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Selenium Determination, Distribution, Behavior, Sources, and Its Relationship to the Physico-Chemical Parameters in Coastal Polluted Lagoon along Jeddah Coast, Red Sea

Ahmed S. A. Ibrahim [1,2] *& Radwan Al-Farawati [1]

[1] Department of Marine Chemistry, Faculty of Marine Sciences, King Abdulaziz University, P.O.Box 80207, Jeddah 21589, Saudi Arabia
[2]Higher Council of Environment, Urban and Rural Promotion, Ministry of Environment, Natural Recourses and Physical Development, Khartoum, Republic of Sudan.

[E-mail: a.saif42@yahoo.com, aabrahim0002@stu.kau.edu.sa*]

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We determined the total dissolved selenium (TDSe) concentrations in the surface and bottom waters of Reayat Al-Shabab lagoon along Jeddah coast, eastern of Red Sea. The lagoon is a heavily polluted body water due to the dumping of untreated sewage effluents. Our aim is to understand the factors that control the vertical and horizontal distribution, behavior, and sources of selenium. The average concentration of the TDSe in the surface and bottom were 1.56 and 0.45 nM, respectively. The surface concentration of TDSe decreased from the eastern lagoon to lagoon mouth with the highest value of 2.51 nM near the eastern part. The annual selenium flux to the lagoon and the average selenium residence time were 1.0 kg/year and 31 days, respectively. Surface TDSe showed significant positive correlation with nitrate ($R^2 = 0.60$, $P < 0.05$), reflecting its role in the biological process. In addition, the TDSe concentration significantly correlated well with chlorophyll a ($R^2 = 0.70$, $P < 0.05$), indicating its role as a nutrient. The TDSe did not reach the toxic level of selenium, thus it can be utilized by marine organisms for normal growth. The bioavailability and mobility of dissolved selenium mainly depend on the dissolved oxygen availability (redox potential), so in this study, we suggest that in the surface water selenium has more mobility and bioavailability in comparison to bottom water. In the absence of strong and negative correlation of selenium with salinity, the concentration of selenium is mainly controlled by biogeochemical processes (conservative behavior) such as utilization by marine organisms, redox potential, and diffusion from sediments.

[**Keywords:** TDSe distribution, Physico-Chemical parameters, Chlorophyll a, Conservative behavior, Jeddah coast, Red Sea]

Introduction

Selenium is a non-metal element in the fourth period of group sixteen of the periodic table. The elements of this group are distinguished by high electronegativity, so they have a strong ability to accept one or more pair of electron[1]. Selenium is an essential element for human health, animal, and some plant at low concentrations. While it considers as a toxic element at high concentrations[2, 3]. Also, it plays an important role in the cell structure and as a protecting agent against oxidative damage. The exposure to selenium leads to poisoning, which causes reproductive failure, a mutation in the egg production of the marine life (fish, birds, and reptiles)[4-7]. And the other Selenium toxicity causes mortality, mass wasting in adults, reduced juvenile progress, and immune suppression[8]. Selenium can be found hydrolyzed as oxyanions Selenite (SeO_3^{-2}), and selenate (SeO_4^{-2}) soluble in the sea water. The solubility and mobility of these forms, increase with the increasing of pH. It is speciation and bioavailability is quite important in the determination of Selenium fate, and its effect in the environment[9]. Selenium toxicity is controlled by the concentration and chemical forms[10, 11]. It has distinguished eco-toxicological features which show very narrow limit between toxicity and nutritionally[12-17].

In the natural environment, selenium exists in three chemical oxidation states; Se (IV), Se (VI), and Se (II), and as Se (0)[18]. These three states control the physiological conduct of the element[11]. Selenite and selenate follow nutrient type as bioactive elements, thus phytoplankton consumes dissolved inorganic selenium in the ocean surface layer[19]. Selenium biogeochemistry consists of inorganic selenium converted to proteins, and the regeneration of the organic selenide from organic detritus[20]. There are many factors dominate selenium distribution involve the sources, selenium paths to the environment, seawater characteristics (include salinity, total organic carbon, depth, temperature, and pH), ocean current, physical, chemical and biological activities[21]. Water circulation can affect the distribution of selenium[22]. Also, the temperature and pH can control the solubility and behavior of selenium in aquatic environments[23]. Salinity and total organic carbon can also explain the selenium sources, distribution, behavior, and speciation[24, 25]. Selenium determination, distribution, behavior, mobility, sources, and toxicity in the marine environments have been conducted to extensive studies.

