



# **Integrated Ecological Assessment of Heavily Polluted Sedimentary Basin within the Broader Industrialized Area of Thriassion Plain (Western Attica, Greece)**

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**Abstract:** The Thriassion Plain, the Saronikos Gulf and Eleusis Bay, Western Attica in Greece, receive pressures from the enormous industrial activity, as well as the Athens metropolitan area and the Piraeus port. Therefore, it is considered as brownfield in the Eastern Mediterranean Sea. The multicomponent industrial activity has impacted the soil, the groundwater of Thriassion Plain and the coastal marine sediments of the adjacent Eleusis Bay, part of Saronikos Gulf as well as a brackish lagoon, Koumoundourou Lake. The industrial activity is expressed by high contents of metals, and oil products. This study presents the pollution record of selected published papers that indicate the temporal evolution of legislated polluting compounds, supporting researchers to provide solutions and policy makers to focus on the whole spectrum of potential policy alternatives.

Keywords: water quality monitoring; heavy metals; PAH contamination; environmental status; groundwater risk assessment; bioavailability index; pollution indicators; Eleusis Bay; Koumoundourou Lake; Saronikos Gulf

# 1. Introduction

Human pressures upon ecosystems through the misuse of resources, pollution and resource over-exploitation, are the main threat to ecosystems and potentially our wellbeing within those ecosystems. The Thriassion Plain is a plain of Attica peninsula, located 25 km west of the metropolitan area of Athens, surrounded by Aigaleo Mt (453 m) to southeast, Parnitha Mt. (1410 m) to the north and Patera Mt (450 m) to the west. It covers a surface of 120 km<sup>2</sup> and comprises the towns Eleusis, Aspropyrgos, Mandra, and Magoula (Figure 1). Eleusis used to be an eminent culture center of antiquity, the remnants of which still dominate in the town.

The areas' vicinity to the capital Athens made it favorable for progress: The urban growth was connected to the industrial spread, reaching its peak in the early 2000's. Since then, a retardation has been observed. Heavy industries, such as two oil refineries, wood/rubber plastics, chemicals and petrochemicals, production of petrol derivatives, iron and steel production, machinery, transport, warehouse and logistics, scrap metals reuse, along with two of the largest shipyards of Greece operate in the broader area. A military airfield, with depots and storage tanks at west, is included in the brownfield profile of the study area. Some agricultural land and livestock/poultry farming units also operate from past years. The biggest landfill of Attica Prefecture is situated at the northeastern boundary of the study area. The modern unit has replaced an older uncontrolled landfill



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**Copyright:** © 2022 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (https:// creativecommons.org/licenses/by/ 4.0/). that operated until 2007 without a background membrane. The waste leakages proved to be contaminated by organic and inorganic substances [1,2]. Evidently, the economic growth occurred under unsustainable conditions with lack of infrastructure. Additionally, unauthorized dumping of industrial by-products or any type of junkyards (used tires, car recycling) has converted the region to a waste sink of Athens.



**Figure 1.** The study area. The Thriassion Plain and the broader region. The towns Aspropyrgos and Eleusis are the main urban areas within the basin. Eleusis Bay, the inner part of Saronikos Gulf, washes the west coastline of the capital Athens and Piraeus port. The two biggest petroleum refineries of Greece, the Refinery of Aspropyrgos and the Refinery of Eleusis are part of the environmental issue. At the southern channel of Eleusis Bay, the Wastewater Treatment Plant in the islet of Psitalia (WWTP) receives the researcher's attention.

Overall, this review comprises the chronicle of the contamination evolution by organic (oil products) and inorganic substances (metals) in soil and groundwater of Thriassion Plain, which is assumed to be among the most severely polluted regions in Greece. Particularly, this study refers to pollution data from seawater and sea bottom sediments from Eleusis Gulf and related ecosystem response [3–7], which washes the study area and, thus, receives its drainage load, as well as significant geochemical data review. It was estimated that selected scientific research on the atmospheric quality should be integrated. The data provided in this review may be combined with other published results of hazardous substances either in all means (air, surface drainage), or in the broader water body of Saronikos Gulf [8–11]. The studies reviewed were chosen among the scientific journals, apart from a few exceptions such as reports of national scientific institutions of the country, or selected conferences, useful to integrate the contamination status. Moreover, we added a few unpublished technical reports that were regarded as helpful background data. Local authorities, researchers or policy makers may be assisted concerning applying further management plans or assessing the effectiveness of any remediation applied in the area. Additionally, it could support the criteria of background concentrations for the amelioration of Europe's legislation. In these terms, it is necessary to shift from the common remediation project to a contingency planning and prolepsis. Waste must be recognized as an integral

part of the total resource stream and of the technology that facilitates the exploitation of natural resources towards viable recovery systems.

### 2. Study Area

# 2.1. Climate and Geology

The prevailing climate of the broader area of Thriassion Plain is characterized as Mediterranean subtropical, the same as Attica's peninsula. The industrial operation combined with the increased air temperatures and the existence of a brackish lake result in a milder microclimate [12]. The geological column includes (a) a basement of a volcanosedimentary complex, (b) Triassic and Cretaceous limestones, (c) clays, marly limestones and conglomerates, aged Plio–Pleistocene and (d) Holocene clay, sands and gravels. The Pleistocene sea-level fluctuations of the broader area have induced the deposition of torrential, lacustrine and lagoon sediments within the study basin (Figure 2), about 400 m thick. Thriassion Plain is surrounded by mountains with steep slopes, though the plain itself reaches 100 m in height. Geotectonic activity has influenced the hydrogeological regime and the groundwater flow. In particular, the Pliocene–Pleistocene deposits are characterized by various hydraulic characteristics. Only a few researchers have conducted pumping tests in the carbonate as well as in Quaternary sediments [13].



**Figure 2.** Geological map of the broader area of Thriassion Plain. The bulk of the studies reviewed involve the processes occurred within the Pliocene–Pleistocene formations.

## 2.2. Eleusis Bay and Saronikos Gulf

Our study area is washed by Saronikos Gulf, the aquatic body of the capital city of Athens and the biggest port of the country, Piraeus. The Gulf is semi-restricted by the Attica peninsula in the Eastern Mediterranean Sea. It is divided into the "inner" part at the north, and the "outer" part, at the south Eleusis Bay; the "inner" part of the Gulf is about 20–30 m deep and connected with Saronikos Gulf through two narrow shallow channels. Along the northern coasts of Eleusis Bay, heavy industrial units, such as the

refineries of Aspropyrgos and Eleusis, iron and steel production, cement production and shipyards operate. This activity occurs combined with all shipping operations of the Port of Piraeus, to the east, and the existence of the Wastewater Treatment Plant (WWTP), located on Psitalia Island. It is worth mentioning that all sewage load from Athens was discharged almost untreated until 1994, affecting both the Eleusis Bay and the Gulf of Saronikos. In past years, Attica's wastewater was primarily treated only, while from 2004 onwards, an integrated sewage treatment plant has been operating properly. The Piraeus port hosting the country's shipping and industrial activities certainly impacts the marine ecosystems of the study area which has been monitored by the program MED-POL/UNEP since 1986. The results of this project have indicated that Eleusis Bay exhibits the highest pollution within Saronikos Gulf [14].

