



## Estimates of atmospheric dry deposition and associated input of nutrients to Gulf of Aqaba seawater

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[1] Dry deposition rates and associated inputs of soluble inorganic nitrogen (N) phosphorus (P) and iron (Fe) were calculated for the north coast of the Gulf of Aqaba in Eilat, Israel, between August 2003 and September 2005. The main inorganic N compounds in the water soluble fraction of aerosol particles were nitrate (60%) and ammonium (38%), with mean concentrations of 35 and 22 nmol m<sup>-3</sup> of air, respectively. Soluble phosphate ranged between 0.09 and 2 nmol m<sup>-3</sup> of air with a mean value of 0.4 nmol m<sup>-3</sup> of air. The soluble inorganic nitrogen and soluble phosphate account for approximately 86% and 69% of total soluble nitrogen and total soluble phosphorus, respectively; the difference is assigned to organic N and P compounds. The mean concentration of soluble aerosol Fe was 0.3 nmol m<sup>-3</sup> of air. Our measurements of the soluble nutrient concentrations are comparable to data previously reported for the eastern Mediterranean area. Dry deposition fluxes of nutrients were estimated for each sampling date using a size-dependent deposition model. The estimated fluxes were highly variable over the sampling period with the mean fluxes of 38, 0.2 and 0.02 μmol m<sup>-2</sup> d<sup>-1</sup> for seawater soluble inorganic N, P and Fe, respectively. The soluble phosphate flux shows a seasonal pattern with higher input during the winter (September to December) than in other seasons. The inorganic N/P molar ratios in the seawater-soluble fraction of the dry deposition (ranging from 32 to 541) were well above the Redfield ratio (N/P = 16), suggesting that atmospheric inputs of nutrients increase the likelihood for P limitation in the Gulf. Atmospheric deposition could contribute a substantial fraction (35%) of dissolved inorganic N to the euphotic zone during the stratification period (April to October), and the N flux from this source could support over 10% of surface primary production and possibly all of the new production during the summer; however, these estimates have a relatively large uncertainty due to error associated with deposition flux calculation and the temporal variability in dust flux. Atmospheric input of seawater-soluble Fe is in large excess compared to that required for the phytoplankton growth driven by the N deposition.

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### 1. Introduction

[2] The atmosphere is an important pathway by which many natural and anthropogenic materials are transported from the continent to the ocean. Estimates of atmospheric deposition fluxes of nitrogen (N), phosphorus (P) and iron (Fe) to the ocean suggest that atmospheric deposition can be

a significant source of these nutrients [Duce *et al.*, 1991; Prospero *et al.*, 1996]. This source of new nutrients to the ocean supports marine productivity and thereby plays an important role in the global biogeochemical cycling of carbon [Jickells, 1995; Paerl, 1997]. The atmospheric input of Fe was found to be important for supporting primary production in high nitrate low chlorophyll (HNLC) oceans [Martin *et al.*, 1994; Coale *et al.*, 1996; Boyd *et al.*, 2000; Tsuda *et al.*, 2003]. Specifically, it is suggested that dust deposition during the last glacial maximum, which was about a factor of 2 higher than at present, may have enhanced the phytoplankton growth and subsequently lowered atmospheric CO<sub>2</sub> level during that period [Martin, 1990; Mahowald *et al.*, 1999; Bopp *et al.*, 2003]. Outside the HNLC zones atmospheric input of major nutrients can have a considerable impact on the productivity and structure

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of oligotrophic oceanic ecosystem [Fanning, 1989; Owens *et al.*, 1992] and of some coastal sites as well [Paerl, 1997]. Model calculations of N deposition have shown that in the year 2000 the deposition of total reactive N exceeds  $2000 \text{ mg N m}^{-2} \text{ y}^{-1}$  in extended parts of the world [Dentener *et al.*, 1996, 2006]. This is about twice as much as the “critical N load” above which changes in sensitive natural ecosystems may occur. Windborne dust was also suggested as an important contributor to regional biogeochemical cycling of P [Okin *et al.*, 2004]. Indeed, marine systems neighboring desert areas are influenced by dust borne P additions for the long-term maintenance of productivity [Okin *et al.*, 2004; Markaki *et al.*, 2003; Bergametti *et al.*, 1992].

[3] The Gulf of Aqaba is an oligotrophic sea [Levanon-Spanier *et al.*, 1979]. It is surrounded by arid areas (30 mm rainfall per year) with the deserts of Sinai and Sahara in the west, Arabia in the east and Negev in the north. The Gulf receives practically no river runoff from land, and the deposition flux of mineral aerosols in this area is significant [Ganor and Foner, 1996]. Accordingly, atmospheric input of nutrients through dry deposition may be one of the major external nutrient sources to the Gulf. The residence time of water in the Gulf is about 1 year, nutrient poor surface water from the Red Sea enters the Gulf at the Straits of Tiran while subsurface water higher in nutrients leaves the Gulf, resulting in net nutrient loss through this circulation pattern [Reiss and Hottinger, 1984; Badran, 2001]. The Gulf is geographically very close to the eastern Mediterranean basin which receives air masses from industrialized and semi-industrialized regions of Europe during most of the year (at least 70% of the time) [Kouvarakis *et al.*, 2001]. Thus it is likely that, in addition to desert dust influence, anthropogenic aerosols originating from Europe and Israel may contribute to the input of nutrient species to the Gulf. Accordingly, it is likely that atmospheric nutrient deposition in this region supports primary production and may constitute a significant fraction of new production. Indeed, it has been reported that surface chlorophyll and primary production in the Gulf are significantly higher than expected from measured nutrient levels during the summer months compared to other oceanic sites with similar conditions [Reiss and Hottinger, 1984; Lindell and Post, 1995]. Several studies have measured the nutrient concentrations and modeled nutrient cycling in the Gulf water column [Badran *et al.*, 2005; Badran, 2001; Rasheed *et al.*, 2002; Niemann *et al.*, 2004]. However, no attempt has been made so far to determine the atmospheric deposition fluxes of nutrients to the Gulf and to evaluate how this external nutrient source influences the Gulf ecosystem.

