

# Radiometric dating and pollutants profiles in a sediment core from the Lagoon of Venice

Datation radiométrique  
Polluants  
Sédiments  
Lagune de Venise

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## ABSTRACT

The  $^{210}\text{Pb}$  radioactive dating technique on two cores from the northern part of the Lagoon of Venice give an average sedimentation rate of 0.7 cm/year. This is used to date the vertical profiles of  $\Sigma$  DDT, Hg, Cd, Pb, Cr, Cu, Ni, Co, Zn and Fe concentrations. A maximum  $\Sigma$  DDT concentration is found at a depth of 10 cm, corresponding to about 1955, in accordance with the time of the largest use of this pesticide in the area drained by the Dese river. Due to antropogenic origin the concentrations of heavy metals in the upper sediment layers are 1.3-12.7 times higher than those in the lower ones. The average concentrations of these elements in the layers below 80 cm are tentatively proposed as a natural background concentrations for the Lagoon of Venice.

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## RÉSUMÉ

Datation radiométrique et profils de polluants dans une carotte de la lagune de Venise

La technique de radiodatation au  $^{210}\text{Pb}$  utilisée sur deux carottes provenant du nord de la lagune de Venise a donné une vitesse de sédimentation moyenne de 0,7 cm/année. A partir de ces résultats, on a daté les profils des concentrations de  $\Sigma$  DDTs, Hg, Cd, Pb, Cr, Cu, Ni, Co, Zn et Fe. La concentration de  $\Sigma$  DDT la plus élevée a été trouvée à une profondeur de 10 cm, qui correspond approximativement à l'année 1955, en accord avec la période d'usage maximal de cet insecticide dans la région arrosée par le fleuve Dese. A cause de leur origine anthropogénique, les strates les plus hautes sont 1,3 à 12,7 fois plus concentrées en métaux lourds que les strates les plus profondes. Les concentrations moyennes de ces éléments dans les sédiments au-dessous de 80 cm sont proposées comme valeurs de base pour la lagune de Venise.

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## INTRODUCTION

Historical records of environmental pollution and pollutant fluxes may be obtained from sediment cores by establishing a time scale against which to plot the pollutant concentrations. For this purpose radioactive dating techniques have been shown very useful and reliable especially when applied to unwarped and unperturbed sediments (Koide *et al.*, 1973).

The importance of sediments as pollution sink or source, depending mainly on the redox conditions, and the scarcity

of informations prompted us to undertake the study of the Lagoon of Venice. In this paper some preliminary results on the vertical distribution of heavy metals and DDTs in a sediment core from the northern part of the lagoon are reported.

During centuries the entire area (Fig. 1) has undergone several intended human modifications to prevent its filling up with solid materials transported by rivers entering the lagoon. At present, the lagoon still receives domestic sewages, mainly from the town of Venice, agricultural drainage through 20 different immission points and a variety of wastes from the industrial zone of Marghera.

