

Priority substances in Adriatic Sea. Case study: Water of Durres and Vlora ports, Albanian coastline

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Abstract

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The elevated activities in the port areas are the main reasons for water pollution by priority substances, which are of growing concern due to their potential toxicity to aquatic organisms and human health. Therefore, this paper reports the concentrations of organochlorine pesticides (OCPs), polychlorinated biphenyls (PCBs), polycyclic aromatic hydrocarbons (PAHs), and BTEX (benzene, toluene, ethyl benzene, and *o*-, *m*-, and *p*-xylenes), analyzed in water samples from two main ports, Durres and Vlora. To assess the pollution status of these areas 24 water samples in total were collected, of which 12 in the port of Durres and 12 in Vlora port, in June 2022. Analyses of this compounds were performed using a gas chromatograph (Varian 450 GC) equipped with ECD and FID detectors. BTEX were analyzed using the HS/SPME technique, followed by the GC/FID technique for their qualification and quantification. Organochlorine pesticides and their degradation products were detected in all water samples. Their presence may be related to their previous uses. PCBs, PAHs, and BTEX were found in almost all analyzed samples. The higher levels were found in stations inside port areas. Their presence is due to anthropogenic factors (intense activity and ship transport) in these areas. The reported levels of these priority substances in water samples from Durres and Vlora's ports were higher and comparable with reported levels of them from other stations in the Adriatic Sea. Although the production and use of these substances have been banned for decades, their presence continues to be reported.

Keywords: Durres and Vlora ports; OCPs; PCBs; PAH; BTEX; GC/ECD/FID

Introduction

Port operations have a significant impact on water quality and the health of marine life. Discharges of wastes from ships and other port activities can harm marine life. In many studies, marine environments have been established as primary sites for the presence of pollutants (Chen et al., 2010, Li et al., 2006). The main types of pollutants entering the aquatic environment are organic compounds, represented mainly by polycyclic aromatic hydrocarbons, pesticides, drugs, dioxins, phenols, and phthalates, or inorganic compounds, which can be represented by heavy metals, nitrogenous forms, and phosphorus. Organochlorine pesticides

(OCPs) and polychlorinated biphenyls (PCBs) are among the most important persistent organic pollutants (POPs), and they are well known for their chronic toxicity, persistence, and bioaccumulation (Lekkas et al., 2004). Persistent organic pollutants have a wide range of industrial, anthropogenic, and agricultural applications. For this reason, the monitoring of marine POPs is of great importance (Yang et al., 2019).

Durres and Vlora ports are located in the Adriatic Sea, on the Albanian coastline. The port of Durres is the largest seaport in Albania. It is an artificial basin that is formed between two moles, to the south of Durres City. In it is handling around 77% of imports and 89% of Albanian exports. The main commodities handled in this port are general cargoes,

grains, minerals, containers, and ferry boats. Since 2014, the port has ranked as the largest passenger port in Albania and one of the largest passenger ports in the Adriatic Sea. Next to the port of Durres is a fishing harbor (at northern part). More in the north of this port (5 km distance) is located the hydrocarbon port of Porto-Romano. In the near future, it is thought that the port area will be re-conceptualized and expanded as one of the largest ports in the Mediterranean Sea. The port of Vlora is the second port in Albania. It is located in Vlora Bay, near the city of Vlora, in southern Albania. The port is considered part of the Lungomare Master Plan of Vlora. Part of this project is the construction of a yacht port and some new roads to make the port area more accessible. Until now, Vlora's port has been mainly used for passengers and commercial shipping. In fact, beside Vlora ports in the area of Vlora Bay are located other ports such as petroleum port (Petrolifera near Zvërneci), Marina port for Delta Force (near Radhima), the military base of Orikumi, and a fishing harbor (near Zvërneci). In the area of Vlora Bay, there are some tourist ships and many small boats (motorized ones) that serve as tourist transportation to visit areas of Sazani Island and Karaburuni Peninsula. Except for this, the geographical position of the bay favors the concentration of pollutants inside of it such as urban pollution and new arrivals from Vjosa River.

The aim of this study is to determine the concentration of organochlorine pesticides (OCPs), polychlorinated biphenyls (PCBs), polycyclic aromatic hydrocarbons (PAHs), and BTEX (benzene, toluene, ethyl benzene, and o-, m-, and p-xylenes) in the seawater collected from two main ports of Albania, and to identify potential sources of these contaminants in the water. For around 50 years (from 1946 to 1992), organochlorine pesticides were widely used in Albania for agricultural purposes. The main agricultural areas are located in the western part of the country, near the Adriatic Sea. The use of pesticides in Albania after 1992 decreased rapidly due to migration and immigration. PCBs were not in use in Albania until the 1990s. They can be found only in some electrical transformers that were used in the early 1990s, but they were reported in many water ecosystems in our country because of their atmospheric depositions. PAHs and BTEX are pollutants, generated by automobile transport, the extraction and processing of oil, coal mining, and other industries. These hydrocarbons could be found in marine water because of ship transport or accidental spills of hydrocarbons. Forest burning and their natural background make them very common in the environment. These pollutants (OCP, PCB, PAH) have high stability, high bioaccumulation capacity, and the ability to spread far away from the application site. Generally, these compounds are difficult to degrade and can persist for many years, particularly in wa-