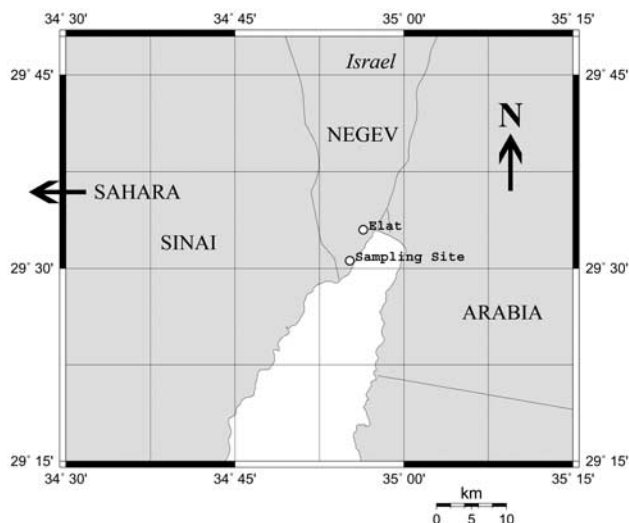
[4] In this study, aerosol samples were collected over a 2-year period (20 August 2003 to 10 September 2005) at the northwest coast of the Gulf of Aqaba in Eilat, Israel. Aerosol loads were determined and the samples analyzed for the concentrations of soluble nutrient species and other soluble components. The dry deposition velocity of aerosols was calculated for each sampling date using a particle deposition model combined with the particle size distribution obtained from Scanning Electron Microscope (SEM) images. Dry deposition fluxes of nutrients to the Gulf were then calculated for each sampling date. These estimates provide unique information on temporal patterns and vari-

ability in the daily fluxes of aerosol deposition in addition to annually averaged values. The measured concentrations and estimated fluxes of nutrients were compared to the data previously reported for neighboring areas. The contribution of this nutrient input to the primary production of the Gulf was assessed. This study provides important data for modeling the nutrient budgets and dynamics in this oligotrophic ocean basin which may be representative of other present-day dust-dominated oligotrophic systems and particularly may represent future conditions of increased aridity and dust fluxes [Tegen *et al.*, 2004; Woodward *et al.*, 2005]. The central hypothesis of this study is that the atmospheric input is an important external source of nutrients to the Gulf and support significant fraction of new production (carbon fixation) in this area.

## 2. Methods

### 2.1. Aerosol Collection

[5] Aerosol samples were collected using a Total Suspended Particle High Volume Sampler (HVS) placed on a roof at the Interuniversity Institute of Marine Sciences (IUI) in Eilat, a few meters off the northwest coast of the Gulf of Aqaba ( $29^{\circ}31'N$ ,  $34^{\circ}55'E$ , Figure 1). The HVS was designed to have four filter holders connected to separate flowmeters, thus collecting four filter samples simultaneously. The airflow path of the HVS and the filter holders were made of all plastic to minimize trace metal contamination. Aerosol particles were collected on a 47-mm polycarbonate membrane filter (Isopore™) which was cleaned using hydrochloric acid and weighted before and after sample collection. The collected filter samples were stored frozen in polystyrene Petri dishes inside plastic bags before being further analyzed. Aerosol samples were taken at least once a week over a 24-hour period with an airflow of  $2.5\text{--}2.8 \text{ m}^3 \text{ h}^{-1}$  between 20 August 2003 and 28 November 2004. However, we suspected that such intermittent sampling may not be able to reflect the average distribution of aerosols over the Gulf because of the episodic nature of aerosol deposition events (dust storms). Aerosols were therefore sampled continuously with an airflow of  $1.2\text{--}1.5 \text{ m}^3 \text{ h}^{-1}$  after 28 November 2004, and new filters were loaded every 72 hours until 10 September 2005. Thus all short-duration (less than a week) aerosol events (e.g., plumes, dust storms) that could be missed or randomly captured by the intermittent sampling at one day per week would be sampled by the continuous sampling scheme used after 28 November 2004. However, the continuous sampling using 72-hour integration time and lower flow rates may mask the impact of any daily aerosol pulses (e.g., results are smoothed), regardless, this sampling strategy should still reflect the seasonal changes of the aerosol concentration in the air. The 72-hour time-resolved measurements of ambient aerosol concentrations should provide more realistic estimates of seasonal and annual average fluxes of atmospheric nutrients to the Gulf. Short-duration aerosol events, on the other hand, which are captured better with the 24-hour sampling, are important to study as these may cause a sudden increase of nutrient deposition fluxes to the Gulf which may trigger phytoplankton blooms. We use the average aerosol nutrient input and its seasonal pattern for modeling regional nutrient dynamics and the storm



**Figure 1.** Aerosol sampling site at the northwest coast of the Gulf of Aqaba (29°31'N, 34°55'E) in Israel.

events captured to determine the potential impact of specific high deposition pulses.

## 2.2. Chemical Analysis

[6] One of the four filter samples collected at a given sampling date was used for pure-water extraction and the extract analyzed for water-soluble aerosol species. The filter sample was placed in an acid-cleaned polypropylene jar with the dusty side facing up. 50 mL of 18.2 mΩ milli-Q water was added to the jar, and the jar was covered using a polypropylene screw cap and then sealed with parafilm. The filter sample was sonicated for 30 minutes to resuspend the aerosol particles into the solution. The extraction solution was then filtered through a 0.4 μm polycarbonate membrane and separated into several portions that were analyzed respectively for the concentrations of cations, anions, total soluble N and other soluble elements. Ions were measured by Ion Chromatography using a DIONEX DX-500 system. Anions (F<sup>-</sup>, Acetate, Formate, MSA<sup>-</sup>, Cl<sup>-</sup>, NO<sub>2</sub><sup>-</sup>, Br, NO<sub>3</sub><sup>-</sup>, PO<sub>4</sub><sup>3-</sup>, SO<sub>4</sub><sup>2-</sup>, Oxalate) were separated and eluted using an AS9-HC anion column (Dionex) using a Na<sub>2</sub>CO<sub>3</sub> eluent, and cations (Li<sup>+</sup>, Na<sup>+</sup>, Ca<sup>2+</sup>, K<sup>+</sup>, NH<sub>4</sub><sup>+</sup>, Mg<sup>2+</sup>) were separated and eluted using a CS12A cation column (Dionex) using a methanesulfonic acid (MSA) eluent. Total soluble N (TSN) was analyzed using a modified persulfate digestion procedure [Delia et al., 1977] followed by Continuous-Flow Autoanalyzer (Alpkem Flow Solution IV) analysis. Total soluble P (TSP) and soluble Fe (SFe) were determined by Inductive Coupled Plasma Optical Emission Spectroscopy (ICP-OES, for samples before 28 November 2004) and by High Resolution Inductive Coupled Plasma Mass Spectrometer (HR-ICP-MS, Finnigan<sup>®</sup> ELEMENT 2, for samples after 28 November 2004) in a matrix of 2% HNO<sub>3</sub>. An operational blank was handled and analyzed with the filter samples. The operational blanks (sample concentration ranges) were 0.1 (0.2–4), 1 (23–162), <0.03 (0.1–3), <0.01 (1–113), <0.05 (30–237), 0.01 (0.2–3) and <0.001 (0.07–3) μM for NO<sub>2</sub><sup>-</sup>, NO<sub>3</sub><sup>-</sup>, PO<sub>4</sub><sup>3-</sup>, NH<sub>4</sub><sup>+</sup>, TSN, TSP and SFe in the water extracts, respectively. These blanks comprise a low fraction of the sample concentrations and were

