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Environmental Pollution 125 (2003) 433–446
ENVIRONMENTAL POLLUTION
www.elsevier.com/locate/envpol

Effect of eutrophication upon radionuclide dynamics in the Sacca di Goro lagoon (Po River Delta, Italy): a combined field, experimental and modeling study

Cristina Bondavalli*

Department of Environmental Sciences, University of Parma, Parco Area delle Scienze, 11/A, 43100 Parma, Italy

Received 10 September 2002; accepted 14 February 2003

"Capsule": *Clues are provided about the dynamics of Cs-137 in terms of pathways, sinks, sources and cycling.*

Abstract

The focus of this paper is on the relationship between eutrophication and radionuclide circulation at the whole ecosystem scale in the shallow estuarine environment of the Sacca di Goro (Po River Delta, Italy). This lagoon is frequently affected by dystrophic crises, due to decomposition of huge amounts of macroalgae (mainly *Ulva rigida*), and critical conditions created at the interface between sediment and water are such that Cs-137 accumulated in the sediment can be mobilized and made available in the water column. The release of cesium from sediment in this ecosystem has been evaluated through a field experiment in which chemical conditions typical of anoxic crises were artificially created in enclosures. Also a lab experiment was carried out to shed light on possible cesium release by decomposing macroalgae. The two experiments allowed drawing conclusions on crucial factors controlling cesium release in the Sacca di Goro, the first objective of this research. The second objective was understanding the fate of radiocesium once transported in the water column. To this end ecological information gathered during the experiments and a yearly sampling campaign, has been converted into whole-system seasonal networks describing ecosystem flow structure for the Sacca di Goro. Analyzed by network analysis this model has provided clues about the dynamics of Cs-137 in terms of preferential pathways, sinks, sources, and cycling activity. Sediment, together with seston and dissolved cesium, appear to be the most significant components in the circulation of Cs-137; while macroalgal biomasses play a crucial role as an indirect causal factor.

Keywords: Biogeochemical cycles; Eutrophication; Ecosystem; Synergism; Radionuclides; Network analysis

1. Introduction

Most of the studies on biogeochemistry of coastal aquatic environments have focused on macronutrients, particularly nitrogen and phosphorous as related to eutrophication processes (Viaroli et al., 1993 a, b, 1995; Christian et al., 1998), but also on sulfur, iron and manganese, which are essential both in plant nutrition and microbial processes (Viaroli et al., 1996). In recent decades, however, the interest toward less common or rare elements has increased, because such chemical species are often tracers of human impacts. This is the case for radionuclides, which can be a dangerous presence for living organisms (Cruz et al., 1997).

Studies on radionuclides have been focused upon their biogeochemical cycles in order to understand their fate in ecosystems (Yu et al., 1998; Maderich, 1999; Matishov et al., 2000; Smith et al., 2000a; Wang et al., 2000). Results of a model-test for radiocesium in continental Lake Uruskul (Southern Urals, Russia), heavily contaminated with Sr-90 and Cs-137, gave satisfactory results to reconstruct the dynamics of this chemical in the lake (Hakanson and Sazykina, 2001). Again, an operational, user-friendly GIS-based tool (Environmental Decision Support System) has been developed to assess the transfer of radiocesium within the entire contaminated area of Ukraine, Belarus, and Russia, with emphasis on geo-chemical, hydrological, and biological processes (Van-der-Perk et al., 2001).

The need to cope with the consequences of significant release of radioactive contaminants into the environment

* Tel.: +39-0521-905-614; fax: +39-0521-905-402.
E-mail address: cristina@dsa.unipr.it (C. Bondavalli).

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doi:10.1016/S0269-7491(03)00076-9

requires a detailed understanding of how such substances distribute within abiotic components and biotic communities. Monitoring their presence, however, does not provide sufficient information to adequately assess their toxic potential. They enter the various matrices of the ecosystem (wet and dry depositions, water inflow, etc.) and the food webs in several ways, where they are transported, diluted or, eventually, concentrated (Pentreath, 1988; Monte et al., 1997). Fundamental to this end is to identify preferential pathways of circulation at the whole ecosystem level, as defining primary sources of contamination and sinks (Monte et al., 1997; Maderich, 1999; Garnier-Laplace et al., 2000; Matishov et al., 2000; Smith et al., 2000a).

In this framework changed chemical conditions and altered biological response of organisms due to other forms of pollution raise further concern, as they can modify patterns of radionuclide cycling (Delaune et al., 1978; Robbins and Edgington, 1975). This paper explores this issue by focusing on eutrophication, the most widespread form of degradation that affects water ecosystems. It is well known, for example, that consequences of anoxic crises, such as oxygen depletion and low redox potential, may accelerate mobilization of radionuclides from the sediment (Delaune et al., 1978; Robbins and Edgington, 1975). Radioactive elements previously buried in the sediment become now available in the water column and they can either remain confined in the water matrix and return to the sediment, or they can enter the food web with various consequences. This latter aspect depends on the type of organisms that dominate the biocoenosis, their properties in relation with radionuclides, specifically their ability to assimilate and release them, and the trophic structure in which they are embedded.

The Sacca di Goro (Po river delta, Italy) is a heavily eutrophied lagoon in which distrophic crises are frequent (Viaroli et al., 1993a, b). In this area the presence of Cs-137, a long-lived radionuclide, traces back to the Chernobyl accident, which is the last massive release of this contaminant (Durec, 1998). This element accumulated in the sediment, but repeated anoxic crises may have created conditions for its gradual release into the water column. The first specific objective of this research was understanding whether eutrophication processes accelerated cesium mobilization from the sediment in the Sacca di Goro. Cesium that is not buried in the sediment is free to cycle, and the second objective was to grasp essential features of cesium circulation at the whole ecosystem level.

To achieve these objectives the role of macroalgae had to be clarified. Given the extremely high abundance reached by these primary producers, direct release could contribute to the observed peaks of cesium in the water column. A field and a laboratory experiment were carried out to shed light on this aspect, that is crucial also for the second objective of this research. In fact, patterns of radionuclide circulation may be completely misunderstood without a detailed knowledge of processes responsible for cesium release in the water.

Once mobilized, cesium can spread in the ecosystem. It can be assimilated by primary producers such as phytoplankton and move up to higher trophic levels through dietary interaction. Decomposition processes occur as well and the detritus chain acts as an alternative route for cesium movement throughout the system. Also the connections between the two branches of the web allow it to cycle, increasing its residence time. The relationship between grazing and detritus chain change dramatically during dystrophic crises with repercussions on the patterns of circulation.

To understand such patterns, all the biological information gathered from field work (sampling) and the experiments cited earlier have been used to build a general simple network model describing cesium circulation in the Sacca di Goro. Such a model was analyzed by network analysis to identify preferential routes along which cesium propagates in the system, components that act as accumulation points (sinks) or sources of contamination, as well as cycling pathways. This knowledge may be useful to understand the dynamics of radionuclides at the whole ecosystem level, with potential benefit in terms of forecasting the impact of radiochemical contamination.