ter ecosystems (Corsi et al., 2010; Nuro et al., 2014; Chen et al. 2010). They can be absorbed easily by suspended particles and organic matter in water and accumulate in sediments at concentrations several times higher than in water (Stogianidis and Laane, 2015; Wang et al., 2009).

Material and Methods

Water sampling in the main Albanian ports

The sampling of water was realized in the end of July 2022. In Durres port, 12 water samples were collected (8 samples inside the port area and 4 samples outside it), and in Vlora port, 12 water samples were collected (6 samples inside the port area and 6 samples outside it), respectively. This period of sampling was chosen due to the intense period for the activities in Albanian ports (from two to three times higher than the normal period). In each sampling point, 2.5 L of sea water were collected in Teflon bottles based in ISO 5667-3:2018. Water samples were stored at 4°C and transported to the laboratory for further analysis. The sampling stations for Durres and Vlora ports were presented in Figure 1.

Treatment of water samples for pollutant analyzes

For the determination of OCPs and PCBs in water samples, liquid-liquid extraction (LLE) was used. One liter of water samples was extracted with n-hexane (2 x 50 ml) in a separatory funnel. After extraction, the organic phase was dried with anhydrous sodium sulfate (Na_2SO_4 (5 g) for water removal. A florisil column was used for the sample clean-up. 20 ml of n-hexane/dichloromethane (4/1) was used for elution. After concentration to 1 ml hexane, the samples were injected into GC/ECD (Yang et al., 2019; Nuro et al., 2014).

Two-step liquid-liquid extraction was used for extracting PAHs from water samples. One liter of water with firstly 50 ml of dichloromethane (first step LLE) and after that 50 ml of hexane (second step LLE) as extracting solvent was added in a separator funnel. After extraction, the organic phase was dried with 5 g of anhydrous Na_2SO_4 for water removal. Extracts were concentrated to 1 ml hexane using Kuderna-Danish and then injected in GC/FID for qualification or quantification of PAHs (Nuro et al., 2014; Wang et al., 2009; Lekkas et al., 2004).

Gas chromatography analysis of pesticides and PCBs

Organochlorine pesticides and PCBs were analyzed simultaneously using a capillary column of type Rtx-5 (30 m long \times 0.25 mm in diameter \times 0.25 μm film thickness) on a gas chromatograph model Varian 450 GC, equipped with electron capture detector (ECD detector). Helium was used as a carrier gas (1 ml/min), while nitrogen was used as a make-up gas