subtracted for the data reported below. The maximum uncertainties (i.e., standard deviations) for determining the nutrient species NO<sub>2</sub><sup>-</sup>, NO<sub>3</sub><sup>-</sup>, PO<sub>4</sub><sup>3-</sup>, NH<sub>4</sub><sup>+</sup>, TSN, TSP (ICP-MS/ICP-OES) and SFe (ICP-MS/ICP-OES) were respectively 0.05, 0.5, 0.1, 2, 3, 0.05/0.7 and 0.03/0.6 μM, which were about 1–19% of the average sample concentrations. We have previously determined that the solubilities of some of the nutrients in seawater may be different (typically lower) from pure water [Chen et al., 2006] specifically for PO<sub>4</sub><sup>3-</sup> and Fe. Accordingly, in our calculations we convert the concentrations we measured in the pure water extractions to those expected to be extracted in seawater on the basis of the relation between the solubility for each component in seawater and pure water as determined for parallel samples collected at this site (see discussion below and Chen et al. [2006]).

## 2.3. Air Mass Back Trajectories

[7] Five-day isentropic air mass back trajectories (AMBTs) were calculated from the National Oceanic and Atmospheric Administration (NOAA) FNL database using the Hybrid Single-Particle Lagrangian Integrated Trajectories (HY-SPLIT) program [Draxler, 2002]. Although the errors associated with these calculations increase as the model steps back in time, the AMBTs still provide useful information about the synoptic situation and general source of the air mass sampled. AMBTs were calculated at 100, 1000 and 3000 m height levels at 1900 UTC (corresponding to the midway of each sampling period) in Eilat (29°31'N, 34°55'E, Figure 2).

## 2.4. Particle Deposition Model

[8] A particle deposition model [Jacobson, 2004] was used to calculate the dry deposition velocities for various sizes of aerosol particles collected for each sampling date. The dry deposition speed ( $V_{d,i}$ , equation (1)) is the sum of particle sedimentation speed in the air ( $V_{s,i}$ ) and its dry deposition against a series of resistances at air-surface interface, which varies with particle size and is also depended on meteorological and physical conditions in the troposphere.

$$V_{d,i} = \frac{1}{R_a + R_b + R_a R_b V_{s,i}} + V_{s,i} \quad (1)$$

where  $V_{d,i}$  and  $V_{s,i}$  are dry deposition and sedimentation speeds of a particle of size  $i$  (cm s<sup>-1</sup>), respectively;  $R_a$  is the aerodynamic resistance between a reference height (10 m) and the laminar adjacent to the surface;  $R_b$  is the resistance to molecular diffusion through the 0.1 to 0.01-cm-thick laminar sublayer.

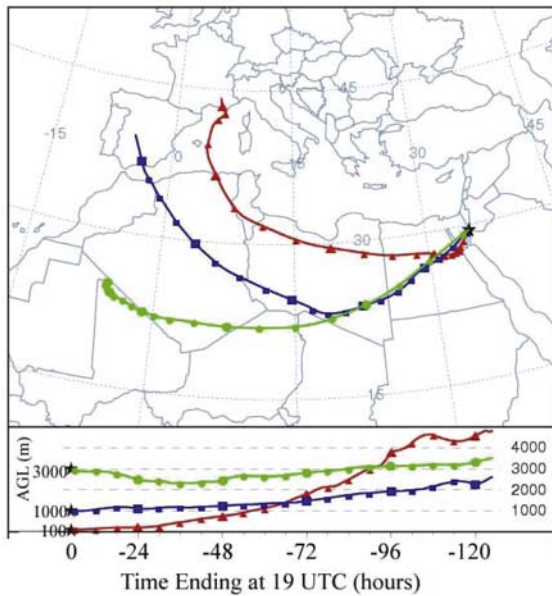
[9] For particle with a diameter of less than 20 μm which is almost the upper size limit of aerosol particles collected in the Gulf, the sedimentation speed can be calculated following equation (2).

$$V_{s,i} = \frac{2r_i^2(\rho_p - \rho_a)g}{9\eta_a} G_i \quad (2)$$

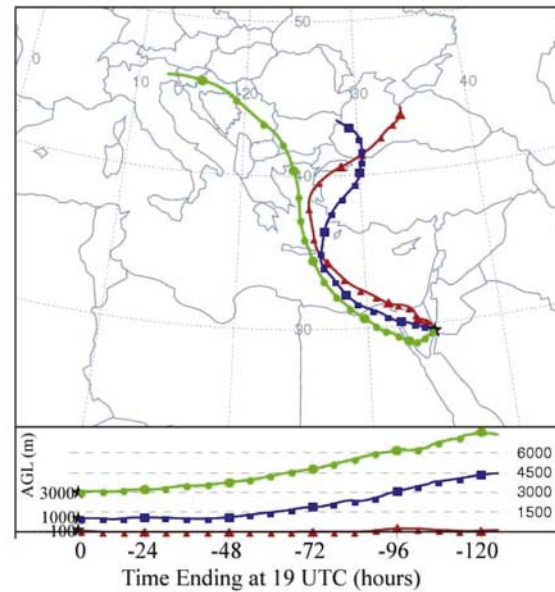
where  $r_i$  is the particle radius (cm);  $\eta_a$  is the dynamic viscosity of air (g cm<sup>-1</sup> s<sup>-1</sup>);  $\rho_p$  and  $\rho_a$  are the densities of