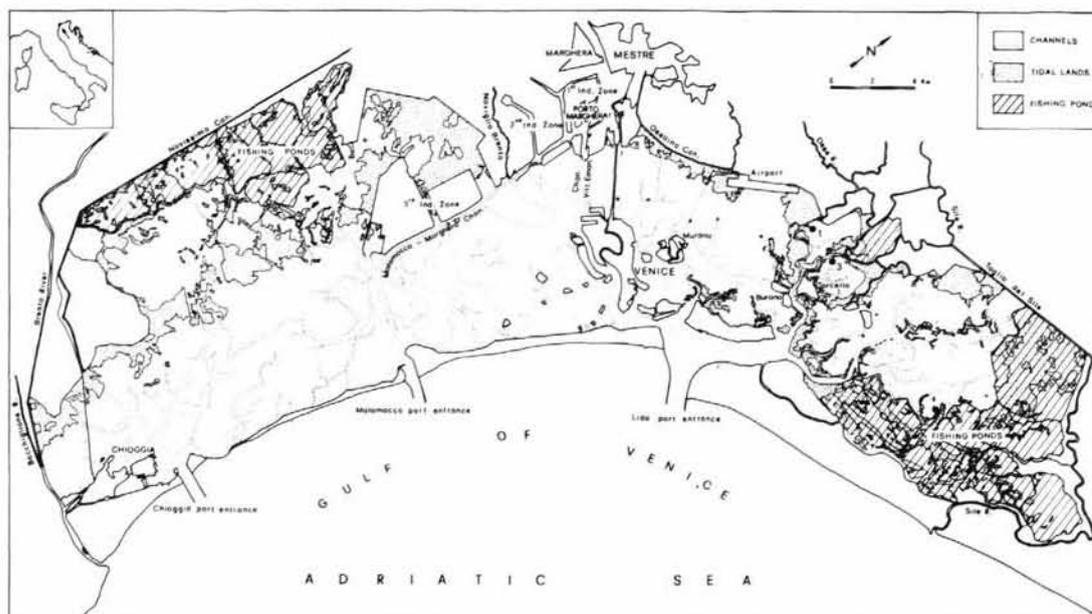


Figure 1  
The Lagoon of Venice and the sampling sites (●).

#### SAMPLING SITE AND ANALYTICAL PROCEDURES

The Venice Lagoon has an area of about 500 km<sup>2</sup>, an average water depth of ~ 0.6 m and a spring tidal excursion of 1 m (Cavaleri, 1980). Due to currents, in some parts the lagoon bed undergoes erosion, whereas in other parts a positive rate of sedimentation is observed. Furthermore, human activities such as fishing or boating are certainly responsible for sediment perturbation in many areas. Since the recovery of undisturbed sediments is very important for the development of the pollution record and the introduction of an accurate time parameter, we chose an area where positive sedimentation could be expected (Cavazzoni, 1977). The cores 1 and 3 were taken in September 1979 near the mouth of the Dese river which is the main fresh water supplier of the lagoon with a maximum water flow of 650 m<sup>3</sup>/sec. (Cavazzoni, 1973). The cores were collected at 45° 30'8" N and 12°25' E in a point where the water depth was 0.5 m, using a 12 cm i.d. open vented plastic corer, which was sealed at the bottom before extraction from the sediment and dry ice-methanol frozen on the boat. Frozen cores were immediately transported to the laboratory, sectioned and dried in Al containers at room temperature after scraping away about 1 cm of the outer surface which was in contact with the plastic walls of the corer. The dry sediment samples were then homogenized in agate mortar and stored in airtight glass jars.

The chemicals and solvents used for analyses were reagent grade or pesticide grade from Baker.

Sediment samples to be analyzed for heavy metals were digested following methods and procedures previously reported (Agemian, Chau, 1975; 1976), and analyzed using a Perkin Elmer Model 5000 atomic absorption spectrophotometer. Instrument settings were as recommended in the instruction manual (Perkin Elmer Corp., 1979) and the background correction was used in all determinations.

The DDTs analyses were performed with the following method. All laboratory glassware for these analyses were treated for few hours with cleaning solution (K<sub>2</sub>Cr<sub>2</sub>O<sub>7</sub> in H<sub>2</sub>SO<sub>4</sub>), rinsed with tap water, distilled water, acetone and finally dried in an oven at 220 °C for three hours. The Soxhlet apparatus for the extraction of pesticides was cleaned by refluxing 200 ml of hexane for 3 hours. A second portion of hexane (200 ml) was refluxed and concentrated to dryness in the round bottomed flask with a rotatory evaporator. The flask walls were then carefully washed with 1 ml of isooctane and this solution was kept for blank control.

