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Multicontamination phenomena occur more often than expected in Mediterranean coastal watercourses: Study case of the Têt River (France)

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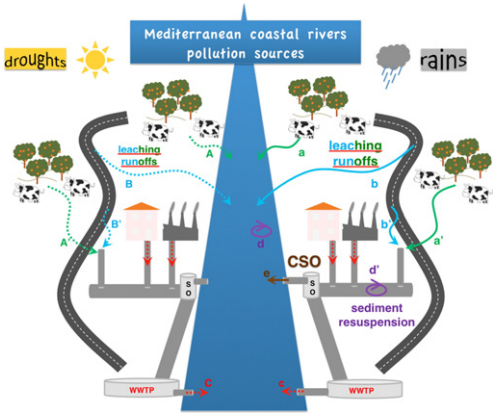
HIGHLIGHTS

- Pollutants cocktails dynamics was studied at river waters during storms and floods.
- Exceptional multicontamination phenomena occur during CSO at heavy rainfall peaks.
- Diffuse pollution sources will be most important at flood flow peaks.
- We can consider these as Mediterranean chronic multiple stressors events.
- Management measures include tackling CSO and monitoring during storm events.

GRAPHICAL ABSTRACT



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ABSTRACT

Contaminants found in watercourses are not only the result of anthropogenic activities but also depend on river's seasonal hydrodynamics. This is particularly true in Mediterranean climate regions where long dry periods are interrupted by strong rainfalls. Storm events remobilize particles from soils and sediments and, as a consequence, the load of particulate matter in rivers can be quite considerable, severely affecting water quality. Nevertheless, an absence of fieldwork studies exists concerning the simultaneous dynamics of mixtures of pollutants in river waters, particularly during strong rainfalls and floods. Our study assessed the concentrations of six families of pollutants, including pesticides, at these events, and compared them to those observed at drought sampling periods. We have used as model a typical Mediterranean coastal river from Southeast France, the Têt River, whose hydrodynamics and major elements fluxes have been fairly investigated. As expected, our results show that chemical mixtures due to human activities occur and that they are particularly relevant during storm events. But the results of our study argue that exceptional multicontamination phenomena actually happen more often than expected because they are linked to recurrent sudden intense rainfall events in the Mediterranean. In particular, combined sewer overflows are responsible for this major issue in urbanized areas, whereas runoff

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and leaching will be the most important sources of pollutant mixtures occurring at flood flow peak. After an overview of the sources responsible for chronic multiple stressors events in regions under a Mediterranean climate regime worldwide, we revisit best management measures to reduce risks from the presence of chemical mixtures in the environment.

1. Introduction

Contaminants found in surface waters are the result of anthropogenic activities such as agriculture, industry and urbanization. Intensive agriculture in particular is still increasing in order to feed the growing population (Tilman, 2002), thus involving higher use of pesticides especially in developing countries (Bourguet and Guillemaud, 2016). Pesticides enter rivers not only via diffuse sources, i.e. soil runoff and leaching of fertilized crops, but also through punctual sources from sewage and WasteWater Treatment Plant (WWTP) effluents. On the other hand, human activities including domestic and industrial sewage systems have a strong influence on the presence of other pollutants in river waters, such as trace metals, nutrients, fecal indicators and industrial or chemical products. Therefore, variation in pollutants concentrations along a river depends on the distribution of human activities all along its course. Besides, contaminants dynamics are influenced by a number of factors, which cannot always be predicted. Thus, river pollution should be monitored at all seasons for better assessment of contaminants levels particularly in areas with a Mediterranean (Med) climate regime (Bernal et al., 2013; Konstantinou et al., 2006). Indeed, Med climate is characterized by long dry periods, during which contaminants accumulate in soils and sediments, and by short but intense seasonal rainfalls (Cowling et al., 2005; Miller, 1983), during which runoff and sediments resuspension occur. As a consequence, in the course of storm events, water quality is known to be impacted by numerous types of pollutants among which pesticides (Boithias et al., 2011; Taghavi et al., 2010), bacterial fecal indicators (Chu et al., 2011; Steets and Holden, 2003), trace metals (Guasch et al., 2010; Oursel et al., 2014), nutrients (Probst, 1985; Ramos and Martínez-Casasnovas, 2006), Polycyclic Aromatic Hydrocarbons (PAHs) (Kim and Young, 2009) and PolyChlorinated Biphenyls (PCBs) (Mandalakis and Stephanou, 2004). This is a global concern because Med climate can be encountered all over the world, not only in the Mediterranean Basin, but also in California, in South Africa, in Central Chile and in Australia (López-Doval et al., 2013). A review of the literature reveals an absence of fieldwork studies concerning the simultaneous dynamics of contaminant mixtures across chemical classes in river waters (Bopp et al., 2016). We believe this is due to the fact that studying how several families of pollutants change with space and time requires vast interdisciplinary expertise (hydrologists, geochemists, ecologists), which adds to the obvious necessary expense of such thorough chemical analyses. As a consequence multiple contamination events are not usually reported even if they are recognized as a major challenge in risk assessment for watercourses (Gregorio and Chèvre, 2014). Even though, it is obvious that pollutants occur as mixtures in the environment, as those observed independently for each pollutant family at storm events (see above). But the scarcity of studies regarding environmental concentrations of pollutants cocktails comes with a consequence: no legislation has yet been implemented for the risk of contaminants mixtures in the environment (Kienzler et al., 2014). Nevertheless, this issue is nowadays pointed as a major concern (Bopp et al., 2016; Kienzler et al., 2014) because chemical dose addition, which is the most accepted measure for the toxicity impact of chemical mixtures (Backhaus and Faust, 2012; Cedergreen et al., 2012; Bopp et al., 2016), can rapidly exceed hazardous concentrations thresholds (Gregorio and Chèvre, 2014; Schwarzenbach, 2009). On top of that, extreme hydrological events are expected to increase due to climate change (Blanchet et al., 2016; Durrieu de Madron et al., 2011; Lespinas et al., 2010). Thus, understanding multiple pollutants dynamics during intense rainfall events is nowadays a key issue for their risk assessment and management (Gregorio and Chèvre, 2014; Kienzler et al., 2016; SCHER, 2012).