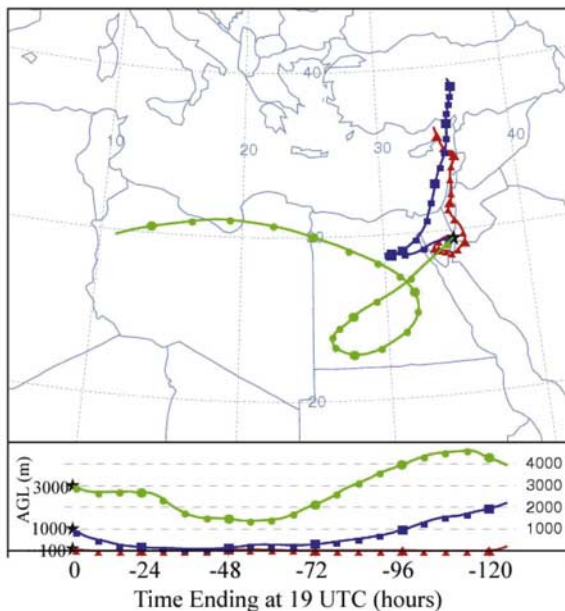
a) January 4, 2004; 19 UTC



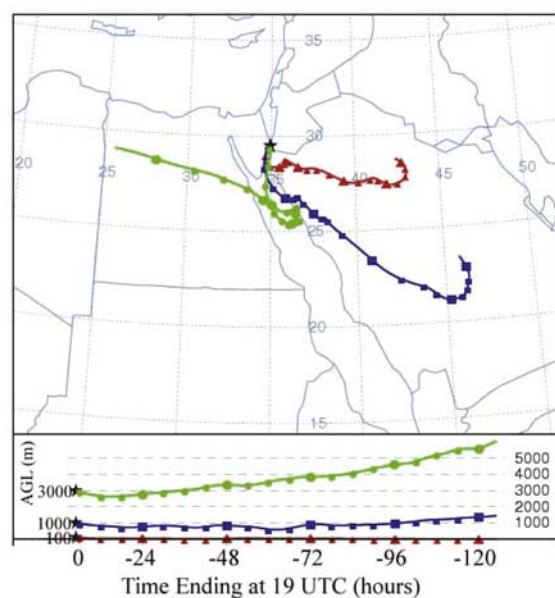
b) August 22, 2003; 19 UTC



c) April 8, 2005; 19 UTC



d) March 2, 2004; 19 UTC

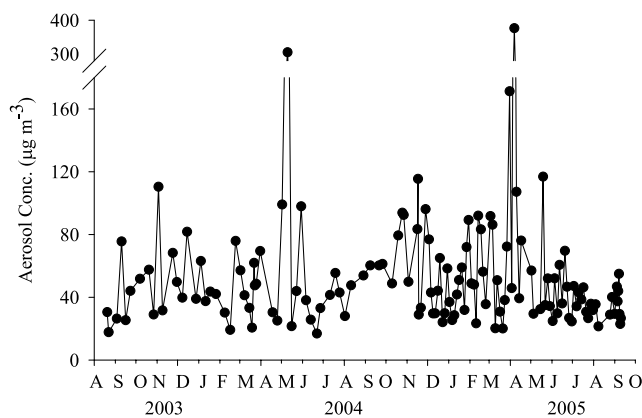


**Figure 2.** Representative 5-day backward trajectories calculated for (a) 4 January 2004, (b) 22 August 2003, (c) 8 April 2005, and (d) 2 March 2004. The starting point is the northwest coast of the Gulf of Aqaba ( $29^{\circ}31'N$ ,  $34^{\circ}55'E$ ) at altitudes of 100, 1000 and 3000 m.

the particle and air ( $g \text{ cm}^{-3}$ ), respectively;  $g$  is the gravitational acceleration ( $\text{cm s}^{-2}$ );  $G_i$  is the Cunningham slip-flow correction factor.

[10] The meteorological inputs for the particle deposition model were obtained from the National Oceanic and Atmospheric Administration (NOAA) archived FNL METEOGRAM database, including surface pressure (HPA), surface temperature (K), temperature (K) and wind

speeds ( $\text{cm s}^{-1}$ ) at reference height (10 m), for each sampling date. The model applied a dust particle density of  $2.6 \text{ g cm}^{-3}$  [Chin *et al.*, 2002] since mineral dust dominates the aerosol particles in this desert-surrounding region. Model output was combined with the particle size distribution analyzed using SEM to estimate the average dry deposition velocity of aerosols ( $V_d$ ) during each sampling date, where the densities were assumed to be the same for



**Figure 3.** Aerosol concentrations ( $\mu\text{g m}^{-3}$ ) observed between 20 August 2003 and 10 September 2005 at the northwest coast of the Gulf of Aqaba ( $29^{\circ}31'N$ ,  $34^{\circ}55'E$ ).

all aerosol particles regardless of size. The dry deposition flux ( $F_d$ ) for each nutrient was then calculated from the concentration of the soluble fraction of aerosol nutrient in the air ( $C_a$ ) and the dry deposition velocity of aerosols ( $V_d$ , equation (3))

$$F_d = C_a \times V_d \quad (3)$$

In addition, for comparison we also calculated the mean deposition fluxes of the nutrients using the more widely used average deposition velocity for these nutrient species [Duce *et al.*, 1991].

[11] Dry deposition of gaseous phase nitrogen ( $\text{HNO}_3$  and  $\text{NH}_3$ ) was not included in the flux estimate of soluble inorganic nitrogen (SIN) in this study. Nonetheless, water-soluble gases like  $\text{HNO}_3$  and  $\text{NH}_3$  can be transferred easily to seawater through air-sea interface and thus may contribute substantially to the atmospheric N deposition to the ocean. Accordingly, the omission of the gaseous phase N can underestimate the total dry deposition of SIN. In the eastern Mediterranean Sea for example ignoring the gaseous phase may have resulted in underestimates by about a factor of 2 [Kouvarakis *et al.*, 2001]. Similarly, our flux calculations may underestimate the whole atmospheric input of SIN to the Gulf. Thus our flux calculations should be considered as the lower range of estimate and further work on atmospheric deposition of gaseous phase nitrogen will be needed for a more complete assessment.

### 3. Results and Discussion

#### 3.1. Aerosol Sources and Loads

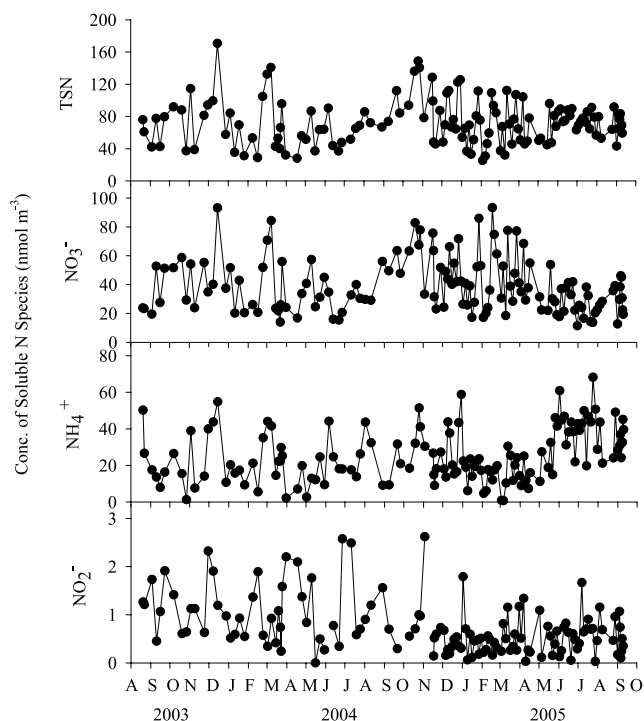
[12] 137 aerosol filter samples were collected at Eilat during the period of 20 August 2003 to 10 September 2005 to study the nutrient fluxes from the atmosphere to the Gulf of Aqaba. The sampling site was located a few meters off the northwest coast of the Gulf, and the collected samples are expected to represent the marine aerosols over the Gulf (Figure 1). Previous studies where samples were collected on shore close to the water line and over the ocean (e.g., southeast Mediterranean Sea [Herut and Krom, 1996; Carbo *et al.*, 2005]) indicate that with the exception of a higher sea salt component the shore based site is representative of the aerosols over the water. This should particularly