Researchers have concluded that Saronikos Gulf and in particular Eleusis Bay is characterized by different morphology and bathymetry [15], along with intense anthropogenic interference [4,8,9,16,17]. The Eleusis Bay marine system is characterized by hypoxic to anoxic conditions developing annually for about five months, due to freshwater inputs and limited exchange that result in strong seasonal density-driven stratification of the water column and consequent limited oxygen circulation into the basin [14,18]. The differences in anoxia magnitude, the accumulation of organic matter and the weak water mass refreshing intensify the problem. Temporal variations in nutrients correspond to a recent pollution decrease rather than climate variations. In contrast, the variation of anoxia intensity seems to be connected to local climate variations [14].

Extreme stratification of the water column, regardless of the pollution status of Eleusis Bay may develop anoxic conditions below 15 m, basically during summer [19,20]. A marine current from the west provides a net water supply of about 240 to 450 m/s<sup>2</sup> per year and estimated sedimentation rates range between 0.5 and 0.8 cm/year [21]. Significant water mass renewal in the eastern part of the Bay has averted ecological decline. On the contrary, in the western part of the bay, which is deeper and less affected by pollutants, the renewal of water is very slow. The operation of the Wastewater Treatment Plant of Psitalia Island has fostered a permanent decrease of dissolved oxygen below 100 m of the western basin. Despite all environmental pressures and due to a sequence of favorable bio-geochemical characteristics, the area's biodiversity remains remarkable, in a way that rare and threatened species (*Pinna nobilis, Pecten jacobaeus*) are present.

The Nested Environmental Assessment Tool (NEAT), a method of assessing different ecosystem features and geographical areas, was applied by [9] in Saronikos Gulf. When combining the integrated habitats with the spatial units the scientists re-confirmed that Psitalia and Eleusis Bay are the most impacted settings of Saronikos Gulf.

# 2.3. Koumoundourou Lake

Koumoundourou Lake is a coastal, shallow, brackish lagoon at the northern shore of Eleusis Bay, in the eastern boundary of Thriassion Plain, with a mean depth of 1.5 m. Submerged springs on the northwest part of the lakeshore recharge with fresh water. When the seawater, the spring freshwater and the rainwater are mixed, a meromictic brackish environment is produced with a 10.50‰ average salinity level [22,23]. The Lake communicates with the sea through an artificial channel (weir) which has outflowed its water to the Gulf of Eleusis since 1994. This construction averted flooding of the adjacent highway Athens–Corinth (Figure 3a). Later, in 1998, the weir was transformed in such a way to permit less water outflow into the sea. This corrective intervention forced the Lake's water level to rise about 20 cm, increased the hydraulic head and reduced the average salinity to 8‰ approximately. Moreover, a horizontal borehole was introduced in 2001, at the northern boundary of the Lake, along with two other pumping valves, aiming at oxygenating the bottom sediments, to accelerate biodegradation (Figure 3b). Additionally, a floating barrier installed nearby restrains any oil products from their diffusion. These corrective measures, though not operating properly, have averted the deterioration of the lake's quality, as described in the following paragraphs.





Koumoundourou Lake



**Figure 3.** (a) A simplified design of the "weir" (channel) that discharges the lake's overflow to Eleusis Bay (left) and a picture of the lake (right). (b) Conceptual sketch of the horizontal borehole (blowing valve) and the cleaning valves installed in Koumoundourou Lake for its protection.

Apart from plankton, benthos, mullets and eels, the Lake hosts about 50 species of birds, mostly migratory ones, whereas wild duck populations as well as swans have sporadically appeared in the area in the past. The Koumoundourou Lake has been denoted as a protected archeological site, as registered in the Official Journal of the Hellenic Republic (5/B/8.1.1974) as well as an area of outstanding natural beauty. It is also comprised in the Mediterranean Wetland (MedWet) database. However, the outstanding scenery has been affected by significant pollution sources: A military camp which serves as a fuel supply station, the biggest oil refinery of the country and a marble-cutting small industry [24].

# 3. Multiple Anthropogenic Impact

#### 3.1. Atmospheric Pollution

The study area is regarded as one of the most arid urban regions in Europe; its climate regime, influenced by the intense urbanism of the four recent decades, is featured by moderate warming, wind speed and direction changes and multiple extreme events [25]. The multi-component industrial rise since 1960, though following a declining trend to date, has determined drastic changes in the use of land, which has led to environmental degradation and a potential impact on the local climate regime, expanding in the adjacent areas [26]. As a result of the aridity, the soils are of low quality as reflected by poor vegetation, which makes the need for sustainable land management mainly focused on soil water maintenance and the further prevention of landscape degradation [27].

In 1992, Asimakopoulos et al. [28] studied the sulphur hexafluoride (SF6) releases from one of the smokestacks of the Hellenic Oil Refineries, also tracking them in the broader area of western Attica. Sulphur dioxide (SO<sub>2</sub>) concentrations gradually decrease over the years thanks to fuel improvement, whereas nitrogen dioxide (NO<sub>2</sub>) concentrations remain almost the same during the examined period with, perhaps, a small decreasing trend during the last few years. Yet, the levels recorded are less than those observed in the Athens area, and below CEC limits: the sea breeze is, potentially, either beneficial as effective ventilation, or detrimental through the transportation of the polluting elements [29]. Concentrations of Ca<sup>2+</sup>, Mg<sup>2+</sup>, K<sup>+</sup>, Na<sup>+</sup>, NH<sub>4</sub><sup>+</sup>, Cl<sup>-</sup>, NO<sub>3</sub><sup>-</sup> and SO<sub>4</sub><sup>2-</sup> were determined in airborne particulates and were attributed to the industrial activity. Specifically, ammonium is likely to originate from the oil refineries and is enriched in the fine particles, chloride is due to a coastal effect and nitrate concentration is due to vehicle emissions. In the urban area the enrichment of secondary aerosols, ammonium, and sulphates, in the fine particle fraction reflects high traffic densities and domestic heating from the highly populated cities [30]. Very recently, Koukoulakis, et al. [31] studied the trace elements in PM<sub>10</sub> (Particulate Matter < 10  $\mu$ m) samples in the air of the town of Eleusis. Only Al, Ba and Zn were abundant while Cr, V, Mn, Pb, Cu, Ni, Ga and Rb were detected in traces. For PM<sub>10</sub>, Al, Fe, Zn and Ba demonstrated the maximum concentrations during winter. The Enrichment Factor (EF) identified the industrial area as the main polluting source, specifically for Cd, whereas Pb originated from combustion processes up to municipal solid waste incineration. Ultimately, the air pollution of the study area has a strong impact on residents' health, as the assessment of the excess lifetime cancer risk revealed that two people out of 30,000 are at risk.

