Bisphenol A, alkylphenol and phtalates in road and car park runoff: from vehicle component emissions to *in-situ* measurement

Bisphénol A, alkylphénols et phtalates dans les eaux de ruissellement de parking et de route : des émissions liées aux composants automobiles aux analyses *in-situ*

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RÉSUMÉ

Cette étude s'inscrit dans le cadre du projet « Roulépur » financé par l'Agence de l'Eau et l'Agence Française de la Biodiversité, et vise entre autres objectifs, à mieux cerner la contamination des eaux de ruissellement par les alkylphénols, le bisphénol A et les phtalates. Ces derniers rentrent dans la composition de nombreux composants et consommables automobiles et suite à la perte directe ou au lessivage de ces éléments par temps de pluie, peuvent être relargués dans les eaux de ruissellement. A ce jour, on ne dispose que de très peu d'éléments pour cerner la contribution de ces apports à la pollution des eaux de ruissellement. Dans le cadre de ce travail, une triple approche a été suivie pour tenter d'évaluer cette contribution. Différentes sources primaires (pièces de carrosserie, pneumatiques, lave-glaces, carburants, huiles moteur, liquides de freinage et de refroidissement) ont été étudiées soit en analysant leurs teneurs en micropolluants soit à travers d'essais de lixiviation au moyen d'un simulateur de pluie. Des mesures ont été également réalisées sur 4 sites expérimentaux contrastés en termes de densité de trafic. Ces essais montrent que certaines pièces automobiles sont susceptibles de relarguer des quantités importantes de polluants au regard des concentrations rencontrées dans les eaux de ruissellement.

ABSTRACT

This study as a part of the "Roulépur" research project funded by the Water agency and the French Biodiversity Agency, aimed at better understanding the contamination of runoff water by alkylphenols, bisphenol A and phthalates. These compounds are used for many automobile components and consumables; they could be emitted into runoff through component wear, leaks or leaching during wet weather periods. To date, very little is known about the contribution of these inputs to stormwater pollution. As part of this work, different parallel approaches were used to attempt to evaluate this contribution. Various primary sources (bodywork pieces, tires, windshield wipers, fuels, motor oils, brake and coolant fluids) were studied either by analysing their micropollutant contents or by leaching tests using a rainwater simulator. Measurements were also carried out on 4 experimental sites contrasted in terms of density of traffic. These tests show that certain car parts are likely to release large quantities of pollutants compared to the concentrations found in runoff water.

KEYWORDS

Organic pollutants, road and parking runoff, leaching of automobile components

1 INTRODUCTION

The “Roulépur” project aims to evaluate innovative solutions for controlling micropollutant loads generated by road runoff. It is essential to improve our knowledge about the nature and the extent of road runoff contamination by micropollutants.

Task 2 of the Roulepur research program, was dedicated to the diagnosis of micropollutant contamination of road and car park runoff. It combined targeted screening of a wide range of metals and organic pollutants in the runoff from 4 road/carpark sites, with a more detailed investigation on the potential emissions from different primary sources (e.g. body car, tires, etc.) for some contaminants.

Among the different pollutants analysed, this work is focused on two families, i.e. alkylphenols and alkylphenols ethoxylates (APnEO) and phthalic acid esters (PAE) and one individual compound, bisphenol A (BPA). Even if some data is now available for APnEO and BPA in runoff (Zgheib et al. 2012; Gasperi et al. 2014) or as to the potential emission of APnEO and BPA from automobile components (Lamprea et al. 2018), knowledge about these contaminants remains scare and little is known about the sources of contamination. Potential sources of these contaminants in runoff are endogenous to the urban watershed, and mainly related to traffic and leaching from construction materials.

APnEO are used as surfactants in different industrial sectors such as the petroleum, polymer, paint and lacquer industries. Based on a short review of vehicle components, APnEO can be found in car bodies, tires, brake fluids and motor oils (Table 1). BPA is used to produce polycarbonate plastic, commonly used for automotive components. PAE are predominantly used as plasticizers and can be found in a large variety of polyvinylchloride products such as automobile instrument panels, sheathing of electrical cables, pipes and doors. Other polymers such as polypropylene and polyurethane are also frequently used in vehicle components. Polypropylene is a thermoplastic polymer and is mostly used for automotive bumpers, while polyurethane is an elastomeric material used for flexible foam seating, elastomeric wheels and tires, automotive suspension bushings or hard plastic parts.

