

Article

Selenium Concentration Levels in Two Polluted Lagoons, Eastern Red Sea Coastal Waters

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Abstract: The importance of selenium in the marine environment is highlighted by its bilateral behavior as a nutrient and toxic element. The cathodic stripping voltammetry validated method was used to determine the selenium concentration. The concentration of total dissolved selenium (TDSe) in Al-Arbaeen and Sharm Obhur lagoons along the eastern Red Sea coastal waters was determined. The total selenium concentration in seven fish species' muscles was determined. TDSe in the surface water of Al-Arbaeen and Sharm Obhur lagoons showed a maximum concentration of 18.56 and 8.38 nM, respectively. TDSe in the surface water of Al-Arbaeen revealed high concentrations near the wastewater discharging pipes in the lagoon. The linear regression between the TDSe and salinity reflected that the discharged water is the source of selenium with a significant negative correlation of ($R^2 = 0.80$, $p < 0.05$). In contrast, TDSe in the surface water of Sharm Obhur showed a significant positive correlation ($R^2 = 0.78$, $p < 0.05$). In the lagoon head, hypoxic and anoxic conditions were dominant. This condition reflected the low TDSe concentration and may affect the selenium chemical forms' abundance in the lagoon. Two fish species *Herklotsichthys punctatus* and *Herklotsichthys* revealed high concentrations of 3.99 and 2.40 $\mu\text{g/g}$, respectively, which exceeded the permissible levels of the WHO, FAO, and ASTDR.

Keywords: total dissolved selenium; Al-Arbaeen lagoon; Sharm Obhur; wastewater pollution; seawater; fish; Red Sea



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1. Introduction

Selenium is a non-metal element located in block b, group sixteen, and period four of the periodic table. Elements of this group are distinguished by high electronegativity, so they have a strong ability to accept one or more pairs of electrons. It was discovered by a Swedish scientist, Jons Berzelius, in 1818 [1]. Its relative abundance in the Earth's crust is around 1.9 ppb [2]. The primary source can be from the ores with different forms (allotropes) [3]. It also reaches the aquatic environment through rocks, petroleum refineries, and coal [4]. In the natural environment, selenium can be found in three chemical oxidation states: Se (IV), Se (VI), and Se (II). These three states control the physiological conduct of the element [5]. Selenium can also be found in the environment and some living organisms at four oxidation states Se^{6+} (soluble as selenate), Se^{4+} (soluble as selenite), Se^{2-} , and Se^0 [6,7]. The solubility and mobility of these forms increase in alkaline pH. Speciation and bioavailability are crucial in determining selenium's fate and its effect on the environment [8].

Selenium is necessary for humans, animals, and some plants at a low concentration. It plays a vital role in the cell structure and as a protective agent against oxidative damage. At the same time, a concentration at higher levels reveals a toxic indication [9,10]. Exposure to selenium leads to poisoning, which causes reproductive failure by a mutation in the egg production of marine life (fish, birds, and reptiles) [11–14]. Selenium toxicity also causes

mortality, mass wasting in adults, reduced juvenile progress, and immune suppression [15]. Selenium toxicity is controlled by the concentration and chemical forms (Se (IV), Se (VI), and Se (II)) [6,16]. It has distinguished eco-toxicological features, which show a very narrow limit between toxicity and nutrition [17–22].

The total dissolved selenium (TDSe) along the eastern Red Sea and Al-Shabab polluted lagoon was investigated. The main goal of this study was to examine selenium concentration levels in two contaminated coastal lagoons. The concentration, sources, and distribution of TDSe were investigated. The studies mentioned above and in this study are considered as a baseline for future studies in Red Sea water.

2. Materials and Methods

2.1. Study Area

Al-Arbaeen lagoon has been subjected to numerous studies [23–28] (Figure 1). It is located southward from Reayat Al-Shabab lagoon, which is shielded by Jeddah Islamic port from the west. This cover is the reason for the low or zero exchange with the open seawater, which explains its low surface water salinity compared with Al-Shabab Lagoon. It is a semi-enclosed lagoon in the (T) shape with two branches extended northeast with a length of 1.5 km and a width of 300 m. Moreover, it has a surface area of 254,000 m² and a total water volume of 1.0×10^6 m³. In addition, it was used for a long time as a sewage dumping area. It receives around 65,000 m³ daily of partially treated wastewater from Jeddah district [29]. It has three land base wastewater sources, one in the lagoon entrance and two in the lagoon branches heads (see Figure 1).

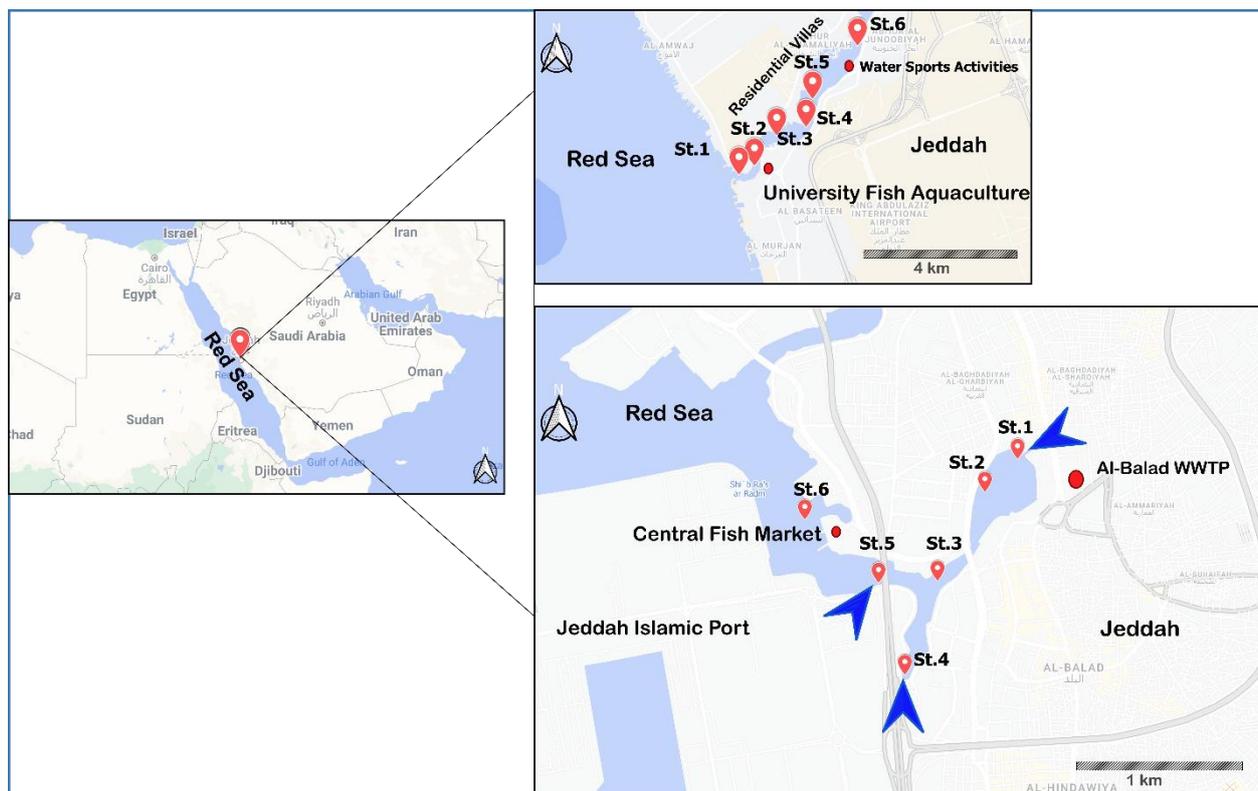


Figure 1. Al-Arbaeen and Sharm Obhur Lagoons, the eastern part of the Red Sea, samples collection sites, and the wastewater dumping points (blue arrows) of Al-Arbaeen.

Sharm Obhur is an erosional lagoon created by tectonic faulting located north of Jeddah city [30,31]. It extends northeast along with the coralline limestone of the Tihama Formation. It has a length and a maximum width of 10 km and 1.5 km, respectively [32]. It is linked with the Red Sea through the small mouth, and its depth increases from the

lagoon head to mouth, with an average depth of 30 m [33]. It is less impacted by pollution than the two polluted lagoons Al-Shabab and Al-Arbaeen [23,25,34] (Figure 1).

2.2. Samples Collection and Preservation

Waters samples from Al-Arbaeen lagoon were collected on 22 March 2016, from six stations along the lagoon (Figure 1). The stations were selected based on the lagoon's physical properties and the pollution sources between latitudes and longitudes of 21.48° N, 21.49° N and 39.17° E, 39.18° E, respectively. Twelve water samples for selenium and nutrients were collected from the surface (0.5 m) and near the bottom (3 m) using a 5 L Niskin bottle attached to a rope after filtration on the boat (MF-Millipore membrane, mixed cellulose esters, hydrophilic 0.45 µm weighted filter paper), and then transferred to 1000 mL high-density polyethylene bottles (HDPEs) and kept in the icebox at a temperature of 4 °C. The filters were kept for the suspended particulate matter (SPM) determination. Dissolved oxygen samples were collected in 60 mL of chemical-oxygen-demand bottles. Then, it was fixed by adding 1 mL of manganese chloride (GPR, BDH, and England) solution and 1 mL of the alkaline iodide (GPR, BDH, and England) solution, which was shaken well and kept closed in the dark in the icebox at 4 °C. Total organic carbon samples were collected in 25 mL glass bottles and kept frozen. Chlorophyll-a samples were collected after filtration using Whatman glass microfiber filters, grade GF/F filter paper, and kept frozen in 20 mL well-tied and foiled tubes. Temperature and pH were measured immediately in the ambient water. Upon reaching the laboratory, selenium water and total organic carbon samples were kept frozen at −20 °C. Chlorophyll a samples were collected, and 10 mL of 90% GC grade acetone was added for each sample and kept in the dark at 4 °C overnight. Dissolved oxygen, salinity, and nutrients were determined immediately upon reaching the laboratory.