# 3.2. Pollution of Soil and Sediments

## 3.2.1. Pollution of Soil from Metals in Thriassion Plain

An early study of the soil pollution from metals was published in 1982 by Nakos [32]. The total concentration values of Pb, Cd and extractable  $SO_4^{2-}$  in surface calcareous soil samples were reported as remarkably higher than those in an allegedly "clean" soil. In addition, total Pb, Cd, Zn, Cr and S concentrations in samples of olive leaves from the same area were reported up to 21 times higher than those tracked in relevant samples from rural sites, where the common concentration ranges of Pb and Cd are 20–50 ppm and 0.03–5 ppm, respectively. Sampling occurred in the vicinity of a steel industry and other metal works. In 1994, the Institute of Geological and Mineral Exploration of Greece carried out a pilot geochemical pollution report [33]. The metals reported according to their concentrations and frequency were Pb > Zn > Cu > Cd > Cr > Ni > Mn > V > Co. The existence of Pb, Zn, Cu and Cd was mainly attributed to the leakage of oil products and the industrial operation as well. The most polluted locations were identified along the coastline of the town of Eleusis, east of the town of Aspropyrgos, from industrial activities and in limited locations around industries. However, the occurrence of Ni, Cr, Co, Mn V and Cd, away from polluting sources, potentially originated from the existence of ophiolitic and bauxite complexes. Massas et al. [34] pointed out that the concentrations, as such, do not reveal the source, the interconnections and the potential effects of target metals and biota. The Geoaccumulation Indicator (Igeo), the Enrichment Factor (EF) and the Availability Ratio  $(A_{Rs})$  when applied, may relate the source and the outcome. According to that study, the concentrations of Cr, Zn, Ni, Pb, Co, Mn, Ba, Cu and Fe were not considered as extremely high. The Igeo revealed a geogenic origin for Fe and Mn, a moderate contamination of the rest of the metals, but a partly high impact for Pb. The calculated EFs indicated the industrial emissions as polluting source. A follow-up was published by Gasparatos et al. [35]. Apart from determining the total and available concentrations of Cu, Zn, Pb, Ni and Cr, their chemical partitioning, was introduced by calculating the mobility factor. The mobility order found was: Pb > Cu > Ni > Zn > Cr. More concentrations of Al, As, Cd, Co, Cr, Cu, Fe, Mn, Mo, Ni, Pb, V and Zn were identified in Antoniadis et al. [36], a research that reported the Potentially Toxic Elements (PTEs) in both soils and plants and the human risk from soil ingestion. To achieve this, the authors calculated the Transfer

Coefficient (*TC*), the Contaminant Factor (*CF*) and the Igeo. The metal concentrations, apart from Al, Co, Fe, Mn, Mo and V, were reported higher than maximum thresholds. However, the metal concentrations in indigenous plant tissues were lower than expected, possibly due to their self-cleaning capability (excluding behavior with time). Among them, Cu and Zn soil-to-plant coefficients were the highest, which can possibly be attributed to recent Cu-Zn-relevant industrial activity. The risk assessment analysis demonstrated that As contributed more than 50% of the health risk related to soil ingestion, followed by Cr, Pb and Mn [36]. The resulting point of this multi-component research was the slow, continuous metal accumulation over time and the need for preventative and/or pollution-restoring actions. Table 1 summarizes the ranges of concentrations of metals in soil and tissues, along with the laboratory method, as provided in the mentioned studies above.

The overall remark from the few published studies about the soil pollution in Thriassion Plain is the existence of high levels of metals that locally have a native contribution from the bauxite formations. Metal and oil industries are evidently the core polluting sources. We cannot find a clear trend since the mentioned studies do not correspond to common sampling locations, though they are in the defined study area of Thriassion Basin. About biota, the environmental indices are useful guides of the contaminants' fate in the soil system and toxicity to live tissues.

## 3.2.2. Pollution of Sediments and Tissues of Eleusis Bay from Hydrocarbons

Oil industries in Thriassion Plain as well as shipping activities in Eleusis Bay are potential pollutants of hydrocarbons and metals of the groundwater and the seawater, respectively. A comprehensive published work on the pollution from Polycyclic Aromatic Hydrocarbons (PAHs) in Eleusis Bay sediments was carried out by Sklivagou et al. [37]. The total  $\Sigma$ PAHs values ranged between 481 and 11,182 ng/g dry weight, indicating a high degree of contamination, parallel to those from Barcelona harbor (Spain), Manakau harbor (New Zealand) and Penobscot Bay (USA). Particularly, Benzo(a)pyrene, a PAH with a serious toxicological profile, ranged from 12.5 to 301 ng/g. The pyrolytic origin of the target compounds was attested by the values of specific ratios (Phenanthrene/Anthracene and Fluorene/Pyrene) widely used in researchers [38]

The PAHs' core profiles were studied in the center of Eleusis Bay by Hatzianestis et al. [39]. From the downcore sediment profile each PAH was determined in every 1 cm layer with a GC-MS system, at up to 32 cm depth. The highest concentrations were detected between 2 and 3 cm core layer, while the surface and the deepest layers reported lower values. A possible decrease of organic pollution may justify the distribution of these values. The PAHs of pyrolytic origin were the main components of PAH mixtures, which is typically expected in marine sediments, as these compounds are more lipophilic and present greater resistance towards biological and/or physicochemical degradation, rather than the petrogenic ones.

In a further study of the PAHs' origin, aliphatic hydrocarbons and certain metals were reported by Sklivagou et al. [40]. In that study, the total PAH (TPAH) concentrations in surface sediments of Eleusis Bay ranged from 481 to 11,181 ng/g. Fluoranthene, Phenanthrene were the dominant PAH members, indicating combustion processes rather than the fossil fuel leakages. About the rest of Saronikos Gulf, the PAH members ranged from low molecular weight, two to three ring compounds, particularly Naphthalene and Phenanthrene, up to Indeno(1,2,3-c,d)Pyrene, compounds with five and six rings, respectively. The values of PAHs and AHC confirm the long-term oil pollution of sediments. The ratios of Phenanthrene/Anthracene and Fluorene/Pyrene revealed more pyrolytic—from combustion activities—rather than petrogenic origin, meaning, a multi-source pattern of PAHs [40]. Investigation of total Fe, As and Sb along with their Sc demonstrated high concentrations in Eleusis Bay sediments, whereas Cr was detected enriched in the sediments near the Athens sewage treatment plant (WWTP).

