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# Origins of biocides in combined sewer overflows in urban areas

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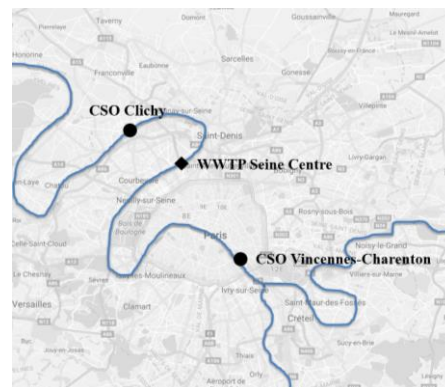
## Context

Historically, pesticides used in agriculture have received much more attention than biocides used in urban areas. However, even if they are used in smaller quantities in cities, biocides loads in receiving waters from rural and urban areas can be in the same order of magnitude (Wittmer et al., 2011). Biocidal substances are largely used and hence contribute to a diffuse contamination of receiving waters. For example, they are used as disinfectant in hospitals and homes or as preservatives in cosmetics, personal care products or building materials (bitumen sheets, paints, renders, tiles, etc.). They are released into receiving waters via discharges of wastewater treatment plant (WWTP) effluents and also via stormwater (separate sewer systems) or combined sewer overflows (combined sewer systems) discharges during wet weather (Bollmann et al., 2014; Bucheli et al., 1998; Burkhardt et al., 2011). As they can have effect at very low concentration on aquatic organisms (Mohr et al., 2008), they are part of emerging concerns but they are not sufficiently monitored in the environment (Dulio et al., 2015) and in urban matrices. However, in order to reduce emissions in the aquatic environment, the identification of their sources is needed. Thus, the purpose of this study is to identify the different urban sources of 18 biocides in combined sewer overflows (CSOs).

## Materials and methods

In that context, the Central Laboratory of Police Prefecture in partnership with the Laboratory of water, environment and urban systems (Leesu) has developed a method to simultaneously quantify 18 biocides, used in building materials but also in household products, at trace level (ng/L- $\mu$ g/L) in diverse urban matrices. The analysed biocides are the following ones: benzisothiazolinone (BIT), chloromethylisothiazolinone (CMIT), dichlorooctylisothiazolinone (DCOIT), methylisothiazolinone (MIT), octylisothiazolinone (OIT), cybutryn, terbutryn, terbutylazine, diuron, isoproturon, carbendazim, iodopropynyl butylcarbamate (IPBC), tebuconazole, thiabendazole, mecoprop and bezalkonium chlorides (C12-C16). First, the sample is filtered through 0.7  $\mu$ m. Both fractions are then extracted: solid phase extraction for the dissolved fraction and microwaves assisted extraction for the particulate fraction. And both extracts are analysed by high-performance liquid chromatography coupled to tandem-mass spectrometry.

Flow-proportional composite samples have been collected in Paris conurbation (figure 1) from two combined sewer overflows (CSOs) and from the Seine Centre WWTP (900 000 connected inhabitants, influents and effluents are sampled) with the help of the Mairie de Paris and the Public service of sanitation in the metropolitan area of Paris (SIAAP). The percentage of stormwater in CSOs has been estimated thanks to the sample conductivity, assuming that CSOs are composed of both stormwater (80-150  $\mu$ S.cm<sup>-1</sup>) and wastewater (1050-1170  $\mu$ S.cm<sup>-1</sup>).



