

## Trace element atmospheric deposition and trends in Venice Lagoon (Italy)

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### ARTICLE INFO

Received 4/12/2020; received in revised form 19/2/2021; accepted 16/3/2021.

DOI: [10.6092/issn.2281-4485/11886](https://doi.org/10.6092/issn.2281-4485/11886)

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### Abstract

Atmospheric deposition data on trace elements from ten bulk sampler stations in the Venice Lagoon, covering a period spanning from 2007 to 2017, are presented and discussed. Results have been statistically analysed and used to estimate fall-out loads on the Lagoon. The Venice Lagoon is not only exposed to long-range transport of contaminants but it is also affected by local urban and industrial emissions. A sharp drop off of the Cd load since 2011 has been observed tied up with decreasing flux recorded at the station in the nearby glass-making district, where decreasing trends have also been detected for other elements. Decreasing long-term trends have been observed for Ni and Sb loads while Zn load has shown a small increase. The data analysis carried out in this study has proven the effectiveness of the bulk-collector-based surveys in detecting temporal and spatial changes in atmospheric depositions. The maintenance of this temporal data series will be a useful tool for evaluating future changes in the fall-out affecting the Venice Lagoon.

### Keywords

*Atmospheric depositions, Temporal trends, Trace elements, Venice Lagoon*

### Introduction

The Venice Lagoon is an example of a complex natural system with a long history of extensive human intervention and anthropogenic pressure. In order to cope with safeguard issues of the historic town and its lagoon, a legal framework consisting of a number of national laws and ministerial decrees has been established since the 1970s (Munaretto and Huitema, 2012). Among the aims of the Special Laws was the abatement of pollution in the lagoon. In the late 1990s quality objects specifically for the Venice Lagoon and its catchment basin were enacted with the aim of regulating the total load of a number of pollutants. In

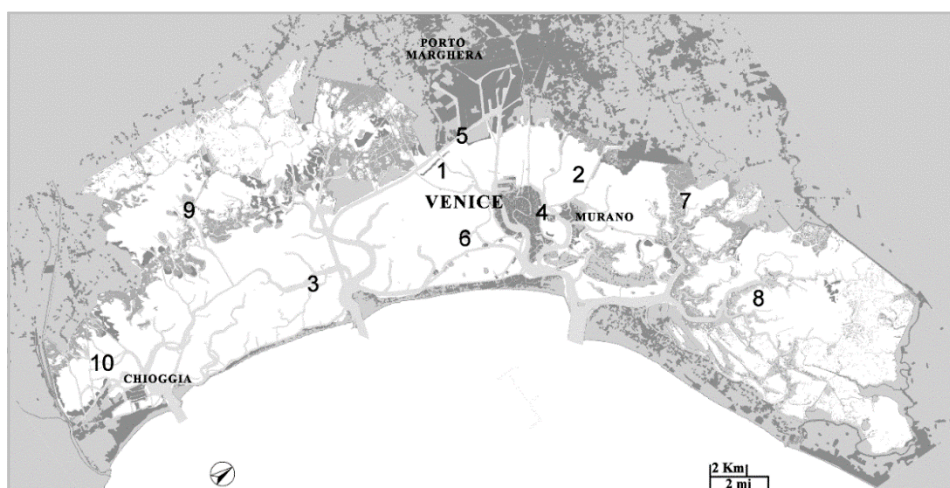
order to implement the regulatory framework, census and controls of water discharge have been conducted and a network of water quality monitoring stations has been established. In addition, a network of atmospheric deposition sampler stations has been deployed with the purpose of updating information required for the assessment of the annual load of pollutants which affect the lagoon. In this study results of the bulk atmospheric deposition of trace elements collected in the Venice Lagoon during a time frame spanning from 2007 to 2017 are reported and discussed, with a focus on atmospheric loads and temporal trends.

