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ScienceDirect
Continental Shelf Research 26 (2006) 1843–1857
CONTINENTAL SHELF RESEARCH
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# Sources and sinks of sediment-bound contaminants in the Gulf of Lions (NW Mediterranean Sea): A multi-tracer approach

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Received 19 October 2005; received in revised form 6 April 2006; accepted 20 April 2006
Available online 25 July 2006

**Abstract**

Surficial sediments collected in 2002 throughout the Gulf of Lions continental shelf (NW Mediterranean) were analysed for trace metals (Cd, Co, Cr, Cs, Cu, Ni, Pb, Sr, Zn and Zr), major elements (Al, Ca, Fe, P and Ti) and the sewage marker coprostanol. In addition, particle size distribution, organic carbon (OC) and carbonates were also determined. Results showed that the metal contamination (Cd, P, Cu, Pb and Zn) is mainly introduced by the local rivers and accumulates—via a regulation by OC and silt fraction (2–63 μm)—in the direct vicinity of the mouths, in high shear stress environments. Here also the signal of sewage contamination is the best preserved, especially off the eastern point sources where local sedimentation rates save the faecal marker from biodegradation processes. It is demonstrated that the shallow prodeltas are the first repository areas for land-derived particles, exposing local ecosystems to both inorganic and organic contaminations. When going seaward, however, sediment dilution, particle sorting and biodegradation processes make that most riverborne contaminants rapidly reach natural levels. Only some metals (i.e. Pb and Zn)—closely associated with the clay fraction—still depict anthropogenic enrichment, which seems to be inherited from man-made aerosols.


*Keywords:* Heavy metals; Coprostanol; Contaminants; Riverborne and aerosol sources; Repository areas; Gulf of Lions

## 1. Introduction

The rapid development of human activities during the last decades within densely populated areas has continuously increased the risk of environmental deterioration, especially in coastal systems (Clark, 2001). Industry, agriculture, urban wastewaters, port activities and tourism release a wide variety of contaminants that mix with the natural constituents of the environment. Both natural and anthropogenic inputs reach finally the coastal zone mainly via river-transport but also by diffuse atmospheric deposition on the sea-atmosphere interface as dust and man-made aerosols. For these reasons, and according to the fact that sediments represent a preferential sink for contaminants, the survey of particulate discharges into sensitive littorals is of great importance.

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doi:10.1016/j.csr.2006.04.010

The Gulf of Lions (NW Mediterranean Sea) is an interesting site for studying the fate of land-derived contaminants in the marine environment because it is bordered by numerous industrialized/cultivated river catchments that discharge important quantities of particles into the coastal zone. Previous investigations on the sedimentary processes at the continent/sea interface highlighted the dynamic feature of the shallower shelf, alternatively acting as sink and source of particulate materials (Roussiez et al., 2005a and references therein). However, even if a large panel of studies have been conducted in this area (e.g. Durrieu de Madron et al., 2000), little is still known about the distribution pathways and entrapment of associated elements such as heavy metals and sewage-borne contaminants.

Improving our knowledge on this has two major interests: (i) on the one hand, the identification of the marine repository areas of terrestrial contaminants may help to combat their negative effects on the biological communities in the coastal areas. Heavy metals behave as conservative pollutants, entailing long-lasting toxic effects for living organisms and an increase of their concentrations in the trophic chain through biomagnification. Concurrently, organic compounds and pathogens deriving from human sewage pose additional environmental risks such as, for example, transmission of infection diseases (Lee and Glover, 1998) and bacterial resistance in aquatic organisms. (ii) On the other hand, anthropogenic compounds can be used for the tracking of particles, a necessary condition to determine the ultimate fate of the continental materials in the marine system.

Our purpose was therefore to identify the dispersion pathways and repository areas of natural and anthropogenic inputs through a large-scale mapping of the geochemical properties in surficial sediments of the Gulf of Lions. The elements we investigated are trace metals (i.e. Cd, Co, Cr, Cs, Cu, Ni, Pb, Sr, Zn and Zr), major oxides (i.e. Al, Ca, Fe, P and Ti) and coprostanol, a sterol associated with sewage solids (Kelly, 1995) widely used to trace the faecal contamination in aquatic environments (e.g. Readman et al., 2005). In addition, general sediment properties (i.e. clay, silt, organic carbon [OC] and carbonate contents) were also considered in order to improve our understanding of geochemical and sedimentological factors that likely control the elemental distributions. To the best of our knowledge, this paper constitutes the most complete survey to date of the extent of both inorganic and sewage contaminations over the continental shelf and gives important information on the particle dispersal throughout this key environment of the Mediterranean Sea.

## 2. Material and methods

### 2.1. Study area

The Gulf of Lions is one of the largest continental shelves of the Mediterranean Sea, receiving various sources of insoluble particles, both of riverine and atmospheric origins. The total river solid discharge to the study area can be estimated to about $6.5–10.5 \times 10^6$ t/yr (from Roussiez et al. (2005a) and references therein). More than 80% of this input arises from the Rhône River (Durrieu de Madron et al., 2000). The other rivers (e.g. Têt, Aude, Orb and Hérault—see Fig. 1) are characterized by smaller watersheds and show a highly variable discharge regime governed by episodic flood events. This makes that suspended sediments of the small coastal rivers are mainly delivered to the marine system in the form of intermittent pulses. The local atmospheric input of particulate matter derives mostly from crust-rich Saharan aerosols, although also anthropogenic-rich European aerosols are introduced into the study area (Löye-Pilot, pers. commun.). By extrapolating a recent estimation of annual aerosol flux in the western Mediterranean basin (Löye-Pilot, pers.com), the overall atmospheric particulate input to the study area can be calculated to about $0.17 \times 10^6$ t/yr. The atmospheric contribution is hence about 50 times lower than the riverine inputs, implying that the sediment particulate matter in the Gulf of Lions is mostly derived from rivers.

In terms of human impact, industrial activity in the Rhône valley is clearly more elevated than in the smaller river basins of the western part of the Gulf, where the socioeconomic activities are mainly related to agriculture and tourism. In terms of geology, the major part of the river catchments is composed of calcareous rocks, except for the Aude and especially Têt rivers, which drain metamorphic and igneous rocks from Pyrenees mountains.

### 2.2. Sampling and geochemical analyses

In November 2002, 51 sediment cores were collected in the Gulf of Lions during the REMORA 3 oceanographic cruise. Sediment samples were