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Parabens in water and sediment: occurrence in river and lakes in different rural and urban environments

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1. Introduction

Parabens are a group of alkyl esters of the p-hydroxybenzoic acid that are widely used as preservatives in pharmaceutical and personal care products due to their broad anti-microbial spectrum and effectiveness. As personal care products, parabens are continuously released into the environment through urban wastewater. The preservatives ethylparaben (EtP) and propylparaben (PrP) belong to category 1 of the endocrine disrupter priority list for wildlife and human health. In order to gain knowledge about presence (or not) of parabens in different water categories, a screening study was performed in France and in 5 overseas territories in 2012 (the long term goal being to provide input for the next review of Water Framework Directive Priority Substances (WFD PS). More than 1700 occurrence data were obtained in water and sediment samples for 3 parabens (methylparaben, ethylparaben and propylparaben).

2. Materials and methods

Samples were collected on more than 160 sites during 3 sampling campaigns (spring/summer/fall). Sampling sites representing different types of landcover were chosen. Water samples were filtered first on GF/F filter (Glass Microfiber; 0.7 µm) and on Iso-disc filter (PTFE; 0.2 µm) and then extracted by on-line SPE with a C18 cartridge (7 µm). Liquid chromatographic/mass spectrometric analyses were carried out with a SL serie 1200 chromatograph (Agilent) equipped with a G6410B mass spectrometric (Agilent) detector based on a double-quadrupole in tandem analyzer and an electron multiplier detector. ¹³C internal standards were added for each compound to ensure accurate quantification.

3. Results and discussion

3.1. Results of the screening study

For water matrix, a high frequency of quantification (~100%) was observed in rivers and lakes for all targeted parabens. The level of concentration was as follow: ethylparaben > methylparaben >> propylparaben (Figure 1). This distribution of concentration is in accordance with parabens uses and degradation rates.

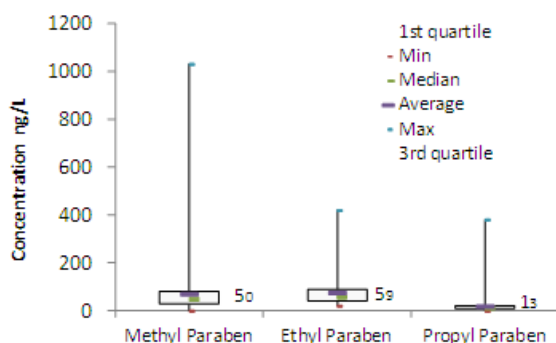


Figure 1: Boxplot displaying concentration trends for the 3 parabens (No. Samples = 336)

Significant differences in terms of concentration profile (highest peaks) were observed between samples collected in France as compared to the ones collected in the overseas territories. In particular for methylparaben, high concentrations (~3 µg/L) were measured in French Guiana in 2 different campaigns (spring and fall). For sediment matrix, a high frequency of quantification was noticed only for methyl paraben. Similar concentration trends were highlighted in the different sites regardless of the anthropogenic pressure (urban, industrial and agricultural), including no-pressure areas (reference sites). Having compared the predicted no effect concentration (PNEC) determined from the ecotoxicity data with 95th centile of maximum measured concentrations at each site (MEC95), a preliminary identification of potential risk was conducted

for these 3 parabens. The results suggest a low risk for the three parabens studied (risk ratio always < 1). The highest risk ratio was observed for ethylparaben (MEC/PNEC = 0.030 when applying MEC95 concentration and MEC/PNEC = 0.0090 when applying median concentration of all quantified data). However, it has to be noted that the currently available PNEC values are derived from chronic data from standard endpoints. They do not take into account non-standard endpoints, such as endocrine disrupting effects.

3.2. Results as compared to literature

This screening study has been conducted on a larger datasets compared to previous reported literature data (column 3 in Table 1).

	Country	No. Samples	Concentration (ng/L) Median (min-max)			LOQ (ng/L)		
			Methyl Paraben	Ethyl Paraben	Propyl Paraben	MtP	EtP	PrP
This study, 2014	France	336	51 (nd-1030)	60 (21-1347)	13 (nd-381)	3	0,5	0,8
S.Esteban et al.,2014	Spain	14	nd	13,5 (11-16)	nd-38	0,7	0,9	0,1
L. Renz et al., 2013	USA	7	nd-17	nd	nd - 12	0,2	0,2	0,2
M. Gorga et al., 2013	Spain	10	1-27	nd - 13	1-15	0,7	0,9	0,1
M. Terasaki et al., 2012	Japan	8	nd - 5	nd	nd - 25	2,1	4,3	4,9
E. Gracia-Lorn, et al., 2012	Spain	11	30 (6-208)	3 (2-4)	15 (81-2)	4,0	2,0	0,9
H. Yamamoto et al., 2011	Japan	19	25 - 676	nd-64	nd - 207	6,1	4,4	2,7
B. Rajendran Ramaswamy et al. 2011	India	29	nd - 22,8	2,5 - 147	nd - 57	1,5	1,8	1,5
IGonzalez-Marino et al., 2009	Spain	8	2-17	nd - 3	nd - 69	0,4	0,1	0,2
N. Jonkers et al, 2009	Switzerland	42	5 (3-17)	0,1 (nd-2)	0,6 (nd-6)	1,6	0,3	0,5
B. Kasprzyk-Hordern et al., 2008	UK	100	nd - 150	nd - 12	nd - 11	0,3	0,5	0,2

Table 1: Parabens data comparison from different studies (nd=not detected)

In the rivers of Spain, median concentrations of PrP, MtP and EtP were lower than in our study and in most of the cases not detected. The lower concentrations observed in Japan and in Switzerland may be explained by more effective removal of parabens by batch-activated sludge treatment and chlorination as a tertiary treatment in WWTPs, leading to 99% and 72% efficiency respectively, as reported by Jonkers et al. 2009 and H. Yamamoto et al., 2011. In the present study, samples were filtered less than 24 hours after the sampling to avoid degradation; this could be a reason of higher frequency of quantification as compared to other studies where samples were stored for up to 6 days.

4. Conclusions

For methylparaben and propylparaben, concentrations are similar in all samples, and no obvious effect could be identified associated to the type of anthropogenic sources. The occurrence of parabens as ubiquitous contaminants in almost 100% of the analysed samples can represent a risk. At the measured levels, acute and chronic toxicity is not expected with any of these compounds tested individually. However, currently available PNEC values do not take into account non-standard endpoints, such as endocrine disrupting effects which need to be considered in assessing the potential risks associated to occurrence of parabens in the aquatic environment. Moreover, a major concern with MeP is its potential to bioaccumulate in aquatic organisms as mentioned by [1]. Parabens are common hygiene product ingredients and special care must be considered for sampling. A quality control of the use of hygiene products during sample collection and preparation should be performed in next screening studies (field blank) to verify the occurrence of any potential sample contamination.

5. References

- [1] John M. Brausch, Gary M. Rand. 2011. Review of personal care products in the aquatic environment: Environmental concentrations and toxicity. Chemosphere 82 pp. 1518–1532

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