Eight fish species were collected on 5 January 2017, using a small boat and gill net from different sites in Al-Arbaeen lagoon. Three fish species from the open seawater were collected as reference samples. They were kept immediately in the icebox, and upon approaching the laboratory, the length and weight were measured using a regular stainless-steel ruler and sensitive balance, respectively. Five fish pieces were used as one sample for each species. Then, they were dissected, and mussels of each fish species were put in a petri dish and kept in dry freezing for 48 h. Afterward, they were ground using an agate mortar and a pestle and kept in a zipped plastic bag.

Water Samples from Sharm Obhur lagoon were collected along the lagoon water body on 6 March 2016, from six stations designed to fulfil the study purposes (Figure 2). Water samples were collected using 5000 mL Niskin bottles attached to a rope between latitudes 21.71° N and 21.75° N in the morning from a one-meter depth. Selenium water samples were directly filtered on the boat using the vacuum pump filtration system and a MF-Millipore membrane, mixed cellulose esters, and hydrophilic 0.45 µm weighted filter paper, into 1 L high-density polyethylene bottles, and kept in the icebox at a temperature of 4 °C. The filter paper was frozen for suspended particulate matter (SPM) determination for each sample. Nutrient samples were collected in 500 mL polyethylene bottles after filtration and kept at a temperature of 4 °C in the icebox. Total organic carbon samples were collected in 25 mL glass bottles and frozen. Dissolved oxygen samples were collected in 60 mL of chemical oxygen demand bottles. Then, they were fixed by adding 1 mL of manganese chloride (GPR, BDH, and England) solution and 1 mL of the alkaline iodide (GPR, BDH, and England) solution, which was shaken well and kept closed in the dark in the icebox at 4 °C. Chlorophyll samples were collected after filtration using Whatman glass microfiber filters, grade GF/F filter paper, and kept frozen in 20 mL well tied and foiled tubes. Temperature and pH were measured immediately in the ambient environment. Upon reaching the laboratory, selenium water and total organic carbon samples were kept frozen at −20 °C. In the same regard, for Chlorophyll-a samples, 10 mL of 90% GC grade acetone was added for each sample and kept in the dark at 4 °C overnight. Dissolved oxygen, salinity, and nutrient were determined immediately upon reaching the laboratory.

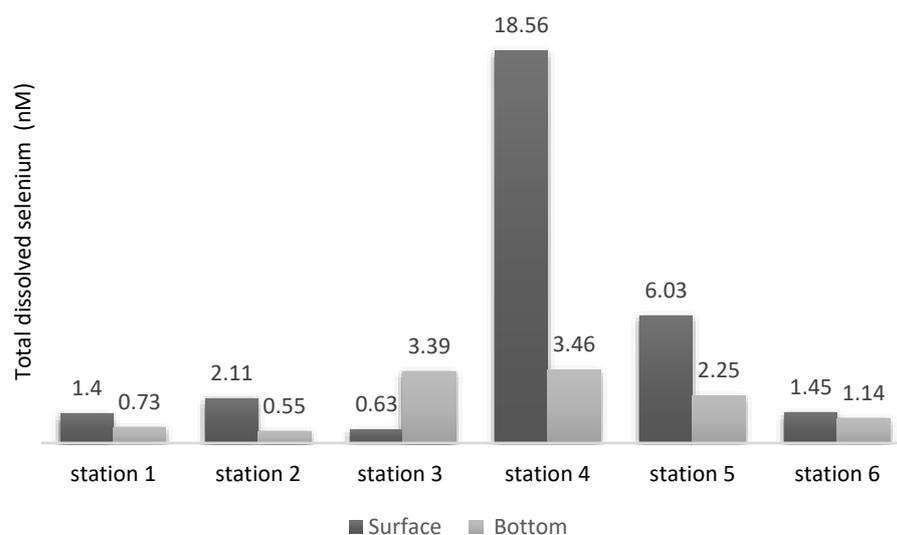


Figure 2. Distribution of the total dissolved selenium in the surface and bottom water of Al-Arbaeen Lagoon.

2.3. Analytical Methods

The total dissolved selenium in the seawater was determined using cathodic stripping voltammetry [34]. Ultraviolet (UV) photolysis under nitrogen purging of the seawater was used to convert selenate SeO_4^{2-} and selenide Se^{2-} to selenite, the electroactive oxidation state of selenium [16]. Briefly, three hours of ultraviolet photolysis using a device designed and assembled by [35], under a high-purity nitrogen (99.999%) flow through 40 mL fused silica tubes filled with filtered seawater sample, was conducted. Then, 10 mL of the seawater sample was pipetted into the cleaned (using 0.1 M HNO_3 65% Suprapur, Merck) electrochemical cell. After that, 40 μL of 50% (*v/v*) HCl 37% (Sigma-Aldrich, ACS reagent) was added using a micropipette. In addition, 40 μL of 10 mM copper sulphate (oxidizing and complexing agent) was added to the electrochemical cell using a micropipette. A Computrace VA 97 Metrohm instrument was used for the determination of total dissolved selenium (TDS_e). Platinum, hanging mercury dropping, and silver–silver chloride electrodes were used as auxiliary, working, and reference electrodes, respectively. The differential pulse mode and the cathodic stripping parameters were adjusted, as shown in the table below (Table 1). Selenium concentration was obtained from the relationship of the applied potential with the produced current (peak height) and the standard solution using the standard addition method. Selenium standard solution (AAS standard from BDH, England) with various concentrations was prepared. Three replication measurements for each sample were implemented.

Table 1. Physical and chemical properties of Al-Arbaeen lagoon water.

Parameters	Surface Water			Bottom Water		
	Max	Min	Mean	Max	Min	Mean
Temperature (°C)	28.9	21.5	26.10	27.5	24.90	26.50
Salinity	37.12	25.80	31.51	37.02	32.60	34.96
pH	8.86	8.17	8.55	8.70	8.20	8.39
DO (ml/L)	18.64	3.62	10.38	11.46	0.62	4.76
Chlorophyll a (µg/L)	83.7	7.82	39.07	176.65	8.0	55.30

Table 1. Cont.

Parameters	Surface Water			Bottom Water		
	Max	Min	Mean	Max	Min	Mean
Nitrate (μM)	50.89	6.77	28.42	26.83	0.35	10.51
Nitrite (μM)	16.36	1.79	7.07	5.44	0.92	2.46
Ammonia (μM)	240.34	34.22	107.2	76.89	20.95	48.88
Phosphate (μM)	23.76	5.57	14.3	10.13	3.26	7.02
TOC (mg/L)	20.37	7.29	11.77	13.81	5.68	10.34
SPM (g/L)	0.16	0.06	0.11	0.17	0.12	0.15

On the other hand, chemical reagents and standard solutions were of analytical reagent grade. Standard solutions and reagents were prepared using Milli-Q water (Resistivity = $18.2 \text{ M}\Omega \text{ cm}^{-1}$). The accuracy of the instrument measurement for this method was evaluated using spiking and recovery (Supporting Materials). The calibration curve of the standard addition was plotted, and linearity was tested (see Figure 3).

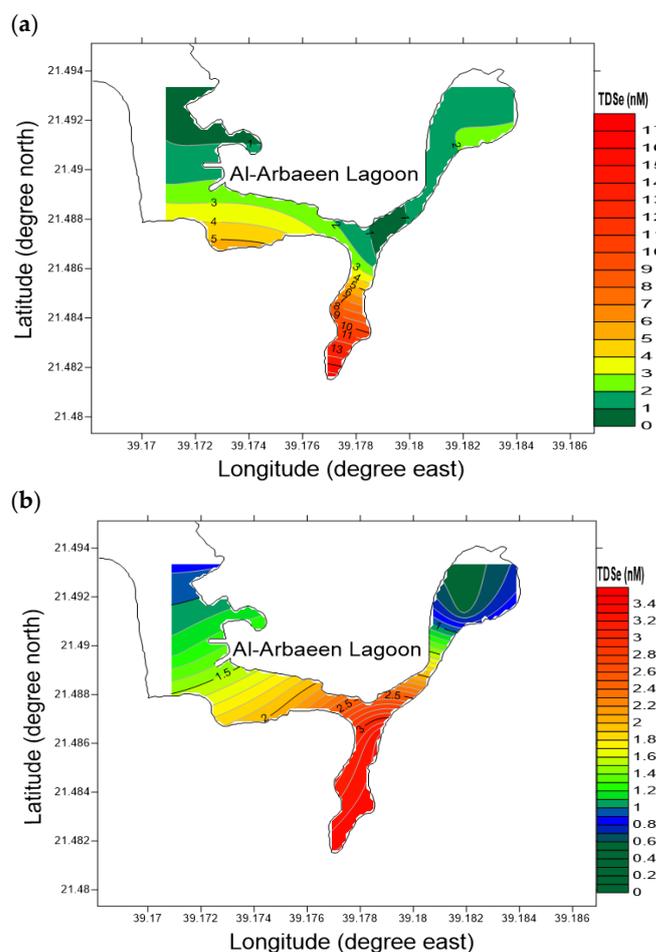


Figure 3. Total dissolved selenium horizontal distribution along Al-Arbaeen lagoon: (a) surface water and (b) bottom water.