	Pb	Cd	Zn	Ni	Cr	Mn	S	Cu	v	Ba	Со	As	Al	Fe	Source		
Soil	14-595	3–30	52-594	52-321	36-425	256-1585	4-1440										
Olive leaves	0-854	0.4-8	10-130	2–17	0.8–22	5-108	0.06–2.1								Nakos [32] Analytical		
Cabbages- µg/g fresh weight	0.82–40														method: Not specified		
Soil ppm	13–5375	LoD-1.6	46-1750	24–740	14–595	245-3305		40–542	9–130		2–26				Kaminari [33] AAS		
Total mg/kg	60.4–693		76.5–690.8	36.4–297.7	42–141	160.3–585.5		12.3–261.6		247.2– 1558.8	5.6–31.5			7.0–24.5 (mg/g)	Massas, Kalivas, Ehaliotis and		
Available mg/kg	1.6-37.4		0.8–32.2	0.5–3.7	0.1–2.5	0.7–24.3		0.5–51.3		4.5-42.8	0.4–2.0			0.9–13.9 (mg/g)	Gasparatos [34] AAS		
Total mg/kg dry weight	67.5–215.8		73–545.5	41.4–175	41.4–148.7			15.4–318.2							Gasparatos, Mavromati,		
Available mg/kg dry weight	2–33.9		0.9–25.6	0.5–3.6	0.1–2.6			0.5–59.9							Massas [35] AAS		
Total mg/kg	15.5–2284.1	0.1–16.9	128.6– 3178.4	37–658	57.0-1244.4	171.3– 1349.9		33.4– 10,682.8			9.5–46	17.1– 110.9	14.1-49.1	7.8–74.5 (%)	Antoniadis, Golia, Shaheen and		
In plant tissues	4.88–11.10	0.57-0.69	33.47– 167.41	1.09-4.96	1.02-4.83	8.35-126.45		13.94–61.89						50.17– 646.39	Rinklebe [36] ICP-OES		
Maximum Contaminant levels (MCLs) in soil	300	3		75	0.01 μg/L in freshwater			140				10 μg/L in fresh- water			86/278/EEC for Cd, Cu, Ni, Pb, Zn. EPA 816-F-01-004 for As		

Table 1. Ranges of metal and other inorganics concentrations in soil and plant tissues in Thriassion Plain.

The PAH research was expanded in surface seawater and tissues of gills and black mussels, *Mytilus galloprovincialis* by Valavanidis et al. [41], a species used as bio-tracer for metal contamination of coastal areas, including the Eleusis Bay. The concentrations of total PAHs ( $\Sigma$ PAHs) in seawater ranged from 425 to 459 ng/L, in locations between Eleusis Bay and Salamis Island, of pyrolytic origin, connected with combustion processes from marine and industrial activities. The  $\Sigma$ PAHs in the gills and mantles of mussels were significantly higher, in the range 1300–1800 ng/g. The dominant PAH members had two-, three- and four-ring compounds in seawater, where all 17 PAH members were identified and measured in mussel tissues. Mussels accumulate PAHs from the water media, whereas the sunlight and temperature reduce them in surface waters. Thus, they have been characterized as "sentinel" organisms concerning monitoring PAH contamination [42].

Elevated concentrations of PAHs in sediments of Eleusis Gulf in two sampling locations were also reported in Botsou and Hatzianestis [43]. All PAH-members were detected, including Benzo(a)pyrene 139–263 ng/g and Indeno[1,2,3-cd]pyrene 87–108 ng/mg. While total PAHs ranged from 1.616 to 3.569 ng/g, values which were lower than previously published levels, Eleusis Bay sediments still displayed a high PAH contamination. These levels approximate past studies and re-depict the industrial activity of the coastal region of Attica, which includes steelworks, oil refineries and shipyards. The origin calculated from PAH ratios suggested fossil, biomass, or coal combustion. Moreover, the comparison of the detected PAH concentrations with other industrial areas such as Kavala port in Greece (reaching the 55,000 ng/g total PAH) and the harbor of Alexandria in Egypt (up to 131,150 ng/gr total PAH) show a high polluted environment. Finally, Phenanthrene, Fluoranthene and Pyrene were found in abundance in Eleusis Bay sediments. Specifically, Benzo(a)anthracene, allegedly carcinogenic, with maximum contaminant level 0.1 ppb  $(0.1 \ \mu g/L)$  in water, ranged from 480 to 760 ng/kg dry weight. In the same study, the PAHs identified in Koumoundourou Lake were Anthracene, Fluorene, Phenanthrene, Benzo(a)pyrene, Fluoranthene and Pyrene [44].

It is worth mentioning the investigation of Persistent Organic Pollutants (POPs) such as PAHs, PCBs, Hexachlorocyclohexanes (HCHs) and DDTs in pellets collected from the beach of the broader area of Eleusis Bay by Karapanagioti et al. [45]. The reported PCB concentrations were elevated and connected with all industrial processes. The relatively lower concentrations of HCHs and DDTs are mostly due to activities other than agricultural. As for PAHs, Chrysene showed the highest concentrations in the sampled pellets as well as Fluoranthene, Phenanthrene, Anthracene and Indeno[1,2,3-cd]pyrene. It was also noted that the PAH values in the pellets were about two times lower than in the mussels. High concentrations of PAHs were also reported in water since, within the PAH group, there are hydrophobic members which are distributed differently in various media (e.g., water versus pellets or mussel gills and mantles). Table 2 presents the ranges of concentrations of the organic pollutants (PAHs, POPs and AHC), as well as the laboratory method applied, of the mentioned studies.

In total, the Thriassion Basin accumulates variable activities, such as oil refineries, shipyards, urban areas and a national highway. This is reflected in PAH diagnostic ratios, which, in total, indicate mixed fuel and coal combustion sources. Based on the data of this study and the International Pellet Watch program, pollution in the broader Saronikos Gulf, Greece, is comparable to other heavily industrialized regions worldwide.

	Eleusis Bay																		
	ΣPAHs	NaP	Acy	Ace	Ant	Flu	Phe	FlA	Pyr	B(a)P	Per	Ba(a)A	InPy	Chr	AHC	DDTs	PCBs	HCHs	Source
ng/g dw	481– 11,182						52.8– 1769	25.4– 1885	22.9– 1408	12.5–301	3.1– 110.3				85.7– 1062				Sklivagou, Varnavas and Hatzianestis [37] GC-MS
ng/g dw			0.3–16.5	0.2–11.3	0.3–27.8	1–18.4	3.3– 111.7	1.9–207	2.5– 201.2	1.6– 203.1	0.6–56.2		6.1– 302.1						Hatzianestis, Rori, Sklivagou and Rigas [39] GC-MS
ng/g dw	481– 11,181	4.2- 364.3	Non de- tectable	2.78– 701.5	6.6– 454.1	14.8– 583.2	54.9– 1769 *	25.4– 1885 *	22.9– 1408 *	12.5–268 *	4.4–98.9	l t	Non de- tectable		85.7– 1062 *				Sklivagou, Varnavas, Hatzianestis and Kanias [40] GC MS * same data as [37]
ng/L In seawater- mean values ± SD	425–459	98 ± 5	Non de- tectable	$36\pm4$	$28\pm2$	$48\pm2$	$78\pm 6$	$66 \pm 7$	$25\pm3$	20 ± 4		l t	Non de- tectable						Valavanidis, Vlachogianni, Triantafillaki, Dassenakis,
ng/g dw gills of mussels ± SD	1300– 1800	$166\pm 8$	Non de- tectable	$127\pm 8$	$43 \pm 3$	$320\pm12$	$285\pm9$	$198\pm22$	$54\pm4$	$26\pm2$			$34\pm3$						Androutsos and Scoullos [41] HPLC
ng/g In sediments			4.7–86	2.4–54	16–57.7	10.2– 62.9	42.6–314	140.9– 449	187–384	139–263	25.7– 68.3		87–154						Botsou and Hatzianestis [43] GC-MS
ng/g pellets for PAHs pg/g pellet for PCBs ng/g pellet for HCHs					36		34	86		3	2		15			2.2-9.9	110– 270,000	3.5	Karapanagioti, Endo, Ogata and Takada [45] GC-Ion Trap- MS for PCBs and PAHs GC-ECD for DDT, HCHs

Table 2. Cont.

