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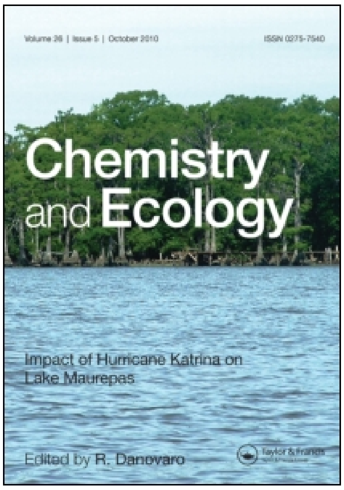

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Potential environmental impact of dissolution of raw phosphate in sea water of the Gulf of Aqaba

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Dissolution of raw phosphate (apatite) in sea water of the Gulf of Aqaba was investigated through lab incubation experiments. Three types from three different sources (Al-Hasa, Al-Abyad and Esh-Shydiya) have been used for these experiments. Impact of quantity, grain size, and source (type) of raw phosphate on dissolution rate were studied. Statistical analysis shows significant differences between the results obtained from comparing each two weights; as weight of apatite increased, dissolved inorganic phosphate-phosphorus (DIP) and fluoride in sea water solution increased. The differences between the dissolution rates of raw phosphate from the three sites were not significant while the differences between the different grain size fractions were significant. Dissolution rates were inversely related to particle size. Using a worst-case scenario, a conservative estimate of the maximum increase in DIP in seawater of the Gulf of Aqaba due to the apatite particles lost to the sea during ship loading resulted in DIP concentrations of 0.03 μM per year. As the residence time of the water in the Gulf of Aqaba is about one year, the DIP concentration will not increase by more than 0.03 μM under the estimated annual quantity of exported phosphate. Fluoride will not increase by more than 0.03 mg/l under the same conditions.

Keywords: apatite; dissolution; phosphate; Gulf of Aqaba

1. Introduction

The water of the Gulf of Aqaba is considered oligotrophic water, containing low inorganic nitrogen (1.0 μM) and phosphate (0.2 μM) [1]. The water of the Gulf is also characterised by high clarity, temperature (20–26 °C) and dissolved oxygen (6–8 $\mathrm{mg\,l^{-1}}$) which create almost ideal conditions for the sustenance of an abundant and diverse ecosystem [2]. At the heart of this ecosystem lies the Gulf of Aqaba fringing coral reef which embraces very diverse, unique and rare species of animals and plants. Unfortunately, this semi-enclosed body of water has witnessed huge environmental pressures due to expansion of urban communities and industrial and shipping activities including the raw phosphate export through the Aqaba Port.

Huge reserves of phosphate rocks are found in three principal phosphate bearing areas at Al-Hasa, Al-Abyad and Esh-Shydiya (Figure 1). The deposits which belong to the uppermost cretaceous [3–5], are mainly carbonate fluoroapatite (Francolite) rocks that contain about 53%