The Venice Lagoon is exposed to local sources of atmospheric pollutants from industrial activities operating in the industrial park of Porto Marghera located on the western shore of the Lagoon, 5 km NW of the historical centre of Venice, including among others a power plant fuelled by coal and refuse-derived fuel (RDF), a gas-fuelled power plant, a glass factory, cracking and petrochemical plants, a refinery recently converted to the production of biofuel from vegetable oil and biomass. In the Murano island is located the world-renowned district of artistic glass where tens glass-makers are still producing glass and others are involved in glass-blowing, engraving and decoration activities. Besides these activities, the Lagoon is exposed to emissions of intense ship and airport traffic, vehicular emissions on the mainland and domestic heating. Atmospheric depositions in the Venice Lagoon have already been investigated for short timeframes or in limited areas. Rossini et al. (2005) measured bulk atmospheric deposition over a period of 13 months between 1998 and 1999. Bettiol et al. (2005) compared atmospheric depositions and river inputs into the lagoon from the watershed in order to evaluate their relative importance. A prevalent atmospheric component was estimated only for Cd (60%), whereas the riverine input

prevailed for Cr, Cu, Fe, Ni, Mn, As (ranging from 70% to 96%) and were similar for Hg, Pb and Zn. Rossini et al. (2010) focused the attention on the Murano glass-making district measuring airborne PM10 and bulk depositions between 2001 and 2003 recognising a significant local source of some elements, in particular for Cd. Morabito et al. (2014) analysed bulk depositions in sampling sites located in correspondence of the three Lagoon inlets over a period of five years from 2005 to 2010. Other studies were aimed at investigating on source apportionment of atmospheric particulate matter (Rampazzo et al., 2008, Contini et al., 2012, Masiol et al., 2012).

### **Materials and methods**

The sampling network for the atmospheric deposition monitoring is made up of ten bulk collectors consisting of an HDPE funnel, with a 0.035 m<sup>2</sup> collection area, directly connected to an HDPE bottle (10L) placed in a PVC structure hanging from a pole 4 m above the mean sea level. The bulk collectors are mounted on stations consisting of a housing unit placed in different sites covering the entire lagoon (figure 1 and table 1).



**Figure 1.** Study area with the position of the sampling sites

**Table 1.** *Geographic coordinates of sampling sites*

| n. | site                | N             | E             |
|----|---------------------|---------------|---------------|
| 1  | Fusina              | 45°24'49.972" | 12°16'26.331" |
| 2  | Campalto            | 45°28'13.915" | 12°20'5.542"  |
| 3  | San Pietro in Volta | 45°19'32.023" | 12°17'8.674"  |
| 4  | Fondamenta Nuove    | 45°26'32.924" | 12°20'33.91"  |
| 5  | Tresse              | 45°25'58.681" | 12°15'40.867" |
| 6  | Sacca Sessola       | 45°24'11.66"  | 12°19'26.608" |
| 7  | Palude di Cona      | 45°30'26.582" | 12°23'50.639" |
| 8  | Palude Maggiore     | 45°30'33.847" | 12°29'14.985" |
| 9  | Valle Millecampi    | 45°17'44.027" | 12°11'17.105" |
| 10 | Val di Brenta       | 45°12'46.379" | 12°14'32.204" |

Bulk atmospheric deposition samples were sent to the laboratory at the end of each sampling period of two months, where they were weighted for the determination of total precipitation. 250 mL representative aliquots were collected and stored in HDPE bottles. HNO<sub>3</sub> (65%) was added to the aliquot for metal determination until pH < 2.

The samples were mineralised by means of microwave assisted acid digestion (Speedwave4, Berghof) according to USEPA 3015A method. Concentrations of As, Cd, Cr, Cu, Fe, Mn, Ni, Pb, Sb, V, Zn were measured by inductively coupled plasma mass spectrometry (ELAN DRC II, Perkin Elmer) according to USEPA 6020B method. The accuracy was checked by analysis of the certified reference materials (Fly Ash CRM019, Sigma Aldrich RTC). A portion of the CRM has been made into the sampling collector filled up with deionized water. The sample thus obtained has been carried throughout the entire sample preparation process.

The measured concentrations were used to calculate the atmospheric deposition of trace elements per unit area in each monitoring cycle. The obtained atmospheric deposition fluxes (mg/m<sup>2</sup>) were then summed up to calculate the cumulative annual deposition flux of trace elements in each station (mg/m<sup>2</sup>/y).