	Eleusis Bay																		
	ΣPAHs	NaP	Acy	Ace	Ant	Flu	Phe	FlA	Pyr	B(a)P	Per	Ba(a)A	InPy	Chr	AHC	DDTs	PCBs	HCHs	Source
ng/g dw sediments					210–240	190–270	1700– 1800	1200– 1500	1350– 1500			480– 760		630–690					Hahladakis, Smaragdaki, Vasilaki and Gidarakos [44] GC-MS
								K	Coumoundo	urou Lake									
ng/g dw sediments					110–115	110–115	50–580	70–150	75–780			760– 900		690–790					Hahladakis, Smaragdaki, Vasilaki and Gidarakos [44] GC-MS

Maximum Contaminant Levels Ba(a)P in drinking water 0.0002 mg/L. Naphthalene (Nap), Acenapthylene (Acy), Acenapthene (Ace), Anthracene (Ant), Fluorene (Flu), Phenanthrene (Phe), Fluoranthene (FlA), Pyrene (Pyr), Benzo[a]pyrene (B[a]P), Perylene (Per), Indeno[1,2,3-cd]pyrene (InPy), Chrysene (Chr), Benzo(a)anthracene (BaA).

#### 3.2.3. Pollution of Eleusis Bay from Metals

In 1977, the primary effects of metals in sediments were published by Grimanis et al. [46]. As, Hg, Cr, Zn, and rare earths were among the first elements investigated in the broader Saronikos Gulf, from Piraeus, Keratsini outfall (the sewage discharge of Athens, before the modern wastewater treatment in Psitalia Island) up to Eleusis Bay. All determined concentrations exceeded their natural backgrounds, with highest values in Piraeus and Keratsini. The following year, Griggs et al. [47] confirmed the same distribution pattern of metals As, hg, Cr, Zn and Co in the sediments of Saronikos Gulf, indicating shipyards, metal manufacturing and process, untreated municipal and industrial waste as the bulk pollutants.

Another pioneer study focused on Eleusis Bay, by Scoullos [21], reported for the first time the ultra-high total Pb concentrations of 500–600  $\mu$ g/g in core top sediments of the bay, while the baseline pre-industrial levels were about 10  $\mu$ g/g. Shipyards and steelworks were then highlighted as polluting sources. The ratio Pb/Al (Enrichment Factor) indicated the man-derived contribution. The down-core pollution investigation demonstrated elevated Pb concentrations > 100  $\mu$ g/g dry weight in the upper 5 cm.

Kersten et al. 1992, identified the  $^{206}$ Pb/ $^{207}$ Pb isotope ratios in both seawater and sediment samples from this area revealed much lower concentrations than those of local background or industrial sources, influenced by gasoline, as Pb source, before being banned [5]. However, the lateral Pb isotope pattern indicates the Keratsini Sewage Outfall (where the pre-treatment of the Wastewater Treatment Plant of Psitalia is operating), with surface runoff from the greater Attica coastline area, as the primary source rather than direct atmospheric origin [5]. The determination of the  $^{206}$ Pb/ $^{207}$ Pb isotope ratios in seawater "tracked" the contaminant plume following the prevailing wind of the bay.

High values of As  $(21-3450 \ \mu g/kg)$  were reported in the sea bottom sediments east of Eleusis Bay, where both the wastewater effluents of the Athens area and industrial discharge of two industries that produce fertilizer and sulphuric acid were considered as polluting sources [48].

The recording of the total metal concentration is not enough to predict their fate in an ecosystem; their toxicity, mobility and availability are dependent on their chemical form combined with basic environmental parameters. Under this concept, Scoullos and Pavlidou [49] indicated that the metal diffusion in the Koumoundourou Lake is regulated by their tendency to make complexes to organic and inorganic ligands as well as pH and salinity values. More specifically, salinity is related with the very labile fraction of the metals, since metals are bonded with chlorine ions; pH increase is analogous to the presence of the less mobile species, whereas the dissolved organic carbon is correlated with the slowly labile and inert fractions of Pb and Zn, respectively. Scoullos et al. [10] examined the pollution from Cd, Cu, Mn, Ni, Pb and Zn in the broader area of Eleusis Bay, the Saronikos Gulf, in connection with the operation of WWTP of Psitalia, starting in 1995. The authors concluded that there is a steady state of metal concentrations, though the Gulf never stopped receiving significant loads of trace metals, specifically in the eastern water body of Saronikos Gulf. Nevertheless, incidents of elevated metal pollution were then, and may be reported in the future, due to other environmental features, such as water circulation. This research could be considered as the baseline for any later assessment of the footprint of the secondary treatment in the Gulf, including the Eleusis Bay. The study pointed out that the values of the year 2004 for dissolved and particulate Cd, dissolved Cu and Pb and particulate Mn and Ni were among the lowest detected in the Saronikos Gulf since the start of the MED-POL monitoring program. Furthermore, spring concentrations slightly decrease with relation to ones in autumn.

As mentioned previously, mussels are good PAH pollution indicators. The mussel's ability is expanded to fingerprinting metals and metallothionein (MT). The spatio-temporal variability of both heavy metals and metallothionein (MT) concentrations in mussels and seawater was studied by Strogyloudi et al [42] in mussels. The highest concentrations of Cd, Cu, Cr, Ni, Zn, Fe, Mn and Metallothionein (MT) were recorded in the mussels residing

industrial locations (steelworks and shipyard), reporting an increased metal bioavailability. Elevated trace metal and MT levels observed in cold periods (winter) were possibly connected to the type of fuel used for heating. There are multiple routes of exposure and other abiotic or environmental factors such as the suspended particulates baring metal traces.

An up-to-date publication from Karageorgis, Botsou, Kaberi and Iliakis [8,50] integrates the major and trace metals from sediments into environmental indices to export the pollution trend in the broader study area, the Saronikos Gulf, for a long period 1999–2018. By setting up the baseline metal values from dated cores, calculating the Enrichment Factors (EFs) and the multi-elemental Modified Pollution Index (MPI) the pollution trend was provided. The MPI is an index that normalizes the sediment against Al and Fe, providing more reliable qualification [51]. The outcome of these two relative studies exhibited the following: (a) Saronikos Gulf sediments are not enriched in V, (b) Cu, Cr and Ni show low to median values in the west part, while elevated enrichment at the east (corresponding to the western coastline of Attica peninsula), (c) Cu and Zn exhibit high enrichment within the Bay of Eleusis, the "inner" Saronikos Gulf, only locally. However, the metal pollution trends in the bay are decreasing, perhaps after ceasing metal-effluent discharge, or implementing environmental measurements. The MPI results and interpretation are in line with a study of Simboura et al. 2015 that provided similar biotic indices from macro-invertebrates and macro-algae.