We have conducted such a type of study on the Têt River, a typical Med coastal watercourse characterized by a high variable flow strongly influenced by drought and rainy seasons (Ludwig et al., 2004). For instance, the average water flow registered over the last 47 years in Perpignan city is 9.46 m^3/s and 3.69 m^3/s during the winter and summer droughts, respectively (Banque Hydro - www.hydro.eaufrance.fr, 2016). But during intense rainfalls, critical flash floods can occur; for example, peak flows of 850 m^3/s in 1999 and 1115 m^3/s in 1992 were observed. In October 1940 a dreadful event called "l'aiguat" occurred at the western Med Basin during which, 750 mm rainfall in one day and a 3620 m^3/s peak flow were attained at the Têt River in Perpignan city (Serrat et al., 2001). The hydrology and biogeochemistry of this river has been largely studied during the last 10 years, confirming that biotic and abiotic factors threaten this watercourse and its ecosystems (Kim et al., 2006; Lespinas et al., 2010; Ludwig et al., 2004; Meyer et al., 2015). For instance, anthropogenic activities are sources of major elements and nutrients fluxes impacting its water quality (Dumas et al., 2015; Garcia-Esteves et al., 2007). Dumas et al. (2015) also studied the impact of storm events on trace metals fluxes finding out the importance of sudden rain events rather than high peak flows to element trace metals weathering from suspended matter. Furthermore, the small size of this watercourse makes dilution phenomena less important; as a result contaminants are easier to detect. This cumulated knowledge makes of the Têt River an ideal model to test how pesticides change with space and time with respect to other pollutants in Med climate coastal rivers.

In this study, we have performed an in-depth analysis of pesticides dynamics at different hydrological regimes from upstream to downstream of the Têt River and surrounding coastal waters. The aims of our study were (i) to conduct a fine comparison of pesticides to other pollutants concentrations, notably nutrients and trace metals, as well as PCBs, PAHs and fecal indicators, along space and time (ii) to determine if multiple contaminations occur and if they are linked to Med coastal rivers hydrodynamics. Only after discerning the potential chronic behaviour of pollutants mixtures (iii) we can provide wise guidance on risk management of pesticides in particular and chemical mixtures in general in Med climate regions worldwide.

2. Materials and methods

2.1. Study site and sampling stations

The Têt River is the longest watercourse of the Pyrénées-Orientales department (Southeast France) with a total length of 115 km and a catchment area of 1417 km^2 (SMBVT, 2012). Two dams partly control the river flow: the Bouillouses dam in the upstream section and the Vinça dam in the plain (see Fig. 1). The Têt River has no major industrial or farm activities along its catchment but is impacted mainly by agriculture and urban activities (see below). It runs through the city of Perpignan, the main city of the department with 120,000 inhabitants. In this study sampling stations were chosen for their contrasted eco-systemic and anthropogenic characteristics along the river course (from R5 to R0 stations) and at the coastal area (from M1 to M3 stations) (Fig. 1). Station R5 is the most upstream station, situated at Serdinya village, 30 km from the source. Upstream there are only little villages and no crop fields. R4 is located at Villefranche village, 4 km downstream R5,