be the case in the Gulf where the water body is narrow thus the air mass composition is not expected to change much during transport. The air masses in this region can be influenced by mineral dust originating from the interior of North Africa (Sahara desert, Figure 2a) and the adjacent Sinai (Figure 2c) and Arabian deserts (Figure 2d), as well as by anthropogenic aerosols transported from the industrialized and semi-industrialized regions of Europe (Figure 2b). The impact of local anthropogenic sources is expected to be small, since Eilat is a tourist town with a population of only  $\sim 50,000$  and has no major industries other than tourism. Moreover, the marine station where the sampler was placed is in a remote location about 10 km south of the city. The general agreement between our data and previous reports for aerosol composition in the area (the Mediterranean Sea) also supports this premise. Although it has been previously suggested that dust storms originating from the Saharan and middle east deserts have a major impact on the eastern Mediterranean and adjacent region, including the Gulf [Ganor and Foner, 1996; Kubilay *et al.*, 2000] to our knowledge no detailed study of aerosols deposition and impact on the Gulf has been carried out.

[13] Total suspended particle concentrations in the air over the north Gulf ranged between 17 and  $376 \mu\text{g m}^{-3}$  with a mean value and standard deviation of  $53 \pm 43 \mu\text{g m}^{-3}$  (Figure 3). These dust concentrations are about a factor of 2 higher than the typical background level ( $28 \pm 30 \mu\text{g m}^{-3}$ ) but comparable to the urban area concentrations ( $51 \pm 33 \mu\text{g m}^{-3}$ ) at the eastern Mediterranean [Gerasopoulos *et al.*, 2006]. They also agree with those observed over the southeastern Mediterranean Sea during 1994/95 ( $57 \pm 31 \mu\text{g m}^{-3}$ ), but are only half of the reported values for the northern ( $112 \pm 58 \mu\text{g m}^{-3}$ ) and central ( $117 \pm 177 \mu\text{g m}^{-3}$ ) coasts of Israel [Herut and Krom, 1996]. The “dusty” Israeli air may be attributed to impacts of local loess soil and anthropogenic pollution in the more heavily populated central regions of the country. The measured aerosol concentrations in the Gulf showed high temporal variability without a clear seasonal trend during the sampling period (Figure 3). Two extreme large concentrations of aerosols ( $304$  and  $376 \mu\text{g m}^{-3}$ ) compared to the normal level ( $<120 \mu\text{g m}^{-3}$ ) were observed on 10 May 2004 and 7–10 April 2005, respectively (i.e., in the spring). These two considerably higher loads of aerosols over a 2-year-long period demonstrate the episodic nature of the vast dust storms over the Gulf, which may be due to special wind condition that lifted the soil dust of the surrounding deserts to up 1000 m above the ground level (Figure 2c). The back trajectories most abundant in the Gulf are those that pass over both the surrounding arid regions and the European continent (Figure 2b), probably because of the dominant northwestern and northern winds in this region. Aerosols from these trajectories may be influenced by anthropogenic sources in addition to mineral dust [Kocak *et al.*, 2004].

#### 3.2. Nutrient Concentrations

[14] Two-year time series concentrations of soluble aerosol  $\text{NO}_3^-$ ,  $\text{NH}_4^+$ ,  $\text{NO}_2^-$  and TSN in the Gulf of Aqaba are shown in Figure 4. High temporal variability in the concentrations but lack of a strong seasonal pattern was observed for all the soluble N species. The individually measured concentrations showed more variation before



**Figure 4.** Atmospheric concentrations ( $\text{nmol m}^{-3}$ ) of water-soluble aerosol  $\text{NO}_2^-$ ,  $\text{NH}_4^+$  and  $\text{NO}_3^-$  and total soluble N (TSN) measured between 20 August 2003 and 10 September 2005 at the northwest coast of the Gulf of Aqaba ( $29^\circ 31' \text{N}$ ,  $34^\circ 55' \text{E}$ ).

28 November 2004 compared to those after 28 November 2004 with considerably less spikes (Figure 4), suggesting that the 72-hour continuous sampling, as expected, better temporally integrated the fluxes than sampling intermittently once a week. One interesting feature that was observed during the continuous sampling period is the opposing behavior between soluble  $\text{NO}_3^-$  and  $\text{NH}_4^+$ ; that is, relatively higher concentrations of  $\text{NO}_3^-$  corresponded to the lower  $\text{NH}_4^+$  during January–May 2005, and vice versa in June–September 2005 (Figure 4). Aerosol loads were also higher in January–May than in June–September corresponding to the higher  $\text{NO}_3^-$  levels (Figure 3), which is probably due to a strong dust impact from African or local deserts over the eastern Mediterranean and the Gulf during the winter and spring [Kocak et al., 2004], while anthropogenically dominated aerosols (with higher  $\text{NH}_4^+$ ) are more common in the summer months. This feature suggested that although the total aerosol load over the Gulf did not show a strong seasonal trend the aerosols were dominated by different sources in the different seasons.

[15] The soluble  $\text{NO}_3^-$  concentrations ranged between 11 and  $93 \text{ nmol m}^{-3}$  with a mean value and standard deviation of  $39 \pm 19 \text{ nmol m}^{-3}$  (Table 1). Soluble  $\text{NH}_4^+$  ranged between 0.7 and  $68 \text{ nmol m}^{-3}$  with the average of  $25 \text{ nmol m}^{-3}$ , and soluble  $\text{NO}_2^-$  has a range of 0.03– $3 \text{ nmol m}^{-3}$  and mean value of only  $0.7 \text{ nmol m}^{-3}$  (Table 1). Soluble  $\text{NO}_3^-$  and  $\text{NH}_4^+$  were the two major components of soluble inorganic N (SIN) in the aerosol. SIN is often considered to be bioavail-