An interesting study concerning the concentrations of Chernobyl-derived <sup>137</sup>Cs in the water column of the Saronikos Gulf and Eleusis Bay was published by Evangeliou and Florou [52]. The activity concentrations of <sup>137</sup>Cs ranged between  $1.0 \pm 0.3$  and  $6.5 \pm 0.7$  Bq m<sup>-3</sup> (mean:  $2.7 \pm 1.6$  Bq m<sup>-3</sup>), whereas the maximum values were registered in the interface between water and sediment during autumn and winter because of the thermocline collapse at the end of winter that caused surface <sup>137</sup>Cs to sink into deeper layers. Moreover, the mean surface residence time of <sup>137</sup>Cs in the Saronikos Gulf was estimated to be  $15 \pm 4$  years, whereas the effective and the ecological half-lives of <sup>137</sup>Cs were  $6.2 \pm 1.5$  and  $72.8 \pm 1.9$  years, respectively. Table 3 summarizes the concentrations of metals in seawater and sediments of study area, along with the laboratory method applied, according to the mentioned studies.

	Eleusis Bay																	
Means/Units	РЬ	Cd	Zn	Ni	Cr	Mn	Sb	Cu	Ba	v	Co	As	Al	Fe	Sc	Hg	MT	Source/ Laboratory Method
Sediments ppm			500		150		2				13	40		10,000		0.5–10		Grimanis, Vassilaki-Grimani and Griggs [46] Neutron Activation Analysis
Sediments ppm			781-1868		175–200							82–97				1.1–1.3		Griggs, Grimanis and Grimani [47] Neutron Activation Analysis
Sediments/ µg/g dry weight (dw)	160-500										12.2–13.7							Scoullos [21] AAS
Seawater/	0.16-0.20 dissolved	0.05-0.1						0.8-3						8-41				Kersten, Garbe-Schönberg, Thomsen,
nM	0.5–1.3 particulate							dissolved						dissolved				ICP-MS
Seawater Eleusis Bay and outer	0.05–0.34 dissolved	12–50 ng/L dissolved	1.6–5.4 dissolved	0.51-1.4 dissolved		0.19-1.2 dissolved		0.06-0.72 dissolved										Scoullos, Sakellari, Giannopoulou,
area/ μg/L	0.03–0.3 particulate	0.22–1.1 particulate	0.18–1 particulate	0.02–0.09 particu- late		0.11–0.5 particulate		0.07–0.23 particulate										AAS
Seawater Saronikos Gulf Waste-Water	0.22-0.39 dissolved	11–24 ng/L dissolved	1–2.6 dissolved	0.35–0.45 dissolved		0.26-0.55 dissolved		0.03-0.46 dissolved										_
Treatment outflow-Psitalia	0.05–0.26 particulate	0.22–1.5 ng/L particulate	0.12-0.65 particulate	0.03–0.11 particulate		0.03–0.34 particulate		0.03-0.13 particulate										
Sea bottom sediments Eleusis Bay/ng/g Fe: mg/g					110-320		1.2–15				11–27	17–140		19–190 mg/g	6.3–11			Sklivagou, Varnavas, Hatzianestis and Kanias [40] Neutron Activation Analysis
Mussels Eleusis Bay/ µg/g dw		0.02–5.36	106–709	3–27.5	1.4–59.9	3.4-60.7		2.4–120						72–2040			73–358	Strogyloudi, Angelidis, Christides and Papathanasiou [42]
Seawater Eleusis Bay/ Dissolved metals		1–3.3	6.7–14.2	6.2–15.8	0.5-4.6	2.8–13		1.8–2.3						7.9–14.3				Spectrophotometry for Metallothionein (MT)
Seawater Eleusis Bay/ Particulate metals		0.5-41.3	0.05-6.38	32–3897	129–2663	0.18-8.59		28-7448						9–56.5				-
Sediments mg/kg dw	85.45-123.57		251.78– 363.78	48.49– 71.23	143.12– 158.46			145.36– 169.32				6.9–7.65						Hahladakis, Smaragdaki, Vasilaki and Gidarakos [44] ICP-MS
Sediments mg/kg dw Saronikos Gulf	5–374		15-982	9–392	19–544	62–5736		7–365		5–121	1–27		0.07–7.31 (%)					Karageorgis, Botsou, Kaberi and Iliakis [50] X Ray Fluorescence
Air /town of Eleusis ng/m <sup>3</sup>	0.04–164	0.01-2.63	37.5-4678	2.2-5.53	11.6–70.7	3.53-44		1.62-83.4	836– 15,942	6.96–39.1		0.2-5.53	6329– 46,216	107–2474				Koukoulakis, Chrysohou, Kanellopoulos, Karavoltsos, Katsouras, Dassenakis, Nikolelis and Bakeas [31]
									Koumoun	dourou Lake								
Sediments ppm	12-220	0.1-0.8	62–715	8-44	21-56	46-800		20-90			9–12			12,332– 11,835				Conides and Parpoura [53]
Water column ppm	9.5–11	1.4-2.10	9.4–19.6		7–12			2.7-22.7						2.2-7.6				

# Table 3. Ranges of metal and other inorganic concentrations in, seawater, sediments, live tissues in Eleusis Bay, air and Koumoundourou lake.

Tab	le 3.	Cont.
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									Elei	usis Bay								
Means/Units	Pb	Cd	Zn	Ni	Cr	Mn	Sb	Cu	Ba	v	Со	As	Al	Fe	Sc	Hg	MT	Source/ Laboratory Method
Sediments µg/g			57-176	16-84	29-186	97–399		10-46		12-70	3–9	13–26						Karageorgis, et al. [54] XRF
Sediments mg/kg dw	31.78-201.28		47.28– 181.97	19.23– 90.17	31.26– 86.81			13.82– 32.87				0.35– 10.86						Hahladakis, Smaragdaki, Vasilaki and Gidarakos [44] ICP-MS

#### 3.3. Water Quality and Sediments Status of Koumoundourou Lake

A pioneer ecological study of Koumoundourou Lake was published by Conides and Parpoura [53] and Conides et al. [24], where heavy metal concentrations, such as Mn, Ni Co, Cd, Zn, Cr and Pb both in sediments and water column of the lake were reported as very low in Koumoundourou Lake in comparison with other areas of Greece. An underground leakage of oil products from a surrounding army camp was also reported. The latter study concluded that water circulation along with the wind effect may avert bioaccumulation of the polluting compounds.

The existence of trace metals and other organic substances along with the groundwater vulnerability of Koumoundourou Lake were correlated by Dimitriou et al. [22]. This study connected the nature of the carbonate aquifers of the broader area with the Lake's vulnerability to pollution from oil and metals stemming from the refinery, the military camp and, mainly, the landfill site northeast of the lake. A vulnerability map demonstrated that a low-permeable formation across the coastline protects, typically, the underground water system from polluting substances. Nevertheless, the industrial activities burden the relatively "protected" groundwater system of the catchment. At the northern hilly side of the lake, the carbonate background delivers a pollution-vulnerable environment.