Dry sediment samples (30 g) were extracted in the Soxhlet apparatus with 200 ml of hexane for 24 hours. To the resulting solution, concentrated to 5-7 ml, 1.5 ml of 96 % H<sub>2</sub>SO<sub>4</sub> were added and the mixture was shaken for 1 hour. The overlying hexane solution was passed through a column (internal diameter 1 cm, length 20 cm) filled with activated copper (1 cm height), 130 °C activated fluorisil (3 cm) and dry Na<sub>2</sub>SO<sub>4</sub> (1 cm), eluted with 60 ml of hexane and concentrated to about 2 ml. This volume was passed through a similar column containing 8 g of prewashed silica gel activated at 220 °C and deactivated with 1 % H<sub>2</sub>O, recovering the 50-130 ml fraction in benzene containing pesticides. Upon addition of heptachloroepoxide as external standard, the solvent was evaporated by a rotatory evaporator and the residue dissolved in 1 ml of isooctane. The final solution was injected in a Hewlett-Packard Model 5840 A gas-chromatograph. A glass column, 6 ft × 4 mm i.d., packed with 1.5 % SP 2250/1.95 % SP 2401 on 100/120 supelcoport and a flow rate of 55 ml/min of argon-methane 95/5 % were used. The injection, column and <sup>63</sup>NiEC detector temperatures were: 250 °C, 200 °C and 300 °C respectively.

Core layers were dated by the <sup>210</sup>Pb technique (Koide, Bruland, 1975). The <sup>210</sup>Pb activity was determined from the activity of the daughter <sup>210</sup>Pb or <sup>210</sup>Pb at the Scripps Institution of Oceanography, La Jolla, California.

#### RESULTS AND DISCUSSION

The total <sup>210</sup>Pb concentrations in the two cores of the Lagoon of Venice (Table 1) are very similar and decrease exponentially with the depth as expected. While core 1 exhibits a maximum <sup>210</sup>Pb activity at the sediment water interface, core 3 has a maximum value at the depth of ~ 5 cm. A decrease or a flattening of <sup>210</sup>Pb activity near the surface of the sediment have been previously reported (Koide *et al.*, 1973; Christensen, Chien, 1981) and attributed to physical or biological mixing of the upper layers associated with anaerobic conditions. Because of the shallow water underlying the sediments in this part of the Lagoon of Venice one would expect high redox potentials at the sediment surface. This is not always verified and dark brown materials and hydrogen sulfide smell were observed at the sediment surface of core 3 during coring operations, indicating a low redox potential. Furthermore, field measurements have shown that in this area anoxic surface

Table 1  
Pb-210 activity as a function of depth in cores N° 1 and N° 3.

Core N° 1		Core N° 3	
Depth (cm)	Pb-210 (dpm/g)	Depth (cm)	Pb-210 (dpm/g)
1.15-1.95	3.34 ± .1	0-3	2.75 ± .1
4.6-5.6	3.15 ± .1	7-10	2.92 ± .1
10.0-11.0	2.90 ± .1	14-17	2.37 ± .1
18.0-19.0	2.67 ± .1	21-24	2.05 ± .1
24.8-25.8	2.26 ± .1	35-38	1.66 ± .1
31.6-32.6	2.41 ± .1	49-52	1.27 ± .1
38.3-39.3	1.90 ± .1	63-66	1.36 ± .07
45.1-46.1	1.63 ± .1	73-76	1.28 ± .04
		84-87	1.26 ± .07
		98-101	1.39 ± .06
		105-108	1.37 ± .05
		108.5-111.5	1.21 ± .08

is simply due to the fact that core 1 was only 45 cm long. The deeper sections of core 3 indicate a constant <sup>210</sup>Pb activity due to the <sup>210</sup>Pb supported by <sup>226</sup>Ra and averaging around 1.31 dpm/g. The lower lines of Figures 2 and 3 have been drawn by subtracting this value from the measured <sup>210</sup>Pb activities. Utilizing the 22.26 yr half-life of <sup>210</sup>Pb average sedimentation rates of 0.81 cm/yr and 0.59 cm/yr are obtained for cores 1 and 3, respectively. Therefore, it seems reasonable to assign to the sediments of this lagoon area an average sedimentation rate of 0.7 cm/yr which can be used to date the profiles shown in Figures 4 and 5. This is in good agreement with the increase of the sediments level in the range 25-50 cm observed by comparing the lagoon bathymetric maps published by the Magistrato delle Acque of Venice in 1934 and 1971. Of course, the solid materials