This study in the Red Sea, Jeddah coast is considered as the first study in the Red Sea region. The main objectives of this study are to investigate the sources, behavior, determination, and distribution, of the total dissolved selenium in Reayat Al-Shabab Lagoon and its relationship to the physico-chemical parameters.

Materials and Methods

Study Area

This study was conducted in the Red Sea coastal water in front of Jeddah city, at Reayat Al-Shabab lagoon and complete depiction can be found elsewhere[26]. It is located in the central part of Jeddah city in the vicinity of Jeddah Islamic Port. Reayat Al-Shabab lagoon is a semi-enclosed lagoon, which receives 35,000 m^3 of treated and raw sewage daily, and in contacts with open seawater[26-31]. The western part is connected to the open water through narrow channel hitch limits water circulation. There are two effluent discharge pipes, were located on the land facing the lagoon water, and the other one was extended under the water with lagoon ground to discharge at lagoon mouth. Sill depth is about 2 m, control the water exchange between the lagoon and open seawater. The subsurface water of the lagoon can be mixed under the rough weather. These conditions induce the accumulation of the organic compounds in the lagoon bottom water.

Sample Collection

Eight stations were designed along Reayat Al-Shabab lagoon. Samples were collected in morning from latitudes 21° 30″ 6.3′ to 21° 29″ 45.6′ during 28 April 2015. Water samples were collected from two depths 0.5 m (surface), and 3 m (bottom) using the peristaltic pump device. The water samples were filtered in the laboratory through 0.45 µm filter, and the filtrates were stored at 4 °C in 500 ml high density polyethylene bottles containing HCl, adjusted to pH = 2. Nutrient water samples were collected in 250 ml polyethylene bottles and stored frozen.

Analytical Methods

Total Dissolved Selenium Determination

TDSe was determined using voltammetry technique, cathodic stripping voltammetry, and differential pulse mode, according to the modified method of[32]. The method for UV photolysis established by[11] was used. Briefly, - 0.4 v deposition potential was used. And deposition time of 10 min was used. The mercury, silver-sliver chloride and platinum electrodes were used as working, reference, and auxiliary electrodes, respectively. 797 VA Computrace Metrohm device was used, triplicate measurements for each sample and standard solutions were performed, and standard addition method was implemented using selenite standard solutions of 10^{-4}, 10^{-5}, 10^{-6}, and 10^{-7} M. Standard solutions and reagents were prepared using Milli-Q water (Resistivity = 18.2 MΩ cm^{-1}). The accuracy of the instrument measurement for this method was evaluated using spiking and recovery. The sample solution of 10 nM Selenium was spiked by 20 nM and 101.3 % recovery was obtained. The reliable precision defined as relative standard deviation was 6.70 % (N = 7) for 0.1 nM of Selenium. The limit of detection based the three times blank standard deviation divided by the calibration curve slope of 0.02 nM was obtained for a deposition time of 10 min.

Nutrients Analysis and Dissolved Oxygen

Nutrients analysis were performed, nitrate was measured using copper coated cadmium metal column to reduce dissolved nitrate to nitrite, and spectrophotometric was conducted by adding of N-(1-naphthyl)-ethylenediamine dihydrochloride[33]. Nitrite was measured by the same method of nitrate. Phosphate was measured by[34]. Ammonia was measured according to[35]. And according to the Winkler method dissolved oxygen was determined[36]. Calibration curve for nitrate and nitrite were plotted using standard solutions of 0.5, 1, 3, 5, 10, 15 µM. Standard solutions and reagents were prepared using Milli-Q water (Resistivity = 18.2 MΩ cm^{-1}).

Physical parameters and Chlorophyll a

Temperature, pH, and chlorophyll a were measured using the multiparameter water quality sonde 6600 V_2 with 650 MDS. Salinity was measured using micro-salinometer MS-310. The instrument sensors were calibrated prior to the field measurements using the YSI calibration methods of the instrument company and their special standard solutions.