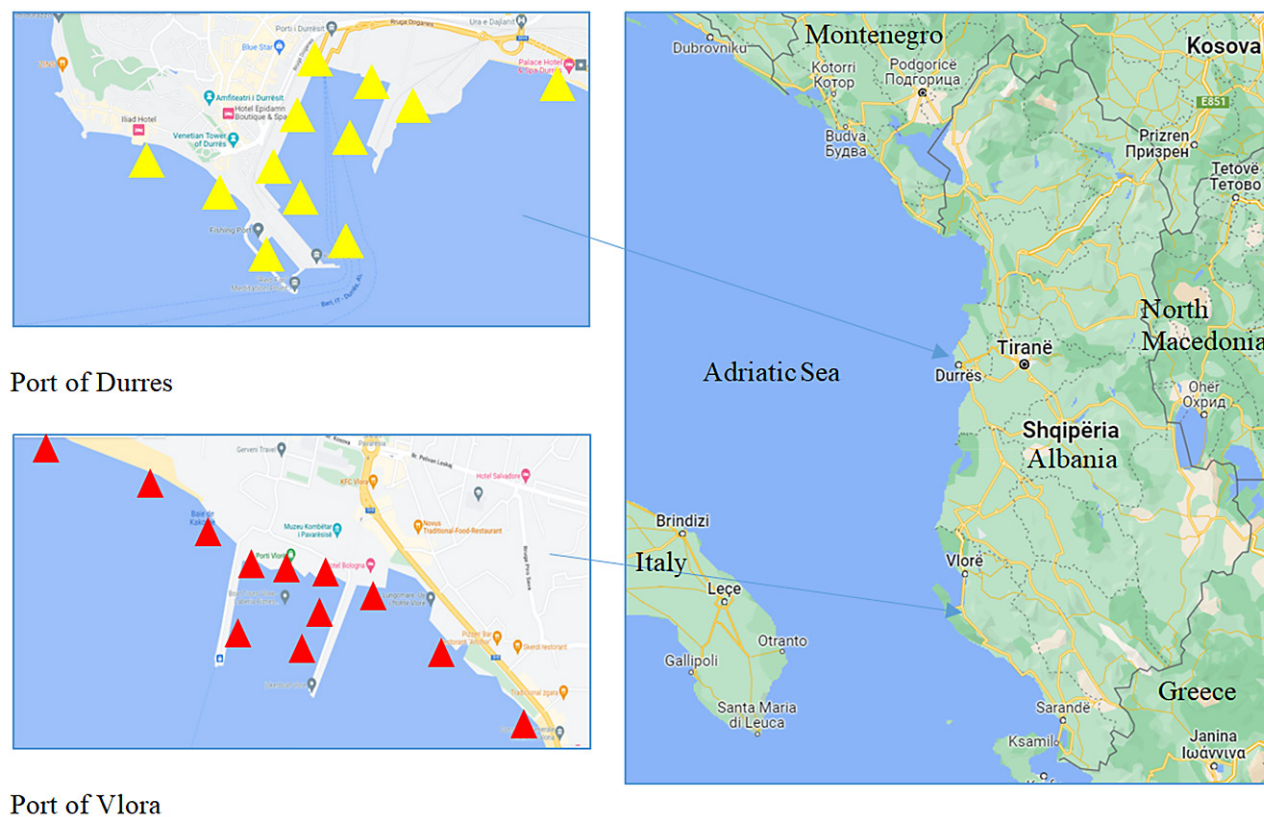


Fig. 1. Map of the study area and sampling sites within ports in July 2022

Source: Authors' own elaboration

(24 ml/min). The manual injection was done in splitless mode at 280°C. The organochlorine pesticides detected were DDT-related chemicals (o,p-DDE, p,p-DDE, p,p-DDD, p,p-DDT), HCHs (α-, β-, γ- and δ-isomers), Heptachlor's (Heptachlor and Heptachlorepoxide), Aldrin's (Aldrine, Dieldrine, and Endrin), and Endosulfanes (Endosulfan alpha, Endosulfan beta, and Endosulfan sulfate). The analysis of PCBs was based on the determination of the seven PCB markers (IUPAC Nos. 28, 52, 101, 118, 138, 153, and 180). The quantification of OCPs and PCBs was based on an external standard method (Li et al., 2006; Lekkas et al., 2004; Nuro et al., 2014).

Gas chromatography analysis of PAHs in water samples

Gas chromatographic analyses of PAHs in water samples were realized with a Varian 450 GC instrument equipped with a flame ionization detector and PTV injector. A VF-1 ms capillary column (30 m x 0.33 mm x 0.25 μm) was used for the qualification and quantification of 13 PAHs according to the EPA 525 Method. Helium was used as a carrier gas at 1 ml/min. The FID temperature was held at 280°C. Nitrogen

was used as the make-up gas (25 ml/min). Hydrogen and air were flame detector gases at 30 ml/min and 300 ml/min, respectively. The EPA 525 Standard Mixture was used for qualitative and quantitative analyses of PAHs. Acenaphthylene, Fluorene, Phenanthrene, Anthracene, Pyrene, Benzo [a] anthracene, Chrysene, Perilene, Benzo [b] fluoranthene, Benzo [k] fluoranthene, Indeo [1,2,3-cd] pyrene, Dibenzo [a, b] anthracene, and Benzo [g, h, i] perylene were determined in seawater samples. The quantification of PAHs was based on an external standard method (Akkanen et al., 2005; Nuro et al., 2014; Wang et al., 2009; Lekkas et al., 2004).

HS-SPME technique for determination of BTEX in water samples

The determination of BTEX in water samples was realized using solid-phase micro-extraction in static head space mode (HS/SPME), followed by the GC/FID technique. 5 ml water sample was put in a 10 ml head-space vial. 100 μm of PDMS fiber was used to extract BTEX from water samples. The adsorption process was realized at 50°C (using a water bath) for 45 minutes. A desorption process (20 seconds at

280°C) was realized in a PTV injector (HS mode was selected) of a Varian 450 GC instrument. A VF-1 ms capillary column (30 m × 0.33 mm × 0.25 µm) was used for the separation of benzene, toluene, ethyl benzene, and xylene isomers. Helium was used as a carrier gas at 1 ml/min. The FID temperature was held at 280°C. Nitrogen was used as the make-up gas (25 ml/min). BTEX Mixture was used for their qualitative and quantitative analyses based on an external standard method (Nuro et al., 2014).

Results and Discussion

The levels of priority substances (OCPs, PCBs, PAHs, and BTEX) were investigated in marine water samples of Albanian main ports based in the Water Framework Directive 2000/60/EC (EC 2000). Water samples from the ports of Vlora and Durres were taken in July. In this period the port activity is higher than other periods. Pollutant analyze was performed by using GC/ECD/FID technique.