2. Study area

The Sacca di Goro is an estuarine environment of the Po River Delta, northern Italy (Fig. 1). Its a large lagoon (26 km^2) with an average depth of 1.5 m and is connected to the sea by a 2 km wide mouth. This embayment is highly eutrophic because of the excess of organic substances and inorganic nutrients flowing into the river from its catchment area. Seasonally, huge amounts of macroalgae (*Ulva rigida* and *Gracilaria sp.*) are produced in the lagoon; during their growing phase they appear to store available nitrogen causing phytoplankton depression (Viaroli et al., 1993 a,b; 1995). The decrease of phytoplankton abundance depresses the grazing food chain, and the detritus chain becomes the main pathway along which matter and energy are transferred. Intense decomposition processes occur at the expense of the macroalgal biomass, with two interlinked consequences: (1) oxygen concentration lowers appreciably; (2) often dystrophic crises occur. Reducing conditions at the top sediment layer accelerate nutrient release in the water column, and a loss of nitrate occurs because of denitrification. The macroalgae–sediment link seems functionally relevant in determining the trophic status of the Sacca di Goro (Viaroli et al., 1995; 1996).

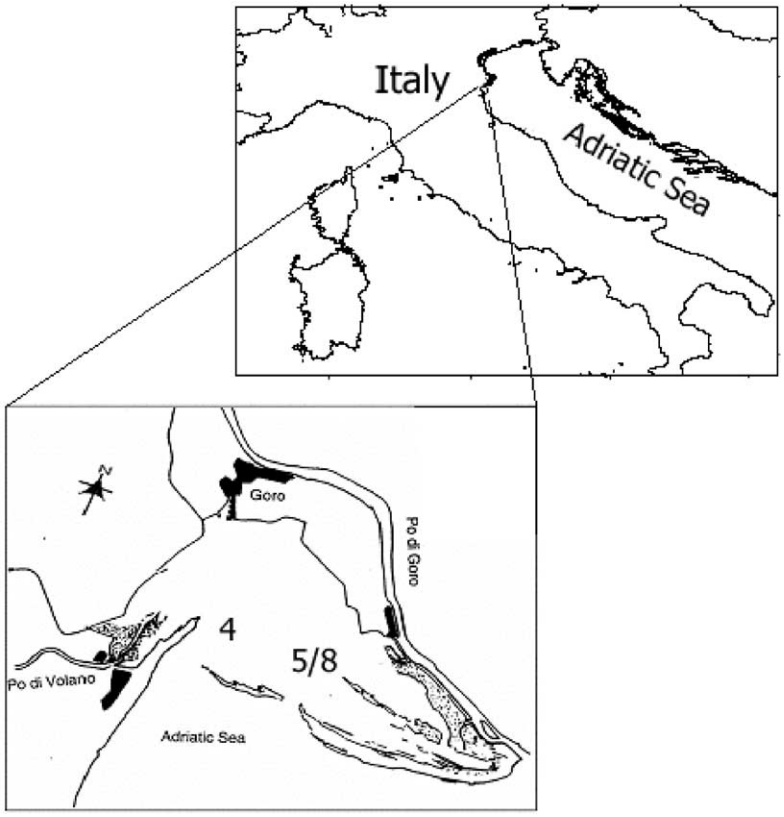

Fig. 1. Study area: Sacca di Goro (Po River Delta, Italy).

Two sites were selected as sampling stations. Each of them presents a peculiar hydrological regime: the one labeled as "station 5/8" (Fig. 1) is characterized by slow water renewal, while the other (station 4) is strongly affected by tides.

3. Materials and methods

The distribution of artificial radionuclides in different environmental compartments, such as water, seston, sediment and macroalgae was first monitored. After this preliminary monitoring phase a first experiment was performed to investigate Cs-137 behavior during macroalgae decomposition; a laboratory experiment was then conceived to study cesium release from the sediment and the role played by organic matter (macroalgae) in this respect.

3.1. Field sampling

The sampling campaign started in March 1992 and continued for 7 years. Here only data collected in 1992 (from March to August) are presented because in that year a systematic approach to sampling was adopted so that all the relevant components of the Sacca di Goro, biotic as well as abiotic, were monitored. Sampling carried out in successive years was related mainly to macroalgae (*Ulva rigida*, *Graciplaria sp.*, etc.) and not regularly accomplished on a monthly bases.

3.1.1. Seawater

Seawater samples, volumes ranging between 25 and 50 l, were collected using a pumping system. HCl (pH = 1) was immediately added after storage to adequately preserve the sample for Cs-137 extraction. This procedure was conducted by ammoniumphosphomolybdate (AMP) technique (Unichim, 1991). To evaluate the radiochemical separation yield, 15 mg of Cs carrier and 10 Bq (1 Bq = 1 event of radiation emission per second) of Cs-134 spike were added to each sample. After a 15–20 min stirring to reach an isotopic equilibrium, 15 g of AMP—$(NH_4)_3PO_4{\cdot}12MoO_3{\cdot}3H_2O$—powder (specific reactant for Cs isotopes) were added to the solution to isolate cesium. After 3 h of slow stirring and 48 h of settling, the clarified surnatant liquid was eliminated. The AMP fraction was then centrifuged and the solid residue was oven-dried at 150 °C and stored into standard Marinelli containers to be counted for determination of Cs-137 content by gamma spectrometry (Bq/m^3). Separation yields ranged between 85 and 95%.

3.1.2. Sediment

Three 30 cm long sediment cores, with a diameter of 5 cm, were collected at each site using a manual corer and

plexiglas cores. A top layer section of about 5 centimeters was removed and dried at 70 °C. Then the samples were homogeneously ground and stored in a Marinelli container for gamma spectrometry determination. Dry/wet percentage, density and organic matter content (AFWD, ignition at 550 °C in a muffle furnace) were also determined.

Additional samples of sediment cores were taken from the study area and Cs-137 vertical profiles were used to gain insights about the sedimentation rate in the lagoon. Each 2.5 cm section of the cores was dried, ground and analyzed by the methods described earlier to obtain a sediment profile of Cs-137 distribution.

3.1.3. Macroalgae

Macroalgae, mainly *Ulva rigida*, were collected at both stations using a rake-like device by raking from 2 to 10 m^2 of sediment surface. Macroalgal thalli were washed in salt water and stored in plastic bags at room temperature. Once in the laboratory, macroalgae were sorted by species, washed again and weighed. Biomass subsamples were dried at 70 °C and weighed to estimate macroalgal standing stock [g(d.w.)/m^2]. These subsamples were powdered, stored in a Marinelli container and analyzed for Cs-137 content. An additional subsample was analyzed for organic matter content (AFDW) by ignition at 550 °C in a muffle furnace.

3.2. Field and laboratory experiments

Field experiments were executed to investigate Cs-137 release from sediment as a consequence of dystrophic crises. Eight dark plastic tubes (i.d. 20 cm, height 1.2 m) were positioned (5–8 July 1993) into the sediment at station 5/8 in locations where the water level did not exceed 60 cm. The tubes were covered at the top with black plastic bags. To elucidate the role of organic matter on cesium release, four of the eight tubes were enriched with 0.1 g/cm^2 of air dried *Ulva*, equal to 10^3 g m^{-2} of fresh material, to simulate the maximum value of biomass that *Ulva* reaches in the lagoon during the dystrophic crises. The experiment lasted about 70 h and temperature (digital thermometer, Hanna Minitherm 8053), dissolved oxygen (Winkler method, A.P.H.A., A.W.W.A., W.P.C.F., 1975), pH [potentiometry, pH-meter TIM 90 and GK 2401 C electrodes (Radiometer)], redox potential (mV meter, Hanna HI 8418) and sulphide concentration [inflection point titration with silver nitrate; mV meter TIM 90, F1212S and K711 electrodes (Radiometer)] were determined every 24 h, inside and outside the tubes. At the end of the experiment Cs-137 content was measured in water and top layer sediment inside and outside the tubes, following the procedure described above for water and sediment sampling.