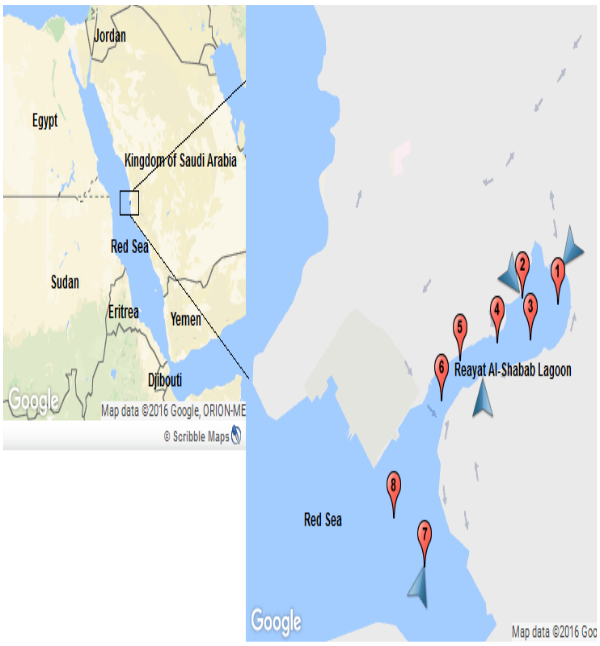


Fig. 1 Red Sea map and the samples sites along Reayat Al-Shabab lagoon including the pollution sources as depicted by blue arrows.

Results and Discussion

Selenium Vertical and Horizontal Distribution

The distribution and concentration of the total dissolved selenium (TDSe) in the surface and bottom of the Reayat Al-Shabab lagoon were obtained as shown in the (Fig. 2). In the surface water, the maximum and minimum concentrations of the total dissolved selenium were 2.51 and 0.63 nM, respectively. The average concentration of the TDSe in the surface water was 1.56 nM. While in the bottom water, the maximum and minimum concentrations of the TDSe were 0.79 and 0.16 nM, respectively. The average concentration of the TDSe in the bottom water was 0.45 nM. In the same regard, in the surface water, TDSe concentrations were decreased from the lagoon head toward the open sea water. This indicates the seawater dilution effect and the anthropogenic activities over there. TDSe in the bottom water did not show clear variation along the lagoon water. According to the lagoon conditions (hypoxia), there are many factors (pH, salinity, dissolved oxygen, and vertical mixing) controlling the distribution of selenium in the lagoon. The TDSe in the lagoon was not showed nutrients type vertical profile, disagreed with many previous studies[37-42]. In the bottom water of the lagoon, the reduction condition is dominated by a high concentration of organic matter, ammonia, hydrogen sulfide, and consumption of dissolved oxygen, thus, all selenium species might be preferred to be in the reducible form or elemental form, settled into the sediment[43, 44]. This also explains why the total dissolved selenium did not follow nutrients type behavior over the lagoon.

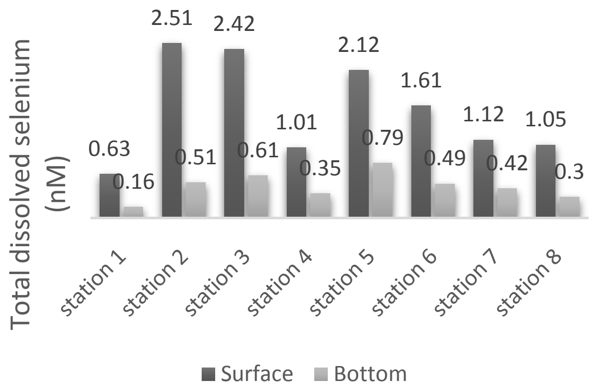


Fig.2 Distribution of the TDSe in the surface and bottom of the Reayat Al-Shabab Lagoon.

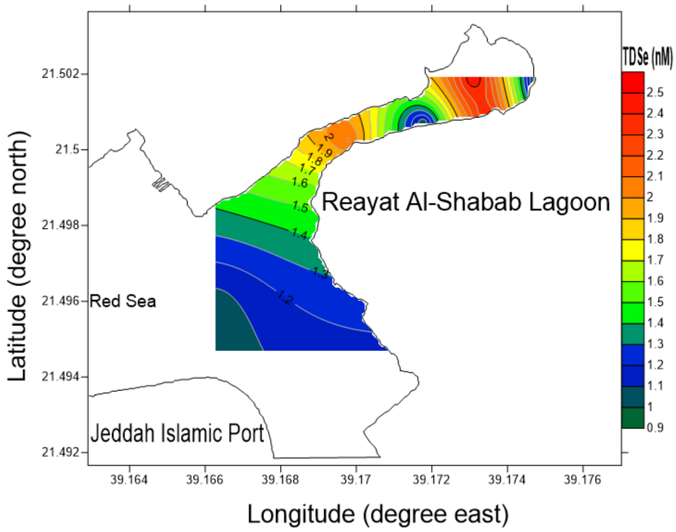
The average concentration of total dissolved selenium over Reayat Al-Shabab lagoon is less than the value found in the Mariut Lagoon in the Mediterranean Sea which reached 12.02 nM[20].

