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Behaviour and fate of urban particles in coastal waters: Settling rate, size distribution and metals contamination characterization

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A B S T R A C T

The evaluation of contaminant net fluxes from the coast to the open sea requires the study of terrigeneous particles behaviour and fate. We studied the particles issued from two small coastal rivers whose waters are mixed with treated wastewater (TWW) coming from the Marseille wastewater treatment plant (WWTP) just before discharge to the Mediterranean Sea. An experimental device was developed and used to investigate particles settling rates, size distribution and metallic contamination when mixing with seawater. The particles were sampled in flood deposits of rivers and outlets during rainy periods and in the outlet water during dry periods. The flood deposits were mainly composed of 50–200 μm-sized particles, higher metals content being observed in the finest fractions. Dry period particles showed the stronger influence of wastewater inputs. Al, Ca, Cs, Li, Rb, Ti, and Tl were mainly of terrigeneous origin, whereas Ag, Ba, Cd, Cr, Cu, Hg, Mg, Mo, Ni, Pb, POC, Sb, Sn and Zn were of anthropogenic origin, issued from non-treated sewage, TWW or industrial waste. In seafloor sediments, all metals exhibited a continuous increase of concentration from the outlet to, at least, 800 m offshore. Implementation of settling particles characteristics in a 3D hydrodynamic and sediment transport model reproduced well the observed deposition of polluted particles in the coastal zone and indicated a non-negligible offshore export of the finest particles and their accompanying pollutants.

1. Introduction

Trace metals are natural constituents of crustal materials. Some of them are essential to the biota, but all have the potential to be biologically toxic if their concentration exceeds certain levels. A wide range of trace metals may be generated throughout human activities, then discharged in rivers by soil erosion processes (depending on the nature of the watershed), throughout wastewaters (treated or not), throughout industrial activities (Azimi et al., 2005) and throughout agriculture (Waeles et al., 2007). Trace metals may be brought by rivers to the coastal area by continuous, diffuse inputs or during flood events, then can accumulate in marine sediments. Their input in the water column and in sediments may be a threat to the quality of the coastal area (Weber et al., 2009; Rocha et al., 2011). Gobeil et al. (2005) studying the Montreal (Canada) WWTP discharge in a large river, the St. Lawrence, have shown that the contribution of the treated wastewater (TWW) was not significant compared to the river metal budget, except for Ag. On the contrary, Oursel et al. (2013) have studied the Marseille (France) WWTP discharge in a small river and showed that, during baseflow periods, the river signature was hidden by the TWW input for most elements or compounds. This difference is due to the larger proportion of TWW discharge in the river stream in the case of Marseille with regard to Montreal (50% vs. 1%, respectively). During flood events, significant amounts of trace metals are rapidly brought to surface aquatic system through runoff processes (Elbaz-Poulichet et al., 2001; Di Leonardo et al., 2009; Weber et al., 2009; Rocha et al., 2011; Nicolau et al., 2012).

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In order to evaluate the impact of such inputs to coastal zones, previous studies have quantified contaminant fluxes at river mouth (Statham et al., 1999; Michel et al., 2000; Ollivier et al., 2011; Nicolau et al., 2012), whereas others have focused on the contaminants fate in the mixing zone, identifying sorption–desorption processes or changes in chemical speciation (Elbaz-Poulichet et al., 1996; Dassenakis et al., 1997; Zwolsman et al., 1997; Millward and Liu, 2003; Waeles et al., 2005). It is therefore necessary to study the fate of particulate pollutants in the coastal zone (Millward et al., 1999), such studies requiring specific experiments.

The settling velocity of river particles in and beneath the river plume is difficult to measure because the fine material can flocculate rapidly in seawater. Indeed, during freshwater/seawater mixing the suspended matter are subjected to important changes in physicochemical properties of the water and their behaviour will depend on their nature, concentration and organic matter content (Thill et al., 2001; Fugate and Chant, 2006). At local scale, the extension and direction of the dilution plume defines where particles settle or are exported depending on the river flow, tide, wind, currents and the morphology of the study area (Naudin et al., 1997; Cugier and Le Hir, 2002; Alliot et al., 2003; Pairaud et al., 2011). Once in the water column, metals can undergo quick desorption or reversely adsorption onto particles. Particles settle directly or after flocculation and accumulate in sediments, which therefore act as a metal sink: the quantification of metal content in surface sediments is consequently a first step to assess the degree of pollution of a marine environment (Bay et al., 2003; Tessier et al., 2011). The grain size is one of the main factors that govern metals contamination in the particulate fraction, as the finest particles contain minerals (e.g. clays, oxides, sulfides, …) and particulate organic matter having strong affinity with metals. There is generally a marked inverse correlation between grain size and metal content in the sediment (Forstner and Wittmann, 1979; Forstner and Patchineelum, 1980). It is therefore critical to quantify the particles size distribution and to assess their settling velocity in the mixing zone. In the last four decades, numerous techniques have been developed for in situ measurement of particles size and settling velocities (Eisma et al., 1996). The Owen tube was for instance used since 1971 to measure the settling velocity distribution of the suspended particulate matter (SPM). It consists in a tube sampler opened at both ends, which is lowered horizontally into the water, closed by trigger at the required depth then rotated vertically (Dyer et al., 1996). The Quisset tube, adapted from the Owen tube, has been developed to determine the settling velocity of particles in the Elbe estuary (Jones and Jago, 1996). These techniques, however, are poorly suited to a shallow plume that is not channelled through an estuary and can move quickly after wind and current changes and their use was, to our knowledge, not associated to the determination of chemical properties in relation with particle size.