Total dissolved selenium in sediment was determined by inductively coupled plasma-mass spectrometry. Before analysis, 0.5 g of a freeze-dried and ground sample from each fish species were weighed into a 25 mL capped Teflon digestion tube. Then, 5 mL of concentrated nitric acid (65% Suprapur, Merck) and 2 mL of hydrogen peroxide (35% GP, Holyland) were added. They were exposed to heating in a hot sand plate for 6 h at 100 °C. After cooling, the digested fish samples were transferred to 25 mL tubes and filled up to 20 mL using ultrapure Milli-Q water (Resistivity = 18.2 MΩ cm⁻¹) [36]. The determination was performed using the Agilent ICP-MS 7700x (see Table 2).

Table 2. Nutrient correlation coefficient against salinity in the surface and bottom water of Al-Arbaeen Lagoon (n = 6 stations).

		Salinity
Surface Water	NO ₃ ⁻	0.94 (<i>p</i> < 0.05)
	PO ₄ ³⁻	0.94 (<i>p</i> < 0.05)
	DIN	0.88 (<i>p</i> < 0.05)
Bottom Water	NO ₃ ⁻	0.20 (<i>p</i> > 0.05)
	PO ₄ ³⁻	0.71 (<i>p</i> < 0.05)
	DIN	0.25 (<i>p</i> > 0.05)

Dissolved oxygen in the seawater was determined by iodometric titration using the improved Winkler method [37–42]. Briefly, the seawater sample was collected using a 5000 mL Niskin bottle and transferred into a 60 mL biological oxygen demand bottle. Dissolved oxygen was fixed by adding 1 mL of manganese chloride (GPR, BDH, and England) solution and 1 mL of the alkaline iodide (GPR, BDH, and England) solution, which was shaken well and kept closed in the dark in the icebox at 4 °C. Upon reaching the laboratory, immediately dissolved oxygen was determined by its titration against the standard solution of sodium thiosulfate (BDH, AnalaR) after the addition of 1 mL of 50% (*v/v*) sulfuric acid 98% (GPR, BDH, and England). An amount of 1 mL of Starch (Fluka, AnalaR) solution was used to determine the titration endpoint. Standard solutions and reagents were prepared using Milli-Q water (Resistivity = 18.2 MΩ cm⁻¹).

Chlorophyll-a pigment was determined using the YSI multiparameter water quality sonde 6600 V₂ with 650 MDS, including the chlorophyll-a sensor based on the optical technique. The device was used after calibration with a standard solution manufactured by the YSI Company to ensure measurement accuracy and precision. In one field trip, chlorophyll-a was determined using the spectrophotometric technique and the equation introduced by [43]. In this method, chlorophyll in the filter paper was soaked in 10 mL, 90% acetone (99%, Sigma-Aldrich, GC grade) for 24 h before spectrophotometer measurement at 663 and 651 nm. The acetone solution was prepared using Milli-Q water (resistivity = 18.2 MΩ cm⁻¹).

Dissolved nitrate and nitrite in the seawater were determined using the spectrophotometry technique [44,45]. The method's main concept is that nitrite reacts with the aromatic amine to produce diazonium salt. This compound reacts with *N*-(1-naphthyl)-ethylenediamine dihydrochloride (coupling reaction, conjugated system, and colored compound). This method transferred 25 mL of seawater sample to the reaction container using a measuring cylinder, followed by adding 1 mL of sulphanilamide (VWR, BDH, AnalaR normapur) and 1 mL of *N*-(1-naphthyl)-ethylenediamine dihydrochloride (BDH, GPR) to the sample solution. Then, the mixture was kept for 15 min before measurement. The absorbance was measured at 543 nm. A series of standard solutions of sodium nitrite (VWR, BDH, AnalaR normapur) were prepared.

In the nitrate case, the reduction of nitrate to nitrite by passing a seawater sample through the granular cadmium (Fluka, 99.99%) coated by a copper-packed column was conducted. In addition, a 2 mL/min flow rate was used with 100% reduction efficiency using

ammonia buffer solution pH 8. Then, all the steps mentioned above were repeated. Standard solutions and reagents were prepared using Milli-Q water (Resistivity = $18.2 \text{ M}\Omega \text{ cm}^{-1}$).

Ammonia was determined using the spectrophotometric technique after the chemical modification (blue indophenol complex formation) was used [46–48]. In this method, phenol (BDH, GPR), sodium nitroprusside (BDH, GPR), a mixture of sodium citrate (BDH, AnalaR) and sodium hydroxide (BDH, AnalaR), and sodium hypochlorite solutions (oxidizing solution) were prepared. The alkaline and sodium hypochlorite solutions were mixed under a volume ratio of 4:1. An amount of 25 mL of filtrated seawater sample was transferred using a 50 mL measuring cylinder into a 100 mL Erlenmeyer flask. An amount of 1 mL of phenol solution was pipetted into the sample container and mixed well. Then, 1 mL of the sodium nitroprusside was pipetted into the sample container and shaken well. An amount of 2.5 mL of oxidizing solution was transferred into the reaction container. The spectrophotometer measurement was conducted after one hour at 640 nm. Ammonium chloride (BDH, AnalaR) standard solutions of 1, 3, 5, and 15 μM were prepared using Milli-Q water (Resistivity = $18.2 \text{ M}\Omega \text{ cm}^{-1}$).

Phosphate in seawater samples was determined using the spectrophotometric technique and phosphomolybdate complex formation [49]. Briefly, 25 mL of filtrated seawater sample was transferred into the reaction container at room temperature. A mixture of 20 mL of ammonium molybdate (Hopkins and Williams, GPR), 50 mL of sulfuric acid (BDH, Aristar), 20 mL of ascorbic acid (BDH, ACS reagent), and 10 mL of potassium antimonyl tartrate solutions (BDH, GPR) was prepared. An amount of 2.5 mL of the mixture was pipetted in the sample container, shaken well, and left closed on the bench for 2 h before the measurement. The samples were measured at a wavelength of 885 nm. Potassium dihydrogen phosphate standard solutions (Sigma-Aldrich, AnalaR) of 0.5, 1, 3, and 5 μM were prepared using Milli-Q water (Resistivity = $18.2 \text{ M}\Omega \text{ cm}^{-1}$).

Total organic carbon was determined using the total organic carbon analyzer TOC-V_{CPH} connected with autosampler ASI. The seawater sample was transferred into a 25 mL screw-capped glass tube and 25% phosphoric acid with a pH < 3 in the sparging step to remove the inorganic carbon. Then, the sample was exposed to zero air and combustion catalytic oxidation using cobalt oxide at 680 °C. The detection was performed using the non-dispersive infrared gas detector (NDIR) [50].

Suspended particulate matter (SPM) was determined using the method with some modifications [51]. In this method, a new and dry MF-Millipore membrane, mixed cellulose esters, and hydrophilic 0.45 μm filter were weighed and kept in a plastic holder. In the field, after the filtration step, the filter with the SPM was kept in a plastic holder at $-20 \text{ }^\circ\text{C}$. The filter was dried at $50 \text{ }^\circ\text{C}$ for 24 h in the laboratory and weighed again. The SPM weight was obtained by subtracting the weight (filter + SPM) from the filter.

Temperature, salinity, depth, and pH were measured in the ambient seawater using the calibrated multiparameter water quality sonde 6600 V₂ with 650 MDS.

3. Results and Discussion

3.1. Al-Arbaeen Lagoon

3.1.1. Selenium Vertical and Horizontal Distribution along Al-Arbaeen Lagoon

The total dissolved selenium (TDSe) distribution in the surface and bottom water along the lagoon is shown in (Figure 2). In the lagoon surface water, the maximum and minimum concentrations of the TDSe were 18.56 and 0.63 nM, respectively. The average TDSe concentration in the surface water was 5.03 nM. In the same regard, the maximum and minimum concentrations of the TDSe in the bottom water were 3.46 and 0.55 nM, respectively. Moreover, the average TDSe concentration in the bottom water was 1.92 nM. The highest concentrations obtained for TDSe were located close to the wastewater pipes at stations 4 and 5 (see Figure 1).