The calculation of the Enrichment Factor (EF) was considered more precise concerning the Koumoundourou Lake's coarse-grain biogenic sediment characterization. According to Karageorgis et al. [54], the identification of the concentrations of polluting compounds does not depict the degree of contamination. The EF for each element revealed a meaningful enrichment in heavy metals up to ten times over threshold limits. More specifically, As is not enriched, Co and Zn are slightly enriched, Cr, Mn and Ni are moderately enriched, V, Cu and Pb are strongly enriched. The reported accumulation of Pb, Cu, and Zn may also be attributed to the atmospheric runoff. When correlating two coastal sediment samples (Eleusis Bay) with the lake ones, the coastal samples appear to be contaminated in all heavy metals and metalloids in the order Cr > V >Mn > Cu > Ni > Pb > Co > Zn > As > Mo. It was assumed that the main source of contamination could potentially be the underground springs that co-discharge hydrocarbons and heavy metals from the broader industrialized area (for V and Ni, specifically). Besides, old leaded gasolines may have potentially been trapped underground. Cr and CU originate from industrial smelting and various types of combustion. This work also considers the Athens Landfill Site as a potential pollutant of the broader area of the lake.

Hahladakis et al. [44] reported the pollution from Cr, Ni, Cu, Zn, As and Pb and PAHs in the sediments of Koumoundourou Lake and the Eleusis Bay. The evaluation of the human influence was achieved with the use of pollution indicators, such as the modified contamination degree (mCd) and the geo-accumulation index (*Igeo*). Additionally, the Sediment Quality Guidelines (SQG) provided the assessment of the biological effects to aquatic biota. The results demonstrated a moderate metal pollution of the sediments of Koumoundourou Lake, while 60% of the samples were impacted by PAHs and are presumably associated to toxic biological effects. The existence of the barrier at the northern side of the Lake, it is assumed, re-feeds the rest of the lake system with these organic compounds. The results were compared with the pollution status of Eleusis Bay.

Water quality of Koumoundourou Lake in terms of nitrates and phosphates was thoroughly examined by Markogianni et al. [55]. Nitrite concentrations ranged from 0.001 to 0.26 mg/L in the water column, phosphate concentrations ranged from 0.003 to 0.019 mg/L with an average of 0.006 mg/L, whereas total phosphorus concentration exceeded 20  $\mu$ g/L, suggesting human impact. According to the OECD classification system through phosphorous values, Koumoundourou Lake is characterized as oligotrophic, occasionally as hyper-oligotrophic.

Similar prospect about the Lake's water quality were published by Mentzafou et al. [56] in a relatively recent study comprising the evolution of important ecological features. The electric conductivity range between 8982 and 27,400 lS/cm depicts a dropping trend when comparing with values since 1984, thanks to the uplifting of the weir. The freshwater of

the springs to the northeast, as well as the groundwater up-flow, may vary significantly, same as the pollution incidents, and the vegetation (macrophytes). The correlation of past values of dissolved oxygen with the current ones, in the range 4.9 and 15.0 mg/L, gives a hint of good aeration. Fish populations have dramatically diminished since the weir was constructed, potentially. Hence, aquatic plants create adverse conditions for the lake's ecosystem. Nutrients and phosphates present a decreasing trend after the year 1998. The soluble concentrations of Cd, Zn, Cu and Fe are lower than the limits defined by EU legislation for drinking water until 2012. Pb, Ni and Mn concentrations surpassed the legislated limits (10.20 and 50  $\mu$ g/L, respectively) occasionally. Certain concentrations showed a decreasing trend in Cu, Cd, Mn, Pb, Zn and Ni. The lake's sediment heavy metal concentrations may be considered as relatively high when compared with respective values from coastal lagoons in the Mediterranean region and worldwide.

## 4. The Groundwater Status—Pollution and Hydro-Geochemistry

First works about the hydrogeological conditions in the broader area of Thriassion Plain were published for the Institute of Geological and Mining Exploration of Greece by Kounis and Siemos [57,58], who suggested the model of two unconfined aquifers: The upper in the Pleistocene sediments and the underlying part in the Mesozoic limestones. Recent studies remodeled the groundwater system considering the existence of a multi-layered aquifer system, which indicated out a more complex underground water flow [13,59].

## 4.1. Groundwater Pollution from Hydrocarbons and Metals

Pollution from petroleum hydrocarbons in Thriassion Plain includes aliphatic and aromatic compounds, such as BTEX (benzene-toluene-ethylbenzene-xylene) and Polycyclic Aromatic Hydrocarbons (PAHs), basically, since they are legislated as dangerous for human health and the ecosystems. According to internal reports of the Hellenic Refinery in Aspropyrgos, routine monitoring of the groundwater quality has been accomplished within its settlement. The authors selected published papers that depict the results of the monitoring.

In 2002, the Bioslurping technology was applied for four years in the facilities of the Hellenic Refinery of Aspropyrgos to remediate the polluted aquifers by Aivalioti and Gidarakos [60]. Despite the evidence of ongoing leakage and the observed capture of LNAPL in the vadose zone, Bioslurping managed to significantly restrict the plume within relatively small parts. Gidarakos and Aivalioti [61] coupled that restoring method with air sparging to achieve better cleaning. The results of frequent groundwater monitoring indicated a remarkable decrease in total petroleum hydrocarbons (TPHs) and BTEX concentrations (up to 99%).

In response to the previously mentioned works, Makri, Stathopoulou, Hermides, Kontakiotis, Zarkogiannis, Skilodimou, Bathrellos, Antonarakou and Scoullos [4] investigated the potential spatial and temporal diffusion of benzene, toluene, ethylbenzene and xylenes (BTEX) in the groundwater system of the broader area of the two refineries. Four restricted plumes of BTEX were reported; three of them located in the vicinity of obvious pollution sources, such as the military airfield and the two refineries and another indicated illegal dumping in wells of oil waste into an abandoned site with no noticeable pollution source. An overall decrease of the concentrations was recorded over three years of monitoring. However, this research was published very recently, after intensive hydrogeological survey that supported the natural attenuation observed.

In Kyriazis et al. [62], significant pollution from heavy metals in groundwater was reported. Specifically, Fe, Mn, Ni, Cd and Pb exceeded the legislated limit (EEC98/83) in most samples. The concentrations of these elements were correlated with geological processes, such as the existence of bauxites, the presence of oxides due to the alteration of rocks and the organic matter of the sediments, which is characterized for its ability to bind the heavy metals. The same paper reported the existence of ions  $Mg^{2+}$ ,  $Na^+$ ,  $K^+$ ,  $Cl^-$  and  $SO_4$ , which were attributed to possible seawater intrusion in the aquifers of the study area.

#### 4.2. The Hydrogeochemical Conditions

The quality of groundwater has been studied since early 1990s by national institutions since over-pumping reportedly led to the water level decline. The measured Cl<sup>-</sup> and Na<sup>+</sup> ions ranged from 300 to 1700 ppm; thus, seawater appeared to intrude at 8 km from the coastline to the inland of Thriassion Plain [57,63]. High concentrations of the Cl<sup>-</sup>, Na<sup>+</sup> and Mg<sup>2+</sup> ions in groundwater of the study area were reported by Bathrellos et al. [64] under the same deliberation. The salinity occurrence was then supported by Christides et al. [65], who measured high values of water hardness (Ca), specific conductivity, dissolved matter and ions of Cl<sup>-</sup> and Na<sup>+</sup> in the groundwater of the study area. Iliopoulos et al. [66] reported moderate values of hardness and low concentration of total salts in the spring water of the carbonate aquifer. The type of this water was characterized as alkaline with good quality and a hydro-chemical type of Ca-HCO<sub>3</sub> and Mg-HCO<sub>3</sub>. In contrast, the groundwater from the phreatic aquifer was identified as degraded because of the occurrence of high concentrations in Na<sup>+</sup>, Cl<sup>-</sup>, SO<sub>4</sub><sup>2-</sup>, NO<sub>3</sub><sup>-</sup> and PO<sub>4</sub><sup>3-</sup>. The origin of hydro-chemical types Na-Cl, Mg-Cl and Ca-Cl in the coastline area is the reverse cation-exchange processes. Seawater intrusion was also concluded from the high the concentrations of  $Cl^{-}$  and Na+ ions, with values in the ranges from 1000 mg/L to 3385 mg/L and 0.5 mg/L to 1050 mg/L, respectively.