Organochlorine pesticides were detected in all water samples analyzed from the ports of Durres and Vlora (Table 1, Figure 2). The total concentration of OCPs in water samples collected in the port of Durres ranged from 1.63 µg/l (station D1) to 6.59 µg/l (station D10) with a mean value of 3.61 µg/l, while their total for samples of Vlora port was between 0.19 µg/l (station V8) to 31.78 µg/l (station V6) with a mean value of 8.16 µg/l. Stations inside both port areas showed higher levels of OCPs than locations around it. The concentration of OCPs in the port of Vlora was found to be higher than in the port of Durres. The higher presence of OCPs in Vlora's stations may be related to their previous uses in agricultural areas near the Vlora Bay and new arrivals by Vjosa River. Note that, these new arrivals can be from other areas of Albania. The geographical position of Vlora Bay, water currents inside/outside of Vlora Bay, punctual sources near the port, etc. can be main factors for levels of OCPs at the port of Vlora. In the port of Durres, OCP levels were lower than Vlora port's. This fact could be supported by port location (it is more exposed to water currents outside and inside the port than Vlora port). The cycle of water in Durres port is shorter than its cycle in Vlora Bay.

The profile of OCPs in water samples of Durres was: HCHs > Heptachlores > Aldrins > DDTs > Chlordanes > Endosulfanes. The presence of HCHs in higher concentrations in the port of Durres could be mainly due to the impact of the Ex-Lindane plant industry in Porto-Romano (5 km north of Durrës's port), which for many years discharged its wastes directly to the Adriatic Sea. The presence of DDT and Heptachlor could be a combination of terrestrial (these pesticides have been stored in the port's warehouses for a long time) and

water currents (new arrivals from the Shkumbin, Erzen, and Ishem rivers). The profile of OCPs in Vlora samples was: Endosulfanes > Heptachlores > Chlordanes > Aldrins > HCHs > DDTs. This profile was different from Durres samples because their origins are different for both ports. Cyclopentadiene pesticides were found in higher concentrations. Endosulfane I, Heptachlor, g-Chlordane and Dieldrin were found at higher levels. This different profile may be due to new arrivals from the Vjosa and Semani rivers and the individual properties of the pesticides (water solubility, persistence, degradation rate, the ability to bind with suspended matter, etc.). Recent use of these pesticides and punctual sources of these contaminants are not excluded. OCP concentrations in Durres and Vlora water samples were in the same range with previous studies performed in the Adriatic Sea. (Corsi et al., 2010; Murtaj et al. 2013; Nuro et al. 2014). The levels of individual pesticides were below acceptable levels according to Albanian and EU standards. (Directive 2008/105/EC).

PCB markers were detected in all analyzed samples from the ports of Durres and Vlora (Figure 4). The total concentration of PCBs in Durres port ranged from 0.145 µg/l (station D1) to 13.59 µg/l (station D11) with a mean value of 1.73 µg/l, whereas in Vlora samples it ranged from 0.19 µg/l (station V8) to 27.47 µg/l (station V7) with a mean value of 5.40 µg/l, respectively. PCBs were detected in high concentrations in Vlora samples. Their presence in seawater samples from both ports may be related to ship transport/activity, industrial activity in the cities of Durres and Vlora, and atmospheric deposition. Oil/waste discharges from some machinery operations and activities (in and out of the port areas) could be possible sources of PCBs and hydrocarbons in seawater samples from both ports. Note that, in this period, several large fires were recorded in several cities (Porto Romano landfill near Durres, Selenica and Narta landfills near Vlora) and forest areas (Karabruni, Llogara, and Selenica near Vlora), and may represent a further source of contamination in both study areas. The profile of PCB markers in Durres samples was: PCB 118 > PCB 32 > PCB 28, while their profile for the port of Vlora was: PCB 118 > PCB 52 > PCB 101 > PCB 138 (Figure 5). PCB 52, a volatile congener, was detected in both areas in high level compare to other congeners. Its origin is mainly due to atmospheric deposition. Relatively, high concentrations of PCB 118 were detected in all water samples. Its origin may be partly terrestrial and partly atmospheric. PCB level and profile were associated with individual concentrations of several congeners. The presence of heavy PCB congeners at some stations could be because of punctual sources of these contaminants in both ports (all of these stations with high concentration of heavy PCBs were within port areas). Ship and car transportation

can affect the PCB content of the analyzed samples. PCB levels were comparable to data reported in previous Adriatic studies (Corsi et al., 2010; Murtaj et al., 2013; Nuro et al., 2014). Levels of PCB markers were lower than permitted by Albanian and EU standards. (Directive 2008/105/EC).