In the surface and bottom water, TDSe did not show a specific pattern of variation, but it was revealed from the histogram (see Figure 2) that the TDSe concentration at the lagoon mouth was slightly higher than at the lagoon head (main lagoon branch). The TDSe

concentration in the surface and bottom of the whole stations reflected high/low pattern concentration, except for station 3 (due to the horizontal diffusion of the bottom water from station 4), which indicates that the TDSe distribution over the lagoon did not show a nutrient-type vertical profile (see Figure 3). The surface TDSe average concentration of 5.03 nM obtained from Al-Arbaeen lagoon was higher than the TDSe average concentration from Reayat Al-Shabab Lagoon [52] and the TDSe surface concentrations of 11.99 nM obtained by [53]. In comparison, it was greater than the TDSe average concentration of 2.27 nM from San Francisco Bay established by [54]. Similarly, it was greater than the TDSe average concentration of 1.48 nM from Southern Chesapeake Bay established by [55]. In addition, it was greater than the average TDSe concentration of 0.06 nM from Marsa Matrouh beaches established by [56]. In the same regard, it was higher than the average concentration of 2.45 nM established by [57].

The horizontal surface distribution of the TDSe over the lagoon revealed high concentrations at stations 4 and 5, which are located close to the wastewater pipes, and it was extended northeast to the lagoon head (main lagoon branch) and northwest to the lagoon mouth (Figure 3a). The highest TDSe concentration was obtained close to station 4 at the surface water. In addition, it was obvious that surface TDSe concentration was high at the lagoon mouth, while in the lagoon head, it was low, which might be due to the low oxygen concentrations (hypoxic and anoxic conditions) over there.

The horizontal bottom distribution of the TDSe over the lagoon showed the same pattern as the horizontal surface distribution. In addition, the extension of the bottom TDSe concentration extended north and northeast toward station 3 and the lagoon head (main lagoon branch), respectively (Figure 3b). This distribution explains the relatively high TDSe concentration value in the bottom water of station 3 compared with the TDSe surface concentration value (Figure 3b). TDSe reaching the lagoon water from station 5 participated in TDSe in the bottom water at station 3 associated with the TDSe from the pipe at station 4 (Figure 3a,b). The vertical cross-section showed that the TDSe concentration was high at stations 4 and 5. The concentration decreased gradually toward station 1 at the lagoon head and toward station 6 on the way to the open seawater (Figure 4). This pattern confirms that the wastewater from stations 4 and 5 is the lagoon's primary selenium source.

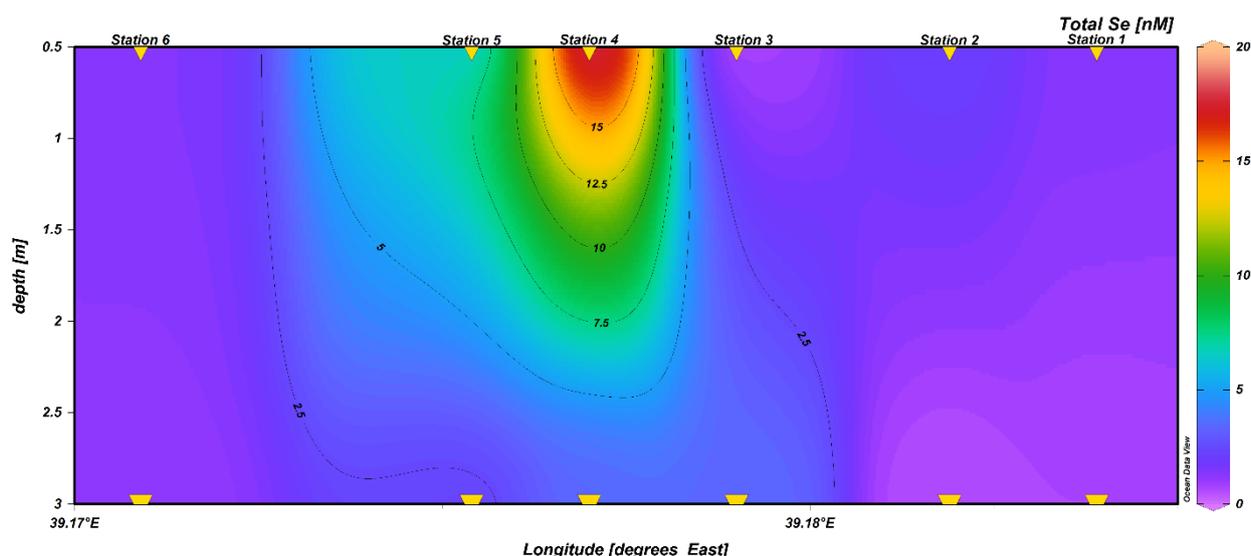


Figure 4. TDSe vertical cross-section along Al-Arbaeen lagoon.

3.1.2. Physical and Chemical Parameters of the Lagoon Water

Temperature, pH, salinity, dissolved oxygen, chlorophyll-a, nitrate, nitrite, ammonia, phosphate, and suspended particulate matter (SPM) were measured in Al-Arbaeen lagoon. The surface water showed a higher average temperature than the bottom water (Table 1). This difference might be referred to as the lagoon's shallow water. In the same regard,

surface water revealed a relatively alkaline pH due to the discharging impact (Table 1). The salinity in the surface and bottom waters was low, reflecting the fresh wastewater discharging effect (Table 1). In the surface and bottom waters, chlorophyll-a showed high concentrations due to the eutrophication conducted by phytoplankton (Table 1). The high nutrient concentration in the lagoon induces the eutrophication process. The total organic carbon showed a high concentration due to the sewage and wastewater dumping in the lagoon (Table 1).

The vertical cross-section of the temperature showed relatively high values (26.9–28.9 °C) in the wastewater discharging points at stations 1, 4, and 5 (Figure 5a) due to the wastewater source. The temperature in the bottom water was higher than that in the surface water due to the shallow lagoon water with no mixing. The temperature at station 6 was low at 21.5 °C, reflecting open seawater temperature. The pH vertical cross-section revealed the high pH value (8.86) in the surface water at station 5 (see, Figure 5b). This effect extended toward the lagoon head at stations 2 and 3. A relatively high pH value at station 6 near the central fish market was observed. The reason behind this high pH refers to fish wastewater. The pH at station 1 revealed a relatively low pH value of 8.31 (Figure 5b). However, the pH values at the surface and bottom water of the lagoon were higher than average open seawater. This gap is evident due to the high nutrient load from dumping pipes that lead to algae growth.

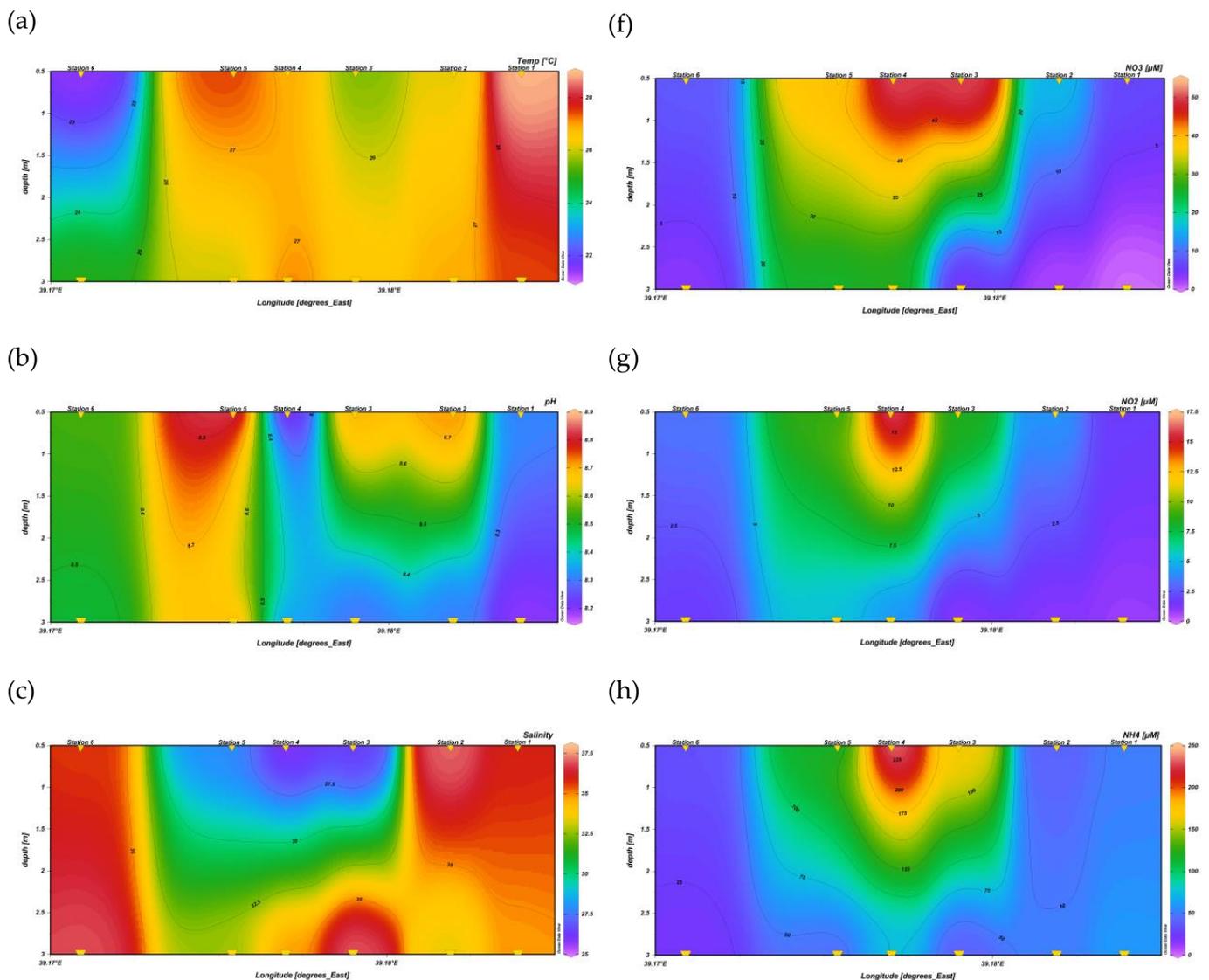


Figure 5. Cont.