Prior to conducting statistical tests, records were visually checked by scatterplots, LOWESS smooth and Box-and-whisker plots of data (Helsel and Hirsch, 2002). Subsequently, records were statistically analysed using non-parametric tests to identify correlations, differences and trends (Helsel and Hirsch, 2002). Non-parametric tests do not assume that the data follow any specific

distribution and are more robust to detect trends than parametric methods are.

Kendall rank correlation test (KRC) between deposition fluxes was applied to study the origin of the elements in the area in further detail. A significant correlation may be reflective of a common source of contaminants. In order to assess the differences between sites Kruskal-Wallis (KW) test was applied. In addition to the KW test, Wilcoxon rank sum (WRS) test was used to calculate pairwise comparison between group levels. The Mann-Kendall trend (MK) test was applied to determine whether a trend has occurred (Yue, 2002). MK test is commonly employed to statistically analyse series of environmental data that depart from normal distribution, in particular with water quality multiple data sets (Mozejko, 2012) and atmospheric depositions time series (Marchetto et al., 2013, Kyllönen et al., 2008).

The inverse squared distance (ISD) method was used to interpolate the atmospheric deposition in the unsampled locations (Hengl, 2009). ISD is the inverse distance weighted (IDW) in which the power p is 2. IDW is a deterministic interpolation method based on the assumption that the value at a given point is a distance-weighted average of data points occurring within a neighbourhood of the given point (Babak and Deutsch, 2009). IDW does not require prior information about spatial relationships except for the basic assumption that values at nearby points are more closely related to the interpolated location than values at distant points. The computation was performed in a grid of cells with a resolution of 200 m overlapped to a

mask of the Lagoon.

Statistical analysis and the spatial predictions were performed using R version 3.6.1.

**Results and discussion**

During the study period (2007-2017) the main wind direction was N-NE, with a mean wind speed of 1.6 m/s (meteorological data from Cavanis Meteorological observatory, Venice). The fluxes of the studied elements in the bulk atmospheric deposition in the Venice Lagoon recorded between 2007 and 2017 are summarised in table 2 where quartiles, expressed as mg/m<sup>2</sup>/y, are reported. The deposition fluxes of the monitored elements in the bulk calculated at different sampling sites followed a similar order: Fe>>Zn>Mn≅Cu≅Pb>Ni≅V>Cr≅As≅Sb≅Cd. This order changed for the sampling site n.4 characterised by a much higher deposition flux of Cd and Pb: Fe>>Pb≅Zn>Mn≅Cu>Cd≅Ni≅V>Cr≅As≅Sb.

Bulk collector n.4 is placed close to Venice and leeward of the glass-making district of Murano. Cd deposition was an order of magnitude higher (1.6 mg/m<sup>2</sup>/y) and Pb deposition was three times higher (21 mg/m<sup>2</sup>/y)

than those observed in other sampling points farther from the glass-making district. In addition to the Pb and Cd deposition, at site n.4 a higher flux of Sb (0.32 mg/m<sup>2</sup>/y) and As (0.52 mg/m<sup>2</sup>/y) were recorded as well. Among the raw materials used by the glass manufacturers are oxides of elements such as trioxide of As and Sb as refining additives to assist mixing of compound and to remove air bubbles, PbO as stabilizer, several metals as colouring agents. As already investigated, emission of elements may be ascribed to glasswork factories (Rampazzo et al. 2008, Rossini et al. 2010). Investigating the geochemical characteristics of PM10, Rampazzo et al. (2008) found a link between As, Cd, Sb, Pb and the lighter fraction of particles emitted by glass factories while coloured glass is being melted. Kendall Rank test showed that Cd was strongly correlated to Sb (p<0.01, τ=0.77) and As was correlated to Pb (p<0.001, τ=0.85). Other strong correlations were found for the couples Mn-Fe (p<0.001, τ=0.78), V-Cr (p<0.001, τ=0.87), Cr-Fe (p<0.001, τ=0.78). Ni was correlated to V, Cr, Fe (p<0.01, τ>0.7), Fe was correlated to Zn (p<0.01, τ=0.74). Cu was not correlated with any other element. The amount of precipitation expressed in mm was not correlated either.