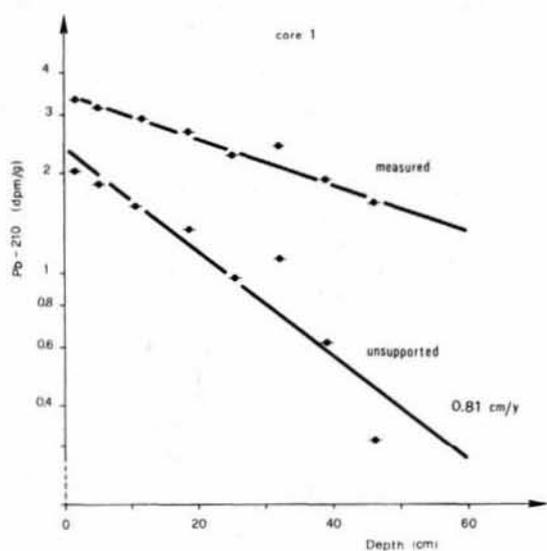


Figure 2  
<sup>210</sup>Pb activity vs depth of the core N° 1.

sediments (Eh values in the range 50 ÷ - 150 mV) are very often in contact with well oxygenated water (Eh values 100 ÷ 350 mV ; D.O. values 3-6.5 mg/l). The plot of the total <sup>210</sup>Pb activity against the depth of the core 1 shows a pure exponential form (Fig. 2). However, from the same plot of core 3 (Fig. 3) it can be seen that this

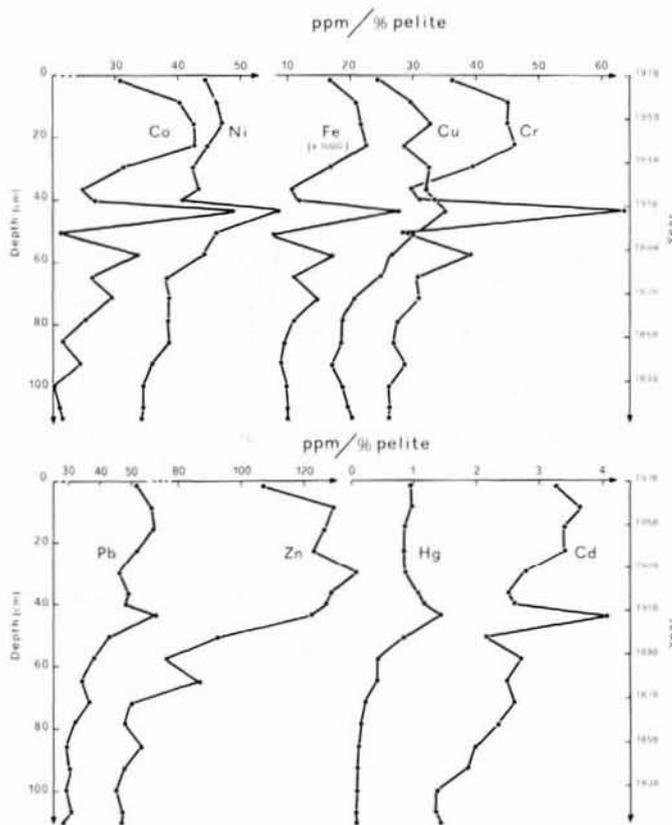


Figure 4  
Heavy metals profiles in core N° 3. The corrected values here reported are obtained from Table 3 using the relationship : corrected values = heavy metal concentration × % pelite.

