OCCURRENCE OF NDL-PCBs AND PBDEs IN ITALIAN RESIDENTIAL DUST AND THEIR RELEVANCE FOR INTAKES IN TODDLERS AND PETS

Brambilla G.¹, Esposito M.², Ferrari A.³, Manni A.⁴*

¹Istituto Superiore di sanità, Viale Regina Elena, 299 I-00161 Rome, Italy; ²Istituto Zooprofilattico Sperimentale del Mezzogiorno, Via della Salute, 2, I-80055 Napoli, Italy; ³Istituto Zooprofilattico Sperimentale del Piemonte, Liguria e Valle d'Aosta, Piazza Borgo Pila; 10 I-16129 Genova, Italy. ⁴Chemical Research 2000 Srl, Via Santa Margherita di Belice,16 I-00133 Roma³ Italy.

Introduction:

Residential dust may represent a relevant source of exposure to persistent organic pollutants (i.e. those released from consumers products and old walls, paintings, furniture and sealed windows), for toddlers and pets, due to their mouthing and licking behavior. The aim of this work was to describe for the first time the occurrence of selected PBDEs (congener numbers congener numbers 28, 47, 99, 100, 153, 154, 183 and 209), Total Σ_{30} PCBs (the sum of the 30 congeners numbers 18, 28, 31, 33, 49, 52, 66, 70, 74, 91, 95, 99,101, 110, 128, 138, 141, 146, 149, 151, 153, 170, 174, 177, 180, 183, 187, 194, 196 and 203), ICES Σ_6 NDL-PCBs (congener numbers 28, 52, 101, 138, 153 and 180) and Σ_7 PCBs (Σ_6 NDL-PCBs + DL-PCB number 118) in Italian houses from different towns, and to evaluate the potential contribution to the overall intake, accounting for available guidance values for chronic alimentary exposures.

Materials and methods:

A kit with weighted pure cellulose floor wipers, a 50 mL PET Falcon labelled tubes, and a questionnaire was distributed to formed volunteers, that were asked to clean the house floor as usual, and to report useful information to correlate analytical findings with the presence of indoor/outdoor sources of emission/release of PCBs and PBDEs, such as consumers products, wall paintings, windows sealings and old electric transformers. The locations of Genoa, Brixia, Padua, Rome, and Neaples were chosen as representative of houses in the Northern, Central and Southern Italy as well as of towns with different anthropogenic and industrial pressures. After the addition of internal standards (PBDEs from CIL, USA; PCBs from Wellington Laboratories Canada), collected wipers were extracted with Accelerated Solvent Extraction with n-hexane/methylene chloride (1:1 v/v). Eluates were reduced to a final volume of 0.2 mL, resuspended in n-hexane and after an acidic attack on Extrelut NT 3 columns, PBDEs and PCBs were cleaned up on activated basic alumina 1 g glass columns. Extracts, resuspended in n-nonane were analyzed for PCBs in high-resolution gas chromatography (HRGC) coupled to low-resolution tandem mass spectrometry (-LRMS/MS) (Thermofisher TSQ Quantum XLSTM) equipped with a J&W 60 m DB5MS GC capillary column. PBDEs were determined using HRGC coupled to high-resolution mass spectrometry (HRMS) (Thermofisher DFSTM) on a VarianVF-1 15 m GC column. Acquisition parameters are extensively described in a previous paper¹.

For dust intake, average and conservative estimates of 16 and 110 mg/day, respectively, were derived from the 1995 German Exposure Factors Handbook. Average body weight of 11.3 kg for Italian toddlers and of 3.5 kg body weight for cats were adopted. Under a deterministic approach, risk characterization accounted for the following guidance value: a proposed TDI of 20 ng/kg bw for NDL-PCBs² and the BMDL₁₀ of 270 and 4.1 ng/kg bw/day proposed by EFSA for BDE no. 47 and no. 99, respectively³.

Results and discussion:

The results about PCBs and PBDEs occurrence in house dust are shown in Table 1, while Figure 1 and 2 illustrates the different analytical profiles of selected PCBs and PBDEs found in the different samples. PBDE no. 47 and no. 99 resulted to have a mean concentration of 35 ng/g dust and a maximum value of 196 and 205 ng/g, respectively. Such mean and median values are in good agreement with the most recent studies carried out in Belgium⁴ with reported ranges of 8 - 62 ng/g and of 9 - 110 ng/g for BDEs no. 47 and no. 99, respectively. For PCBs, the not harmonized set of congeners taken into account by the different authors and the different upperbound *vs* lowerbound computing of left-censored data do not allow an immediate comparison of our data with those from the literature. Anyway, minimum and maximum values referred to ICES \sum_{6} NDL-PCBs congeners fall within the same range of \sum_{5} PCBs (congener numbers 105, 118, 138, 153, or 180) indicator

reported in a recent review⁵. Highest PCBs concentration were found in houses built around 1960 in Northern Italy, while PBDEs were higher in houses from an industrial town with several ferrous and non ferrous smelters plants. Lowest values in dust were always referred to houses from the Southern Italy (Neaples).

The risk characterization has been carried out considering the mean and the maximum level of dust contamination, under the conservative scenario of 110 mg dust daily ingested by toddlers and cats. For \sum_{30} PCBs, an Italian toddler would intake around the 15% of the 20 ng/kg bw/day TDI, while a cat the 50%, accounting for the mean contamination of 332 ng/g (Table 1); considering the highest occurrence value of 1,236 ng/g, such exposure would rise up to the 60% and the 195% of the TDI for toddlers and cats, respectively.