Laboratory experiments (31 May–28 July 1993) were planned to investigate the possible role of macroalgae decomposition on Cs-137 release into the water column. Water (50 l, not filtered) taken from the lagoon (st. 5/8) and fresh macroalgae [7 kg (wet weight)], roughly cleaned from main epiphytes, were stored in four large tanks. They were kept in the absence of light at room temperature for a period of about 60 days. On days 0, 4, 8, 18, 34 and 61 from the beginning of the treatment about 300 g (wet weight) of macroalgae were taken away to determine dry weight (d.w., desiccated at 70 °C), ash free dry weight (AFDW, ignited at 550 °C in a muffle furnace) and the amount of Cs-137 still present in decomposing thalli. Because of the very low activity of Cs-137 in water, any determination would required a consistent volume (50 l). Due to this problem Cs-137 was determined only at the beginning and at the end of the experiment.

3.3. Gamma spectrometry

Water, sediments and macrolagae samples were analyzed, qualitatively and quantitatively for Cs-137 by gamma spectrometry. Nondestructive analysis was performed in a high resolution (FWHM 1.8 KeV a t1.33 MeV) Ge (Li) γ-detector (Selena) with a multichannel analyzer. Counting times ranged between 1 day and 1 week depending on the sample quantities and signal intensity. The data, decay corrected to sampling date, were processed with a IBM AT Personal Computer equipped with an EG&G ''quantitative analysis software program for gamma spectrometry''. In addition, quality control was assured by several analyses made on certified biotic and abiotic samples (IAEA 307, IAEA 367, IAEA 368) supplied by the IAEA (International Atomic Energy Agency).

3.4. Network analysis

The formal analysis of the model constructed and described here comes under the general heading of network analysis (Ulanowicz, 1987; Ulanowicz and Kay, 1991). This modeling approach requires designating individual compartments, flows of matter/energy between compartments, import and export from outside the system. The connections of the ecosystem to the surrounding environment consist of processes such as advections, primary productions, respirations and permanent sedimentation. The creation of an ecosystem network begins with the identification of the key components that comprise the ecosystem; the next step is to designate a medium of exchange and connect these compartments to one another via feeding and detrital pathways determined from information on diet, primary production, respiration, etc. The work reported herein followed Cs-137 as the medium of exchange, and any information available in the literature, collected in the field and obtained from the experiments was used to

specify the topology of the transfers. Compartmental standing stocks of Cs-137 were quantified as Bq per square meter (Bq/m^2), and flows in Bq per square meter per day (Bq/m^2/day).

The systematic analysis of the ecosystem flow network is comprised of several techniques. Ulanowicz assembled the four primary methods used in Network Analysis into a single software package, NETWRK (Ulanowicz and Kay, 1991), including input–output analysis, trophic level analysis, cycling analysis and the calculation of indices that characterize the entire system. The constituent algorithms are based upon linear algebra and information theory and are explained in detail elsewhere (Ulanowicz, 1986). Below are outlines of the basic ideas behind the methods used for this paper.

Input–output analysis allows one to quantify how any one compartment depends on any other compartment to obtain its requisite medium. In particular, a total dependency matrix, TDM (Szymer and Ulanowicz, 1986) provides information on the fraction of the total amount entering compartment j (column designation) that has been provided by compartment i (row designation) over all pathways, both direct and indirect. A coefficients on matrix diagonal ($i=j$) represent the fractions of throughput that recycle back to that compartment.

Hannon (1973), Finn (1976), Levine (1980) and Patten et al. (1976) give various examples of how one may employ input–output analysis. One very useful such application is the decomposition of the graph according to each input. That is, the eventual fate of each of the nonzero inputs to the system is traced independently of the other inputs to the system. Not only does this decomposition portray the isolated effects of the various inputs, but these sub-networks can be linearly recombined to recreate what the effects of any other combination of inputs would be, if the flow structure were kept the same.

Most networks of ecosystem flows contain cycles of material or energy, and the magnitude and structure of these cycles are analyzed in detail by NETWRK. The program enumerates all of the simple cycles in the given matrix of exchanges and then calculates the fraction of total activity that is devoted to cycling—what is known as the Finn cycling index (Finn, 1976).

4. Results

4.1. Field work and experimental approach

The lagoon receives water from Po di Goro, the southernmost branch of the Po River Delta and from the Po di Volano. In spring, the flows into the lagoon reaches its maximum of 631×10^6 m^3/month. During this period the concentration of dissolved oxygen is high (143% oxygen saturation) as a result of both the freshwater inflow and the presence of large quantities of *Ulva rigida* and *Gracilaria* sp. (Fig. 2) which are responsible for most of the high primary production rate. In late spring and during summer, oxygen concentration lowers because macroalgal biomass is decomposing. At station 5/8, where *Ulva rigida* reached the highest biomass [500 g(dw)/m^2], oxygen concentration was close to zero from the end of July to the beginning of August. It is during this period that characteristic signals of the dystrophic crises can be detected: oxygen depletion, anoxia at the sediment level, reducing conditions and nutrient release from the bottom (Viaroli, et al., 1993 a,b; 1995; 1996).

Cs-137 concentration in the water did not change significantly from March to June 1992. In July its concentration increased, reaching a peak during the period of macroalgae decomposition (August, Fig. 3). Also the quantity of Cs-137 in *Ulva* thalli sampled at station 5/8 reached the maximum value in this period (Fig. 4). Cesium content in the sediment top layer was higher at station 4 than at station 5/8 (Fig. 5). Maximum value for Cs-137 at station 4 was about 70 Bq/Kg(dw), whereas at station 5/8 it never exceeded 30 Bq/kg(dw).

During the field experiment, oxygen demand (Fig. 6) increased at a faster rate in tubes treated with organic matter: oxygen was completely consumed in <70 h. Both pH and redox potential declined much more in the enriched tubes, reaching minimum values of pH 6.3 and -293 mV, respectively, at the end of the incubation. Production of free sulfides in the treated tubes began

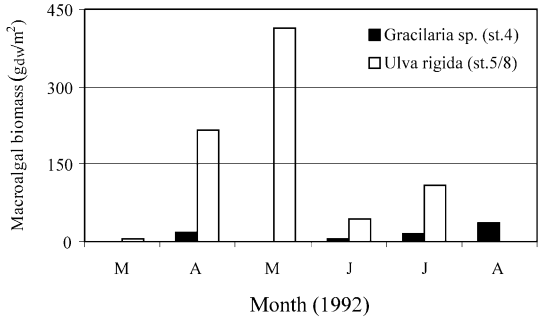

Fig. 2. Seasonal trend for macroalgal biomass [g(dw)/m^2]: *Gracilaria* sp. (st. 4) and *Ulva rigida* (st. 5/8).