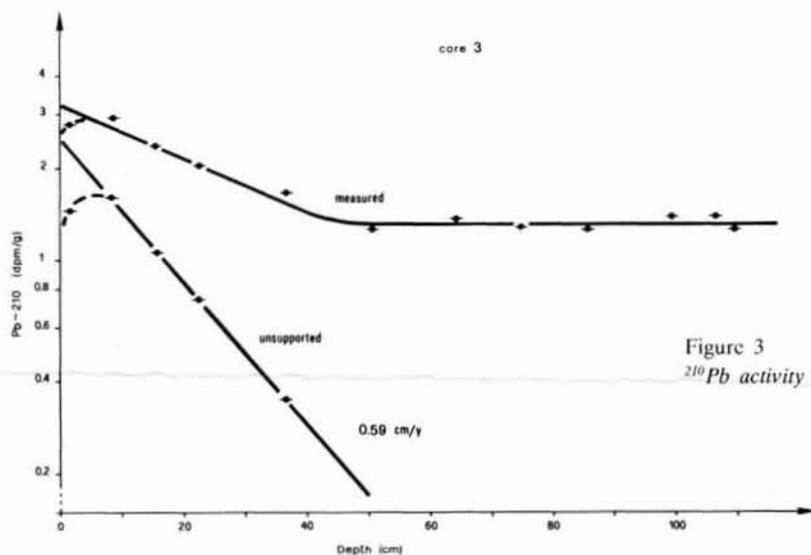


Figure 3  
<sup>210</sup>Pb activity vs depth of the core N° 3.

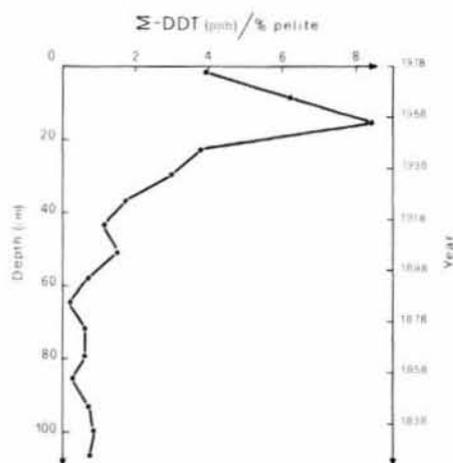


Figure 5  
 $\Sigma$ -DDT profile in core N° 3. The corrected values here reported are obtained from Table 3 using the relationship: corrected values = heavy metal concentration  $\times$  % pelite.

transported by the Dese river are mainly responsible for the accumulation of sediments in this part of the lagoon.

The concentrations of a set of metals in core 3 and the percentage of pelitic fraction ( $> 63 \mu\text{m}$ ) are given in Table 2. A decrease of concentration from maximum values at or near the surface to constant values at a depth of about 80 cm is shown by all profiles. This is a general trend characteristic of polluted areas (Chow *et al.*, 1973).

The decrease of concentration of all elements at the surface, also observed by  $^{210}\text{Pb}$  (Fig. 3), can be caused by sediment mixing. Since the heavy metals are known to be associated with the sediments finer fraction the ratios between the amount of element and the percentage of pelite has been reported against the depth in Figure 4. Quite surprisingly, for all elements except Zn, a well pronounced maximum is present at a depth of 42 cm or about 1916. One can observe that this correspond to the period of time (1916-1917) in which the first world war operations were most intense in

this area with the fighting line on the Piave river, about 20 km north of the coring point. The fallout of polluted aerosol due to war activities can be responsible of these elevated values. On the other hand, this observation further supports the  $^{210}\text{Pb}$  dating.

The average concentrations of heavy elements in the strata below 80 cm can be assumed as natural background values for this area of the lagoon of Venice. Except for Cr, Ni and Hg they are in a reasonable agreement (Table 3) with the average values found in surface sediments from the Adriatic Sea (Selli *et al.*, 1977) and except for Fe, with those assumed as maxima for non polluted sediments (Prater, Anderson, 1977). The textural character and the mineralogical composition of the sediments are not identical in the entire lagoon (Barillari, 1978; Barillari, Rosso, 1975; Hieke Merlin *et al.*, 1979). However, most of them have more than 75 % of pelitic fraction ( $< 63 \mu$ ) as have cores 1 and 3, and to this fraction are mainly associated the heavy metals (Gibbs, 1977). On this basis the heavy metal concentrations of the deeper layers of core 3 can be tentatively assumed as background values for the Lagoon of Venice. This is of particular interest if a precise evaluation of the extent of pollution due to toxic elements in the lagoon sediments is needed.

Enrichments on top layers of all metals are evident from Figure 4. The ratios between the surface concentration and that of the layers below 80 cm are: Hg, 12.7; Cd, 2.4; Pb, 1.8; Co, 1.9; Ni, 1.3; Cu, 1.5; Zn, 2.0; Fe, 2.0; Cr, 1.6. The higher ratio for Hg can be understood taking into account that the Dese river drains the water of about 300 km<sup>2</sup> of heavily cultivated land where pesticides containing Hg have been widely used in the past.

