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Baseline

Temporal evolution of cadmium, copper and lead concentration in the Venice Lagoon water in relation with the speciation and dissolved/ particulate partition

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ABSTRACT

In order to study the role of sediment re-suspension and deposition versus the role of organic complexation, we investigated the speciation of cadmium (Cd), copper (Cu) and lead (Pb) in samples collected in the Venice Lagoon during several campaigns from 1992 to 2006. The increment in Cd and Pb concentration in the dissolved phases, observed in the central and northern basins, can be linked to important alterations inside the lagoon caused by industrial and urban factors. The study focuses on metal partition between dissolved and particulate phases. The analyses carried out in different sites illustrate the complex role of organic matter in the sedimentation process. While Cd concentration in sediments can be correlated with organic matter, no such correlation can be established in the case of Pb, whose particulate concentration is related only to the dissolved concentration. In the case of Cu, the role of organic complexation remains unclear.

The Venice Lagoon is the largest Italian lagoon and is densely inhabited. Since the 13th century, it has been subjected to substantial human-caused changes, mainly in order to maintain certain hydraulic characteristics and to stop the silting up. Industrial and agricultural activities, together with the population increase that marked the last century, have caused serious problems of chemical pollution, considerably reducing the environmental quality of the Venice Lagoon (Pavoni et al., 1987; Scarponi et al., 1998; Capodaglio et al., 2005; Chapman et al., 2009). Studies to evaluate and quantify the lagoon's environmental chemical contamination, which started > 30 years ago, mainly focused on the analysis of sediments and emphasized the presence of organic and inorganic pollutants (Donazzolo et al., 1983, 1984; Albani et al., 1989; Molinaroli et al., 2013).

Previous attempts to evaluate pollutant dispersion were generally based on the assumption that the phenomenon is largely controlled by sediment movement as a consequence of tidal currents (Alberotanza et al., 1987; Basu and Molinaroli, 1994). Although these studies showed a general agreement with the distribution of pollutants in bottom sediments, they also highlighted a few anomalies. An in-depth distribution of pollutants may be described taking into account the processes affecting their partitioning between water/atmosphere and sediment/ water interfaces and the influence of the chemical-physical parameters

(Martin et al., 1994; Millward, 1995).

Studies carried out on the lagoon sediments showed that their toxicity was not correlated to the total content of pollutants but to their speciation (Scarponi et al., 1998); consequently, the differentiation between the forms in which components are present (Iyer and Sarin, 1992; Achterberg and Van Den Berg, 1997) is fundamental.

Benthic fluxes of trace elements studied in the Venice Lagoon (Turetta et al., 2005) were prevalently controlled by chemical and diffusive processes, while benthic fluxes of organic pollutants tended to be remobilized from the sediments by re-suspension and partly trapped as particulate matter in the microlayer at the water/atmosphere interface (Manodori et al., 2006, 2007).

Improvements in sampling procedures, sample treatments and analytical techniques enabled the reliable quantification of pollutants and the speciation study of trace elements (Martin et al., 1994; Capodaglio et al., 1996; Cescon et al., 1996) to evaluate their bioavailable fraction (Pavoni et al., 1998; Scarponi et al., 1998).

Furthermore, the concentration of elements in the dissolved phase, especially for Pb, Cu, Cd and Chromium (Cr), may be a function of the geographical area and tide level (Martin et al., 1994; Cescon et al., 1996). The increase in re-suspension in lagoon waters since 1994, due to the reduction of macroalgal coverage, which was combined with a

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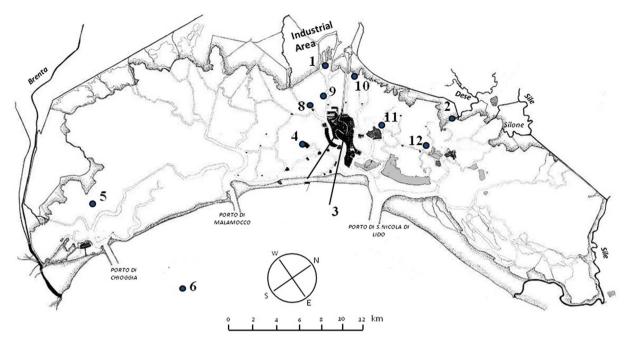


Fig. 1. Map of the Venice Lagoon with the sampling sites: Marghera (1), Dese (2), Historic centre of Venice (3), Sacca Sessola (4), Chioggia (5), Adriatic sea (6), Punta della Salute (7), San Giorgio in Alga (8), Tresse (9), Campalto (10), Murano (11), Mazzorbo (12).

significant decrease in organic carbon (OC), and to the mechanical dredges used for harvesting the invasive Manila clam (*Ruditapes philippinarum*) (Sfriso et al., 2005) got the sediments mix down to 30 cm (Pranovi et al., 2003, 2004). Resuspension phenomena and increased navigation in the lagoon contributed to the net loss (erosion) of sediments observed.