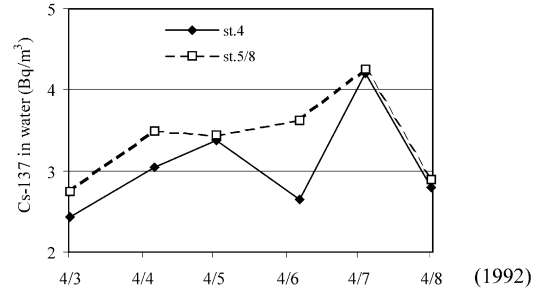

Fig. 3. Cs-137 concentration in lagoon water measured at station 4 and 5/8 (Bq/m^3).

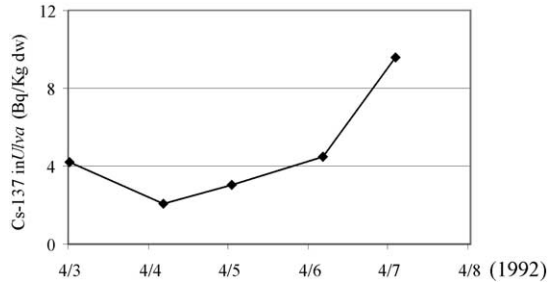

Fig. 4. Cs-137 in *Ulva* thalli collected at station 5/8 in the Sacca di Goro [Bq/kg(dw)].

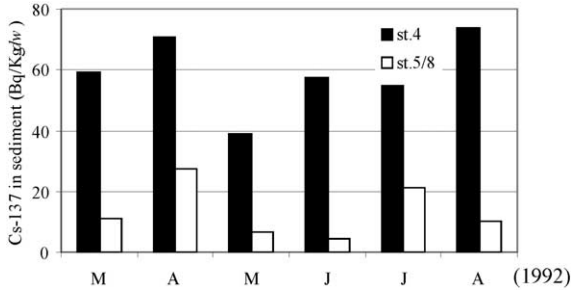

Fig. 5. Cs-137 activity in top sediment from station 4 and 5/8 [Bq/kg(dw)].

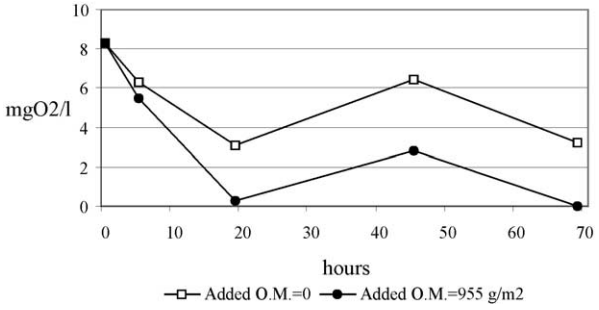

Fig. 6. Field experiment: oxygen demand measured during the experiment inside the tubes (mgO_2/l).

after a few hours of incubation, reaching 4.4 mgS/l after about 70 h, whereas no production was observed in the control tubes. The regeneration of inorganic nutrients was also higher in the treated tubes: as an example inorganic phosphorus increased from 62 to 224 μgP/l.

Table 1
Average values of Cs-137 activity detected during filed experiment in superficial sediment [Bq/kg(dw)] and water (Bq/m^3) collected outside and inside the eight dark plastic tubes

Treatments	Top layer sediment (0–5 cm) [Bq/kg(dw)]	Water (Bq/m^3)
Outside tubes	9.40	3.35
In tubes (Added O.M. = 0)	9.72	8.26
In tubes (Added O.M. = 955 g/m2)	8.63	8.59

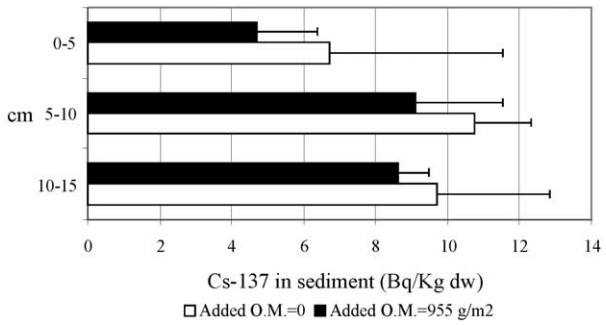

Fig. 7. Field experiment: Cs-137 activity [Bq/Kg(dw)] measured at different depth (0–5, 5–10, 10–15 cm) in sediments collected inside the tubes at the end of the experiment.

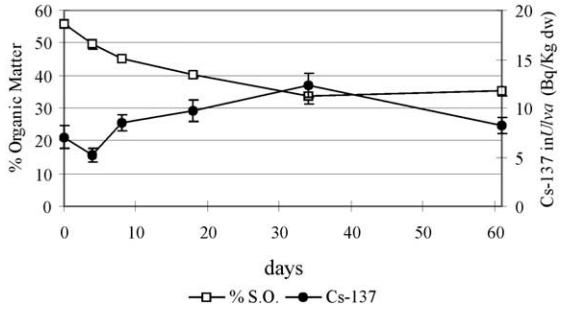

Fig. 8. Lab experiment: Cs-137 concentration [Bq/Kg(dw)] and organic matter percentage (OM%) in *Ulva* thalli, each measure is an average of four.

Differences in Cs content between samples of top layer sediment (0–5 cm) were very small (Table 1; Fig. 7).

Cs-137 activity was much higher in water samples coming from inside the tubes. Reducing conditions appeared earlier in the tubes added with organic matter but the final amount of cesium in the water was almost the same in treated and not treated tubes. This means that the effect of organic matter upon benthic metabolism of Cs-137 was quantitatively negligible. Using the data in Table 1 a daily release of Cs-137 of about 0.067 Bq/Kg(w.w.) was calculated.

Ulva biomass [average content of Cs-137 equal to 7.05 (±1.20 S.D.) Bq/kg] was employed in the laboratory to artificially reproduce dystrophic conditions, typical of the Sacca di Goro. During decomposition the percentage of organic matter decreased in the thalli, passing from 56% (±0.97 S.D.) to a minimum of 34% (±2.56 S.D.) of the dry weight after 34 days. In this period, Cs-137 concentration in the remaining thalli increased, and reached a maximum value of about 12.5 Bq/kg (Fig. 8). This trend is similar to that observed in the Sacca di Goro at station 5/8 (Fig. 4).

Cs-137 measured in the water at the beginning and the end of the experiment, provided an estimate of the release rate of 0.003 Bq kg^{-1} day^{-1} by macroalgae into the water column.

4.2. Modeling construction

Focusing on station 5/8, for which a more complete data set was available, ecological information obtained from field data and experiments was organized and used for the construction and analysis of a network model. Previous studies on nitrogen dynamics in the Sacca di Goro (Naldi et al., 1994, Christian et al., 1998) suggest two main seasonal phases related to the life cycle of *Ulva rigida* (Fig. 2): one dominated by assimilatory processes, connected to *Ulva* production, and the second in which macroalgae are decaying and decomposing.

The concentration of Cs-137 in the water did not change from March to June (Fig. 3); it increased in summer, reaching a peak in July, when macroalgal decomposition takes place. A similar trend was observed by Christian et al. (1998) for dissolved nitrogen. The beginning of the decomposition phase was than taken as a separator for two distinct periods in which processes and environmental conditions markedly differ.