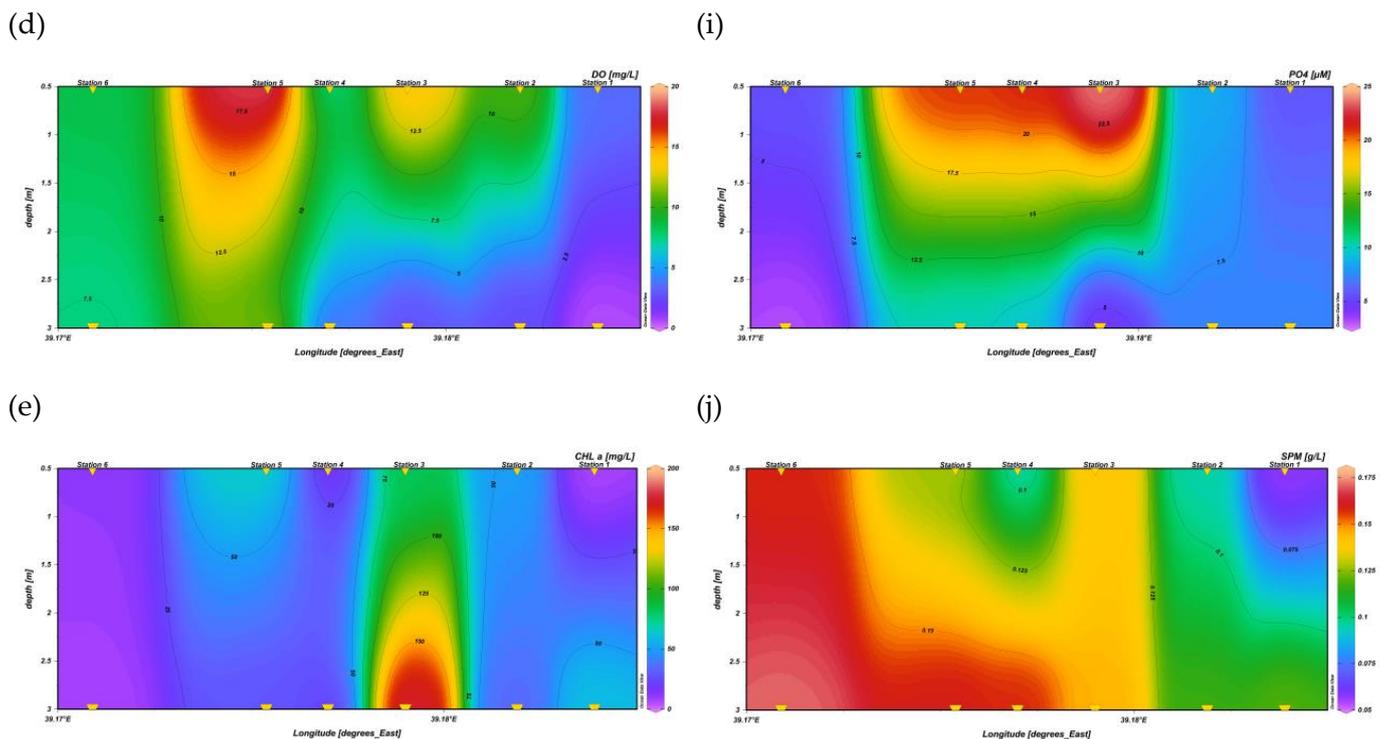


Figure 5. Physical and chemical parameters vertical cross-section along Al-Arbaeen lagoon.

The vertical cross-section reflected low salinity at the wastewater discharging points at stations 4 and 5 (25–26) extended to station 3, and gradually increased toward the lagoon head at about 36 (see Figure 5c). However, the salinity over the lagoon water is less than open seawater salinity, and the fresh wastewater's effect is apparent. The vertical cross-section of the dissolved oxygen along the lagoon reflected a high dissolved oxygen concentration of 18.6 mg/L at the wastewater discharging pipe at station 5 extended to station 3 and decreased gradually toward the lagoon head (see Figure 5d). In the lagoon head, the dissolved oxygen in the bottom water reached low concentrations (0–2.5 mg/L) (see Figure 5d). This deficiency can be explained by a high nutrient load that leads to the high growth of algae and oxygen consumption.

The chlorophyll-a vertical cross-section showed a relatively high concentration in the lagoon's inner part and a low concentration toward the lagoon mouth (Figure 5e). This phenomenon is due to the high nutrient load coming from the pipes. Chlorophyll-a at station 3 showed high concentrations in the surface, and bottom water (Figure 5e), and this might be due to the high concentration of nutrient reached over there from the two pipes at stations 4 and 5. The nitrate vertical cross-sections revealed high nitrate concentrations (45–50 μM) in the wastewater discharging pipes. This confirms that wastewater is the primary source of pollution in stations 4 and 5 (Figure 5f). The fresh polluted water extends from stations 4 and 5 toward the lagoon head with a nitrate concentration of about 5 μM at station 1 (see Figure 5f). In the lagoon head-bottom water, the nitrate concentration was very low at 0.3 μM , while the hypoxia and the anoxic conditions were dominated by relatively high pH (Figure 5b,f). These conditions induced nitrate reduction (denitrification) to ammonia, which is confirmed by a high concentration of nitrite and ammonia (Figure 5g,h) in the lagoon head-bottom water. A high concentration of ammonia and nitrite near the wastewater discharging points was obtained (stations 3, 4, and 5) (Figure 5g,h). This reflects that wastewater is the lagoon's primary source of nitrite and ammonia. Phosphate revealed a similar pattern to nitrate. The vertical cross-section of the phosphate showed a high concentration at the wastewater discharging pipes ($\text{PO}_4^{3-} > 20 \mu\text{M}$) and decreased gradually toward the lagoon head (5–7 μM) (see Figure 5i). The suspended particulate matter (SPM) was high at the wastewater discharging points and gradually decreased

toward the lagoon head (Figure 5j). A high concentration of SPM at station 6 was observed. This is due to the fish market wastewater (see Figure 5j). The vertical cross-section of SPM confirms that wastewater pipes are the primary source of SPM in the lagoon.

The vertical cross-sections of the whole parameters revealed that the wastewater discharging over Al-Arbaeen lagoon is the primary source of pollution. The surface nitrate and phosphate versus the salinity along the lagoon showed negative correlation coefficients of 0.94 and 0.94, respectively. This is strong evidence that wastewater is the source of pollution (see Table 2). The bottom phosphate versus salinity showed a negative correlation coefficient of 0.71 (see Table 2); this was attributed to the substantial contribution of the wastewater discharging in the lagoon. In contrast, the bottom nitrate versus salinity did not show any variation pattern (Table 2); this may be due to the hypoxic and anoxic conditions, which lead to nitrate reduction and produce nitrite and ammonia. On the other hand, the surface dissolved inorganic nitrogen (DIN) as the sum of (nitrate, nitrite, and ammonia) against the salinity showed a negative correlation coefficient; this indicated the wastewater contribution to the lagoon water (Table 2). However, the DIN in the lagoon bottom water did not show any correlation (Table 2).

3.1.3. Selenium Behavior, Sources, and Its Relationship to the Physical and Chemical Parameters

Total dissolved selenium (TDSe) in the lagoon surface water against the nitrate and phosphate revealed a positive correlation coefficient of 0.88 and 0.64, respectively (Table 3). This was attributed to selenium's function as a marine ecosystem nutrient [58]. The surface chlorophyll-a concentration against the TDSe did not show a specific variation pattern (Table 3), whereas surface salinity versus the TDSe showed a negative correlation coefficient of 0.80 (Table 3), which proves that the wastewater discharging is the primary source of selenium in the lagoon. Contrary to [53,58], it reflected a conservative distribution behavior in a wide salinity range of 26–37. This indicates that selenium species in the lagoon might be exposed to different reduction processes. It could be removed from the sediment (selenide associated with heavy metals) and/or atmosphere (as hydrogen selenide) [59] through biological uptake [60,61] or released into the lagoon water from the degraded organic compounds. The surface dissolved oxygen against the TDSe did not show an apparent variation (Table 3). The surface dissolved inorganic nitrogen (DIN) against the TDSe showed a positive coefficient of 0.97 (Table 3), indicating its role as a nutrient. The bottom DIN did not show a correlation against the TDSe (Table 3). The surface total organic carbon against the TDSe did not reveal an apparent linear correlation coefficient (Table 3). This can be attributed to the wastewater origin source. The bottom phosphate, salinity, and dissolved oxygen did not show any certain correlation with the TDSe in the Al-Arbaeen lagoon except for the nitrate, chlorophyll a, DIN, and total organic carbon, which revealed a positive correlation coefficient of 0.88, 0.56, 0.66, and 0.62, respectively (Table 3). The reason behind the significant positive correlation coefficient of the latter chemical parameters can be attributed to the organic matter regeneration and selenium released into the lagoon water [62], while nitrate and DIN can be referred to as a selenium nutrient role in the biological processes.