The horizontal distribution of the TDSe in the surface and bottom of the lagoon revealed high concentrations over stations 2, 3, and 5 (see Fig. 3a and Fig. 3b) which are located close the discharging pipes (see Fig. 1). While the other discharging pipes did not show high concentration. In the bottom water, TDSe may reach zero which dissolved oxygen is zero and hydrogen sulfide is high. The vertical cross section of the TDSe along the lagoon also confirmed that the higher concentrations closed the discharging sources. The TDSe concentration was 1 nM at about 1.5 m depth and after that decreased to reach very low values (see Fig. 4). The selenium input from the water discharging to the lagoon is very low as represented by the stations 2, 3, and 5 concentrations values, compared with the open seawater selenium (Fig. 3 and Fig. 4), and also was obvious from the positive correlation between the TDSe and salinity (Figure 9d).

Physical and Chemical Parameters of Seawater

We measured the temperature, pH, salinity, dissolved oxygen, chlorophyll a, nitrate, nitrite, ammonia, and phosphate of sea water in Reayat Al-Shabab lagoon. The surface temperature ranged between 26.2 and 28.1 °C, whereas the bottom temperature ranged between 25.4 and 27.7 °C with a slightly different between the surface and bottom temperature due to the shallow water column along the lagoon. The pH of the surface was 8.10 to 8.38, with an average of 8.31, and the bottom was 8.01 to 8.36, with an average of 8.23. It is considered within the normal pH range of marine water. The salinity of the surface was ranged from 34.22 to 38.01 with an average of 37.10 which is lower than bottom salinity that ranged between 38.31 and 38.44 with an average of 38.37, indicating the effect of the wastewater discharge. The salinity at station 7 was 34.22 located over the wastewater discharge pipe. The dissolved oxygen at the surface was ranging from 5.50 to 9.23 ml/L with an average of 6.57 ml/L under the oxic condition. In contrast, the dissolved oxygen at the bottom was lower, ranging from 1.41 to 6.62 ml/L with an average of 3.29 ml/L. The chlorophyll a ranged between 10 and 25 µg/L with an average of 18.18 µg/L and highest concentrations were evident in the eastern part of the lagoon compared to the western part. The surface nitrate concentrations ranged from 1.677 to 16.885 µM with an average of 9.01 µM. The surface nitrite concentrations ranged from 0.273 to 11.739 µM with an average of 2.62 µM. The bottom nitrate and nitrite concentrations varied between 0.044 and 0.652 µM, 0.063 and 0.273 µM with an average of 0.40 µM, 0.19 µM, respectively. The surface phosphate concentrations were in the range between 0.912 and 6.624 µM with an average of 2.65 µM, and the bottom phosphate concentrations ranged from 1.008 to 4.416 µM, with an average of 2.66 µM. The surface ammonium concentrations ranged from 0.602 to 6.062 µM with an average of 2.32 µM, and bottom ammonium concentrations ranged from 0.064 to 5.464 µM with an average of 2.73 µM. The nutrients concentration showed high concentrations due to the waste water discharging over the lagoon. The surface nitrate and surface phosphate versus the salinity (Fig. 5a and Fig. 5b) showed a negative correlation coefficient 0.80 and 0.90 respectively, which confirm that fresh wastewater is the source of the nutrients. Bottom phosphate against salinity displayed a negative correlation coefficient 0.90, (Fig. 6b) which support that fresh wastewater is the source of the nutrients. In contrast, bottom nitrate versus salinity showed a weak positive correlation coefficient, and this is might be due to its seawater origin.