We divided the data set in two parts: Phase A, from 4 March to 23 May, related to the growing season, and Phase B, from 24 May to 4 August, associated to *Ulva* decomposition. A network for each phase was constructed.

Five main components for the ecosystem were identified (Christian et al., 1998). They included two primary producers: phytoplankton (compartment 1—PHY) and *Ulva rigida* (compartment 2—ULVA); a unique compartment grouping zooplankton species, detritus and the microbial components associated to it (compartment 3—SESTON); sediment (compartment 4—SED) and dissolved Cs-137 (compartment 5—DCs) completed the network as non-living compartments. Detailed information on components included in compartment 3, SESTON, was not available, so this catchall compartment was created to represent these poorly resolved elements of the ecosystem.

Compartments 1, 3 and 5 receive cesium from the outside environment, whereas all the five state variables have been assumed to export it. A particular kind of "dissipation" (Ulanowicz, 1986) has been included in the model to take into account radionuclide decay. For each compartment quantification of this flow was done considering the half life of cesium.

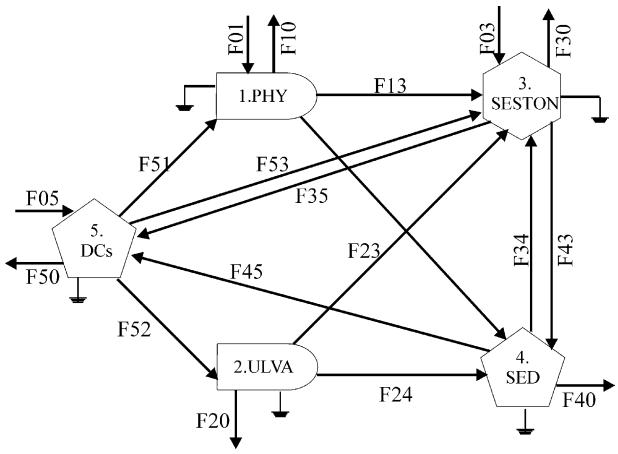

Fig. 9. Compartments and flows representing the network diagram for the Sacca di Goro lagoon.

Compartments are connected by 11 flows listed in Table 2. The network is shown in Fig. 9. Next in network construction is the estimation of standing stocks and flow values for the system in the two different phases. Direct separate determinations of Cs-137 in phytoplankton and seston were not available: these two quantities were estimated by apportioning the entire amount of cesium in particulate matter to these components, as a function of their mass concentration (details about this procedure are in the web page www.cbl.umces.edu/~bonda/ATLSS.html- Bq/m^3). Dissolved cesium was measured directly by analytical techniques (see Section 3). Multiplying these values for cesium by the average depth at station 5/8 (0.8 m) the Cs content for each compartment was converted in the required units (Bq/m^2). The amount of Cs-137 in *Ulva* was calculated multiplying cesium content in thalli by total biomass. Direct determination provided Cs-137

Table 2
List and description of network compartment interactions

Interaction	From	To	Process represented
F_{13}	Phytoplankton	Seston	Phytoplankton degradation
F_{14}	Phytoplankton	Sediment	Phytoplankton sedimentation rate
F_{23}	*Ulva*	Seston	*Ulva* degradation
F_{24}	*Ulva*	Sediment	*Ulva* sedimentation rate
F_{34}	Seston	Sediment	Seston sedimentation rate
F_{35}	Seston	Dissolved Cs-137	Seston mineralization
F_{43}	Sediment	Seston	Sediment resuspention
F_{45}	Sediment	Dissolved Cs-137	Sediment release
F_{51}	Dissolved Cs-137	Phytoplankton	Net phytoplankton uptake
F_{52}	Dissolved Cs-137	*Ulva*	Net *Ulva* uptake
F_{53}	Dissolved Cs-137	Seston	Net seston uptake

content in sediment; only the five centimeter top layer was considered it is directly involved in the circulation of the radio-nuclide (Christian, personal communication). Standing stock values in both phases are reported in Table 3.

Inputs of Cs-137 to compartments PHY, SESTON and DCs were calculated from water inflow data from Po di Goro and from the sea. Concentration values related only to the total amount of cesium transported by the Po river. To split Cs-137 concentration between dissolved and particulate fractions, we used the ratio 12:88, according to Queirazza and Martinotti (1987). For PHY and SESTON compartments inflow from the sea were obtained from tide measurements and using the concentration determined at the sampling stations. Since direct separate determinations of Cs-137 in the two compartments were not available, to apportion these two quantities we followed the calculation used for state variables.

Exports from PHY, SESTON and DCs were obtained assuming water inflow to be equal the outflow, and using the concentrations determined at station 5/8. Export from Ulva in phase A was estimated as 20% of the standing stock; the same value was used to quantify Ulva export in phase B (Viaroli, personal communication).

For each seasonal phase the amount of cesium stored in the sediment had to be considered. To examine recent sedimentation rates in the delta, Cs-137 fallout stratigraphy was used, searching for the peak of Cs-137 in cores (Appleby, 1997; Hart et al., 1998; Carroll et al., 1999; Delaune et al., 1978; Dominik et al., 1981; Fuller et al., 1999; Sharma et al., 1987). The Chernobyl accident (April-May 1986) injected a considerable amount of radionuclides into the environment. The fallout over the Po River area that followed shortly after the event, represents the most recent and consistent input of radioisotopes in this area. Accordingly, this event could be reasonably associated to the highest Cs-137 content detected in the sediment, so that the observed peak of Cs-137 profile could be dated 1986. Such peak in the cores collected from the Sacca di Goro was found at a depth of 12–15 cm (Fig. 10).

Considering this value and the time range from 1986 to 1992, the sedimentation rate resulted equal to about 2 cm year^{-1}, a results in agreement with other values

Table 3
Standing stocks (Bq/m^2) for the five compartments exchanging Cs-137 in the Sacca di Goro

Compartments	Phase A (Bq/m^2)	Phase B (Bq/m^2)
Phytoplankton	0.001	0.018
Ulva	0.540	1.570
Seston	0.043	0.650
Sediment	414	389
Dissolved Cs-137	2.210	2.01

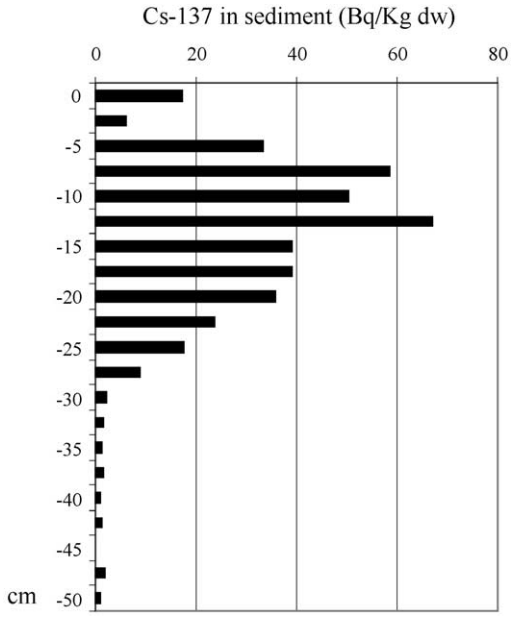

Fig. 10. Cs-137 vertical profiles in a sediment core collected in the Sacca di Goro [Bq/kg(dw)].

obtained for other lagoons of the Po river (1–3 cm year^{-1}, Albertazzi et al., 1984). This quantity appears in the model as an export, because it remains buried in the sediment and does not take part in exchanges. Export flow from the sediment was calculated using sedimentation rate, density and Cs-137 activity values.