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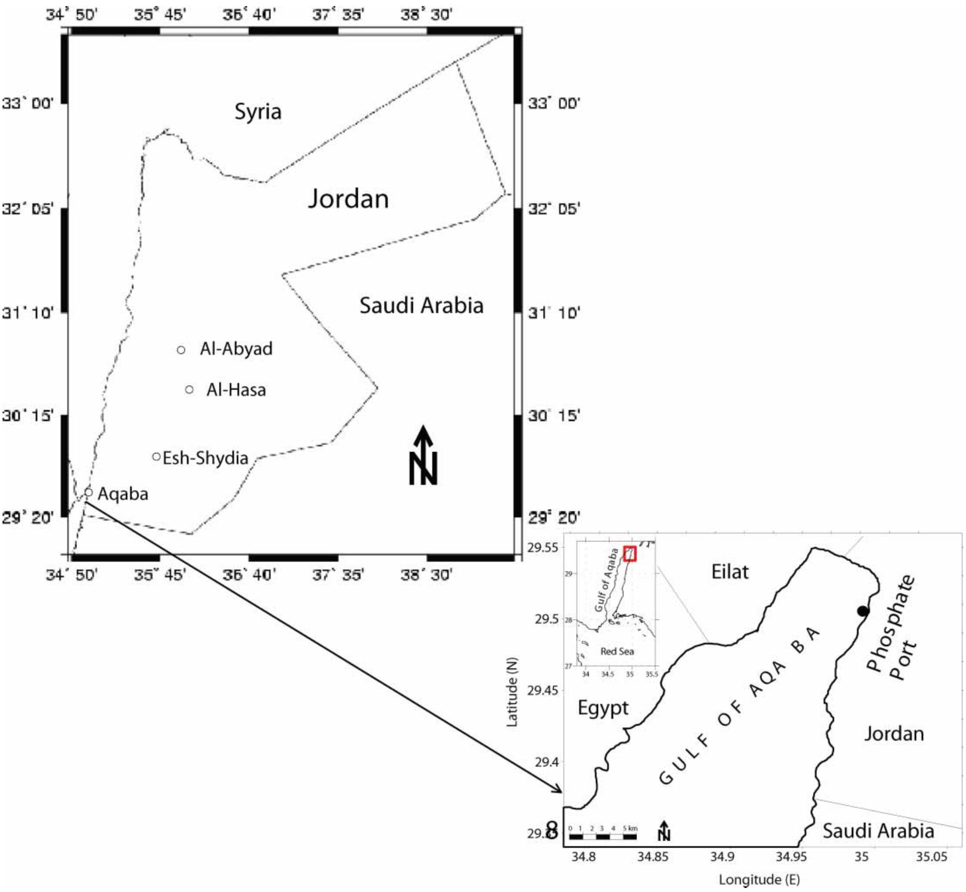

Figure 1. Map of Jordan showing the source areas of phosphate rocks (Al-Hasa, Al-Abyad and Esh-Shydya) and the Gulf of Aqaba where raw phosphate is exported.

calcium as CaO ($CaO = Ca \times 1.3392$), 32% phosphorus as P_2O_5 ($P_2O_5 = P \times 2.2916$) and 4% fluorine. Explorations have proved the existence of large deposits in the above mentioned areas but particularly in Esh-Shydiya 35 km south of Ma'an city in south Jordan (Figure 1). According to these explorations, phosphate formation covers about 60% of the total area of Jordan (Jordan Phosphate Mines Company, personal communication). This is why phosphate rock is considered the main natural resource in the country. The powder of this rock has been marketed internationally for the past 60 years. The marketable raw material is transported to Aqaba by trains and trucks and exported through Aqaba Port at the most northeastern tip of the Gulf of Aqaba (Figure 1). The annual exports ranged between 3.5–4.0 million tons during the past four years. On average, more than 3.5 million tons are exported through Aqaba Port each year (Jordan Phosphate Mines Company, personal communication). Phosphate to be exported is stored in six warehouses at Aqaba Port, with a total capacity of 0.34 million tons. Two berths are available for shipment, where phosphate is loaded onto ships by the use of one stationary and two mobile ship loaders. The two berths can handle up to 8 million tons of phosphate rock per a year. Unfortunately, during transportation, storage and loading some phosphate is lost to the atmosphere, to the land around the storage areas and near the loading berths, and to the sea. Estimation of the lost quantities varies between 0.05% [6], 0.1% [7] and 1% [8]. In all cases, however, substantial quantities of phosphate particles are lost to the water of the Gulf of Aqaba during the loading process. The lost raw phosphate particles which enter the sea water may partially dissolved and form DIP [9, 10] or may settle and deposit onto the sea floor as a particulate phosphate. The phosphate particles whether fluoroapatite $Ca_5\ (PO_4)_3$ F or hydroxyl apatite $Ca_5\ (PO_4)_3$ (OH) dissolve in sea water until an equilibrium is reached according to the equations