(a)



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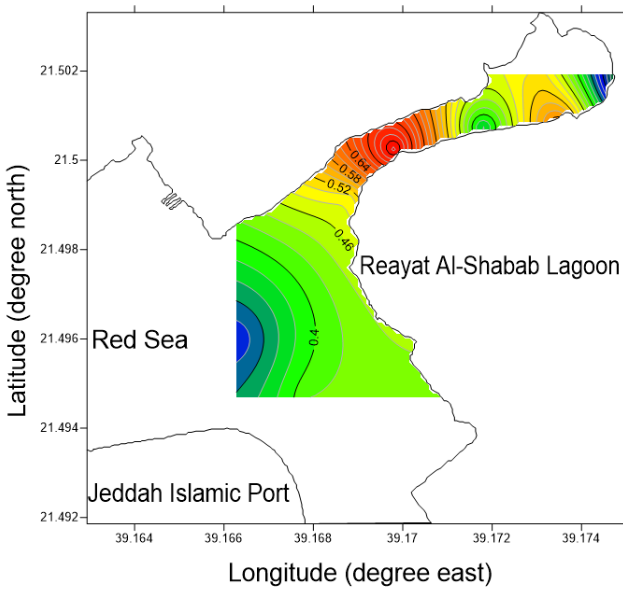


Fig. 3 Total dissolved selenium horizontal distribution along the Reayat Al-Shabab lagoon (a) surface water and (b) bottom water.

Also, the surface and bottom dissolved inorganic nitrogen (as the total sum of NO_3^-, NO_2^-, and NH_4^+), versus salinity along Reayat Al-Shabab lagoon (Fig. 7a and Fig. 7b) showed a negative correlation coefficient 0.80 and 0.90, respectively. These indicate that fresh waste water is a source of nutrients.

Physico-chemical parameters (salinity, and temperature) (see Fig. 8a), over the lagoon, reveal that four sources of waste water discharging, two pipes close to the lagoon head (station 2, 3, and 5). The fourth one lies at the lagoon entrance. The discharging from pipes in lagoon head is not too much (salinity = 37.5), compared with the third pipe that takes place at the lagoon entrance (salinity from 34 to 36.5). Nitrate concentration is high at the same positions (see Fig. 8) where salinity is low, this is a good prove that waste water discharging is a source of nitrate. Also, phosphate vertical section shows the same fact as nitrate (see Fig. 8). Nitrite concentration is low in the lagoon bottom water, and surface water closed to the lagoon head (see Fig. 8). On the other hand, it shows high concentration in surface water in the lagoon entrance, where pipe takes place. For ammonium, there are two main sources (see Fig. 8), first one is the pipe lies at the lagoon entrance, and the second one is the lagoon head bottom water, and it increases up to the lagoon surface water, where denitrification process dominants (dissolved oxygen is very low and reach zero). The total dissolved selenium is high over the lagoon surface water compared to the bottom water (see Fig. 4) where ammonium is high and oxygen is low (hypoxia condition). Chlorophyll a concentration is high in lagoon head bottom water (see Fig. 8), at the depth of low oxygen (see Fig. 8), high ammonium, and low total dissolved selenium where the respiration takes place. So, might be, most of the selenium species will be in selenium elemental form in the sediment as we mentioned before.

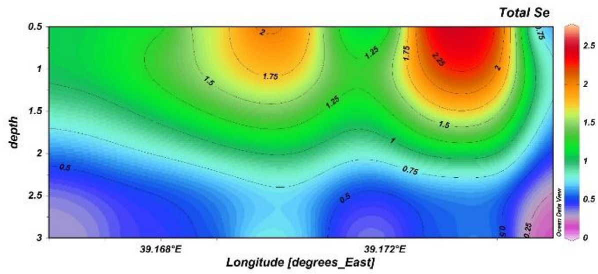
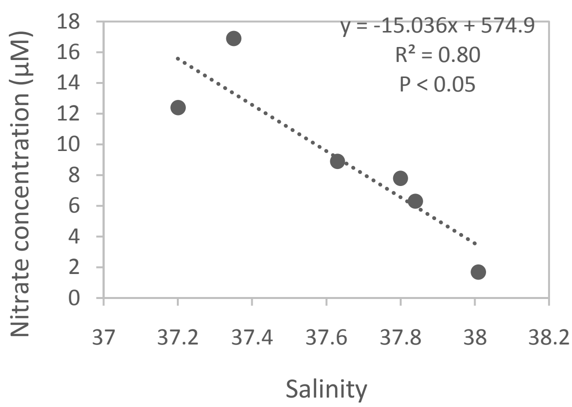


Fig. 4 Total dissolved selenium vertical cross section along the Reayat Al-Shabab Lagoon.