Dissipation values were obtained from the diminution of the standing stocks of Cs-137 due to decay. To express inputs and exports in correct units (Bq m^{-2} day^{-1}—Table 4) values calculated were corrected considering the entire surface of the study area (12×10^6 m^2).

Internal exchange values for the two phases are summarized in Table 5.

The system could not be considered at steady-state because during the growth phase *Ulva* standing stock increases. It had to be assumed that the outflow from this compartment was less than the inflow; the opposite occurs during the second phase where decomposition of macroalgae prevails. Also, in both periods total input to sediment is greater than total output, suggesting a predominance of releasing processes.

Of the variety of analysis computed by the network software (Ulanowicz, 1987), we found the "Structure Analysis" and the "Biogeochemical Cycle Analysis" to be the most informative for Cs-137 cycling models.

In the Structure Analysis the TDM is calculated to evaluate the fraction of the column compartment's throughput that resided at some point in row compartments (Table 6).

Here both direct and indirect contributions are considered. In many cases the sum of the dependencies down a column cases exceeds 100%: this means that the currency is visiting more than one compartment on its

Table 4
Compartment import, export and dissipation ($Bq/m^2/day^{-1}$) from and to outside the system, for the two considered time period

Compartments	Phase A (Bq m^{-2} day^{-1})			Phase B (Bq m^{-2} d^{-1})		
	Import	Export	Dissipation	Import	Export	Dissipation
Phytoplankton	0.018	0.001	6×10^{-8}	0.027	0.002	2×10^{-6}
Ulva	–	0.004	3×10^{-5}	–	0.004	1×10^{-4}
Seston	0.181	0.010	3×10^{-6}	0.078	0.064	6×10^{-5}
Sediment	–	0.083	0.026	–	0.545	0.024
Dissolved Cs-137	0.156	0.510	1×10^{-4}	0.062	0.218	1×10^{-4}

Table 5
Compartment internal exchange ($Bq/m^2/day^{-1}$) for the two considered time periods: values, procedure and source for calculation

Interaction	Phase A		Phase B	
	Value	Procedure and sources	Value	Procedure and sources
F_{13}	0.017	Phytoplankton mass balance	0.157	Phytoplankton mass balance
F_{14}	0.001	Standing stock per day (Billen and Lancelot, 1988);	0.018	Standing stock per day (Billen and Lancelot, 1988)
F_{23}	0.004	20% of *Ulva* standing stock	0.024	Total loss of *Ulva* biomass ($F_{20}+F_{23}+F_{24}$) minus the export F_{20} divided by 2 because $F_{23}=F_{24}$
F_{24}	0.004	See F_{23}	0.021	See F_{23}
F_{34}	0.039	See F_{14}	0.558	See F_{14}
F_{35}	0.158	Seston lost in one day (total community respiration) times average cs-137 concentration in seston	0.030	Seston lost in one day (total community respiration) times average cs-137 concentration in seston
F_{43}	0.004	Patel et al. (1978)	0.183	Direct estimate (field experiment)
F_{45}	0.228	See F_{43}	0.499	See F_{43}
F_{51}	0.001	Minimum estimate	0.150	Net phytoplankton production times average Cs-137 concentration in phytoplankton
F_{52}	0.030	Sum of growth (exponential model) and losses ($F_{20}+F_{23}+F_{24}$)	0.013	
F_{53}	0.001	Compartment 5 mass balance	0.210	Compartment 5 mass balance

Table 6
Total dependency matrices for phase A and phase B: coefficients represent the fraction of a compartment throughput (column designation) that resided at some point in another compartment (row designation), over all pathways, both direct and indirect

	Phase A					Phase B				
	PHY	ULVA	SES	SED	DCs	PHY	ULVA	SES	SED	DCs
PHY	0.16	2.96	8.31	1.28	2.96	13.40	4.18	34.00	16.70	15.80
ULVA	0.06	1.16	1.97	1.40	1.16	3.99	1.25	7.60	5.13	4.71
SES	1.8	34.10	1.20	11.80	34.10	36.70	11.50	35.90	45.30	43.30
SED	2.26	43.00	3.16	0.87	43.00	74.80	23.40	75.30	35.10	88.20
DCs	5.26	100.00	2.88	1.52	1.48	84.70	26.50	61.80	29.20	27.80

way through the network. As an example of how indirect contributions increase dependency over 100%, consider *Ulva* in phase A: it receives material directly only from DCs (F_{52}), hence it completely depends on DCs (100%). Nevertheless *Ulva* depends indirectly for a 43% from sediment (Table 6). Because dissolved cesium components receives material from sediment (F_{45}) part of the output from DCs to *Ulva* (F_{52}) can be traced back to sediment; that is, some of the material that was present in the sediment first spent time like dissolved Cs-137 and then passed on to *Ulva*. Considering all (direct and indirect) pathways that reach *Ulva*, one discovers that its dependency coefficients sum up to 181%.

Both the primary producers included in the model get cesium mainly from DCs and sediment, but they show considerable dependency also on seston (37% for phytoplankton in phase B). On the other hand, seston in phase A depends mainly upon phytoplankton with a percentage equal to 8%; for phase B seston, like the primary produces, depends mainly on DCs and sediment.

The latter compartment relies mostly on seston (12% in phase A and 45% in phase B) but also by a significant amount on sediment and dissolved cesium. The other non living compartment, dissolved cesium, depends largely on seston but primarily on sediment (43% in phase A and 88% in phase B).

The "input analysis" computes how a input unit is distributed among internal flows; also it describes how such flows enter in the production of an export units. Results of this analysis are described in Fig. 11, where only flows accounting for more than 1% of each input are represented.

First input to phytoplankton is considered. In phase A this amount goes mainly to seston (90%) and then to DCs compartment (69%) from where it leaves the system (Fig. 11a). In phase B this input follows a different main path: after reaching seston it goes mostly to sediment (124%) and then to DCs (54%) (Fig. 11d).

Input analysis of seston compartment shows that in phase A most of the cesium travel to dissolved cesium (77%) and than exit the system (Fig. 11b): in phase B, it reaches sediment (134%) before leaving the system from DCs (Fig. 11e).

