



Remobilization of polycyclic aromatic hydrocarbons and organic matter in seawater during sediment resuspension experiments from a polluted coastal environment: Insights from Toulon Bay (France)[☆]



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ABSTRACT

Polycyclic aromatic hydrocarbons (PAHs) and organic matter contents were measured in seawater during resuspension experiments using sediments collected from Toulon Bay (Northwestern Mediterranean Sea, France). The studied sediments were very highly contaminated in PAHs, especially in 4-ring compounds emitted from combustion processes. The sediments used for resuspension experiments were collected at 0–2 cm (diagenetically new organic matter, OM) and 30–32 cm depths (diagenetically transformed OM). They were both mostly composed of fine particles (<63 μm), enriched in organic carbon (8.2 and 6.3%, respectively) and in PAHs (concentration of Σ34 PAHs: 38.2 and 35.7 × 10³ ng g⁻¹, respectively). The resuspension of these sediments led to an increase in concentrations of dissolved Σ34 PAHs, dissolved organic carbon (DOC) and dissolved humic- and tryptophan-like fluorophores in seawater up to 10-, 1.3-, 4.4- and 5.7-fold, respectively. The remobilization in seawater was higher for 4–6 ring PAHs, especially benzo(g,h,i)perylene, whose concentration exceeded the threshold values of the European Water Framework Directive. This noted the potential harmful effects of sediment resuspension on marine biota. From these sediment resuspension experiments, we determined OC-normalized partition coefficients of PAHs between sediment and water (K_{OC}) and found that during such events, the transfer of PAHs from sediment particles to seawater was lower than that predicted from octanol-water partition coefficients (K_{OW}) (i.e., measured K_{OC} > K_{OC} predicted from K_{OW}). The results confirmed the sequestration role of sedimentary OC quality and grain size on PAHs; the OM diagenetic state seemed to impact the partition process but in a relatively minor way. Furthermore, differences were observed between 2–4 ring and 5–6 ring PAHs, with the latter displaying a relatively higher mobility towards seawater. These differences may be explained by the distribution of these two PAH pools within different OM moieties, such as humic substances and black carbon.

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1. Introduction

Polycyclic aromatic hydrocarbons (PAHs) are among the most widespread organic pollutants in marine environments. Due to

their hydrophobicity and low water solubility, PAHs entering marine waters tend to sorb onto particulate organic matter (OM) (Gustafsson et al., 2001; Kennish, 1992; May et al., 1978) that is then deposited to the sediments via vertical sinking. This mechanism is recognized as a major pathway for the removal and global cycling of hydrocarbons in the ocean (Adhikari et al., 2015; Berrojalbiz et al., 2011; Gustafsson et al., 1997). However, due to natural (waves, currents, storms) and anthropogenic (dredging, trawling, ship traffic) forcing, coastal sediments are frequently resuspended, refocused and transported along the continental shelf (Durrieu de

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Madron et al., 2008; Schoellhamer, 1996) and thus may turn into a potential source of organic pollutants, including PAHs, for the water column (Dong et al., 2015; Eggleton and Thomas, 2004). While natural forcing generally induces the resuspension of surface sediments (the first centimeters), human activities, particularly dredging or harbor improvements, may lead to the resuspension over sediment depths of several dozens of centimeters. Dissolved PAHs originating from sediment resuspension may then have harmful effects for organisms living in the water column (Gewurtz et al., 2007; Varanasi et al., 1985; Woodhead et al., 1999).

The partitioning of PAHs between water and sediment evaluated through the determination of the sediment-water or sediment OM-water partition coefficients (K_d or K_{oc} , respectively) is largely driven by grain size and OM content of sediments and may vary during resuspension events (Cornelissen et al., 2006; Karickhoff et al., 1979; Rockne et al., 2002; Tremblay et al., 2005). However, little is known about the association between PAHs and sedimentary OM subjected to transformations during burial and diagenesis (Berner, 1980; Henrichs, 1993); that is, the change in K_d and K_{oc} values with sediment depth or OM aging. Further, studies dealing with PAH partitioning between water and sediment should better consider the interactions between dissolved PAHs and dissolved OM (DOM) (Akkanen et al., 2005; McCarthy and Jimenez, 1985; Yang et al., 2016). Over the last decade, the dynamics of DOM in coastal waters has been evaluated through the study of its fluorescent fraction (FDOM). Recently, FDOM associated with marine or river sediments has been characterized, i.e., the FDOM from sediment pore waters (Burdige et al., 2004; Dang et al., 2014; Sierra et al., 2001) and from sediment particles (Brym et al., 2014; Dang et al., 2014; He et al., 2016), as well as FDOM released by coastal sediment resuspension (Komada et al., 2002). Hence, due to the influence of OM in PAH partitioning processes, it is generally observed that theoretical K_{oc} , i.e., K_{oc} predicted from their octanol-water partition coefficients (K_{ow}), are lower than measured K_{oc} (Accardi-Dey and Gschwend, 2002; Feng et al., 2007; Fernandes et al., 1997). As a corollary, concentrations in water that are determined from sediment concentrations and theoretical K_{oc} are frequently overestimated. This also leads to an over-estimation of uptake processes and/or effects of those chemicals on exposed organisms (Arp et al., 2009; Hawthorne et al., 2007). Measuring K_{oc} is thus essential for more accurate predictions of PAH toxicity in the water column.