Table 1: Presence of APnEO, BPA and PAE in automotive components

	Material which may contaminate water by leaching	**Material which may contaminate water by wear or leaks**
APnEO	Bodywork car, tires	Tires, brake fluid, motor oils
BPA	Bodywork car	-
PAE	Bodywork car (paint and plastic), tires	Tires

2 MATERIALS AND METHODS

Three different approaches were applied. The first two approaches focused on vehicle components and aimed at (i) evaluating the APnEO, BPA and PAE contents in several vehicle components (Approach 1) and (ii) examining the potential emission of both families by leaching (Approach 2). For Approach 1, tires and 7 fluids or consumables, i.e. windshield washing fluid, coolant, brake fluid, new and used engine oils, gasoline, diesel and, finally, gas exhaust particles were analysed in triplicate. For Approach 2, leaching tests of different automotive body parts (i.e. plastic and metallic bodywork, seals, automobile optics and tires) were carried out in triplicate, using a portable rainfall simulator. Diluted mineral water was used as a synthetic rainwater. Simulated rainfall experiments were conducted for 90 - 120 min and the rain intensity ranged from 3 - 6 mm/h. The last approach (Approach 3) aimed at evaluating the levels of APnEO, BPA and PAE in runoff for 4 real sites (Paris: Heavily trafficked expressway along the banks of the Seine river, Compans: Highway RD 212, with heavy traffic and near an international airport, Rosny-sous-Bois: road with moderate traffic in a suburban town centre and Villeneuve-Le-Roi : residential parking lot). Depending on the site, between 3 and 12 events were sampled. Analyses were performed by Leesu Lab. APnEO (7 congeners, i.e. NP, NP1EO, NP2EO, OP, OP1EO, OP2EO, NP1EC) and PAE (6 congeners, DEHP, DMP, DEP, DBP, DiBP, DNP) were quantified using internal standards. APnEO were analysed by liquid chromatography coupled to the tandem mass spectrometry (LC-MSMS) and PAE by gas chromatography coupled to the MS (GC-MS).

3 RESULTS AND DISCUSSIONS

3.1 Automobile fluids and components

Results for PAE (∑6), BPA and APnEO (∑7) in automobile fluids and components are illustrated in

Figure 1. For liquid samples, concentrations in µg/l are provided whereas contents in mg/kg dry weight are plotted for solid matrices. For fluids and liquid consumables, concentrations typically ranged between 2 and 145 µg/l for PAE and between from 9.5 to 873 µg/l for APnEO. For BPA, besides brake fluid, for which extremely high values were observed (up to 43 000 µg/l), concentrations ranged from 0.15 to 53 µg/l. For APnEO, high values are especially observed for gasoline and diesel, and to a lesser extend in used engine oil. Gas exhaust particles and tires exhibited high levels of PAE (6 053 and 14.0 mg/kg respectively), followed by APnEO (0.4 and 1.7 mg/kg respectively).

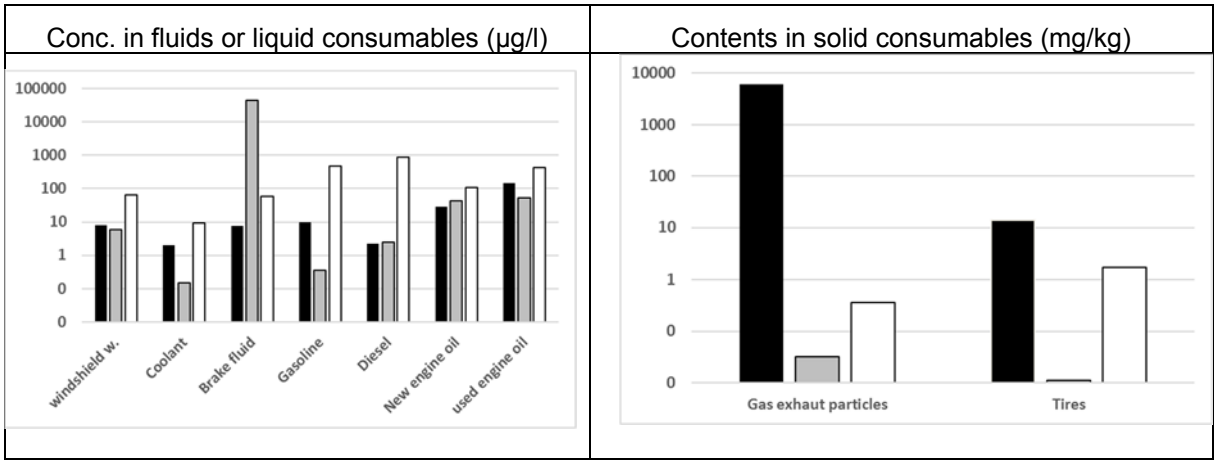

Figure 1 : Mean concentrations or contents of PAE (∑6, black), BPA (grey) and APnEO (∑7, white) in different automotive fluids or consumables