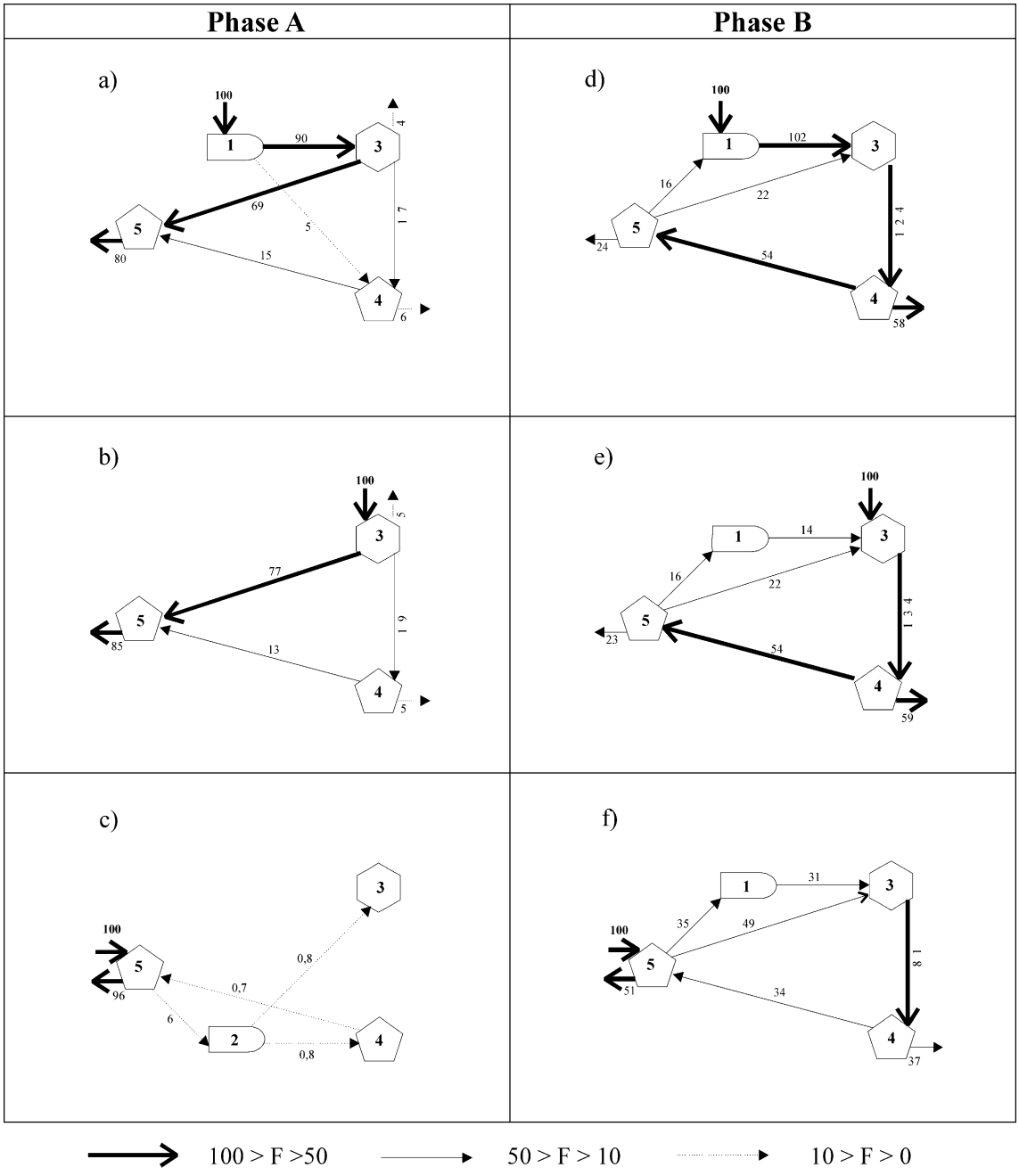

Fig. 11. Input analysis results for the Sacca di Goro network (only flows accounting for more than 1% of each input are represented).

Most of the radionuclide entering as dissolved cesium in phase A leaves the system from the same compartment (96%) without significant contributions to the internal flows (Fig. 11c). In phase B the contribution is more consistent and the main flow is the one connecting seston to sediment (81%) (Fig. 11f).

In the biogeochemical cycle analysis cyclical paths are identified and the Finn Cycling Index (FCI) calculated. This index indicates the percentage of Cs-137 which is recycled in the system: its value is higher in phase B (53%) than in phase A (3%).

5. Discussion

Field data together with seasonal trends of the monitored variables provide a preliminary picture of the contamination pattern in the lagoon. Cesium concentration in the water column increases passing from growing to decomposing period (see Fig. 3). This could be due either to an augmented release from sediment or by macroalgae, the two matrices in which this element is mainly stored.

As the season approaches the decomposition period, *Ulva* biomass decreases but Cs-137 in macroalgae reaches its maximum value (see Fig. 4). During these processes plants release cesium (Bondavalli et al., 1996) but thalli that are still in good conditions tend to accumulate this element. This it has been confirmed also by the lab experiment, which provided clues about the role of macroalgae in cesium cycling. The concentration in *Ulva* thalli increased during the experiment, as shown in Fig. 8, but the net release of the isotope to the water column of laboratory tanks was only 0.003 Bq kg^{-1} day^{-1}. The result of the experiment suggests that during phase B release from plants is not as important as accumulation, and the bi-directional process is unbalanced in favor of the intake. Because only water and *Ulva* were in the tanks, one may conjecture that the peak of cesium in water is not due to release form magroalgae Likely, cesium released by decomposing macroalgae is passed directly to the microbial community living on them. A release in the water and successive accumulation in the remaining thalli seems an unlikely phenomena: these thalli are progressively decomposing and their ability to accumulate or concentrate chemical species is diminishing. On the other hand the ability of microorganisms to accumulate cesium, mobilize and recycle it has been well documented (Avery, 1995, 1996).

Because sediment is an important deposit and sink for Cs-137 (Avery, 1996; Smith et al., 2000b), the field experiment carried out in the Sacca di Goro at station 5/8 was conceived to assess its role in Cesium release. Research shown that macroalgal degradation processes altered the chemical equilibrium at the sediment-water interface. In particular, cesium was mobilized at an estimated daily rate of about 0.067 Bq/Kg(w.w.). These results confirm what observed in other studies (Delaune et al., 1978; Robbins and Edgington, 1975). In freshwater lakes, for example, periods of intensive Cs-137 recycling coincided with the occurrence of reducing conditions and anoxia (Dominik and Span, 1992; Hilton et al., 1994).

These experiments led to conclude that the peak of cesium observed in the water column during summer was likely due to mobilization from sediment, rather then from direct release by decomposing macroalgae. However an overall picture of cesium circulation in the Sacca di Goro comes from the network analysis which indicates compartments and flows that are mostly responsible for cycling activity in the lagoon.

For both seasons the total dependency matrices (TDM, Table 6) show that SESTON, SED and DCs are the main suppliers of cesium to the rest of the system (highest dependency coefficients). In particular, DCs is the main source of the isotope to *Ulva* and phytoplankton because it is the only component from which primary producers receive medium. Thus the connections from DCs to plants are the channels through which all the contribution to primary producers travels.

Also seston is an important source of cesium for the system, especially for sediment and dissolved cesium. In phase B the indirect cointribution to seston by DCs ($F_{52}\rightarrow F_{23}$; $F_{52}\rightarrow F_{24}\rightarrow F_{43}$; $F_{51}\rightarrow F_{13}$; $F_{51}\rightarrow F_{14}\rightarrow F_{43}$) is much higher than the direct flow (F_{53}): 38% vs. 5%. Primary producers depend indirectly on seston as well. For phytoplankton the dependency in phase B reaches 37%, whereas *Ulva* shows higher dependency in phase A: 34% of the Cs-137 entering this compartment once resided in seston.