$$Ca_5(PO_4)_3F \longleftrightarrow 5Ca^{2+} + 3PO_4^{3-} + F^-$$

$$Ca_5(PO_4)_3OH \longleftrightarrow 5Ca^{2+} + 3PO_4^{3-} + OH^-$$

This equilibrium depends mainly on many factors that include the nature of the rocks, and the acidity and temperature of the water [9, 10]. The dissolution of the lost particles in seawater and the potential formation of DIP, which may increase the natural concentration of this nutrient in the water of the Gulf, was always a major concern to the scientists as well as coastal managers in this region. Therefore, phosphate export has been always considered one of the activities that may cause serious effects on the coral reefs of the Gulf of Aqaba, and many scientists, interested groups and decision makers were involved in long and historical debates on its potential environmental effects. The debates are based on the theory of any activity that may increase the concentration of phosphate and or nitrogen micronutrients can adversely impact coral reefs and other biota in the Gulf of Aqaba.

The present study was initiated to investigate the dissolution of phosphate rock particles of various grain size and various sources (types) when these particles enter the Gulf waters and to estimate the effect of the resulting DIP on the total mass of this nutrient in the Gulf of Aqaba. Such information is necessary and up most urgent for the decision makers and coastal zone managers in Aqaba. This paper represents the first report on the results that have been obtained from this study.

2. Material and methods

2.1. *Sample source and processing*

Representative raw phosphate powder samples from the three mining areas of Al-Hasa, Al-Abyad and Esh-Shydiya to be exported through Aqaba Port (Figure 1) were provided by the Jordan Phosphate Mines Company. Some of the major chemical constituents of these deposits are presented in Table 1. In addition to their sources, the raw phosphates are coded according to their tricalcium phosphate (TCP) content (e.g. Al-Abyad 70/72, Al-Hasa 73/75 and Esh-Shydiya 73/75) where $TCP = \%P_2O_5 \times 2.185$. Samples were analysed for grain size distribution using a set of calibrated stainless steel analytical sieves. Phosphate powder fractions of <0.63, 63–125, 125–600, 600–2000 μm in diameter were collected, dried and then stored in pre-cleaned glass or polypropylene containers for future use.

Table 1. Source area (type) and percentage concentrations of some major constituents of the Jordan phosphate rocks. Source: Jordan Phosphate Mines Company reports, personal communication.

Site	H_2O (%)	TCP (%)	SiO_2 (%)	Al_2O_3 (%)	Cl_2 (%)
Al-Hasa	2.43	73.81	2.00	0.17	0.017
Al-Abyad	1.73	71.82	3.50	0.35	0.038
Esh-Shydia	0.72	74.51	4.30	0.35	0.030

TCP: Tricalcium phosphate.

2.2. *Dissolution experiments*

2.2.1. *Effect of raw phosphate weight and source*

For this purpose, nine aliquots of 0.25, 0.5, 1.0 or 2.5 gm from each type of raw phosphate were placed into 500 ml Erlenmeyer flasks containing 250 ml of seawater that has been filtered through Whatman GFC filter paper. In addition, three 250 ml aliquots of filtered seawater samples (reference samples) were placed in three similar flasks for each weight set. The 12 flasks were then closed tightly and shaken continuously for 1, 24 or 72 h.

At the end of each shaking time three sample solutions from each weight and each type (and one reference seawater sample) were filtered through Whatman GFC filter papers. DIP and fluoride concentrations were determined and compared with their concentrations in the reference seawater samples.

2.3. *Effect of grain size*

Aliquots of 0.25, 0.5, 1.0 or 2.5 gm of the <63, 63–125, 125–600 and 600–2000 μm size fractions of each type of raw phosphate were placed into 500 ml Erlenmeyer flasks containing 250 ml of filtered seawater. A total of 12 flasks for each size fraction, in addition to 3 flasks containing filtered seawater as reference samples were shaken continuously for 1, 24 or 72 h.