Table 3. Physical and chemical parameters correlation coefficient against total dissolved selenium in the surface and bottom water of Al-Arbaeen Lagoon ($n = 6$ stations).

		NO ₃ [−]	PO ₄ ^{3−}	DIN	Chlorophyll a	Dissolved Oxygen	Total Organic Carbon	Salinity
Surface Water	Total Dissolved Selenium (TDSe)	0.81 ($p < 0.05$)	0.64 ($p < 0.05$)	0.97 ($p < 0.05$)	0.08 ($p > 0.05$)	0.006 ($p > 0.05$)	0.02 ($p > 0.05$)	0.80 ($p < 0.05$)
Bottom Water		0.88 ($p < 0.05$)	0.04 ($p > 0.05$)	0.66 ($p < 0.05$)	0.56 ($p > 0.05$)	0.02 ($p > 0.05$)	0.62 ($p < 0.05$)	0.06 ($p > 0.05$)

The estimated annual selenium flux of the lagoon, calculated at the effluent source (fresh wastewater), was 3.50 kg/year based on [63]. The accumulation of this amount in the lagoon environment for a long time might significantly threaten the lagoon ecosystem. The selenium average residence time in the lagoon was 53.4 days based on [63], which was calculated based on the average selenium concentration and the lagoon volume. This short time leads to the absence of the TDSe species in reduced forms due to its utilization by microorganisms, a reduction due to the massive amount of organic compounds and very low dissolved oxygen concentration (remineralization). The maximum TDSe concentration in the lagoon surface water exceeded the permissible concentration level (0.13 nM) introduced by the Canadian drinking water guidelines [64]. The TDSe average concentration in the lagoon water of 3.5 nM exceeded the permissible level of 1.3 nM of the world health organization [65].

3.1.4. Selenium Levels in Some Fish Species Collected from the Lagoon

Total selenium was investigated and determined in the muscles of seven fish species *Sardinella albella*, *Monodactylus argenteus*, *Ulua mentalis*, *Ambassis urotaenia*, *Herklotsichthys punctatus*, *Herklotsichthys*, and *Caranx melampygus* from Al-Arbaeen lagoon. Total selenium in four fish species from the Red Sea open water near the coral reef barrier was determined, as shown in Table 4. The maximum total selenium was 3.99 µg/g for the *Herklotsichthys punctatus*, which exceeded the permissible limit established by [66–68]. In the same regard, total selenium in the *Herklotsichthys* revealed 2.40 µg/g, which exceeded the acceptable level stated above.

Table 4. Fish species' biological, physical, and chemical properties, collected from www.fishbase.com (accessed on 10 Jan 2017).

Scientific Name	Feeding Habits	Biotype Complex	Number of Fish	Length (cm)	Weight (g)	Total Se (µg/g) d.w	Trophic Level
<i>Sardinella albella</i>	zooplankton, phytoplankton	Reef-associated	2	11.5	18.00	1.98 ± 0.09	2.7 ± 0.30
<i>Monodactylus argenteus</i>	Algae, Invertebrate	brackish	1	7.0	7.00	0.83 ± 0.72	3.0 ± 0.33
<i>Ulua mentalis</i>	benthic crustaceans	Reef-associated	2	14.0	58.00	ND	3.7 ± 0.51
<i>Ambassis urotaenia</i>	Ostracods, Gastopods, euphausiids	Brackish, reef-associated	5	7.0	5.00	1.60 ± 0.18	3.4 ± 0.4
<i>Herklotsichthys punctatus</i>	pelagic-neritic	Lagoons, reef-associated	5	8.5	7.00	3.99 ± 1.51	3.1 ± 0.3
<i>Herklotsichthys</i>	Zooplankton	Lagoons, estuaries	5	9.5	6.00	2.40 ± 0.39	3.6 ± 0.0
<i>Caranx melampygus</i>	Small fish, crustaceans	brackish reef-associated	1	15.0	44.00	ND	4.5 ± 0.80
<i>Cephalopholis hemistiktos</i>	Fish	Reef-associated	1	17.0	60.00	1.60 ± 0.18	4.1 ± 0.60
<i>Cheilio intermis</i>	Crustaceans, mollusks, sea urchins	Reef-associated	1	30.0	173.00	0.80 ± 0.74	3.5 ± 0.54
<i>Hologymnosus annulatus</i>	Small fish, Crustaceans	Reef-associated	1	29.0	226.00	1.60 ± 0.18	4.2 ± 0.73
<i>Scomberoides lysan</i>	scales and epidermal tissues	Brackish, reef-associated	1	26.0	105.00	ND	4.0 ± 0.67

Sardinella albella revealed a total selenium concentration of 1.98 µg/g, almost equal to the permissible concentration of 2.0 µg/g introduced by [66–68], which can be considered a warning risk for the marine organisms in the lagoon. The *Ambassis urotaenia* showed a relatively high total selenium concentration of 1.6 µg/g. Total selenium concentration in the *Cephalopholis hemistiktos* and *Hologymnosus annulatus* species collected from open

seawater showed a relatively high concentration of 1.6 $\mu\text{g/g}$. In contrast, the *Cheilio intermis* showed a normal concentration of 0.80 $\mu\text{g/g}$.

The investigated fish species revealed trophic levels based on the food items 2.7–4.5. The variations pattern of the selenium concentration with the trophic level and the bioaccumulation factor (Figure 6) reflected a directly proportional relationship between the trophic level and the bioaccumulation factor. This can be attributed to the increase in selenium concentration due to its accumulation through species at the beginning of the food web (low trophic level) to the higher trophic level species (see Figure 6). Meanwhile, the trophic level and the selenium concentration revealed an inversely proportional relationship (see Figure 6). This might refer to the fast metabolism in the species within the low trophic level compared to the species' metabolism at the high trophic level [69].

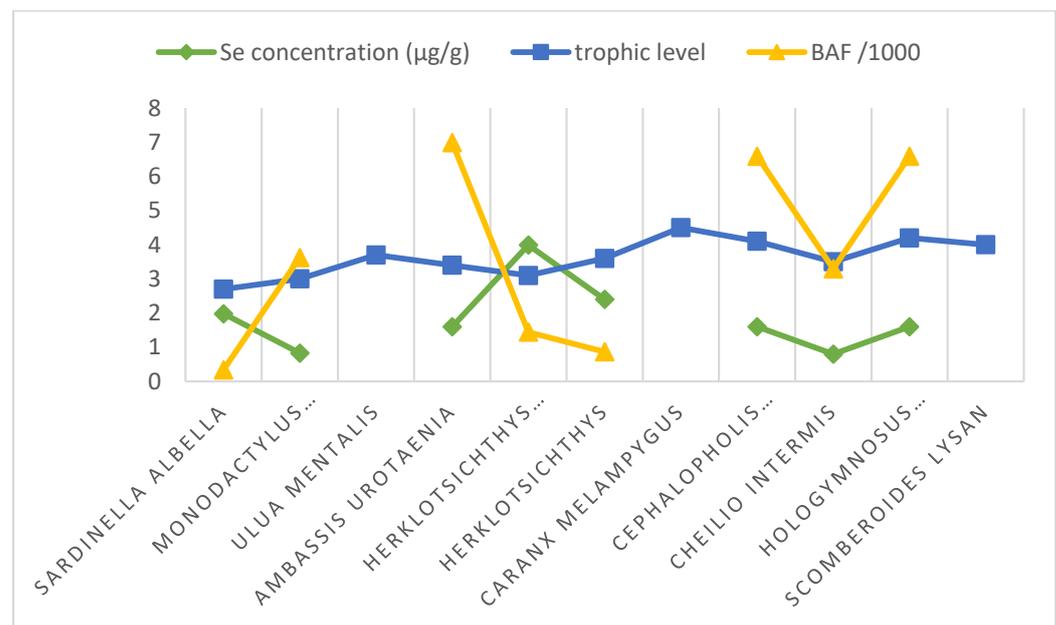


Figure 6. Comparison of selenium concentration among fish species using the bioaccumulation factor and trophic level.

3.2. Sharm Obhur

3.2.1. Selenium Horizontal Distribution

The distribution of the surface TDSe along the Sharm Obhur is shown in Figure 7. The maximum and minimum concentrations were 8.38 and 1.54 nM, respectively, with an average of 3.86 nM, which exceeded the permissible level introduced by [65]. The concentration of the TDSe in the Sharm entrance and the head was higher than the Sharm's middle part (Figure 7). TDSe showed a high concentration in the lagoon head, which decreased toward the lagoon entrance except for stations 1 and 2; relatively high concentrations were observed. This can be attributed to the waste of the university fish aquaculture. The high TDSe concentration at the lagoon can be attributed to water sports activities and villas' domestic waste (see Figure 8).