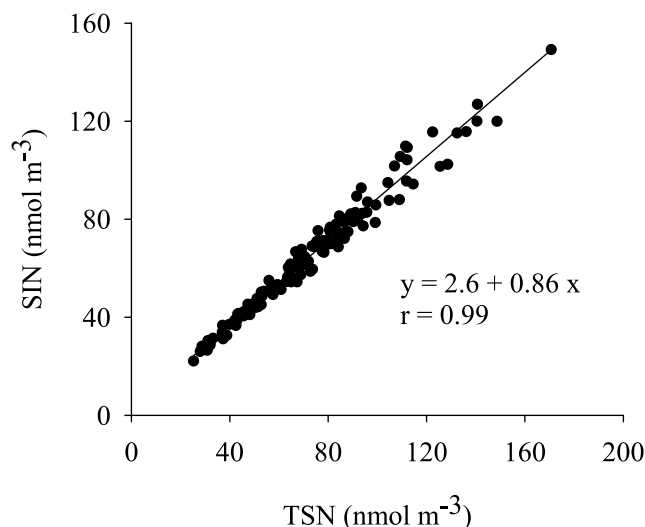
able in the water column and consequently used in this study to estimate the dry deposition flux of N nutrient to the Gulf. The TSN ranged from 25 to  $171 \text{ nmol m}^{-3}$  of air and the soluble organic N (SON) defined as the difference between TSN and SIN ranged between 0.05 and  $29 \text{ nmol m}^{-3}$  with a mean value of  $8 \text{ nmol m}^{-3}$  (Table 1). The SIN concentrations were found to be strongly correlated with the TSN ( $r = 0.99$ , slope = 0.86, Figure 5) consistent with the dominance of the inorganic species which accounted for 86% of aerosol TSN over the Gulf.

[16] To our knowledge, no data of aerosol  $\text{NO}_3^-$ ,  $\text{NH}_4^+$  and  $\text{NO}_2^-$  contents in aerosols over the Gulf of Aqaba were previously reported. However, our observations of the soluble  $\text{NO}_3^-$  and  $\text{NH}_4^+$  concentrations are in reasonable agreement with the values reported for various coastal sites in the eastern Mediterranean (Table 2). The measured mean  $\text{NO}_3^-$  ( $39 \text{ nmol m}^{-3}$ ) is in the middle of the range reported for the three eastern Mediterranean sites Finokalio [Bardouki et al., 2003; Kouvarakis et al., 2001; Mihalopoulos et al., 1997], Erdemli [Mace et al., 2003; Kocak et al., 2004; Ozsoy et al., 2000] and Tel-Shikmona [Herut et al., 2002]. Finokalio has the lowest  $\text{NO}_3^-$  concentrations, probably because it is a small village on the northern coast of Crete Island with limited human activities within 20 km of its range. Tel-Shikmona, on the other hand, is adjacent to the industrial city of Haifa and therefore has a mean  $\text{NO}_3^-$  concentration twice as great as that observed in the Gulf. The measured  $\text{NH}_4^+$  concentrations in our samples are about a factor of 2–5 lower than those reported from the eastern Mediterranean sites [Bardouki et al., 2003; Kouvarakis et al., 2001; Mihalopoulos et al., 1997; Mace et al., 2003; Kocak et al., 2004; Herut et al., 2002], which may be a result of a dominant mineral dust component and a relatively low anthropogenic fraction in the Gulf aerosols (see section 3.1). Indeed, Herut et al. [2002] reported that the mean  $\text{NH}_4^+$  concentration was about a factor of 4 lower in desert type air masses ( $29 \text{ nmol m}^{-3}$ ) than in “normal air” ( $129 \text{ nmol m}^{-3}$ ) at Tel-Shikmona, and our observation of soluble aerosol  $\text{NH}_4^+$  ( $25 \text{ nmol m}^{-3}$ ) was comparable to that in the desert type air ( $29 \text{ nmol m}^{-3}$ ).

**Table 1.** Statistical Characteristics of Concentrations of Water-Soluble Aerosol  $\text{NO}_2^-$ ,  $\text{NO}_3^-$  and  $\text{NH}_4^+$ , Soluble Inorganic Nitrogen (SIN), Total Soluble Nitrogen (TSN), Soluble Organic Nitrogen (SON), Soluble  $\text{PO}_4^{3-}$ , Total Soluble Phosphorus (TSP), Soluble Organic Phosphorus (SOP), and Soluble Iron (SFe) Measured at the Northwest Coast of the Gulf of Aqaba ( $29^\circ 31' \text{N}$ ,  $34^\circ 55' \text{E}$ ) Between 20 August 2003 and 10 September 2005 ( $n = 137$ )

Nutrients	Concentrations, $\text{nmol m}^{-3}$				
	Range	Median	Mean	SD	95% Confidence
$\text{NO}_3^-$	11–93	35	39	19	3
$\text{NH}_4^+$	0.7–68	22	25	14	2
$\text{NO}_2^-$	0.03–3	0.6	0.7	0.6	0.1
SIN	22–149	64	65	24	4
TSN	25–171	69	73	28	5
SON	0.05–29	6	8	5	0.9
$\text{PO}_4^{3-}$	0.09–2	0.3	0.4	0.2	0.04
TSP	0.04–3	0.5	0.6	0.4	0.06
SOP	0.008–0.9	0.2	0.2	0.1	0.02
SFe	0.04–1	0.2	0.3	0.2	0.04





**Figure 5.** Strong correlation between the water-soluble inorganic N (SIN) and the total soluble N (TSN) concentrations ( $\text{nmol m}^{-3}$ ) in the Gulf of Aqaba atmosphere.

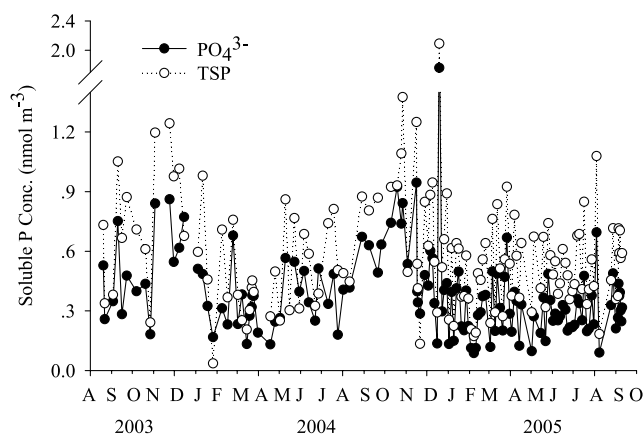
[17] Our time-integrated sampling of aerosols with the undenuded HVS and sample handling may lead to errors in aerosol-nitrate measurement, due to the evaporation loss of the semivolatile ammonium nitrate or adsorption of nitric acid gas. The volatilization impact may result in underestimation of the true nitrate concentrations and thus deposition. The amount of nitrate loss is dependent on the filter material, loaded particle mass and ambient meteorological conditions such as temperature and humidity [Wang and John, 1988; Chang *et al.*, 2000; Schaap *et al.*, 2004]. Evaporation loss of ammonium nitrate was only found when Teflon and quartz filters were used (here we use polycarbonate filters), and the amount lost was substantial only at high temperature ( $>25^{\circ}\text{C}$  [Schaap *et al.*, 2004]). It was also found that the nitrate loss could be as high as 85–95% at low mass loading (93–208  $\mu\text{g}$ ) but was less than