At the end of each shaking time four sample solutions (and one reference seawater sample) were filtered through Whatman GFC filter papers. DIP and fluoride concentrations were determined and compared with their concentrations in the reference seawater samples.

2.4. *Chemical analysis*

DIP was determined in aliquots of the sample solutions according to the colorimetric method of Murphy and Riley [11] as described by Strickland and Parsons [12]. Fluoride concentration was determined spectrophotometrically following the colorimetric method of Greenhalgh and Riley [13]. In order to check for p adsorption to glassware that was used in the experiments, each flask was washed and rinsed with 10 ml of distilled water at the end of the experiment. The flask was then rinsed with 10 ml of 1 M HCl followed by another 30 ml of distilled water to remove any adsorbed phosphate. DIP concentration was determined in the combined washing solution. DIP concentrations were undetectable in all experiments.

3. Results

3.1. *Grain size analysis*

The results indicate that highest percentages of the grains were between 125–250 and 250–600 μm (Figure 2). However, it was obvious that the small size fractions (<250 μm) were higher in Al-Hasa (64.1%) than in Al-Abyad (44.1%) and Esh-Shydiya (34.2%).

3.2. *Effect of weight and type (source) of raw phosphate*

Figure 3 shows that when each of the three types of raw phosphate was mixed with seawater, the phosphate particles dissolved only partially and caused an increase in the concentration of DIP in the test solution. It is apparent and logically accepted that as the weight of raw phosphate used in

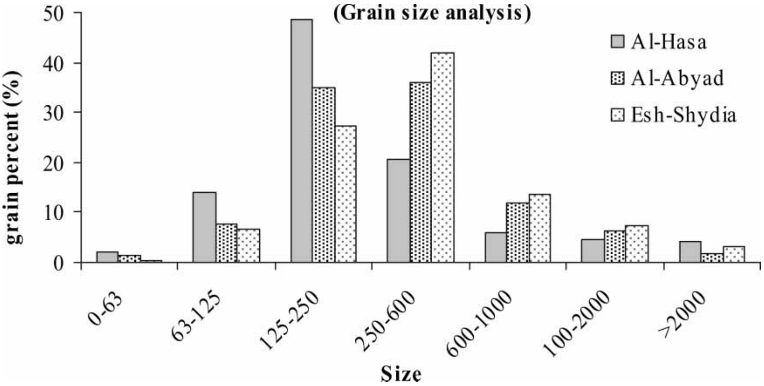

Figure 2. Average grain size distributions for the three types of raw phosphate used in this study.

the experiment increased, the concentrations of the DIP and fluoride increased. It is clear that the increase in the dissolution rate of the three types of raw phosphate and consequently the relative increase in the concentration of the DIP and fluoride in the test solution was higher during the first hour compared to the following 24- and 72-h periods. Table 2 shows the one-hour dissolution rate as the initial rate (Ri) and the 72 h as the final rate (Rf). The results presented in the table indicate that the (Ri) of phosphate was about 23, 16 and 15 fold of the final dissolution rate (Rf) for Al-Hasa, Al-Abyad and Esh-Shydiya, respectively. Fluoride (Ri) was 99, 132, and 40 times higher than (Rf) for the same types of raw phosphate in the same order. The dissolution decreased as the