Moreover, only few studies focused on the Mediterranean area despite its high population density and its climate specificities. Marseille is an example of large Mediterranean city, where the city WWTP effluents are mixed with small rivers before being rapidly discharged at sea, without passing through an estuary (Oursel et al., 2013).

In this context, through the adaptation of an instrumental laboratory device to separate particles accordingly to their settling velocity in seawater followed by the analysis of the elemental (minor, major and trace) content of each particle size fraction, the purposes of this study were (1) to investigate the physical and chemical characteristics of river/urban particles discharged to the coastal environment, (2) to relate the obtained results with the surface sediment quality in the coastal area and finally (3) to implement a 3D hydrodynamic (Pairaud et al., 2011) and sediment transport model (Verney et al., 2013) within the area to investigate the role of heavy rainfall events on the fate of contaminated particles.

2. Material and methods

2.1. Study site

The Huveaune River extends over 48.4 km long and runs through a watershed with an area of 523 km^2 which consists in karstic formation (60%) and detrital sediments. Land-use in the downstream part of the watershed is urban and industrial. The Jarret River extends over 21 km with a 102 km^2 watershed mainly urban and industrial.

In dry period, these two rivers merge in Marseille; the resulting water is mixed with the Marseille City WWTPs effluent, and then channelled by the Outlet 2 (Out2) to the sea at the Calanque of Cortiou (Fig. 1 and Fig. SI-4.1). This treatment plant, one of the largest in Europe ($1.7 \cdot 10^6$ inhabitant eq.), uses both physical and biological treatment processes. During dry periods ($\sim$300 days), around 100 and 80 $Mm^3\ y^{-1}$ of TWW and river water, respectively, are discharged to the sea (Le Masson, 1997). Oursel et al. (2013) have evaluated the annual global SPM discharge to be around 3900 t, whose 840 t coming from rivers, highlighting the main contribution of the TWW in such conditions. During flood events, the Huveaune and Jarret river flows can overpass 60 and 16 $m^3\ s^{-1}$, respectively. As the maximal outlet discharge capacity cannot overpass 30 $m^3\ s^{-1}$, a significant part of rivers waters is channelled through the Huveaune former bed to the Prado's beach (Fig. 1) during extreme events, i.e. when rainfall overpass $\sim$40 mm h^{-1}. During rainy periods, around 4.1 and 9 $Mm^3\ y^{-1}$ of TWW and rivers waters, respectively, are discharged to the sea. Le Masson (1997) has evaluated the corresponding SPM discharge to 6500 t, 94% coming from rivers. On average, during flood events, 90% of Huveaune and Jarret waters (and SPM) are channelled through the outlet and only 10% are deviated through the Huveaune former bed. Additionally, discharge of untreated wastewater brought to the coastal zone between 456 and 1450 t y^{-1} of SPM, during the period 2001–2007 (Jany et al., 2012).

Statistics on the 01/01/2009 to 31/08/2011 studied period (Météo-France) showed an average of 609 mm y^{-1} of rain distributed as follow: 304, 41, 12, 5 and 3 d y^{-1} where rain was <1, 1–10, 10–20, 20–40 and >40 mm d^{-1}, respectively, which are typical values for a Mediterranean coastal area.

2.2. Sampling and sample conditioning

Two types of samples were considered: (1) flood deposits sampled after a rain with the aim of characterizing most of the particles discharged to the sea during flood events and (2) suspended matter collected using a sediment sampler placed in the river flow (Phillips et al., 2000) in order to characterize the particles brought to the sea during base-flow periods.

2.2.1. Rainy periods sampling

In order to characterise particle inputs to the Calanque of Cortiou, flood deposits were sampled after a rainy event on the 11/05/2009 at Outlet 1 (FDOut1) and on the 14/04/2011 at Outlet 2 (FDOut2). Outlet 1 is only active when the WWTP is by-passed (Fig. SI-4.1) due to an overflow of sewage/rain waters mixture due to the fact that the old Marseille centre has a non-separated sewage network which collects both wastewaters and runoff. The FDOut1 sample is thus likely to contain non-treated sewage particles, as also occurs in many southern Mediterranean coastal cities. In order to characterise particle inputs upstream the WWTP, flood deposits were sampled on 14/04/2011 after a rainy event in the