PAHs were detected in all water analyzed samples from Durres and Vlora ports (Figure 6). Their average value for the port of Durres was 4.57 µg/l and for Vlora's port, it was 5.48 µg/l. The PAHs levels for Durres samples ranged from 1.97 µg/l (D12) to 8.61 µg/l (D3) and for Vlora samples, they ranged from 0.78 µg/l (V10) to 11.49 µg/l (V4). Again, PAHs were found in higher concentrations in Vlora samples. PAH presence in both ports could be mainly because of elevated ship transport inside port areas. Automobilit transport and any possible accident involving hydrocarbons (in the port areas or near them) could be another source of PAH pollution in marine water samples from Vlora port. Mechanical activities near the ports of Durres and Vlora, as well as some massive fires near these areas, could be important sources of hydrocarbons in marine water. Note that the presence of some individual of PAHs was at a higher level in both study areas. Benzo[k]fluoranthene, Benzo[b]fluoranthene Pyrene, Acenaphthylene, Fluorene, and Chrysene were found at higher levels in Durres samples, while Anthracene and Chrysene were most abundant in Vlora stations (Figure 7). Their profile in both areas is a combination of their origins in hydrocarbons (transport emitting, spillage of hydrocarbons, mechanical activities, etc.) and pyrogenic (forest and urban waste burning, transport emitting, industrial activities, etc.). The differences between Durres and Vlora ports could be connected mainly with geographical position, water currents, and the characteristics of industrial/mechanical activity near each area. Momentum values of PAHs for analyzed samples are not excluded. PAH levels in seawater

samples were in the same range/higher than the reported levels for other stations in the Adriatic Sea, Albania (Nuro et al., 2014; Murtaj et al., 2013; Marini and Frapiccini, 2013; Mandić and Vrančić, 2017; Magi et al., 2002). The presence of PAH individuals (Anthracene) at some stations was higher than the permitted level according to Albanian and EU norms (Directive 2008/105/EC).

BTEX were found almost in all water samples from Durres and Vlora ports. The average BTEX for Durres port (2.73 µg/l) was almost the same as for Vlora samples (2.51 µg/l). Their total was slightly higher in Durres, because of the abundant level of Toluene at D3 station (Figure 8). The total amount of BTEX ranged from 0.24 µg/l (D9) to 11.11 µg/l (D11) at Durres samples and from 1.02 µg/l (V10) up to 4.89 µg/l (V4) at samples taken in the port of Vlora. The profile of BTEX in Durres stations was: Toluene > *m*-Xylene > *o*-Xylene > Benzene > *p*-Xylene > Ethylbenzene and their profile for Vlora samples was: Benzene > Toluene > *m*-Xylene > *o*-Xylene > *p*-Xylene > Ethylbenzene (Figure 9). Benzene and Toluene were the most frequently found in the analyzed samples, but the presence of Xylene isomers was found at higher concentrations in some stations (D1, D6, D12, V1, V2, V3, V4, V11, and V12). The presence of BTEX in seawater samples could be due to elevated ship transport, automobilit transport, or any possible accident involving hydrocarbons near the areas of Durres and Vlora ports. The momentum values of BTEX for both ports are not excluded. Benzene concentrations were found in higher/comparable concentrations than reported levels for other stations in the Adriatic Sea, Albanian part (Nuro et al., 2014). The benzene level was lower than the permitted level according to Albanian and EU norms (Directive 2008/105/EC), which excludes its level for some stations in Durres and Vlora.

Table 1. Data on priority substance concentrations from the ports of Durres and Vlora, July 2022

Compounds	Port of Durres						Port of Vlora					
	Mean	Median	Minimum	Maximum	STDV	Count	Mean	Median	Minimum	Maximum	STDV	Count
HCHs (µg/l)	1.37	0.54	0.09	5.65	1.67	12	0.94	0.32	0.14	3.26	1.12	12
DDTs (µg/l)	0.47	0.23	N.D.	2.15	0.58	12	0.90	0.20	N.D.	6.62	1.90	12
Aldrin's (ug/l)	0.61	0.43	0.05	2.10	0.56	12	1.34	0.44	N.D.	6.50	1.87	12
Heptachlors (µg/l)	0.49	0.28	N.D.	1.72	0.56	12	1.68	0.42	N.D.	12.26	3.45	12
Chlordane's (µg/l)	0.38	0.17	N.D.	1.36	0.44	12	1.44	0.12	N.D.	14.29	4.09	12
Endosulfanes (µg/l)	0.27	0.12	N.D.	1.25	0.35	12	1.86	0.19	N.D.	16.85	4.81	12
Pesticides (µg/l)	3.61	3.56	1.63	6.59	1.51	12	8.16	4.65	0.19	31.78	9.40	12
PCBs (µg/l)	1.73	0.66	0.15	13.79	3.82	12	5.40	1.40	0.19	27.47	7.95	12
PAHs (µg/l)	4.57	4.86	1.97	8.61	2.22	12	5.48	4.85	0.78	11.49	3.08	12
BTEX (µg/l)	2.73	2.14	0.24	11.11	2.96	12	2.51	2.29	1.02	4.89	1.36	12