**Table 2.** Quartiles of deposition fluxes expressed in mg/m<sup>2</sup>/y and precipitation expressed in mm

| Qs  | As    | Cd    | Sb    | Pb   | Ni   | Mn  | V    | Cr   | Cu   | Fe   | Zn  | mm   |
|-----|-------|-------|-------|------|------|-----|------|------|------|------|-----|------|
| 0   | 0.046 | 0.030 | 0.086 | 0.65 | 0.38 | 3.0 | 0.35 | 0.16 | 0.43 | 29   | 6.2 | 280  |
| 25  | 0.14  | 0.067 | 0.13  | 4.4  | 0.76 | 5.1 | 0.61 | 0.30 | 2.9  | 64   | 11  | 610  |
| 50  | 0.25  | 0.12  | 0.16  | 7.0  | 1.1  | 7.1 | 0.79 | 0.37 | 4.0  | 96   | 14  | 750  |
| 75  | 0.46  | 0.23  | 0.22  | 10   | 1.5  | 10  | 1.0  | 0.52 | 6.4  | 141  | 18  | 900  |
| 100 | 1.1   | 6.0   | 0.61  | 27   | 6.1  | 180 | 1.2  | 2.4  | 39   | 1400 | 83  | 1100 |

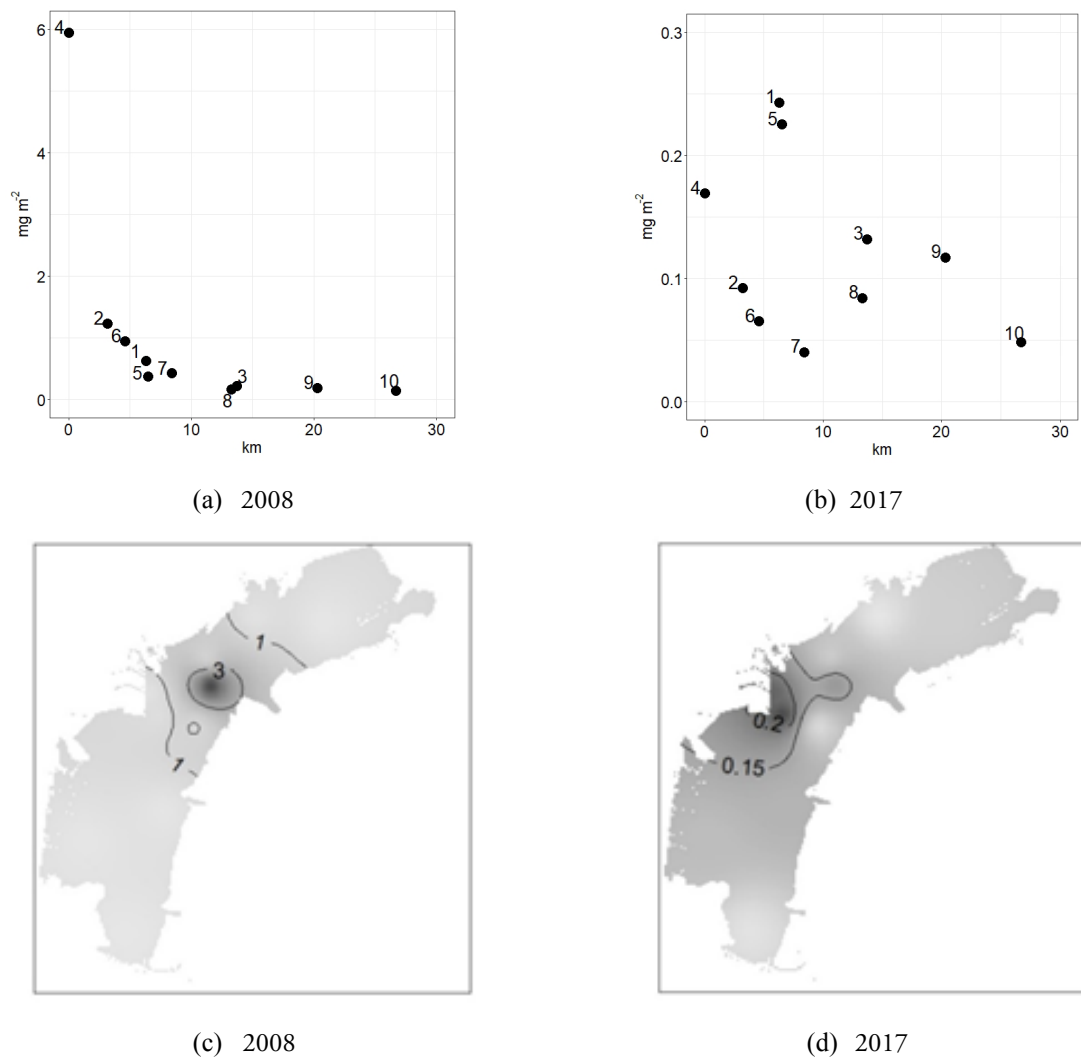
From the output of the Kruskal-Wallis test by rank it was possible to identify significant differences between sites for Cd, Sb, Pb, Fe (p<0.001), As, Mn (p<0.01), V, Cr, Cu, Zn (p<0.05). The pairwise comparison using Wilcoxon rank sum test showed that station n.4 was significantly different to any other site for Pb (p<0.001), as well as for Cd and Sb (p<0.05), with the exception of site n.2 located 2 km NE from Murano. A significant difference of Sb fluxes was also detected between station n.2 and bulk collectors n.3,7,8,10 (p<0.05) and n.9 (p<0.01), showing higher fluxes. Fe deposition fluxes were higher at the bulk collector n.5 and WRS test confirmed that there was a significant difference between site n.5 and 7 sites out of 10 (sites