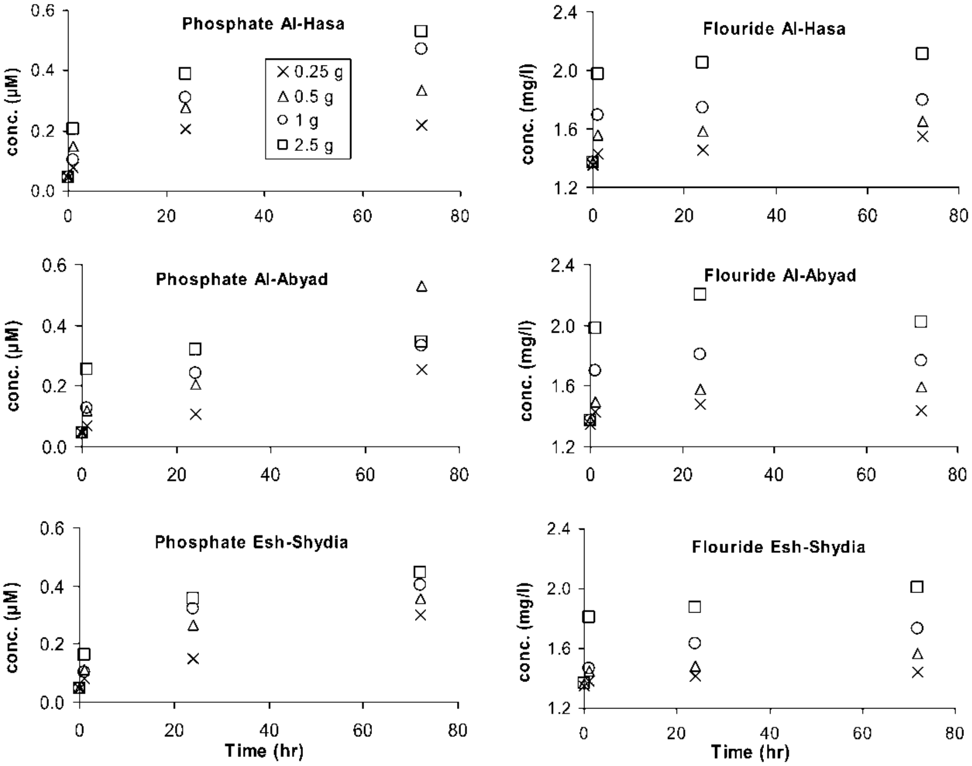

Figure 3. Time course of DIP (μM) and fluoride (mg/l) concentrations in the seawater of the incubations with different weights and types of the raw phosphate.

Table 2. Initial (Ri) and final rate (Rf) ($\mu g\,l^{-1}\,h^{-1}\,g^{-1}$) of increase in DIP and fluoride due to dissolution of different types of Jordan raw phosphate.

		DIP ($\mu g\,l^{-1}\,h^{-1}\,g^{-1}$)		Fluoride ($\mu g\,l^{-1}\,h^{-1}\,g^{-1}$)	
Site	Rate	Average	SD	Average	SD
Al-Hasa	Ri	0.890	0.539	73.399	16.281
	Rf	0.039	0.019	0.742	0.685
Al-Abyad	Ri	0.769	0.243	66.452	10.651
	Rf	0.049	0.042	0.459	0.630
Esh-Shydia	Ri	0.610	0.374	29.511	14.716
	Rf	0.048	0.035	0.739	0.306

reaction time increased until equilibrium is reached and a steady state of saturation is attained. Table 2 shows also that the differences between the initial rates (Ri) as well as between the final rates (Rf) of dissolution of the three types of phosphate, and consequently the increase in the concentrations of DIP and fluoride are not wide.

3.3. *Effect of grain size*

The concentrations of DIP and fluoride in solution increase with decreasing grain size of the raw phosphate fraction used in the experiment (figure not shown), which suggests that the dissolution of the raw phosphate particles increases with decreasing grain size of the particles.

4. Discussion

The results indicate that the dissolution of the raw phosphate was high at the beginning of the experiment causing a high increase in the concentration of DIP and fluoride in the test sample. These results are in agreement with those of Ossenkamp [14] who reported that dissolution of apatite was fast during the first 24 h with the concentration of fluoride increasing rapidly and thereafter there was more drift towards equilibrium. This happened even after the apatite was washed thoroughly and repeatedly to remove ultrafine particles which are suspected to cause the high initial solubility. Similar results were reported by Lovgren and Bengtsson [15]. Using batch experiment, initial release of phosphate was rapid and then decreased with time Welch et al. [16].