A clear distinction between the metals, e.g. Co, Fe, Ni, generally associated with soil and rocks debris, and those, e.g. Pb, Cu, Zn, etc., frequently present in aerosol originating from fuel or coal combustion and from steel industries is reported in literature (Christensen, Chien, 1981; Dexter, Pavlou, 1973; Bruland *et al.*, 1974; Goldberg *et al.*, 1976). Such a distinction is based on the fact that the metals belonging to the first group show constant vertical profiles in the sediments, whereas the concentrations of the others decrease with the depth. In our case such a distinction does not seem to be applicable since all examined metals, except

Table 2  
 Heavy metal (ppm),  $\Sigma$ -DDT (ppb) concentrations and pelite (%) in sediments on dry weight basis.

Depth (cm)	Pelite	Hg	Cd	Pb	Co	Ni	Cu	Zn	Fe	Cr	$\Sigma$ -DDT*
0-3	78.6	0.72	2.57	40.8	24.2	34.8	21.0	84.3	13 200	28.6	3.08
7-10	78.7	0.73	2.85	45.6	31.6	36.5	23.2	102.0	16 500	35.7	4.84
14-17	73.9	0.59	2.50	42.9	31.5	34.8	24.2	93.1	15 800	33.3	6.28
21-24	84.5	0.66	2.87	43.1	36.4	38.1	24.3	104.2	19 200	39.3	3.20
28-31	77.9	0.68	2.15	35.8	24.2	33.1	25.4	106.5	13 300	30.9	2.32
35-38	72.5	0.77	1.80	35.4	17.9	31.5	23.2	93.1	7 700	21.4	1.26
38.5-41.5	73.5	0.85	1.90	35.1	19.5	29.8	24.3	93.6	8 700	22.6	
42-45	70.5	0.96	2.87	40.6	34.4	39.7	24.9	86.5	19 700	45.2	0.77
49-52	72.0	0.60	1.53	31.0	15.0	33.1	22.1	66.5	5 400	20.2	1.06
56-59	79.5	0.34	2.15	30.8	26.7	34.8	21.0	59.9	13 800	30.9	0.56
63-66	74.0	0.31	1.84	25.2	19.5	28.1	20.1	64.3	8 100	22.6	0.08
70-73	81.5	0.18	2.12	29.8	24.2	31.5	16.5	53.2	12 100	25.0	0.48
77-80	77.8	0.12	1.79	24.9	19.5	29.8	14.3	58.8	8 400	21.2	0.50
84-87	76.9	0.09	1.50	22.8	16.5	29.8	14.2	52.3	7 100	20.5	0.19
91-94	78.3	0.06	1.45	23.7	19.1	28.9	13.2	48.8	6 900	22.3	0.64
98-101	78.6	0.06	1.06	22.7	14.9	26.9	14.6	47.1	7 700	19.8	0.68
105-108	78.5	0.05	1.05	24.3	16.1	26.9	15.5	48.7	7 800	20.6	0.57
108.5-111.5	78.4	0.05	1.10	21.8	16.6	26.5	15.6	48.0	7 700	20.3	

\* $\Sigma$ DDT = sum of p,p' DDT: 1,1,1-trichloro-2,2-bis(p-chlorophenyl)ethane  
 o,p' DDT: 1,1,1-trichloro-2-(o-chlorophenyl)-2-(p-chlorophenyl)ethane  
 p,p' DDD: 2,2-bis(p-chlorophenyl)-1,1-dichloroethane  
 o,p' DDD: 2-(o-chlorophenyl)-2-(p-chlorophenyl)-1,1-dichloroethane  
 p,p' DDE: 1,1-dichloro-2,2-bis(p-chlorophenyl)ethylene

Table 3

Natural background values of some heavy metals in sediments of different areas.

Element	Lagoon of Venice	Middle Adriatic Sea (1a)	Non polluted sediments (1b)
Hg	0.1	0.30	—
Cd	1.2	0.71	—
Pb	23	32.0	< 40
Co	17	12.3	—
Ni	20	49.8	< 20
Cu	15	26.1	< 25
Zn	49	88.7	< 90
Fe	7500		< 17000
Cr	21	44.8	< 25

a) From Selli et al., 1977; b) from Prater, Anderson, 1977.