N.D. – Not detected or lowers than Limit of Detection (LOD)

Source: Authors' own elaboration

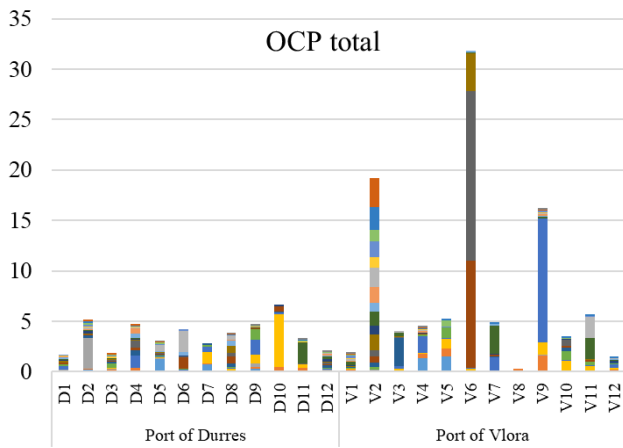


Fig. 2. Total of organochlorine pesticides in Durres and Vlorë ports

Source: Authors' own elaboration

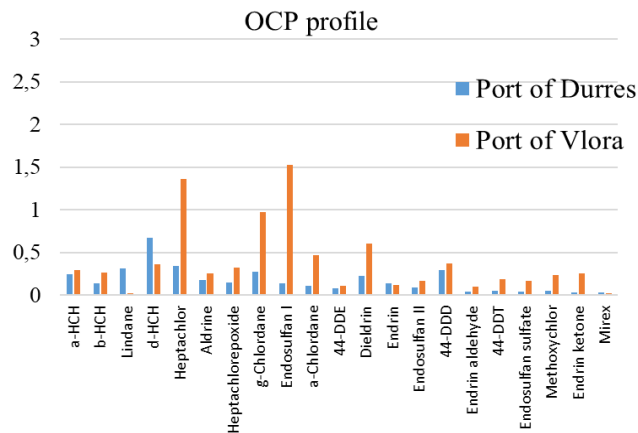


Fig. 3. Profile of organochlorine pesticides in Durres and Vlorë ports

Source: Authors' own elaboration

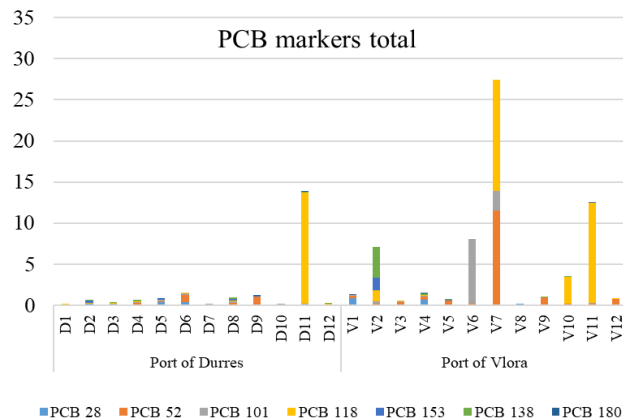


Fig. 4. Total of PCBs in Durres and Vlorë ports

Source: Authors' own elaboration

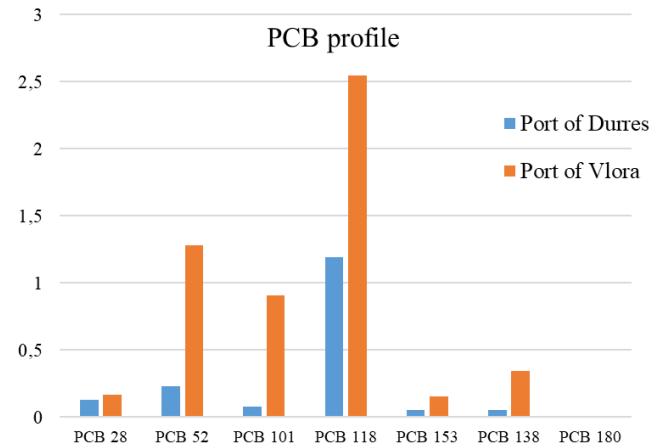


Fig. 5. Profile of PCBs in Durres and Vlorë ports

Source: Authors' own elaboration

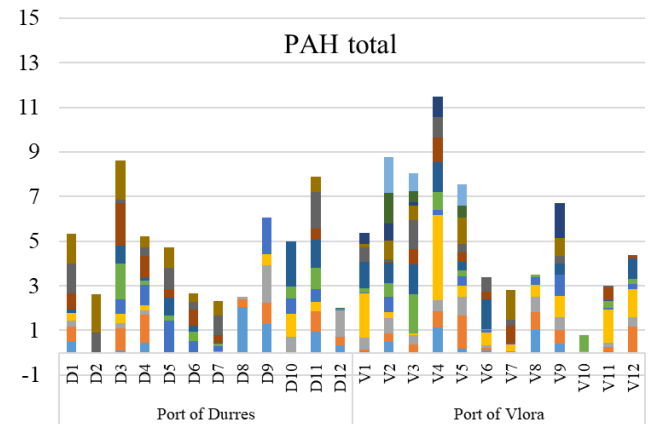


Fig. 6. Total of PAHs in Durres and Vlorë ports

Source: Authors' own elaboration

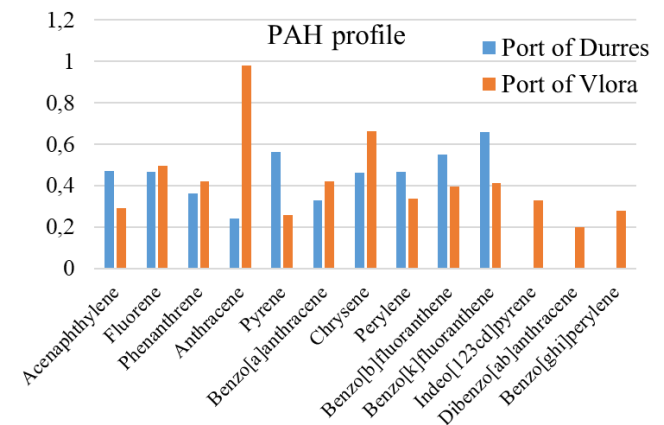


Fig. 7. Profile of PAHs in Durres and Vlorë ports

Source: Authors' own elaboration

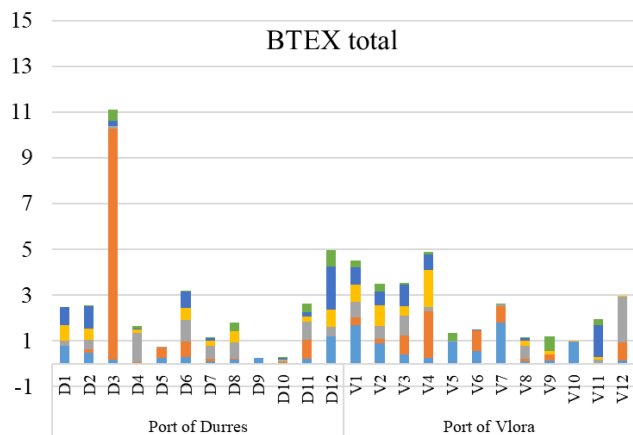


Fig. 8. Total of BTEX in Durres and Vlora ports

Source: Authors' own elaboration

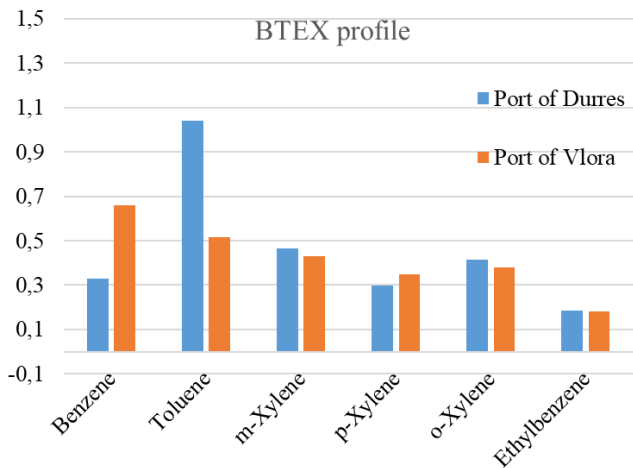


Fig. 9. Profile of BTEX in Durres and Vlora ports

Source: Authors' own elaboration

The data on priority substances in the ports of Durres and Vlora have been processed with multivariate cluster analysis. From the processing data for the main groups of priority substances for both ports, it was noted three main groups (Figure 10). The greatest similarity in the first group shows HCHs, DDTs, and Aldrins. Their similarity exceeds 70%, followed by Heptachlors, which have a lower similarity. In the second group, Chlordanes and Endosulfans show higher similarity, with a similarity of over 80%. This group has similarity with Group 1, with confidence level of more than 40%. In group 3, the greatest similarity is for PAHs and BTEX, with greater than 50% similarity. The similarity of Group 3 with other groups is about 30%. The data for Endosulfans (15%), total pesticides (2%), and PCBs have the least similarity with the other groups. The similarity of these groups can be attributed

to the nature of the substances (organochlorine compounds show similarity, hydrocarbons show similarity), the timing of their use, and their respective physicochemical properties (solubility, degradation rate, etc.). A cluster analysis of contamination levels at each station (Figure 11) revealed seven