With the same considerations, Christides et al. [67] reported that the relatively elevated values of  $Cl^-$ ,  $Na^+$ ,  $Ca^{2+}$ ,  $Mg^{2+}$  and  $SO_4^{2-}$  determined in groundwater, attesting to sea water intrusion. This consideration was re-evaluated by Hermides and Stamatis [13] and Makri et al. [68], who reconstructed the origin of groundwater salinity through the mass ratios of halides. According to [4], the distributions of the chloride/bromide mass ratio and of fluoride concentrations were used as environmental tracers. Based on published research, the  $Cl^-/Br^-$  ratio in groundwater fluctuates from 102 to 1510 as follows: The values ranging from 100 to 200 indicate the existence of fresh water, and the value approximating to 292 indicates sea water influence. In the same pattern, ratio values between 300 and 600 indicate human intervention, such as domestic sewage and livestock farming, and ratio values between 939 and 1510 have geogenic origin, such as the dissolution of the mineral halite (NaCl). The evapotranspiration of irrigated water may also impact groundwater quality. The identified elevated fluoride concentrations are possibly due to the decay of organic material within the Neogene–Quaternary sediments, or the water enclosed in the Palaeozoic-Middle Triassic complex, connected to hydrothermal fluids.

Recently, Hermides, Makri, Kontakiotis and Antonarakou [3] combined all available geochemical data of the study area, resulting in the construction of the groundwater geochemical evolution: The two end-members are Ca-Mg-HCO<sub>3</sub> and Na-Cl, while the groundwaters of Ca-Mg-HCO<sub>3</sub> type from the mixing processes evolve to Ca-Mg-Cl- HCO<sub>3</sub> type. The reverse cation exchange induces the Na-Cl-type waters into Ca-Cl type. Certain refreshing of Na-Cl waters to Na-HCO<sub>3</sub> type also exists.

Finally, the synthesis of the hydrogeological and hydrogeochemical data gave rise to a reconstruction of the hydrogeological model for the Thriassion Plain presented in Hermides et al. [69].

## 5. Conclusions

The temporal review of the datasets and the results obtained from field studies about the pressures exerted on the Thriassion Plain, Koumoundourou Lake and Eleusis Bay may be concluded as follows:

The investigation of the pollution from petroleum products in the study area, such as the Polycyclic Aromatic Hydrocarbons (PAHs), started in the mid-1980s. To date, relevant research has been produced by institutions; limited works have been published. The concentrations and distributions of these toxic compounds indicates that the degradation of sediment quality, along with the living organisms affected, e.g., mussels, is restricted to marine areas, close to the point sources. The multi-type activities, such as oil refineries, shipyards, urban areas, and the national highway, is reflected in PAH diagnostic ratios, which in total indicate mixed fuel and coal combustion sources. Since agriculture shrinks, any organic pollutant originates from industrial-relevant activities. More actions of source control and sediment quality protection should be taken by national, global, and European strategies and plans (e.g., Land-Based Protocol of UNEP, or the Horizon 2020 Initiative) with focus on the locations that receive the anthropogenic impact. In contrast, volatile organic compounds, such as the BTEX, show good attenuation, evidently, though their investigation should be repeated. Regular monitoring in all means, sediments (surface and downcore), groundwater and biota by authorities is indisputably required.

The overall estimation of the pollution from metals in the seawater, the bottom sediments, and the live tissues of Eleusis Gulf, from the ultra-high concentrations of metals in the mid-1970s, to date shows a stable or even a decreasing trend in the concentrations. Bioacumulation–bioavailability indices, geoaccumulation index, enrichment factor and modified pollution index are useful tools for the assessment of a pollutant's impact in sediments, flora, and fauna. Presumably, any decrease of the dissolved metal concentrations is mainly attributed to the combined effect of dilution and metal removal due to coagulation, precipitation and formation of fine particles. Furthermore, the severe economic crisis in Greece has obligated some polluting industries to close down. The environmental policy implementation coupled with technological progress by big pollutants like heavy industries have promoted certain sediment recovery from metals. European or national programs should fund integrated ecological research to provide holistic environmental status and necessary interventions.

The general remark from the few published studies about the soil pollution is the existence of high levels of metals. We cannot conclude there is an increasing trend since the mentioned studies do not correspond to common sampling locations. The environmental indices also yield information about the degree of mobility, partitioning and availability of the target elements and may be used to cause the contaminants' evolution fate in the soil system and toxicity to live tissues.

Koumoundourou Lake, a brackish Mediterranean lagoon close the seashore of Eleusis Bay, has also received pressure from the broader area's uncontrolled industrialization, the severe change of land use and the influence of the landfill site of Athens. Despite the environmental burden, it seems that the lake's water quality, as concerns the chemical parameters recently measured, remains stable. This may be attributed to the artificial channel—weir—constructed, uplifting the water surface and, consequently, the water volume. The long-lasting existence of metals in the sediments has certainly made the lake's ecological conditions worse. However, of the last decade a significant decrease in heavy metal concentrations is noted, which can be partially attributed to the reduction in pollution pressures due to recent environmental legislation as well as the modification of the hydrologic regime of the lake. Much attention should be given to a new adjustment of the weir, to allow the entrance of new fish species. This would result in the restoration of the food dynamic by the consumption of algae and detritus, improving the aquatic parameters overall.

The industrial and, to a lesser degree, the agricultural activities have significantly affected the groundwater quality. Metals and oil hydrocarbons deteriorate the ecosystems that depend mostly on groundwater flow. On the other hand, the hydrogeological conditions of the study area have influenced the pollutants' fate and behavior. A new conceptual hydrogeological model was proposed for the Thriassion Plain aquifer system after reconsidering the hydro-geochemical processes. A revised conception about the aquifers' salinity of the study area is the seawater entrapment prior Pleistocene–Holocene clay deposition. An ongoing monitoring in all means of Thriassion Plain (sediments, marine ecosystems, and groundwater) is strongly recommended.

To sum up, this review has presented the evolution of the contamination of a multistressed region in the Mediterranean basin, in soil and groundwater of the mainland of the Thriassion Plain, as well as the sediment and seawater of the Eleusis Bay, which is the closely affected sea body. Apart from our concluding comments, as above, we suggest more research under a holistic environmental prospect and more dynamic policy from authorities, allowing for both restoration and prolepsis.

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