10% when the loaded particle mass was high ( $>2500 \mu\text{g}$ ) as at our site [Wang and John, 1988]. The concentration enrichment of particulate-phase nitrate could result in a decrease of evaporation losses from 30–80% to less than 5% for undenuded Teflon filters [Chang *et al.*, 2000]. Although the hot and dry weather in Eilat tends to increase the evaporation of particulate ammonium nitrate, our filter type and the sampled aerosol masses on the filters which were all heavier than 2500  $\mu\text{g}$  would significantly decrease the rate of evaporation. Moreover, mineral dust is the major aerosol component in this desert-surrounding region and nitrate in such samples is often associated with the coarse mode dust particles [Savoie and Prospero, 1982; Spokes *et al.*, 2000] and thus present in the form of stable compounds like  $\text{Ca}(\text{NO}_3)_2$ . In addition to the potential evaporation artifact, the nitrate concentration can also be overestimated because of adsorption from the gas phase. This also depends on the filter type; specifically, cellulose filters were found to adsorb significant amount of nitric acid gas which is then assigned to aerosol nitrate [Savoie and Prospero, 1982; Schaap *et al.*, 2004]. The use of polycarbonate filter (the thinnest filter type) in our sampling will not cause such severe overestimation of aerosol nitrate. Although we cannot quantify the impact of these evaporation and adsorption processes on our nitrate concentration estimates we expect them to be small (and compensating each other) because of the high aerosol load and filter type used in our samples. This is the first attempt to investigate the concentrations and fluxes of aerosol nutrients over the Gulf of Aqaba, further studies on aerosol nitrate using a denuded sampler should be done in the future, however the 2-year field data reported in this study still provides critical information for modeling the nutrient dynamics of this oligotrophic marine system.

[18] Concentrations of soluble aerosol  $\text{PO}_4^{3-}$  and TSP are plotted against the sampling dates in Figure 6. Like for the N species there was no strong seasonal trend for either P species, however, relatively higher concentrations of soluble  $\text{PO}_4^{3-}$  and TSP are seen during September to January (mean 0.5 and 0.7  $\text{nmol m}^{-3}$  respectively) compared to other months (mean 0.3 and 0.5  $\text{nmol m}^{-3}$ , Figure 6). This periodic change of aerosol P concentration is consistent

**Table 2.** Comparison Between the Measurements Obtained During This Work and Those Reported in the Literature for the Eastern Mediterranean Area Regarding Aerosol  $\text{NO}_3^-$  and  $\text{NH}_4^+$  Concentrations

Value	Location	Period	Reference
<i>Aerosol <math>\text{NO}_3^-</math>, <math>\text{nmol m}^{-3}</math></i>			
$39 \pm 19$	Eilat, Israel	Aug 2003 to Sep 2005	this work
$25 \pm 4$	Finokalia, Crete Island	Jan 2000	Bardouki <i>et al.</i> [2003]
$27 \pm 13$	Finokalia, Crete Island	Oct 1996 to Sep 1999	Kouvarakis <i>et al.</i> [2001]
$24 \pm 18$	Finokalia, Crete Island	Mar 1994 to Apr 1995	Mihalopoulos <i>et al.</i> [1997]
$36 \pm 20$	Erdemli, Turkey	Mar to May 2000	Mace <i>et al.</i> [2003]
$54 \pm 40$	Erdemli, Turkey	Jan 1996 to Dec 1999	Kocak <i>et al.</i> [2004]
44	Erdemli, Turkey	Oct 1991 to Dec 1992	Ozsoy <i>et al.</i> [2000]
$93 \pm 29$	Tel-Shikmona, Israel	Apr 1996 to Jan 1999	Herut <i>et al.</i> [2002]
<i>Aerosol <math>\text{NH}_4^+</math>, <math>\text{nmol m}^{-3}</math></i>			
$25 \pm 14$	Eilat, Israel	Aug 2003 to Sep 2005	this work
$43 \pm 5$	Finokalia, Crete Island	Jan 2000	Bardouki <i>et al.</i> [2003]
$53 \pm 21$	Finokalia, Crete Island	Oct 1996 to Sep 1999	Kouvarakis <i>et al.</i> [2001]
$54 \pm 32$	Finokalia, Crete Island	Mar 1994 to Apr 1995	Mihalopoulos <i>et al.</i> [1997]
$49 \pm 38$	Erdemli, Turkey	Mar to May 2000	Mace <i>et al.</i> [2003]
$36 \pm 23$	Erdemli, Turkey	Jan 1996 to Dec 1999	Kocak <i>et al.</i> [2004]
$117 \pm 88$	Tel-Shikmona, Israel	Apr 1996 to Jan 1999	Herut <i>et al.</i> [2002]



**Figure 6.** Atmospheric concentrations ( $\text{nmol m}^{-3}$ ) of water-soluble aerosol  $\text{PO}_4^{3-}$  and total soluble P (TSP) measured between 20 August 2003 and 10 September 2005 at the northwest coast of the Gulf of Aqaba ( $29^{\circ}31'N$ ,  $34^{\circ}55'E$ ).

