), between 100 and 1,000 ng/g (MIT, OIT, BIT, THB, TEB, IPBC, TB) and over 1,000 ng/g (BAC). Spatial variability was found in both the grab and integrated samples. Our results allowed us to calculate an exposure level between 0.02 (TBZ) and 20 (BAC C16) ng/kg bw/day.

Table 1: Frequency of quantification (FQ, %), quantification limits (LOQ, ng/g), contents (ng/g) and exposure (ng/kg bw/day) for 12 samples of settled indoor dust from Ile-de-France.

Biocides	*FQ (%)*	*LOQ (ng/g)*	*Median contents (min-max) (ng/g)*	*Maximum exposure (ng/kg bw/day)*
Methyl-isothiazolinone (MIT)	100	18	490 (59-1,400)	0.97
Octyl-isothiazolinone (OIT)	100	9	270 (47-1,300)	0.96
Dichloro-n-octyl-isothiazolinone (DCOIT)	17	18	34 (31-36)	0.03
Benzisothiazol-3(2H)-one (BIT)	100	27	890 (130-3,500)	2.50
Chloro-methyl-isothiazolinone (CMIT)	58	19	50 (23-150)	0.11
Diuron (DIU)	100	9	67 (25-300)	0.21
Isoproturon (IPU)	0	5.9	< LOQ	-
Thiabendazole (THB)	100	5.9	150 (18-1,100)	0.75
Tebuconazole (TEB)	92	12	170 (32-2,100)	1.50
Benzalkonium chloride C12 (BAC C12)	100	92	1,800 (260-12,000)	8.40
Benzalkonium chloride C14 (BAC C14)	100	92	1,100 (160-7,300)	5.20
Benzalkonium chloride C16 (BAC C16)	17	92	16,000 (4,500-28,000)	20
Mecoprop (MCPP)	42	12	34 (17-110)	0.08
Carbendazim (CBZ)	100	12	44 (23-260)	0.18
Iodopropynyl butylcarbamate (IPBC)	100	19	110 (32-920)	0.66
Cybutryne (CBY)	0	12	< LOQ	-
Terbutryn (TB)	50	12	210 (110-1,100)	0.78
Terbuthylazine (TBZ)	58	5.9	19 (18-24)	0.02

4 CONCLUSION

For the first time, some widely used biocides were quantified in household dust. The highest contaminations were found for quaternary ammoniums with 28,000 ng/g (BAC C16) and isothiazolinones with 3,500 ng/g (BIT). The next steps will be to increase the number of samples and to analyse biocides in the indoor air and greywater to establish mass balance in dwellings.

ACKNOWLEDGEMENT

This work was supported by the Biocid@Home research program (ANR-20-CE34-0001-01).

5 REFERENCES

Darney, K. *et al.* (2018) ‘Aggregate exposure of the adult French population to pyrethroids’, *Toxicology and Applied Pharmacology*, 351, pp. 21–31. doi:10.1016/j.taap.2018.05.007.

Hwang, J. *et al.* (2021) ‘Skin irritation and inhalation toxicity of biocides evaluated with reconstructed human epidermis and airway models’, *Food and Chemical Toxicology*, 150, p. 112064. doi:10.1016/j.fct.2021.112064.

Jones, I.A. and Joshi, L.T. (2021) ‘Biocide Use in the Antimicrobial Era: A Review’, *Molecules*, 26(8), p. 2276. doi:10.3390/molecules26082276.

Mahler, B.J. *et al.* (2009) ‘Fipronil and its Degradates in Indoor and Outdoor Dust’, *Environmental Science & Technology*, 43(15), pp. 5665–5670. doi:10.1021/es901292a.

Paijens, C. *et al.* (2020) ‘Determination of 18 Biocides in Both the Dissolved and Particulate Fractions of Urban and Surface Waters by HPLC-MS/MS’, *Water, Air, & Soil Pollution*, 231(5), p. 210. doi:10.1007/s11270-020-04546-6.