In the Mediterranean Sea, Toulon harbor (Southern France, Northwestern Mediterranean Sea) hosts the main French Navy structure. It is located deep inside Toulon Bay and enclosed by a sea wall. Because of this separation, and also the irregular freshwater inputs and the low tide, water circulation in this part of the Bay is limited, leading to low water regeneration and potentially strong accumulation of chemical contaminants such as PAHs in the sediments (Benlahcen et al., 1997; Misson et al., 2016; Wafo et al., 2016). To maintain a navigable water depth, harbor dredging is regularly required in accordance with the current legislation. Such dredging may induce a sediment resuspension over 100 cm sedimentary depths, thus allowing surface and deep sediments to be resuspended. Since surface and deep sediments have different OM contents (from potentially different origins along with diagenetic transformations with time/depth), their resuspension could generate various remobilization kinetics of pollutants in seawater. Several studies have reported high concentrations of trace metals, metalloids and organometallics during Toulon Bay seabed disturbances (Cossa et al., 2014; Dang et al., 2015a; Pougnet et al., 2014; Tessier et al., 2011). In addition, Dang et al. (2015b) demonstrated that Pb was significantly released in seawater and further transferred to organisms such as mussels during resuspension experiments on surface and deep sediments from Toulon Bay. Nevertheless, to our knowledge, no data are available describing

PAH remobilization during sediment resuspension events in such a strongly dredged and multi-contaminated area.

Therefore, in the present study, we carried out supplementary experiments simulating the resuspension of surface and deep sediments in Toulon Bay, as proposed by Dang et al. (2015b). Our objectives were (i) to assess the contamination level and origin of PAHs in a sediment core from Toulon Bay, (ii) to evaluate the effect of surface and deep sediment resuspension on the remobilization of PAH and OM, as well as on the water quality, and (iii) to compare theoretical and measured K_{oc} to better understand the factors controlling PAH remobilization during sediment resuspension experiments (SRE). To our knowledge, this study represents the first assessment of remobilization kinetics of both dissolved PAHs and DOM during SRE involving surface and deep sediments.

2. Materials and methods

2.1. Study area

Located on the French Northwestern Mediterranean coast, Toulon city is a part of a large urban area of approximately 500×10^3 inhabitants. Toulon Bay is divided into two unequal basins, a small basin (9.8 km², semi-enclosed) submitted to various anthropogenic inputs (the French Navy, commercial traffic, raw sewage from the urban area, industries) and a large basin (42.2 km²) that is less impacted and open to the sea (Fig. 1). Toulon harbor is situated in the small bay, which is the discharge area for the watershed. Low tides in the Mediterranean Sea associated with weak currents in this area of Toulon Bay have significant implications for the accumulation of contaminants in sediments (Dufresne et al., 2014; Tessier et al., 2011).

2.2. Sampling and sample treatment

Sediments and seawater were collected at the Missiessy (MIS) site within the nuclear submarine harbor of the French Navy, on May 5th, 2014, with the support of the French Navy (ship, material, divers) (Fig. 1).

2.2.1. Sediment core

A sediment core of ca. 50 cm was collected through the use of an interface corer (Plexiglas tube, 10 cm diameter and 1 m long), by Navy divers keeping undisturbed the bottom water column and the upper sediment column, and so preserving the water-sediment interface as described in Dang et al. (2014, 2015a), Tessier et al. (2011). The collected sediment core was maintained vertically and was carefully transferred (by boat and then van) to the laboratory before being installed on a home-made slicing table under glove box. The core was sliced with a 2-cm resolution under inert atmosphere (N₂) to preserve oxidation-reduction (redox) conditions. Each slice was then homogenized in a 150 mL high-density-polyethylene bottle and split into 3×50 mL polypropylene centrifuge tubes under N₂ atmosphere. Then, porewater was extracted by centrifugation (15 min, 20 °C, 400 rpm, Sigma 3–18 K), recovered under N₂ atmosphere by filtration (0.2- μ m on-line syringe filters, cellulose nitrate, Sartorius) and stored in required vessels depending on further chemical analyses. Such methodology was successfully applied to study depth profile variation of main diagenesis tracers and OM characteristics (Chen and Hur, 2015). In accordance with previous studies on trace element sedimentary dynamics in the same area (Dang et al., 2015b), two slice samples were selected from this sediment core to perform SRE: the 0–2 cm sediment layer (denoted hereafter “0–2 cm sediment”) and the 30–32 cm sediment layer (denoted hereafter “30–32 cm sediment”) (see § 2.3). The PAH concentrations were determined in

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