n.4,6,8,9 p<0.01; sites n.1,3,10 p<0.05). V fluxes at station n.5 were higher and statistically different from the depositions measured at 5 sites out of 10 (sites n.5,8,10 p<0.01; sites n.6,9 p<0.05). Bulk collector n.5 is placed close to the industrial park of Porto Marghera. Trend analysis in the annual fluxes were estimated using Mann-Kendall test on data from all of the stations. Decreasing trends were detected mainly at station n.4 (As, Cd, Sb, Ni, Mn, Cu p<0.01, τ<-0.7; Fe, Zn p<0.05, τ<-0.4). Statistically significant decreasing trends were also found in other stations nearby industrial and urban sources of emissions. In particular, station n.6 that lied on the leeward side of Venice and Murano followed a similar pattern of trends observed at station n.4 (As,

DOI: [10.6092/issn.2281-4485/11886](https://doi.org/10.6092/issn.2281-4485/11886)

Cd, Ni, V  $p < 0.01$ ,  $\tau < -0.7$ ; Sb, Pb  $p < 0.05$ ,  $\tau < -0.5$ ). At the stations n.1 and 5, facing the industrial area of Porto Marghera, decreasing trends were detected for Ni ( $p < 0.01$ ,  $\tau < -0.6$ ), Pb and V ( $p < 0.05$ ,  $\tau < -0.6$ ). A decreasing trend of Sb flux was also detected at the bulk collector n.10 ( $p < 0.05$ ,  $\tau = -0.53$ ). It is worth pointing

out the weak but statistically significant positive trends detected for As and Zn fluxes at sampling site n.3 ( $p < 0.05$ ,  $\tau > 0.5$ ). Stations n.8 and 9 did not show any significant trends of the studied elements during the considered time frame.



**Figure 2.** Fluxes at the different stations in 2008 (a) and 2017 (b), in relation to the distance from the station n.4 ( $\text{mg}/\text{m}^2/\text{y}$ ). Interpolation maps of the depositions in 2008 (c) and 2017 (d) ( $\text{mg}/\text{m}^2/\text{y}$ ).