**Figure 1:** Sampling sites

## Results and discussions

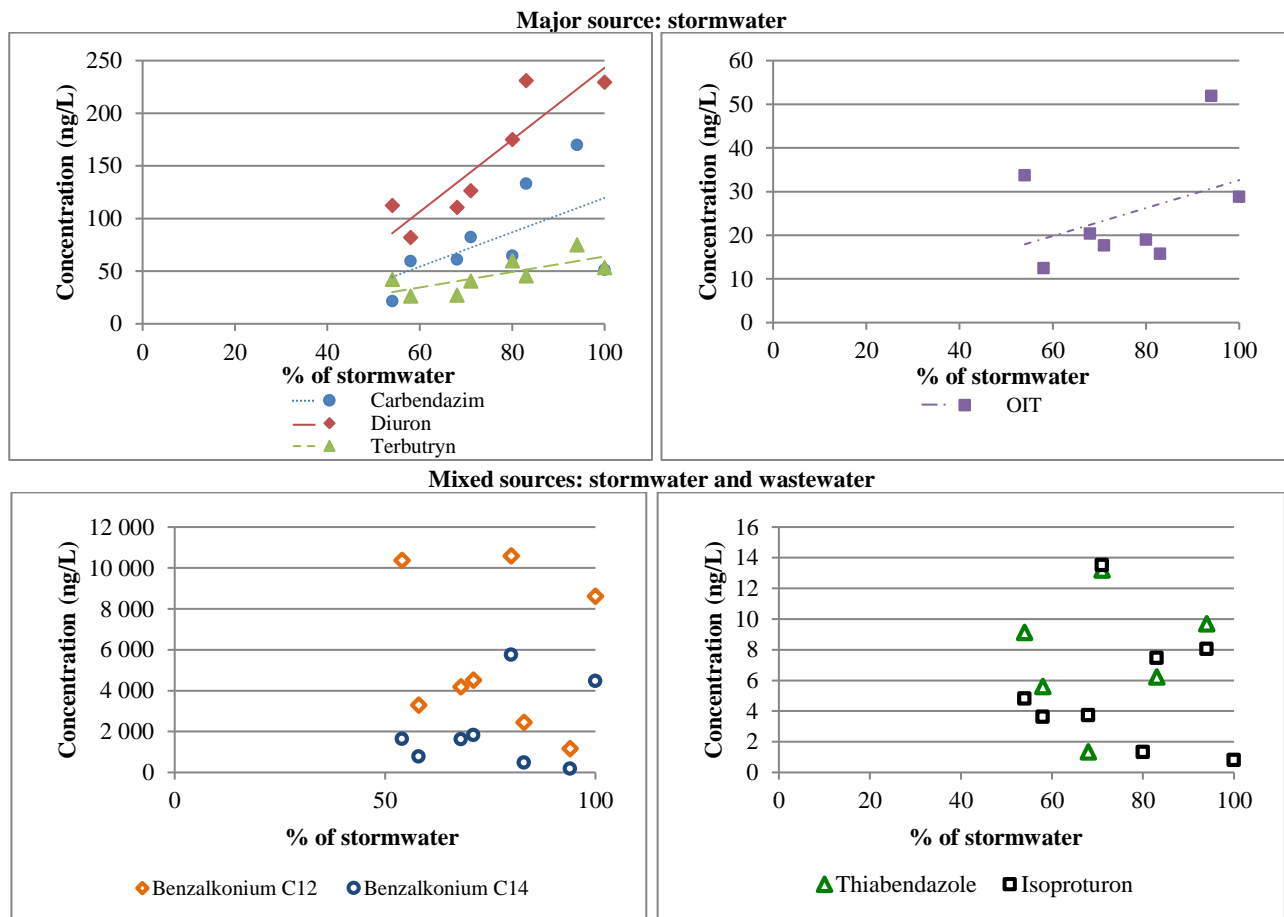
The results on dissolved fraction have shown a contamination of CSOs by the 18 targeted biocides. Concentrations are in the same order of magnitude for the two CSOs. They range from few ng/L to tens of ng/L for most of biocides but can exceed 100 ng/L (MIT, bezalkonium C16 and diuron) or even 1  $\mu$ g/L (bezalkonium C12 and C14).

First, compared to WWTP influents, concentrations of most biocides in CSOs are higher or in the same order of magnitude, which highlights that stormwater do not dilute biocides in wastewater but is also a source of biocides. Also, concentrations in CSOs can vary depending on the percentage of stormwater (figure 2). Three cases have been observed:

- Biocides for which stormwater substantially contributes to the contamination of CSOs. As shown on figure 2 (top charts), for carbendazim, diuron, OIT and terbutryn, the higher the percentage of stormwater is and the higher the concentrations are. Hence their major source in CSOs seems to be urban surfaces, and more specifically building materials (bitumen sheets, paints, renders, tiles, etc.) from which biocides have leached during rain events.

- Biocides for which the contribution of wastewater seems more important. For instance, BIT, CMIT and MIT have been detected and quantified in Clichy CSOs only when stormwater percentage was below 60%. These compounds are widely used in household products, cosmetics and personal care products and thus, these uses are their major source in CSOs.
- Biocides for which no clear trend was observed (bottom charts on figure 2) and then both contributions are significant and cannot be discriminated.

Finally, annual mass loads of the 18 targeted biocides have been estimated in both CSOs and in the WWTP effluents of Seine Centre. Compared to WWTP effluents, CSOs seem to be a significant source of benzalkonium C12 and C14, CMIT, cybutryn, and OIT as their mass loads in CSOs are in the same order of magnitude or higher than in WWTP discharges. Mass loads of BIT, MIT, thiabendazole and benzalkonium C16 in CSOs represent less than 10% of mass loads in WWTP effluent. Thus, the contribution of CSOs to the contamination of surface waters seems to be minor. Mass loads of other biocides represent from 11 to 32% of those in WWTP effluents.



**Figure 2:** Concentrations of few biocides as a function of stormwater percentage in two CSOs (Clichy and Vincennes-Charenton).

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