High dependency coefficients characterize also sediment which is the second most important source of radionuclide to primary producers, although it establishes no direct connections with them. The indirect pathway is due to the presence of dissolved cesium which is taken up by plants. As for se ston, phytoplankton and *Ulva* show the higher dependency on sediment differently in the two seasons: macroalgae in phase A (43%) and phytoplankton (75%) in phase B. Sediment provides medium to seston and DCs, especially in phase B, when 88% of Cs_137 that passes through the water column once resided in the sediment. For seston the percentage is 75.

The remaining two compartments, phytoplankton and *Ulva*, showed a different behavior. Overall, the system slightly depends on cesium stored in primary producers. In particular, exchanges involving macroalgae show very small coefficients. Therefore *Ulva*'s contribution to the rest of the system is very low. This applies also to internal recycling (total dependency matrix diagonal), for which *Ulva* shows the lowest value. Phytoplankton shares

the same behavior as *Ulva*, but in phase B it contributes to internal recycling by 13.4%, while seston depends on it for a considerable amount of medium. In conclusion SESTON, DCs and SEDIMENT appear to be mostly responsible for cesium dynamics in the system; in particular the latter component play a pivotal role in cesium circulation. More details about this are provided by the input analysis.

Input analysis reveals that in phase A the main pathway through which cesium reaches phytoplankton involves only two compartments, namely seston and DCs. Cesium entering the system through seston for the most part flows into DCs and then leaves the system. Finally most of the cesium entering the lagoon in a dissolved form leaves the system directly and only a small fraction participates in internal flows. The larger part of this amount reaches *Ulva*, but it is only a small percentage of the total inflow (Fig. 11).

In phase B internal circulation increases: the number of intercompartmental exchanges created by each input augments as much as the magnitude of flows. For example, in comparison with phase A, the main quantity of cesium that reaches phytoplankton passes though seston, SED, DCs (Fig. 11d), whereas seston is fed by a main pathway that involves SED and DCs (Fig. 11e). Input to DCs compartment instead of exiting straight away the system, reaches the other compartments, except for *Ulva*, and flows are of significant magnitude (Fig. 11f). Overall for all three inputs a noticeable contribution to internal recycling is given by the path that comprises seston, sediment and dissolved cesium ($F_{34} \rightarrow F_{45} \rightarrow F_{53}$; Fig. 11).

This picture is confirmed by the Flin cycling index. Its value is 3% in phase A, but goes up to 53% in phase B, in accordance with other cases in which an increased circulation of cesium during the decomposition phase was observed (Christian et al., 1998).

Network analysis results suggest that pattern of cesium release from the sediment completely change on an ecosystem scale between the two phases. According to the "Input Analysis", a large amount of cesium is released into the system and circulate during phase B; on the contrary in phase A it remains trapped in the sediment. During phase B large quantities of this radionuclide enter the water column (F_{45}) and from there it can be taken by primary producers and seston. This creates a contamination pathway that can propagate the radionuclide to the open sea and other ecological systems, with the potential to reach higher trophic levels, including man. This path is much more intense in the mid summer phase when seston and phytoplankton show the highest dependence on sediment.

The overall conclusion of this study is that, at present, cesium dynamics in the Sacca di Goro has been changed with respect to the recent past. Radionuclides in general are removed from the water column by physical and biological activity (Fowler et al., 1987) and transferred to the sediment. The high sedimentation rate calculated for the Sacca di Goro suggests that most of the cesium that entered the lagoon were stored in the sediment, with a higher accumulation capacity shown by clay sediment at station 4 with respect to sandy particles that predominate in the sediment at station 5/8 (Avery, 1996; Bergeijk et al., 1992). By acting as a trap for radionuclides, sediment was able to accumulate much of the radioactivity that reached the lagoon ecosystem (especially after the Chernobyl accident). The repeated dystrophic crises have reversed this trend and sediment behaves now more as a source of Cs-137 than a sink. It follows that the effect of future contamination events will be further amplified by this greater circulation, with implication at the whole ecosystem scale.

On the other side, Cs-137 stored in *Ulva* was not directly released to the water column. The total dependency matrix provides further insights to support this view: transfers in and out are always very low, as a consequence system dependence on *Ulva* is generally very small and so is its internal recycle (TDM diagonal, Table 6). All these evidences suggest a slow turnover rate for the cesium stored in this compartment. It must be kept in mind, however, that in this network the *Ulva* compartment comprises macroalagae and their associated microbial community: a complete picture of *Ulva* contribution to cesium circulation the system needs that the role of microbes in cesium cycling and the interplay between these organisms and the macrooalgae be clarified. In fact the microbial community living on algae may play an important role in cesium accumulation and release, especially during decomposition, when the activity of microbes greatly increases (Avery, 1995, 1996). Also previous studies indicate the ability of dead algal cells in retaining cesium suggesting the persistency of cellular structures where cesium have been stored (Davis, 1961; Gutknecht, 1965).

This study focuses upon lower levels of the trophic chain and detritus; higher trophic level organisms such as benthic fauna and fish have not been explicitly considered because of the lack of information and data. To my knowledge, no other work of this kind has been conducted, but a more complete model for an area of the lagoon of Venice, similar to the Sacca di Goro, has been constructed and analysed by Carrer and Optiz (1999), using energy as a currency. Results of this model showed that a large transfer of matter and energy occurred at the lower levels, concentrated in a few flows. Furthermore, in the lagoon of Venice energy is barely passed up to the trophic web: a large amount of it is recycled to the detritus pool directly from the first and the second trophic levels, underlying the importance microorganisms. The two systems, which are very similar, share the same behavior as for trophic dynamics and this suggest that the network for the Sacca di Goro, although rather simple, is ecologically reasonable.

Although it seems that in these lagoons upper trophic levels do not to play a key role in cesium circulation, by network analysis contamination pathways for economically important species or species that are rare or play a pivotal role in the functioning of this ecosystem could be identified. As a consequence of this effort one could forecast economic loss, potential impact on human health or extinction risk due to contamination events. To this end, however, it is necessary that the network is enlarged to include all the variables (species) of interest. In turn this implies that more ecological information on these species are collected.

6. Conclusions

Understanding the mechanisms by which radionuclides are cycled is essential for predicting their distribution and concentration in the various components of the ecosystem. Because of the complex interactions that characterize estuarine environments, such understanding requires that detailed monitoring study, laboratory experiments, and modeling effort be combined.

Previous study on the Sacca di Goro investigated only the presence and distribution of radionuclides (Mezzadri and Parisi, 1991; Nonnis Marzano and Triulzi, 1994, 2000) but, to date, this environmental database has never been used to identify and quantify processes controlling the distribution and rates of transfer of Cs-137. In this paper I tried to contribute in this direction by investigating the relationship between radinuclide circulation and eutrophication, a common condition in this environment. The experiments conducted both in laboratory and in the field have suggested that decomposition of macroalage biomass, by altering chemical conditions at the water-sediment interface, increase cesium mobility. As a consequence higher amounts of this chemical species become free to disperse and circulate in the lagoon. Also, by organizing present ecological knowledge of the ecosystem of the Sacca di Goro in a network of flows, hypotheses concerning the fate of cesium once it is released in the water column have taken shape. Further research is needed to clarify patterns of circulation of contaminants in this estuarine ecosystem, especially in the framework of multiple contamination events. In general this seems a promising approach that can provide clues for the understanding and, eventually, the management of the relevant presence of radioactive elements that may occur because of human activities.

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