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

In the surface, TDSe versus nitrate (see Fig. 9a) showed a positive correlation coefficient (R^2 = 0.60), indicating that it involves in the biological processes in the lagoon. In contrast, negative correlation coefficient (R^2 = 0.53) between phosphate and TDSe was obtained (Fig. 9b). Chlorophyll a versus TDSe displayed a positive correlation coefficient (R^2 = 0.70), reflect that it is associated with chlorophyll a as a nutrient (Se IV), and thus, it associates with the growing of phytoplankton in the lagoon (Fig. 9c)[45].

(a)



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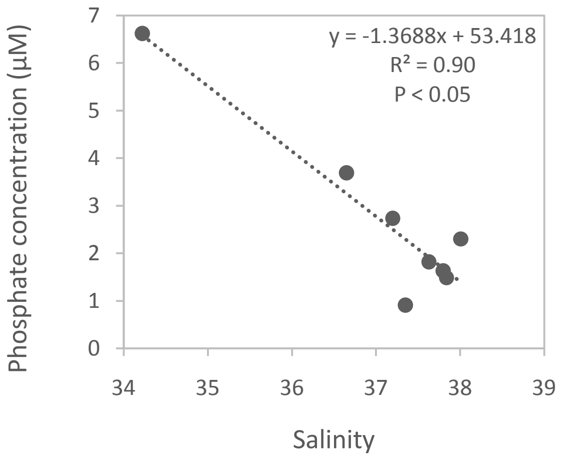


Fig. 5 Correlation coefficients of the surface (a) nitrate and (b) phosphate against the salinity in the lagoon.

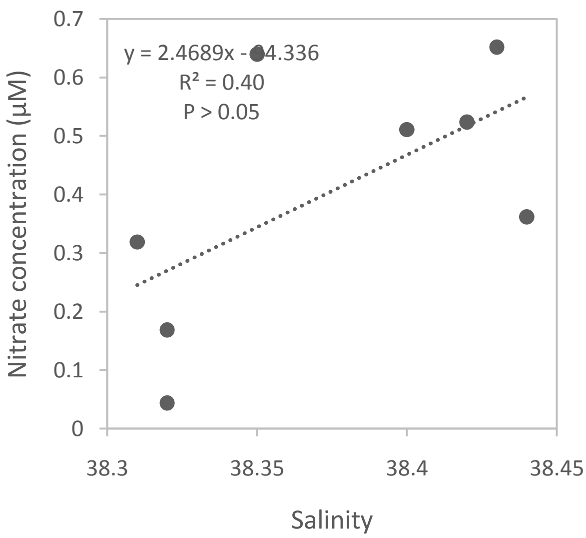
TDSe against salinity along Reayat Al-Shabab lagoon displayed a positive correlation coefficient ($R^2 = 0.45$), indicating that seawater is a source of selenium (conservative behavior), and there is no selenium originates from waste water dumping except selenium concentrations from stations 2, 3, and 5. Pipes closed to these stations might contribute to the selenium total input in the lagoon (Fig. 9d). In the surface water, TDSe showed a positive correlation coefficient against dissolved oxygen ($R^2 = 0.74$), hence, by the increasing of dissolved oxygen (see Fig. 9e), TDSe is being available for marine organism due to high redox potential[46, 47], and its transfer through the food chain is easier. Result to the accumulation of selenium through the food chain may lead to toxic symptoms.

In the bottom water, TDSe did not show any significant correlation coefficient against nutrients, chlorophyll a, and salinity. TDSe in this study did not reach the toxic level in the fresh water, thus, it can be utilized by marine organisms for normal growth[13, 48, 49]. Also, the TDSe concentration at the surface and bottom of Reayat Al-Shabab was lying under the acceptable concentration level of Canadian drinking water guidelines 1978[50]. The estimated annual TDSe flux to the lagoon calculated based on the TDSe average concentration in the lagoon and wastewater annual flux is 1.00 kg/year and the TDSe average residence time based on the TDSe average concentration and the lagoon volume is 31 days. The selenium annual flux from the wastewater discharging is low compared with the selenium annual flux calculated by Abdel-Moati [20] which is less than flux may lead to the toxic effects. The residence time value calculated above causes the selenium remove from the seawater by reduction (hypoxia and anoxia conditions) of the dissolved species to elemental selenium or selenide in the sediment and volatile species in the atmosphere.