The concentration of TDSe in the Sharm is high compared with its concentration in the Red Sea open water [70,71] and Reayat Al-Shabab Lagoon [52]. In addition, its average concentration (3.86 nM) is high compared with the average concentration established by [53], 2.94 nM during the same season. Similarly, it is high compared with total selenium concentrations found by [57,72,73].

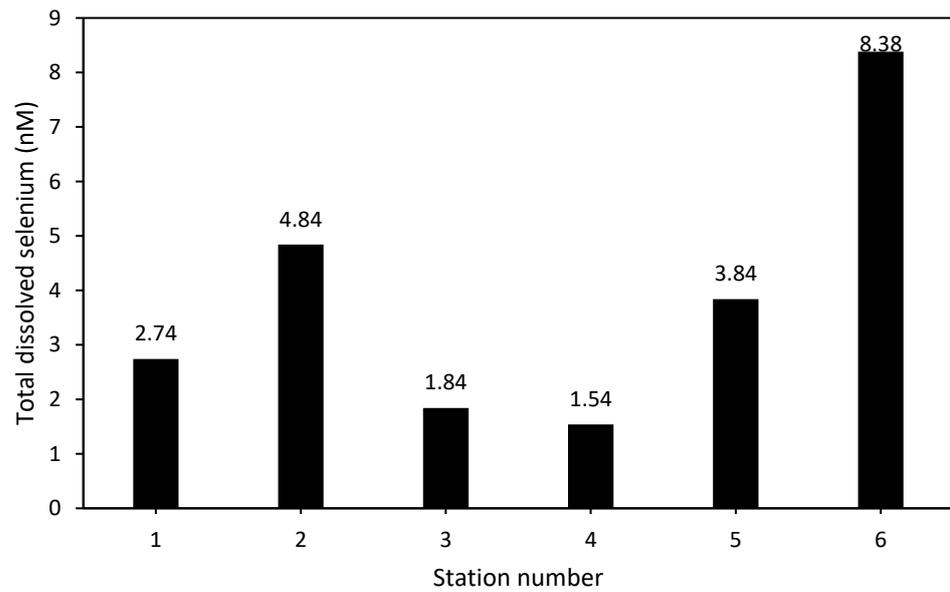


Figure 7. Distribution of the total dissolved selenium in the surface water of the Sharm Obhur.

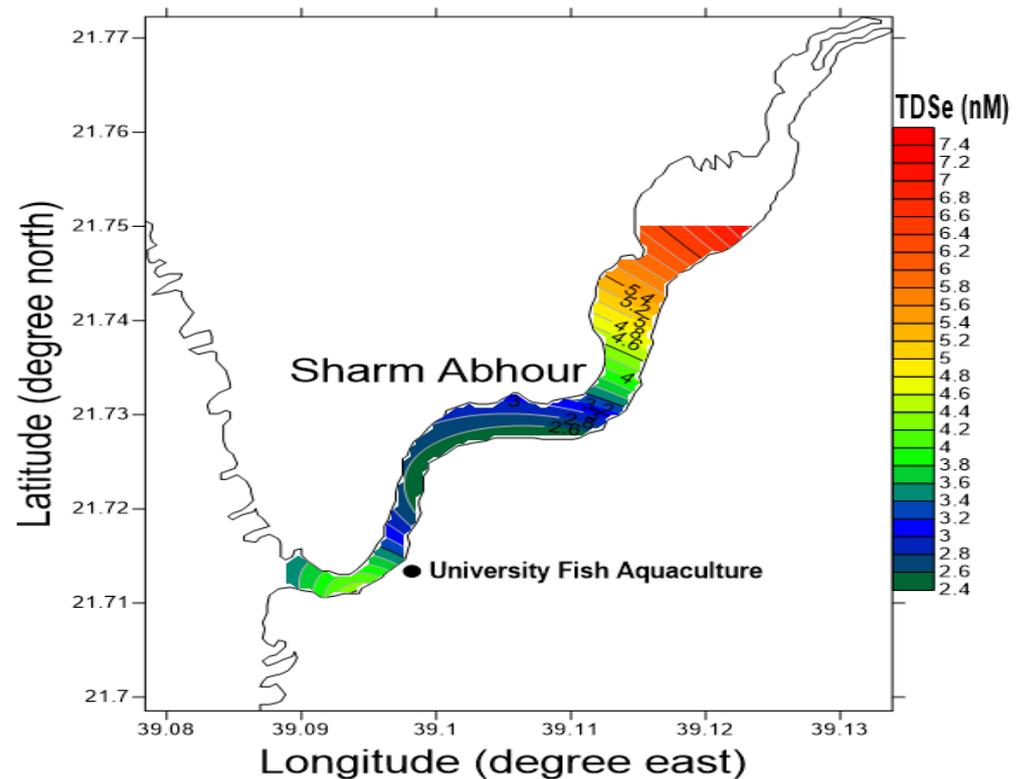


Figure 8. Total dissolved selenium horizontal distribution along the Sharm Obhur surface water.

3.2.2. Physical and Chemical Parameters Properties of the Sharm Water

Eleven physical and chemical parameters were measured: pH, salinity, temperature, dissolved oxygen, suspended particulate matter, nitrate, nitrite, ammonia, dissolved inorganic nitrogen (DIN), phosphate, and chlorophyll a. The surface temperature showed almost the same values with little variations along the Sharm (Table 5). In the Sharm surface water, salinity revealed values within the average seawater salinity (Table 5). The pH in the Sharm surface water showed slightly alkaline pH values. The pH alkaline values (see Figure 9a) can be attributed to the phytoplankton or algal productivity in the Sharm [74] or might be due to the villas domestic wastewater discharging (Table 5). Chlorophyll-

a showed low concentrations compared to Al-Arbaeen and Reayat Al-Shabab lagoons (Table 5) (see Figure 9b). The nutrients revealed low concentration comparably with the seawater nutrient concentration (Table 5). The total organic carbon showed higher concentrations than in Al-Arbaeen and Reayat Al-Shabab lagoons (Table 5). The nitrate depletion at stations 2 and 3 due to the phytoplankton or/and algae utilization (Figure 9c) supports the high chlorophyll-a concentration (Figure 9b). The nitrite reflects low concentration at the Sharm entrance and increases gradually toward the Sharm head (Figure 9e). The ammonia reveals a high concentration near the university fish aquaculture (Figure 9f). A source might be from the aquaculture discharging. The dissolved inorganic nitrogen (DIN) showed a pattern similar to the nitrate and nitrite (Figure 9g).

Table 5. Physical and chemical properties of Sharm Obhur water.

Parameters	Surface Water		
	Max	Min	Mean
Temperature (°C)	26.90	26.80	26.85
Salinity	38.18	37.97	38.04
pH	8.26	8.07	8.21
DO (ml/L)	6.30	5.75	6.12
Chlorophyll a (µg/L)	2.65	1.54	1.85
Nitrate (µM)	4.202	0.114	1.31
Nitrite (µM)	0.798	0.063	0.315
Ammonia (µM)	0.147	0.052	0.091
Phosphate (µM)	0.273	0.081	0.1535
TOC (mg/L)	465.2	162.8	342.03
SPM (g/L)	0.0246	0.0125	0.0194

The phosphate showed a high concentration at the Sharm Entrance and decreased toward the Sharm head. In addition, it revealed apparent depletion at station 4, which might be due to the phytoplankton or/and biodiversity divergence (Figure 9h). The surface temperature in the Sharm inner part was higher than the outer part. This can be attributed to the low tide [75,76] (Figure 9i). The salinity revealed that the relatively high salinity in the Sharm head decreases gradually toward the Sharm entrance. This is due to the high evaporation in the Sharm head [75] (Figure 9j). The suspended particulate matter (SPM) showed relatively low suspended materials (Figure 9k). On the other hand, it showed high values near the discharging points at stations 2 and 4 (Figure 9k).

In the Sharm surface water, nitrate showed a significant positive correlation coefficient of 0.89 versus the salinity (Table 6), which indicates a conservative distribution even in the water close to the university fish aquaculture. Similarly, the dissolved inorganic nitrogen showed a significant positive correlation coefficient of 0.90 against the salinity (Table 6), indicating a conservative distribution in the Sharm water. In addition, phosphate showed a significant positive correlation coefficient of 0.2 against the salinity (Table 6), reflecting a conservative distribution. The total organic carbon (TOC) maximum and minimum concentrations were 465.2 and 162.8 mg/L, respectively, with an average of 342.03 mg/L.

The horizontal surface distribution of the TOC showed a high concentration in the Sharm outer part (Figure 9l) close to the university fish aquaculture and accommodation villas, which might be a source of selenium.