The highest flux of Cd was recorded in 2008 at the station n.4 ( $6.0 \text{ mg}/\text{m}^2/\text{y}$ ). Representing graphically the fluxes calculated at the different stations in 2008, in relation to the distance, a decreasing trend appeared evident moving away from the station n.4 (figure 2a). As shown in the interpolation map created on the ISD method basis in the 2008 (figure 2c) the whole central part of the Lagoon had fluxes higher than  $1 \text{ mg}/\text{m}^2/\text{y}$ , being affected by the hot spot in correspondence

of Murano. Northern and southern Lagoon, far from the glass-making emission source, appeared to be less affected by Cd depositions ( $< 0.2 \text{ mg}/\text{m}^2/\text{y}$ ). By matching the spatial distribution of Cd fluxes in 2008 with that in 2017 (figure 2c and 2d) the drop of the deposition flux occurred in the decade stood clearly out. In the 2017 the deposition pattern (figure 2d) was very different showing higher deposition in the western shore of the Lagoon ( $> 0.15 \text{ mg}/\text{m}^2/\text{y}$ ), particularly around

DOI: [10.6092/issn.2281-4485/11886](https://doi.org/10.6092/issn.2281-4485/11886)

the industrial area of Porto Marghera ( $>0.2 \text{ mg/m}^2/\text{y}$ ). The highest flux of Cd recorded in 2017 was  $0.24 \text{ mg/m}^2/\text{y}$  at the station n.1.

The outputs of the statistical tests found confirmation in the evidence of the aforementioned studies focused on the Venice Lagoon which agree in identifying two main groups of elements. The first group included Cd, Sb, As, Pb representing highly anthropogenic enriched elements. A second group included V, Ni, Fe, Cr, Cu, Mn, Zn associable to a mix of sources having a different nature both crustal and anthropogenic, such as combustion of fossil fuels (Ni, V), traffic emission from diesel (Cu), tyre wear and road dust (Zn).

The element deposition fluxes in the Venice Lagoon calculated in the present survey were within the range found in other regional sites less affected by anthropogenic emissions (Olmo et al., 2015 and literature therein), with the exception of As, Cd and Pb whose deposition fluxes were similar to those found in other urban and industrial sites (Olmo et al., 2015).

Comparing the results of this study with those reported for remote areas far from pollutant sources (Kyllönen et

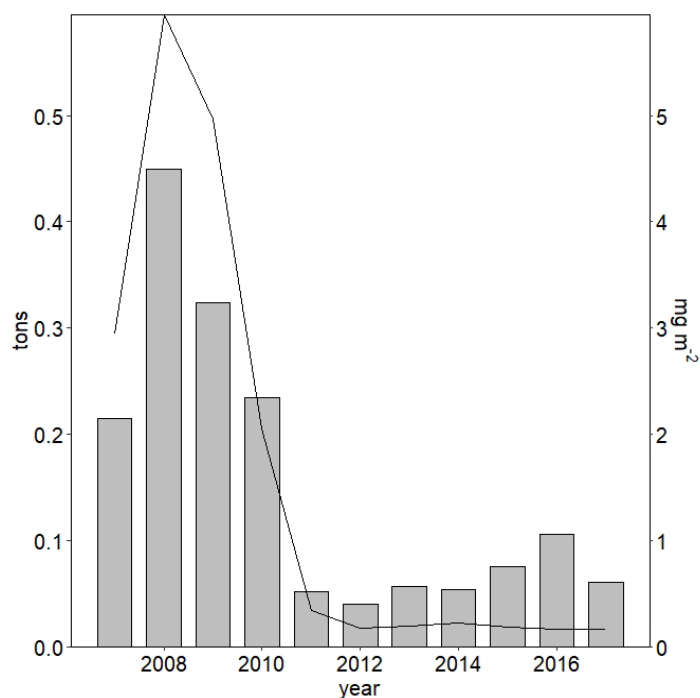
al., 2008), the annual deposition flux of trace elements in the Venice Lagoon was much higher, ranging from ten times for V, Cr, Cu to twenty times for As, Cd, Ni, Mn, Fe, Zn to forty times for Pb.

The yearly deposition fluxes calculated for the different stations were used to estimate values in unsampled locations using ISD interpolation method. The interpolated values were then summed up to estimate the total loads of each element in the Lagoon (table 3). The most remarkable evidence was the sharply decrease of Cd loads after 2010 that followed the plummeting flux at station n.4 (figure 3). Mn and Fe loads showed a peak recorded in 2012, due to a single event of unknown origin, occurred at the station n.7 during the 2012 autumn campaign.

Loads of the other elements are fairly variable, nevertheless decreasing trends were detected for Ni loads (MK test:  $p<0.01$ ,  $\tau=-0.75$ ) and rather weakly for Sb loads (MK test:  $p<0.05$ ,  $\tau=-0.51$ ). It is worth pointing out the small but positive trend of Zn loads (MK test:  $p<0.05$ ,  $\tau=+0.49$ ).