Relation between initial rates of DIP and fluoride increase and raw phosphate weight (Figure 4a) shows that there is a positive correlation between the weight and the rate ($r^2 = 0.86$ & 0.98 for DIP and Fluoride, respectively). ANOVA analysis at 95% confidence interval was used to determine the significance of difference in DIP and fluoride concentration due to the dissolution of various types of raw phosphate. The analysis of the results obtained from the use of different weights shows significant differences between the results obtained from comparing different weights (p values <0.05). This result is in agreement with that reported by Atlas and Pytkowicz [9] who investigated apatite dissolution using different weights (0.01–1.0 gm) and reported higher (DIP) when using larger weights of apatite. The results are anticipated, as it is known that the quantity of the products in a reaction (DIP and fluoride in the present case) depends on the quantity of the limiting reactant (the reactant with the least molar quantity) [17]. In the dissolution reaction, the product depends mainly on the quantity of the compound (apatite in the present case), dissolved in a solvent. However, any solvent has usually an ability to dissolve a limited quantity of the solute. This fact can be used to explain why the increases in DIP and fluoride are not linear as shown in Figure 3. Their concentrations increase as reaction time increases, until equilibrium is reached at specific concentrations [9,10,18] when the dissolution rate of raw phosphate particles is the same

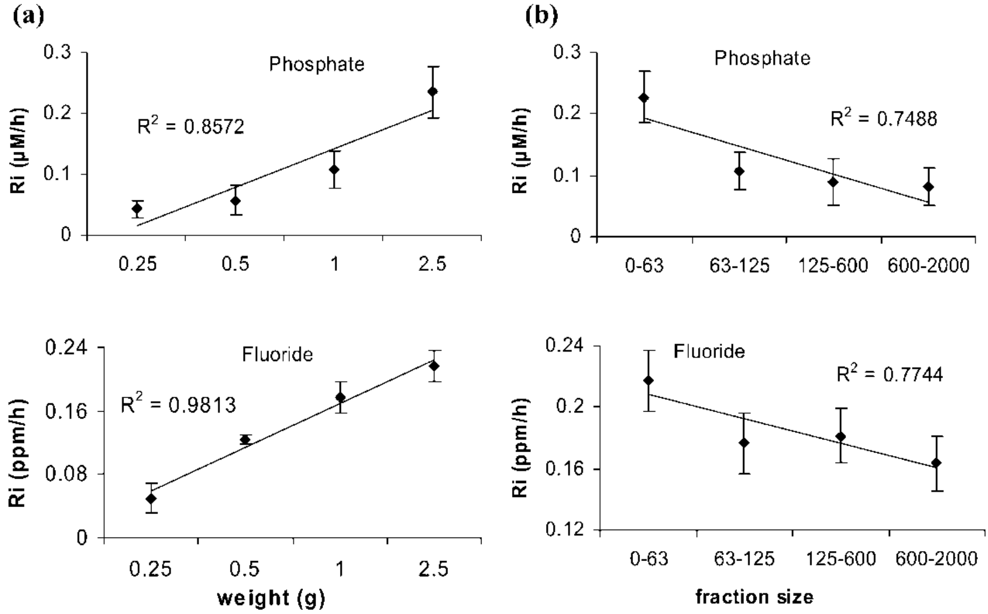

Figure 4. Relations between (a) initial rates and weights, and (b) between initial rates and size fractions of raw phosphate used in the experiments for phosphate and fluoride.

as the rate DIP sorption on the particles [19]. However, dissolution results demonstrate that the rates of phosphate dissolutions are generally lower than the rate of fluoride dissolution (Table 2). This is in agreement with Atlas and Pytkowicz [9] and Dorozhkin [20]. The first authors found different rates of phosphate and fluoride changes and attributed this to different process altering each component. Dorozhkin [20] found higher rates of fluoride dissolution and suggested that fluoride is dissolved first and phosphate is the last to be detached from apatite surface.