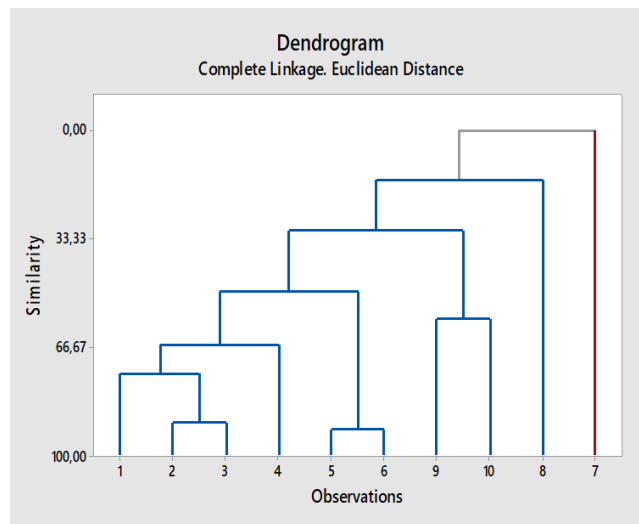


Fig. 10. Cluster analysis for primary substances in water samples from Albanian ports

(1 = \sum HCHs, 2 = \sum DDTs, 3 = \sum Aldrines, 4 = \sum Heptachlors, 5 = \sum Chlordanes, 6 = \sum Endosulfanes, 7 = \sum Pesticides, 8 = \sum PCBs, 9 = \sum PAHs and 10 = \sum BTEX).

Source: Authors' own elaboration

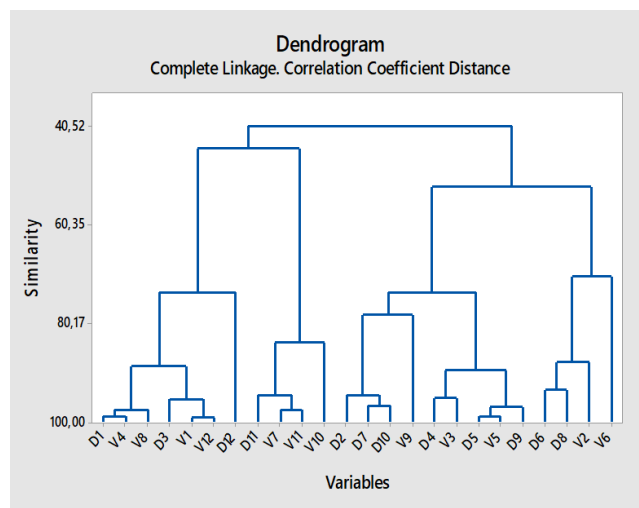


Fig. 11. Cluster analysis for the pollution of each station for the ports of Durres and Vlora

(1 = \sum HCHs, 2 = \sum DDTs, 3 = \sum Aldrines, 4 = \sum Heptachlors, 5 = \sum Chlordanes, 6 = \sum Endosulfanes, 7 = \sum Pesticides, 8 = \sum PCBs, 9 = \sum PAHs and 10 = \sum BTEX).

Source: Authors' own elaboration

groups sharing a similarity greater than 90%. Five groups show similarities above 80%, and two major groups show similarities above 40%. The main groups mainly consist of sampling stations inside and outside the port. There are no visible differences between the Durres and Vlora stations. This could be related to the same origin of pollution in the marine waters of Durres and Vlora ports. Individual levels of some pollutants lead to differences between stations and between ports.

Conclusions

Priority substances were detected in all water samples from Durres and Vlora ports. Due to the geographical location of Vlora Bay, high levels of organochlorine pesticides, PCBs, and PAHs were detected in water samples from Vlora port. Pesticides were found due to past use in agricultural areas near both ports, new arrivals from rivers, water currents, and punctual sources near ports, etc. Pesticide degradation products were found in high concentrations compared to their active products. This fact is related to the previous use of pesticides in Albania and their degradation processes. The presence of PCBs, PAHs, and BTEX in seawater samples from both ports may be related to intense shipping traffic, industrial activity in the cities of Durres and Vlora, and atmospheric deposition. Mechanical activity may be a terrestrial source of PCBs and hydrocarbons in seawater samples from Durres and Vlora. Some large fires were recorded near these areas during this period, which could affect the contamination levels in both study areas. Levels of individual organochlorine pesticides, PCB markers, PAHs, and Benzene in water samples for both ports were lower than permitted levels for surface waters according to EU Directive 2013/39 and Albanian norms. The exception was for some individuals (in some stations), who were found in relatively higher concentrations than others. Monitoring of organic pollutants in the waters of Durres and Vlora ports should be ongoing, as these areas can be affected by many sources of pollution.

Acknowledgments

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