**Table 3.** Deposition loads of trace elements expressed in tons.

|      | As   | Cd   | Sb   | Pb  | Ni   | Mn  | V    | Cr   | Cu  | Fe  | Zn  |
|------|------|------|------|-----|------|-----|------|------|-----|-----|-----|
| 2007 | 0.17 | 0.22 | 0.14 | 7.1 | 0.98 | 4.4 | 0.57 | 0.32 | 2.5 | 60  | 7.3 |
| 2008 | 0.27 | 0.45 | 0.14 | 4.6 | 0.95 | 4.7 | 0.63 | 0.31 | 3.6 | 66  | 7.6 |
| 2009 | 0.12 | 0.32 | 0.10 | 4.0 | 0.82 | 3.5 | 0.50 | 0.25 | 2.7 | 60  | 8.8 |
| 2010 | 0.11 | 0.24 | 0.10 | 3.7 | 0.62 | 3.0 | 0.41 | 0.16 | 1.8 | 38  | 8.1 |
| 2011 | 0.11 | 0.05 | 0.08 | 4.0 | 0.50 | 3.2 | 0.44 | 0.18 | 1.8 | 53  | 7.0 |
| 2012 | 0.14 | 0.04 | 0.08 | 4.1 | 0.75 | 14  | 0.50 | 0.29 | 2.5 | 124 | 7.8 |
| 2013 | 0.14 | 0.06 | 0.10 | 3.3 | 0.53 | 3.0 | 0.35 | 0.18 | 2.2 | 48  | 11  |
| 2014 | 0.21 | 0.05 | 0.07 | 2.0 | 0.63 | 3.6 | 0.33 | 0.20 | 2.2 | 46  | 7.8 |
| 2015 | 0.17 | 0.08 | 0.08 | 4.5 | 0.53 | 8.1 | 0.49 | 0.25 | 5.8 | 104 | 12  |
| 2016 | 0.23 | 0.11 | 0.08 | 3.7 | 0.49 | 5.8 | 0.40 | 0.24 | 7.5 | 75  | 13  |
| 2017 | 0.19 | 0.06 | 0.08 | 4.6 | 0.48 | 7.1 | 0.48 | 0.27 | 3.9 | 91  | 11  |



**Figure 3.** Annual loads of Cd expressed in tons (bars) and fluxes at station n. 4 expressed in mg/m<sup>2</sup> (line).

## Conclusions

In the last few decades, it has been demonstrated that the atmosphere is a significant pathway for the transport of contaminants to aquatic ecosystems. Once trace elements reach aquatic systems, their behaviour is highly dynamic: variations in the chemical speciation, inorganic scavenging, sedimentation, complexation affect the biogeochemical cycle of trace elements (Khan et al., 2014). Although some elements are trace nutrients, they become toxic to organisms at enhanced concentrations. Among the pollutants that have detrimental effects on aquatic organisms are elements such as As, Cd, Pb which may be transferred to humans through dietary intake of seafood products.

Bulk collectors have proved to be a valuable tool for the monitoring of atmospheric depositions. The analysis of a comprehensive data set on trace elements in atmospheric depositions, collected during monitoring campaigns from 2007 to 2017 in the Venice Lagoon, has revealed the effectiveness of such monitoring programs to detect rapid changes in the contaminant fall-out, like the sharp decrease observed for Cd deposition, as well as long-term trends, like those detected for elements such as Ni, Sb and Zn.

Even if the resuspension of contaminated sediments stored in the canals of the industrial area is recognised

as the major potential source of contaminants, including harmful trace elements, the atmosphere represents a significant route by which contaminants could be transported to the Lagoon (Bettiol et al., 2005). The Venice Lagoon is also well known for its shellfish harvesting activity and fish breeding whose extensive production can be an easy target for chemical contamination (Berti et al., 2015).

Owing to the importance of the atmosphere as a pathway for the contaminant transport, the monitoring activity of the fall-out needs to be carried on to supply updated information on the annual load of contaminants delivered to the Lagoon.

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