Hg, show a similar trend, i.e. due to anthropogenic contributions, they show higher concentrations in the upper layers than in the deeper ones. The enrichments are within a narrow range (1.3-2.4) and anthropogenic fluxes (Table 4) are of the same order of the natural ones. This leads to the assumption that, since all these elements are utilized and therefore mobilized in the Marghera industrial zone (~ 20 km SW of the coring point), from this area can originate the polluted aerosol falling directly into the sediments or transported by water runoff from the adjoining land. Industrial wastes during the past decades have been discharged into the lagoon mainly in the areas close to industries. Therefore, the possibility for the heavy metals to be transported to the northern part of the lagoon by water currents originated by tides can not be completely disregarded.

The total fluxes, calculated assuming the weight of 1.08 g of dry sediment per cm<sup>3</sup> of deposit (Table 4) are usually one order of magnitude higher than those of the California Basins (Bruland et al., 1974), but much lower than those

observed in more polluted areas such as the Palace Moat in Tokio (Goldberg et al., 1976) or the Narrangasett Bay (Goldberg et al., 1977). This may not be surprising if one considers that the preferential direction of the pollutant diffusion plumes recorded in the lagoon sediments is from Marghera toward the Lido Port Entrance, i.e. far from the point in which the cores were collected.

The  $\Sigma$ -DDT profile (Fig. 5) shows a maximum of 6.28 ppb at the depth of 16 cm or about 1955 and decreases toward the sediment surface due to the lower use of this type of pesticide in recent years. The concentrations in the upper layers do not necessarily represent the fluxes of these compounds reaching the sediment by aerosol fallout. More probably they are the average  $\Sigma$ -DDT concentrations in the solid materials transported here by the Dese river.

Although DDT has been used in Italy after the second world war, non-zero concentrations are found in the layers below 23 cm or about 1945. This may be due to the vertical mixing due to physical and biological activities in this area of the lagoon. In addition the possible cross contamination of

Table 4

Fluxes of heavy metals to sediments of some areas.

Element	Origine	Lagoon of Venice	California Basins (1a)	Palace Moat Tokyo (1b)	Narrangasett Bay (1c)
Hg	A	0.47			
	N	0.04			
	Total	0.51			
Cd	A	1.15			
	N	0.81			
	Total	1.96	0.21	1.7	
Pb	A	14.2			124
	N	17.3			2.6
	Total	31.5	2.1	130	126.6
Co	A	10.4			
	N	12.0			
	Total	22.4	0.53	7.7	
Ni	A	5.8			
	N	20.2			
	Total	26.0	2.3	19.0	
Cu	A	6.3			193
	N	11.5			3.1
	Total	17.8	2.9	130	196.1
Zn	A	37.8			230
	N	36.2			14
	Total	74.0	7.3	600	244
Fe	A	5 900			
	N	5 800			
	Total	11 700	1 800	22 000	
Cr	A	10.1			152
	N	15.3			14
	Total	25.4	8.1	43	166

a) from Bruland et al., 1974, b) from Goldberg et al., 1976; c) from Goldberg et al., 1977.

the samples during their exposure to the atmosphere in the drying room can not be completely excluded. The  $\Sigma$ -DDT level at the sediment surface is higher than the concentrations observed in 1977 in sea sediments of the Gulf of Venice (CIESM, 1978). However, more recent and extended studies indicate that in the areas in front of the Lagoon and opposite the Brenta, Adige and Po di Levante rivers mouths  $\Sigma$ -DDT levels are up to about ten times higher (Pavoni, private comm.). Much larger amounts are observed in more polluted localities such as the Saronikos Gulf (up to 1900 ppm) (Dexter, Pavlou, 1973) and the Palos Verdes shelf in California (up to 30 ppm) (Young *et al.*, 1976). The areal distribution and sources of these pollutants into the

lagoon and their transport to the sea by water exchange is under investigation.

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