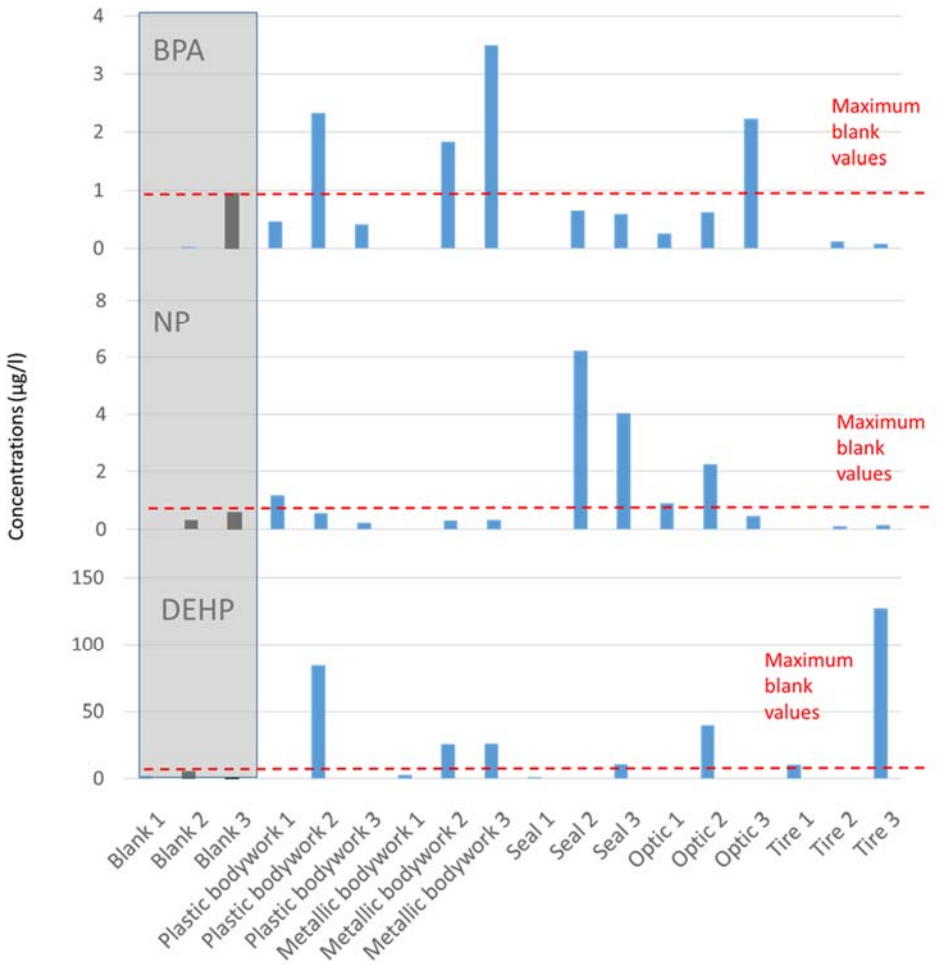

Figure 2 : Concentrations of BPA, NP and DEHP (µg/l) in synthetic water after leaching of vehicle components

3.2 Leaching of bodywork pieces using rainwater simulator

Concentrations of BPA, NP and DEHP (µg/l) in synthetic water after rainfall simulation on vehicle components are illustrated in figure 2. Blanks are also provided. For NP, no clear emissions in comparison to the blanks are observed except for seal pieces. Leaching of some plastic and metallic bodywork pieces can lead to the release of DEHP and BPA with concentrations up 80 and 2 µg/l respectively.

3.3 Road and parking runoff

To date, only results on Paris and Compans sites are available and concentrations of BPA, NP and DEHP (µg/l) in runoff for both sites are provided in Table 2. Other analyses are in progress. For APnEO, nonylphenol (NP) is predominant and similar orders of magnitude can be observed for between literature and our study. Indeed, NP concentrations (0.42 – 1.95 µg/l for Paris, 0,86 – 5.82 µg/l for Compans) are in the same order of magnitude than those reported in literature (Björklund et al. 2009; Stachel et al. 2010; Zgheib et al. 2012; Wicke et al. 2015). The NP Environmental quality standard is fixed at 0.3 µg/l.

For PAE, DEHP is generally predominant, with a contribution ranging from 40 to 85% of total PAE. High concentrations of DEHP are observed in runoff, with mean concentrations of up to 46.50 and 14.48 µg/l for Paris and Compans site respectively. A high inter-event variability is observed as previously observed in runoff by Stachel et al. (2010), i.e. 6 – 78 µg/l or by Zgheib et al. (2012), i.e. 3 – 58 µg/l. Runoff concentrations are higher than Environmental quality standards, which are established at 1.3 µg/l.

Table 2: Concentrations of BPA, NP and DEHP (µg/l) in runoff (min – max, mean)

	Paris	**Compans**
BPA	0.74 – 1.91, 1.46	0.23 – 0.96, 0.44
APnEO (∑7)	1.94 – 4.32, 2.88	1.21 – 8.13, 2.85
DEHP	20.32 – 83.17, 46.50	4.56 – 125.38, 14.48

4 CONCLUSIONS

These first results provide information as to the contribution of vehicle components in runoff contamination. Leaching of different vehicle components such bodywork pieces or seals can lead to significant emissions of BPA, NP or DEHP. A deeper exploitation is, however, required to better evaluate the contributions of vehicle components in comparison to other urban sources, as the targeted pollutants can be also emitted from various road construction materials.

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