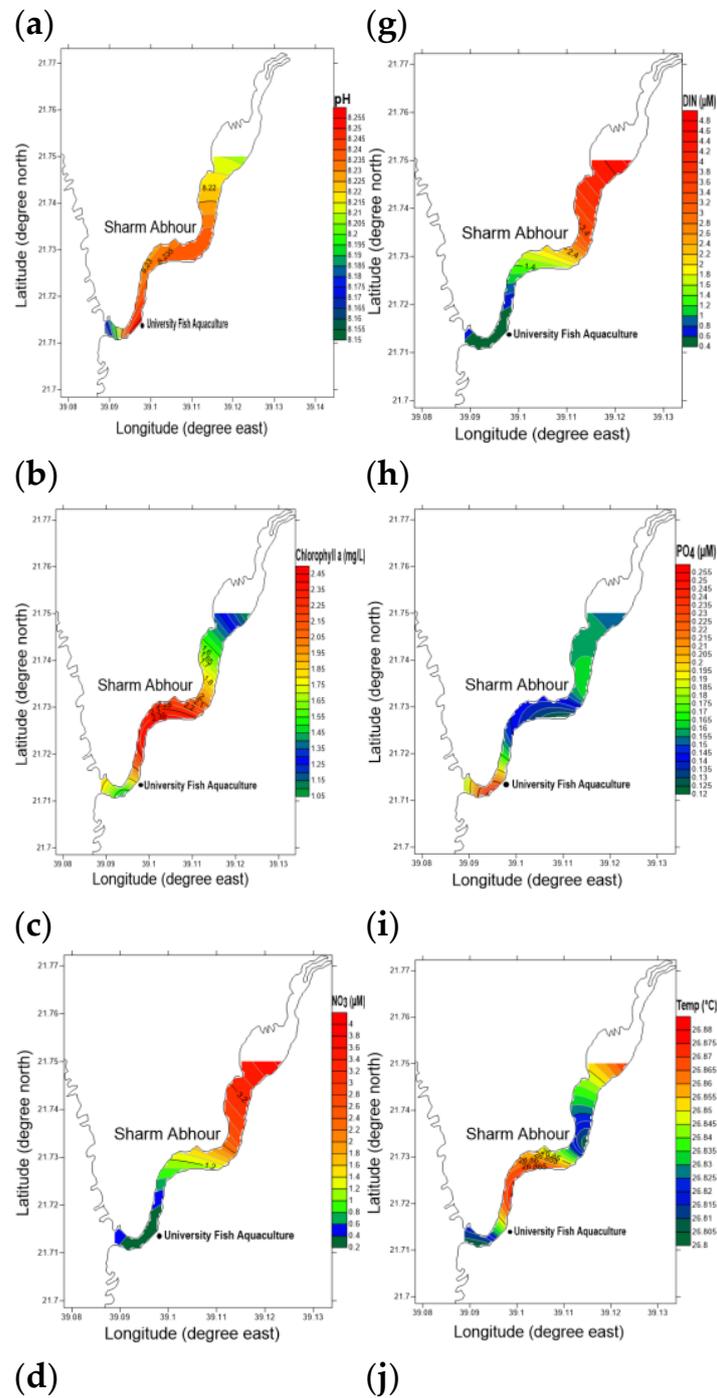


Figure 9. Cont.

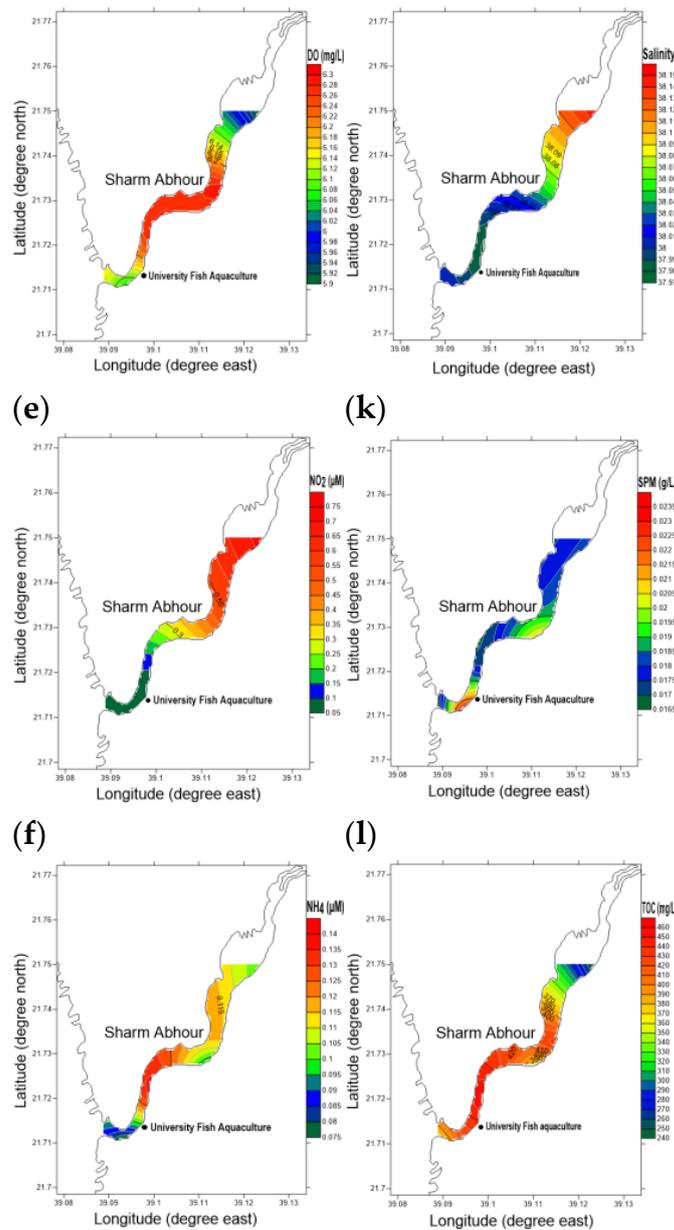


Figure 9. Physical and chemical parameters horizontal distribution along the Sharm Obhur surface water.

Table 6. Nutrient correlation coefficient against salinity in the surface and bottom water of Sharm Obhur ($n = 6$ stations).

		Salinity
Surface Water	NO_3^-	0.89 ($p < 0.05$)
	PO_4^{3-}	0.20 ($p < 0.05$)
	DIN	0.90 ($p < 0.05$)

3.2.3. Selenium Behavior, Sources, and Its Relationship to the Physical and Chemical Parameters

TDSe in the surface water of the Sharm reflected a significant positive correlation coefficient (Table 7) against the salinity, which indicates a conservative distribution behavior. This does not mean that there is no anthropogenic selenium, due to human activities because there are many hotels, water sports activities, and the university fish aquaculture.

These activities can be considered as selenium sources to the Sharm. In the same regard, TDSe in the surface water showed a positive correlation coefficient of 0.68 against nitrate, which reflects its role as a nutrient (Table 7). TDSe in the surface water reflected a positive correlation coefficient of 0.85 versus phosphate, which indicates its role as a nutrient (Table 7). TDSe in the surface water of the Sharm revealed a significant negative correlation coefficient versus the chlorophyll a, which verifies its utilization in the biological process (Table 7). TDSe against the TOC showed a negative correlation coefficient of 0.6 (Table 7). This reflects that TDSe in the Sharm is utilized by biological processes [60,61] and removed from the water due to the high TOC concentration.

Table 7. Physical and chemical parameters correlation coefficient against total dissolved selenium in the surface and bottom water of Sharm Obhur ($n = 6$ stations).

Surface Water	Total Dissolved Selenium (TDSe)	NO_3^-	PO_4^{3-}	Chlorophyll a	Total Organic Carbon	Salinity
		0.68 ($p > 0.05$)	0.85 ($p < 0.05$)	0.91 ($p < 0.05$)	0.61 ($p > 0.05$)	0.78 ($p < 0.05$)

4. Conclusions

This study investigated selenium concentration levels, distribution patterns, and sources in Al-Arbaeen and Sharm Obhur Lagoons. The dumping of wastewater in Al-Arbaeen lagoon started a long time back, and the wastewater pipes have been determined and are well-known. On the other hand, Sharm Obhur is considered an unpolluted lagoon. However, there are various sources of pollution such as domestic wastewater, fish aquaculture wastewater, and water sports activities. TDSe selenium showed high concentrations compared with Sharm Obhur. However, in the two lagoons, selenium revealed that the high concentration in some stations exceeded the permissible concentration introduced by many organizations. In Al-Arbaeen lagoon, wastewater was the main source of selenium based on the relationship of selenium with salinity, while in Sharm Obhur, the wastewater from fish aquaculture, residence villas, and water sports activities might be the main source of high selenium concentration. Nutrient (nitrate, nitrite, and phosphate) concentration reflected the high concentration in Al-Arbaeen lagoon. The wastewater from pipes was the main nutrient source based on the relationship of nutrient with salinity. The nutrient concentration from Sharm Obhur showed a normal concentration similar to the open seawater concentration. The selenium concentration in fish muscles revealed a risky concentration. Factors that substantially affected the two lagoons' health status were lagoon size and shape, wastewater load, and water circulation with open seawater. The algae growth and oxygen concentration level might dominate the biogeochemistry of different selenium forms in Al-Arbaeen lagoon, while Sharm Obhur is dominated by water circulation and algae growth.

Supplementary Materials: The following supporting information can be downloaded at: <https://www.mdpi.com/article/10.3390/w15040687/s1>.

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