Comparison of total dissolved selenium concentration with some previous studies

The average concentration of TDSe in the lagoon was 1.01 nM, while Cutter, found that the average total selenium was 2.27 nM from San Francisco Bay[51]. On the other hand, Takayanagi established that the average total selenium was 1.48 nM from Southern Chesapeake Bay[52]. Also, from the pristine river-estuarine system, Patrick found that total dissolved selenium was 2.45 nM[53]. While Abdullah found that total selenium in Vest Fjord varied from 0.89 to 1.51 nM, and in the same study, from Bunne Fjord, he found that total selenium ranged from 1.22 to 1.70 nM. Dao 1991, found that the average total selenium from Rhone River was equal to 2.30 nM, and from the Gulf of Lion off the Mediterranean Sea varied from 0.5 nM at the surface to 0.9 nM in deep water[54]. Safa 2015, established that the average concentration of total dissolved selenium was equal to 0.00006 µM from Marsa Matrouh Beaches, northwestern Mediterranean coast[55].

(a)



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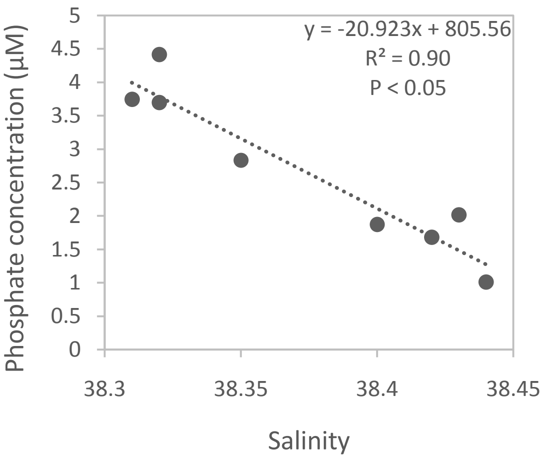
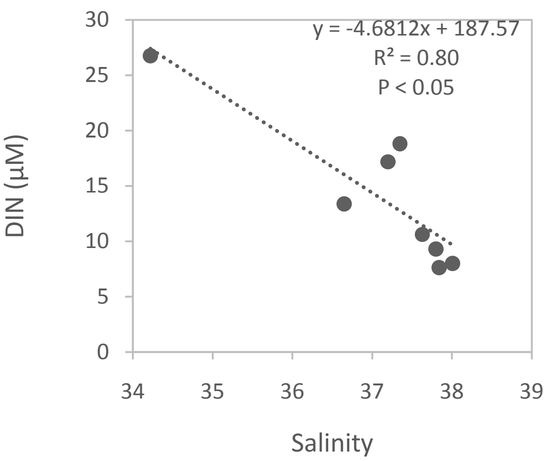


Fig. 6 Correlation coefficients of the bottom nitrate and phosphate against the salinity in the lagoon.

(a)



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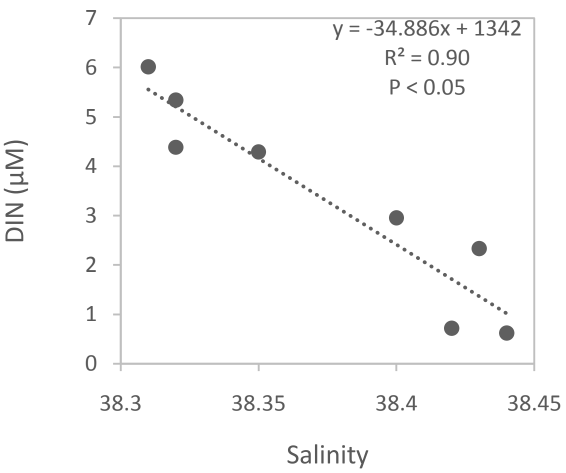

Fig. 7 Correlation coefficients of the (a) surface and (b) bottom dissolved inorganic nitrogen (DIN) against the salinity in the lagoon.