The modeled contaminant transport from the industrial area of the Venice Lagoon (Sommerfreund et al., 2010) highlighted that the resuspension and the erosion of the sediments, together with the present industrial discharges and the tributary sources, are an important source of pollutants to the entire lagoon. However, the model did not take into account the effects of the significant decrease of organic matter observed in the sediment.

In this paper, we will study the spatial distribution of cadmium (Cd), copper (Cu) and lead (Pb) and their temporal evolution from 1992 to 2006 in different sites of the Venice Lagoon. The study focuses on the metal speciation and the metal partition between dissolved and particulate phases, in order to evaluate the mobility and the partition of Cd, Cu and Pb in the transition and lagoon water ecosystems.

As shown in Fig. 1, in order to study the temporal evolution of trace element speciation in the Venice Lagoon, sub-surface waters were sampled across several sites and in different years (from 1992 to 2006). In more detail, in July 1992, six sub-surface water samples were collected in six different sites; in October 1992, twelve sub-surface water samples were collected at the San Marco dock (Punta della Salute, every 2 h); in 1994, from March to the end of June, twelve sub-surface water samples were collected at Sacca Sessola: in 1997, from March to the end of September, nineteen samples were collected of the island of San Giorgio in Alga; finally, in 2005-2006, ten sub-surface water samples were collected at Campalto and at Sacca Sessola. In 2001 and 2002-03, in relation with the two benthic chamber experiments carried out in Tresse and Campalto (Turetta et al., 2005), trace metal speciation was also studied in sub-surface water samples; in relation with the sampling of microlayer in the Venice Lagoon, trace metal speciation was investigated in eight samples of sub-surface water collected at Sacca Sessola and Murano in 2001-2003 (Manodori et al., 2006) and on nine sub-surface samples collected at Mazzorbo in 2005-2006 (Manodori et al., 2007). All the samples were collected under neap tidal conditions, except for those sampled in October 1992, which were collected

under spring tidal conditions in order to evaluate the possible variability of the trace metal concentrations as a function of tide level. Furthermore, in 2005–2006, in order to assess the OC concentration, campaign surface sediment samples were collected at Campalto and Sacca Sessola.

The procedure adopted for collecting all the samples was the same. In order to minimize contamination, the sampling apparatus and all the containers and materials were acid-cleaned, appropriately rinsed and conditioned following previously reported procedures (Scarponi et al., 1996b) in a clean chemistry laboratory (Class 100) at the Environmental Science Department of the Ca' Foscari University of Venice, where the handling, treatment, and analysis of the samples were carried out, as well. After being collected with a Teflon pump and stored in a 50 l polyethylene tank, water samples were filtered through a suitable filtering apparatus (Sartorius, SM 16540, with acid-clean cellulose nitrate membrane filters 0.45 μ m pore, Sartorius, SM 11306, 142 mm diameter). The filtered samples were collected directly in 2 l FEP bottles and stored at -20 °C until analysis.

Total OC concentration was established by applying a Shimadzu TOC SSM-5000A analyzer directly on the sediment sample. A portion of the dried sample was treated with HCl 2,5N and heated at 120 $^{\circ}$ C for 30 min to remove inorganic carbon.

The analyzer operated through the high-temperature conversion to carbon dioxide of all carbon in the treated sample. The carbon dioxide was quantified by infrared detection, and the results were reported as mg C on g of dried-weight.

To assess the total dissolved concentration of cadmium (C_{Cd}), lead (C_{Pb}) and copper (C_{Cu}) in the samples collected up to 1994, the filtered samples were previously irradiated with a high-intensity UV lamp and then acid-digested (HCl Ultrapure 30%, Merck or Romil); the treated samples were then analyzed by Differential Pulse Anodic Stripping Voltammetry (DPASV) with a Multipolarograph Analyzer EG & G mod. 384 B. Details of the analytical method are described elsewhere (Capodaglio et al., 1989, 1991, 1994, Scarponi et al., 1996). For the samples collected after 1994, the analysis of total metal concentrations was performed by Inductively Coupled Plasma Sector Field Mass Spectrometry (ICP-SFMS, ElementThermoguest, Bremen, Germany), following the method developed by Turetta et al. (2004). To assess the total concentration of Cu, Cd and Pb in particulate matter, samples were

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