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Pathways of inorganic and organic contaminants from land to deep sea: The case study of the Gulf of Cagliari (W Tyrrhenian Sea)

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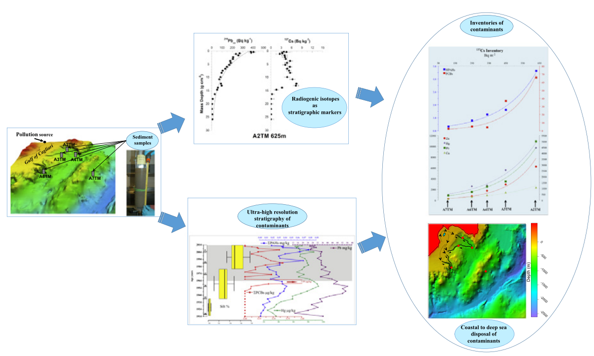
HIGHLIGHTS

- Bottom morphology reveals a complex and articulated system of submarine canyons.
- High-resolution stratigraphic markers for sedimentary marine records
- Canyons as primary pathway conveying sediments and associated contaminants.

GRAPHICAL ABSTRACT



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ABSTRACT

In continental margins, canyons appear to act as natural conduits of sediments and organic matter from the shelf to deep basins, providing an efficient physical pathway for transport and accumulation of particles with their associated land-produced contaminants. However, these mechanisms have not been yet sufficiently explored by geochemical markers. The continental slope of the south Sardinia has been used as a natural laboratory for investigating mechanisms and times of transfer dynamics of contaminants from land to sea and from shelf to deep sea through an articulated system of submarine canyons. Here, dynamics of contaminants have been investigated in a pilot area of the central Mediterranean basin (Gulf of Cagliari, S Sardinia) where important industrial plants are sited since beginning of the last century. Five sediment cores dated by ^{210}Pb and ^{137}Cs reveal: i) a complex dynamics of organic and inorganic contaminants from point source areas on land to the deep sea and ii) a crucial role played by canyons and bottom morphology as primary pathway conveying sediments and associated contaminants from sources to very far deep sea environments. In particular, this study provides new integrated tools to properly understand mechanisms of connection between coastal sectors and deep sea. This is challenging mostly in regions where coastal pollution could represent critical threats for larger areas of the Mediterranean Sea.

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1. Introduction

Coastal marine areas are under increasing pressure due to anthropogenic activities introducing significant amount of contaminants into the sea with consequent concerns for the fragile marine ecosystems and, indirectly, human health. The complex biogeochemistry of inorganic and organic contaminants in the marine environment combined to their persistence and limited potential for degradation, produces long-term residence time, long-range potential transport and relevant effects of bioaccumulation and biomagnification in the trophic web (e.g., Lohmann et al., 2007; Scheringer et al., 2009; Romero-Romero et al., 2017). Contaminants in the marine sediments undergo a combination of chemical (e.g., adsorption/desorption, water/particle exchanges, biodegradation), early diagenetic and sedimentological processes (e.g., re-suspension and re-deposition), which make difficult to track, in terms of chemical and physical dynamics, their evolution in the marine environment.

Recent investigations demonstrated that many chemical contaminants reach the deep-sea (e.g. Storelli et al., 2009; Jamieson et al., 2017) representing, with the reduced physical and chemical dynamics of this environment, a long-term risk with unpredictable effects for the deep ecosystem (Froescheis et al., 2000; Covaci et al., 2008). Recent studies (e.g. Louvado et al., 2015; Astrahan et al., 2017; Paluselli et al., 2018) explored in some detail mechanisms of contaminants transfer from coastal to deep sea areas (>200 m), canyons, in particular, act as natural conduits of sediments and organic matter from the shelf to the deep basins, thus providing an efficient physical pathway for transport and accumulation of sediments including contaminants (Martin et al., 2013; Tubau et al., 2015; Fernandez-Arcaya et al., 2017; Cai et al., 2018). Due to the tectonic settlement and/or to the sediment supply, canyons may act as delivery systems or as sink for contaminants (de Stigter et al., 2007, 2011), depending on the uplift of the source region, the subsidence of the basin area or to the amount of sediments available.

Several recent multidisciplinary projects have focused on the study of canyons, and have considerably increased our understanding of their ecological role, the goods and services they provide to humans, and the impacts that anthropogenic activities have on their ecological condition (Fernandez-Arcaya et al., 2017). Being a link between coastal areas and deep oceans, they serve as conduits for the transport of sedimentary material from the surface to the bottom of the sea (Fabres et al., 2008; Costa et al., 2011; Mil-Homens et al., 2013). Also, the role played by canyons in transporting contaminants to the deep sea is enormously reinforced when they are the locations of highly dynamic shelf to basin export processes (e.g., Dense Shelf Water Cascading-DSWC in the NW Mediterranean Sea; Canals et al., 2006). In this context, the continental slope of the south Sardinia is an interesting natural laboratory for exploring mechanisms and timing of transfer dynamics of contaminants from land to sea and from shelf to deep-sea through an articulated system of submarine canyons and specific source areas. An ensemble of chemical analyses of radionuclides, inorganic and organic compounds from sediment cores sampled between 625 and 1153 m depth provides an unprecedented opportunity to investigate dynamics of contaminants from the land sources to the marine sinks.