Conclusions

The TDSe at the surface of Reayat Al-Shabab lagoon showed a decreasing from the eastern lagoon to the lagoon mouth. While in the bottom part, TDSe did not show clear sequences. In this study, selenium vertical distributions did not show the nutrient type behavior. In the bottom water also the concentration of dissolved oxygen is low, due to the high concentration of organic matter, thus, all selenium species preferred to be in reduced form. This was proved by the short average residence time of 31 days and the low annual selenium flux of 1 kg/year.

Chlorophyll a and total dissolved selenium displayed a significant positive correlation coefficient, and that confirms its association in the biological process in the lagoon. Selenium distribution in Reayat Al-Shabab lagoon is controlled by physical and chemical properties of seawater to a certain degree. The positive correlation coefficient between total dissolved selenium and nitrate confirmed that selenium is utilized by the biological processes in the lagoon. Our future research will concern with the determination of vertical and horizontal distribution for each selenium species, and how it is linked to the physical and chemical characteristics of the lagoon seawater.

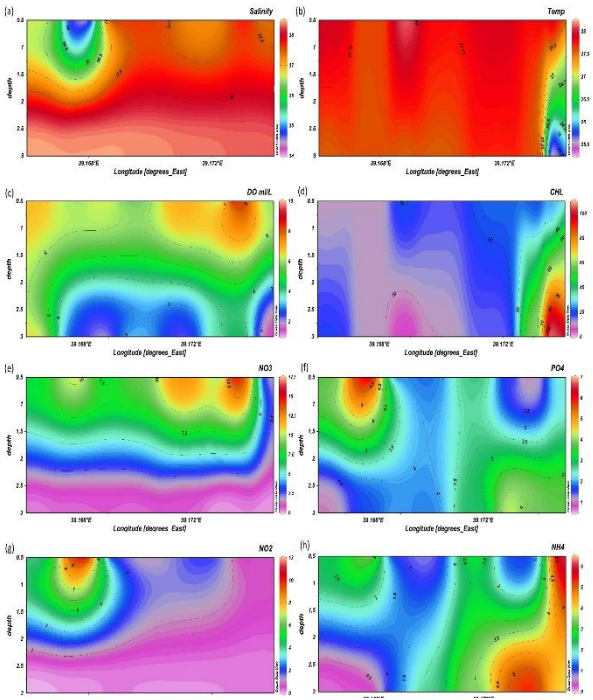

Fig. 8 Vertical cross sections of (a) temperature, (b) salinity, (c) nitrate, (d) nitrite, (e) ammonia, (f) phosphate, (g) dissolved oxygen, and (h) chlorophyll a along Al-Shabab lagoon.

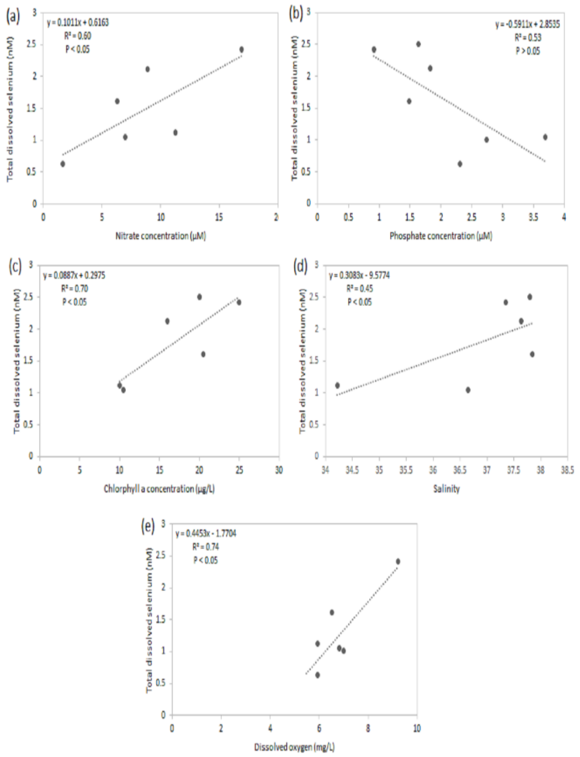

Fig. 9 Correlation coefficients between the surface total dissolved selenium and (a) nitrate, (b) phosphate (c) chlorophyll a, (d) salinity, and (e) dissolved oxygen.

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