Relation between initial rates of DIP and fluoride increase and raw phosphate fraction size (Figure 4b) shows a negative correlation between the two parameters. This is in agreement with Mavropoulos et al. [21] who found that the release of phosphorus and calcium was mostly originated from sample fine fraction. This is more likely caused by the relatively larger surface areas of the smaller grains [22,23]. The present findings are in agreement with the results reported by Atlas and Pytkowicz [9] who found that the final DIP and fluoride concentrations increased with increasing surface area of apatite. To explain their findings, they suggested that the larger the surface area the larger the surface exposed to collision with sea water particles which in turn facilitates the dissolution reaction and increases the dissolution rates. It is worthy of mention that the dissolution of raw phosphate occurs mainly at the surface of the particles through chemical reactions between the surface of apatite and the solution [20,24,25]. In contrast, statistical analysis indicates however, that there are no significant differences between the dissolution of the three types of raw phosphate from the three sites. However, higher dissolution rates were estimated for Al-Hasa site compared to the other sites from other origins which may be attributed to the higher content of the small grain size (<250 µm) fraction in Al-Hasa apatite.

4.1. *Equilibrium and total load of DIP and fluoride: significance and potential impact on marine environment*

The main concern in the present study is with DIP concentrations, which may increase as a result of dissolution of raw phosphate in seawater of the Gulf of Aqaba because DIP is a key factor in the eutrophication process, which occurs in the Gulf water and affect its coral reefs. In addition, many workers have shown that calcification in reef corals can be suppressed by large increases in DIP in the surrounding waters [26,27]. Accordingly, we tried to estimate the increase in DIP concentration in seawater of the Gulf of Aqaba due to apatite particles dissolution applying the worst-case scenario and therefore, using the initial rates of dissolution for the smaller size (63–125 μm) and most soluble fraction. The calculations were based on the type of the raw phosphate that may reach the sea during ship loading. If the previously reported 1% maximum loss of raw phosphate during transportation [8] is considered , and that 50% of the lost particles are reaching the sea and the exported phosphate is about 3.5 million tons/year (Phosphate Port, personal communication), then about 0.18 million tons/year is reaching the marine environment of the Gulf each year. According to the phosphate equation for the fraction 63–125 μm, the final concentration of DIP will be 9.954×10^{13} μM for 1 year. However, since the total water volume of the Gulf of Aqaba that mixed well throughout the year is about 2.9×10^{15} l [1,28,29] and the residence time of the water in the Gulf of Aqaba is about one year [28,30,31], the DIP concentrations resulted from the dissolution of the lost raw phosphate in the Gulf of Aqaba will be 0.0.03 μM per year. Using the same calculations fluoride will not increase more than 0.03 mg/l per year.

DIP concentrations in the water of the Phosphate Port area range between 0.02 and 0.50 μM which are comparable with the other sites and even with offshore waters [1,32,33]. Accordingly, the increase in DIP due to the partial dissolution of the raw phosphate reaching the water of the Gulf would not cause a substantial or significant increase in the prevailing levels of DIP in the water. Similarly, the increase of fluoride would not cause a significant increase the prevailing fluoride concentrations of 1.38 mg/l in the water of the Gulf of Aqaba [34]. However, this conclusion is based on the results of the dissolution under the experimental conditions. It is important to point out that many other factors can affect the dissolution of phosphate particles in the marine environment, and consequently the DIP concentrations. The factors include temperature, pH of seawater and CO_2 and other carbonate species in that water [9]. In addition, conditions prevailing in bottom sediments, and properties and composition of the sediments can also affect the solubility of phosphate particles reaching these sediments. Some of these factors will be dealt with in a future work.

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