Considering PBDEs, congeners no. 47 and no. 99 only have guidance values for chronic intakes. For this reason, such congeners are discussed separately from the overall PBDE sum. From Figure 2, it appears evident that in most of samples they result among the most contributing congeners to the overall \sum_{7} sum. Their intake, under the mean dust contamination scenario, would result close to 0.3 and 1.1 ng/kg bw/day in toddlers and cat, respectively, while under the worst case scenario, the intake would rise up to 2 and 6 ng/kg bw/day. Considering the $BMDL_{10}$ of 4.1 ng/kg bw/day for BDE no. 99, as already seen for PCBs, cats would result potentially overexposed, independently from the aggregate contribution from food/petfood intake. For BDE 47, the intakes from dust seem do not deserve any toxicological concern as matter of the higher BMDL₁₀ than that of BDE 99 (270 vs 4.1 ng/kg bw/day). The uncertainties of such estimates rely on the expert judgement about the dust intake in cats, that has been considered equivalent to that attributed to toddlers, despite the wider time spent in close contact with the floor by cats than by toddlers. Another variability to be considered is the possible seasonal variation of dust contamination: dust samples were representative of the spring period. Associated toxicological outcomes represented by the contemporary intake of PCBs and PBDEs, both sharing a neurodevelopmental and thyroid gland toxicity have been not considered. The upperbound approach suggested for intake estimates has been adopted: this affects data from lowest contaminated dust, whose associated uncertainty is directed towards lower levels.

Analytes	unit	min	mean	sd	max
$\sum_{30} PCBs$		28.9	332	312	1,246
\sum_{6} NDL-PCBs	ng/g	10.0	98.4	78.3	279
\sum_{6} NDL-PCBs + PCB 118		11.8	116	103	394
∑ ₃₀ PCBs		0.27	8.96	20.6	82.9
\sum_{6} NDL-PCBs	ng/m ²	0.09	2.36	4.56	18.5
\sum_{6} NDL-PCBs + PCB 118		0.11	3.01	6.49	26.2
\sum_{8} PBDEs	ng/g	115	1,053	1,852	7,607
\sum_{7} PBDEs		16.6	93.1	124	500
\sum_{8} PBDEs	ng/m ²	0.29	16.1	17.9	70.7
\sum_{7} PBDEs		0.14	1.59	1.86	6.61

Table 1. Occurrence descriptors of PCBs and PBDEs in Italian house dust	samples
---	---------

Conclusions:

With respect to dust contamination, on average, the residential environment can be considered safe for most sensitive and vulnerable group of living organisms, such as cats. In general, pets represent the sentinel of the overall quality of the indoor environment. Recorded background dust contamination will represent the starting point to evaluate the PCBs and PBDEs time trends. Further work is in progress on a wider characterization of dusts with respect to pesticides and perfluoro-alkyl substances and on bio-monitoring data in indoor/outdoor cat groups, with or without thyroid gland disorders.

Acknowledgements:

The authors acknowledge funding from the Italian Ministry of Health [grant number 2009–1534860] 'ENVIFOOD' project. Authors wish to thank Drs Vittorio Abate, Nicola Iacovella, Francesco Serpe and Micaela Tiso for the sampling and analytical activities.

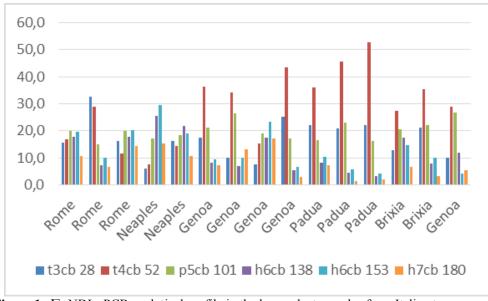


Figure 1. \sum_{6} NDL- PCB analytical profile in the house dust samples from Italian towns. Data normalized as percentage on the sum.

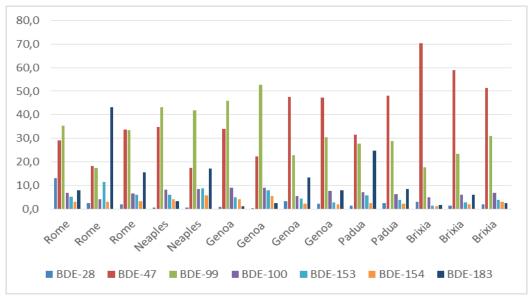


Figure 2. \sum_7 PBDE analytical profile in the house dust samples from Italian towns. Data normalized as percentage on the sum.

References:

- 1. Brambilla, G., Abate, V., Esposito V., et al., (2015) *Food Additives & Contaminants: Part A*, http://dx.doi.org/10.1080/19440049.2015.1029993
- 2. Agence Française de Sécurité Sanitaire des Aliments (2007) Available from: http:// www. anses.fr.
- 3. European Food Safety Authority (2011). EFSA Journal 9:2156 [274 pp.].
- 4. D'Hollander, W., Roosens, L., Covaci, A., et al., (2010) Chemosphere 81: 478-487.
- 5. Whitehead, T., Metayer, C., Buffler, P., Rappaport, S.M. (2011) J Expo Sci Environ Epidemiol. 21:549-64.