2. Material and methods

2.1. Site description

The Gulf of Cagliari (CG) is placed in the SE sector of Sardinia (Fig. 1A), where fluvial sedimentation since Pleistocene caused favourable conditions for the development of coastal lagoons separated by the rhodalgal limestones Miocenic hills of Cagliari (Cossellu, 2007; Kalb, 2008). Holocenic sandbars complete the evolution of the coastal margin. Clayey, sand and siliciclastic carbonate sediments characterize the latest Pleistocene-Holocene stratigraphy of Cagliari Basin. Because of sediment supply, sea-level changes and tectonics, CG has a pronounced shelf that extends about 15–20 km south-eastward (Lecca et al., 1998). The shelf abruptly interrupts with a marked break in slope located at 60/80 m below sea level (bsl). Extensive canyons (and several tributary incisions) dissect the offshore-continental slope margin of CG, named Pula (PC), Sarroch (SC), S. Elia-Foxi (SEC) and Carbonara (CC), respectively (Fig. 1B). In this area, also two major seafloor relative highs are present: the Carbonara Ridge (CR) and the Banghittu Knoll (BK; Fig. 1B and C). CR is a 85 km long, 25 km wide NE-SW striking seamount rising >1000 m from the surrounding seafloor (Fig. 1C). The deeper flanks of CR slopes are located at 1250 m bsl, while its apical sector reaches 156 m bsl. BK is located in the SW sector of CG, on the uppermost portion of the slope, where it acts as a physiographic boundary between SC and PC. BK shows an almost flat, SW gently dipping top (ca 0.5°) on its apical part, which is located between 110/140 m bsl.

The presence of two major industrial complexes dominate the potential inputs of contaminants at sea from the coastal area of the CG where also urban activities of the Cagliari city, with its 500.000 inhabitants and the Is Arenas waste water treatment plant occur. The industrial area behind the Cagliari town (Macchiareddu-Grogastu conglomerate) additionally includes plants of i) fluorine treatment (Fluorsid S.p.A.), ii) generation and distribution of propane (IsGas) and iii) a petrochemical plant (Syndial S.p.A.); in the westernmost part of the gulf the refinery industrial plant of Sarlux-Saras S.p.A. (oil refining and production of electricity) occurs.

2.2. Data and methods

Seafloor bathymetry and sediment cores were acquired during the Anomcity_2014 oceanographic cruise on-board of the R/V Minerva Uno (National Research Council, CNR). Bathymetric data were used to deploy a Digital Terrain Model (DTM) covering an area of 17′000 km^2 in the 2670 m to 1 m bsl bathymetric range.

Five sediment cores were collected using a box-corer, at depths ranging from 625 and 1153 m (Fig. 1A and B; Table 1). The A2TM and A3TM cores are located at a depth of about 600 m bsl in the SEC and SC canyon branches, respectively; these two sampling sites were selected where a slope decrease in the seafloor morphology has been observed. The A4TM sets at the confluence of SEC and SC active branches, at a depth of about 900 m bsl, while the A6TM was sampled at the confluence of the PC active branches (SW from A2TM and A3TM, at about 780 m bsl). Finally, the A7TM lies SE from the apical sector of the CR main axis, at about 1150 m bsl depth. A full description of data and methods can be found in the additional materials (see text in Supplementary Material). Core sub-samples were prepared for grain size, geochemical and radiometric analyses. Cores analysis included: radionuclide measurements, grain-size fraction, concentrations of Al, Fe, Mn, Cd, Pb, Co, V, As, Cr, Cu, Ni, Zn, Hg, TOC, TN, PAHs (16 US-EPA priority congeners) and ΣPCBs. Results are reported in Supplementary Table S1.

3. Results

3.1. Seafloor morphology

The morphology shows the existence of several abandoned canyon branches, partially filled by sediments and presently unlinked to the active sections of SC and SEC (Fig. 1B). Contrary to the active canyon thalwegs, minor incisions tend to disappear toward deeper sectors of the slope. Since their intrinsic sediment dynamic, the CG active set of canyons and incisions rules the mechanism of sediment distribution from land to deep sea (e.g., Puig et al., 2014). The overall emplacement of uppermost branches of canyons is mainly controlled by the slope direction, i.e., ca NW-SE (Fig. 1B and C). On the contrary, the presence of CR, located at the slope foot, constraints the pattern of deeper segments of canyons (Fig. 1B).