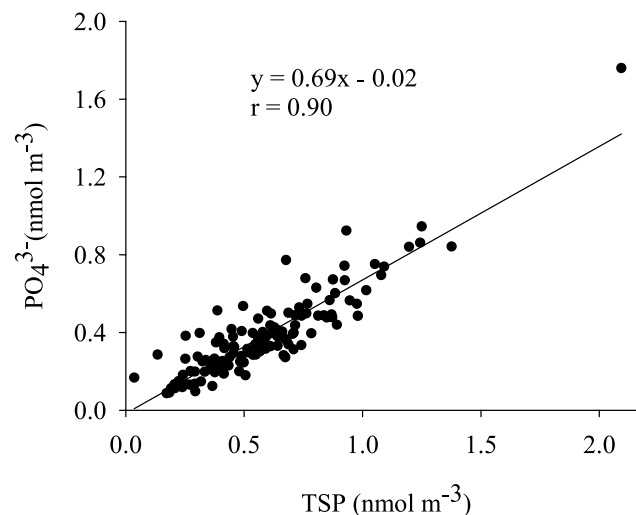
with the higher frequency of Saharan dust outbreaks (which contains  $\sim 0.09\%$  of particulate P [Ridame and Guieu, 2002]) over the eastern Mediterranean during the winter and fall compared to other seasons [Kocak et al., 2004; Swap et al., 1996]. The measured  $\text{PO}_4^{3-}$  and TSP have ranges of  $0.02\text{--}2$  and  $0.04\text{--}3$   $\text{nmol m}^{-3}$  and averages of  $0.4$  and  $0.6$   $\text{nmol m}^{-3}$ , respectively (Table 1). Soluble  $\text{PO}_4^{3-}$  and TSP were found to be strongly associated with each other ( $r = 0.90$ , Figure 7), suggesting that soluble  $\text{PO}_4^{3-}$  was a major component of TSP in aerosols or that the two species have similar origins. Unlike soluble aerosol N for which the organic N fraction was relatively low (14% of TSN), both soluble  $\text{PO}_4^{3-}$  (69%) and soluble organic P (SOP, 31%) contributions to the TSP are important (Figure 7, slope = 0.69). SOP ranged between  $0.008$  and  $0.9$   $\text{nmol m}^{-3}$  with a mean value of  $0.2$   $\text{nmol m}^{-3}$  (Table 1). The mean soluble  $\text{PO}_4^{3-}$  ( $0.4$   $\text{nmol m}^{-3}$ ) in our samples was about a factor of 2 lower than the seawater leachable  $\text{PO}_4^{3-}$  ( $0.8$   $\text{nmol m}^{-3}$ ) reported by Herut et al. [2002] at Tel-Shikmona. The lower value at the Gulf may correspond to the relatively lower aerosol loads (about 1/2 of those at the Israeli Mediterranean coast; see section 3.1). Like in the case of  $\text{NH}_4^+$ , this could also be explained by the lower anthropogenic “industrial” component and higher mineral dust component of aerosols over the Gulf compared to central Israel. Specifically, phosphate in mineral dust is predominantly bound to Fe oxides or associated with Ca, Mg, Al and Fe minerals which are known to be weakly soluble [Bergametti et al., 1992; Ridame and Guieu, 2002]. Anthropogenic phosphate sources may be associated with more soluble components, thus variable solubility may influence the concentrations of soluble  $\text{PO}_4^{3-}$  measured in this study [Herut et al., 2002]. To evaluate the  $\text{PO}_4^{3-}$  contribution to the Gulf via dry deposition we corrected the measured concentrations from our pure water extractions to those expected to occur in seawater (see discussion below).

[19] The Fe concentrations in the aerosols sampled before 28 November 2004 were more variable and slightly higher

than those measured after 28 November 2004 (Figure 8). The different variability is attributed to the different sampling strategies used during the two periods (see method section), and the slightly higher concentration of soluble Fe is probably due to the lower detection precision of the ICP-OES compared to the HR-ICP-MS that was used for analyzing the samples after 28 November 2004. The measured soluble Fe ranged between  $0.04$  and  $1$   $\text{nmol m}^{-3}$  with a mean value of  $0.3$   $\text{nmol m}^{-3}$  (Table 1). No soluble aerosol Fe data was previously reported for the Gulf and neighboring area. Total Fe concentrations of  $14$  and  $12$   $\text{nmol m}^{-3}$  were observed at the eastern Mediterranean sites at Erdemli [Kubilay and Saydam, 1995] and Tel-Shikmona [Herut et al., 2001]. Using an aerosol Fe solubility of 1–10% in an aqueous solution [Jickells and Spokes, 2001], the calculated soluble Fe based on the above total Fe is expected to range from  $0.1$  to  $1$   $\text{nmol m}^{-3}$  at Erdemli and Tel-Shikmona. Our mean SFe ( $0.3$   $\text{nmol m}^{-3}$ ) falls in the range of these calculated values. Like in the case of  $\text{PO}_4^{3-}$  we correct the measured pure water soluble Fe concentrations to account for differences in solubility between pure water and seawater using the relation from Chen et al. [2006], when evaluating ecological effects of atmospheric Fe input (see discussion below).

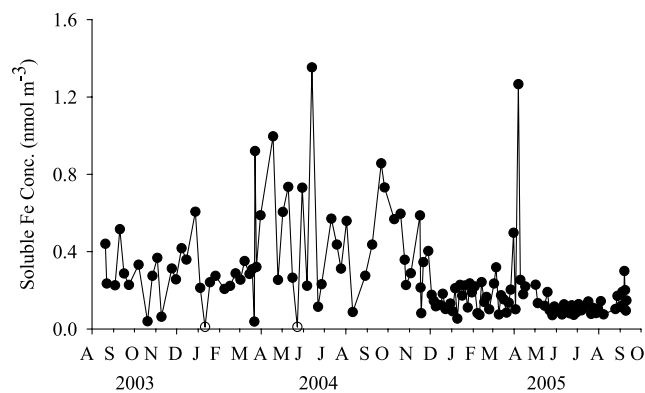
### 3.3. Nutrient Fluxes

[20] Only dry deposition fluxes of nutrients to the Gulf of Aqaba were estimated in our study because the climate of this desert-surrounding region is extremely dry throughout the year with almost zero rainfall. Deposition velocity which is needed for the calculation of input fluxes depends on various parameters including the aerosol particles size. Although aerosol particles were sampled with no size resolution, the size distributions of the aerosols were analyzed separately using a GT-321 particle monitor (Met One Instruments<sup>®</sup>) for the various sampling dates. It was found that about 60% of the total volume was contributed by particles with an aerodynamic diameter greater than  $5$   $\mu\text{m}$ , and the particles between  $2\text{--}5$   $\mu\text{m}$  and  $<2$   $\mu\text{m}$  accounted for the 35% and 5% volume respectively. In addition we



**Figure 7.** Strong correlation between the water-soluble  $\text{PO}_4^{3-}$  and the total soluble P (TSP) concentrations ( $\text{nmol m}^{-3}$ ) in the Gulf of Aqaba atmosphere.

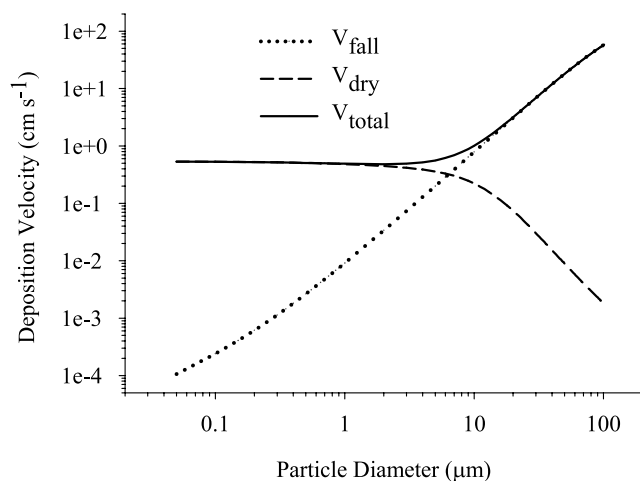




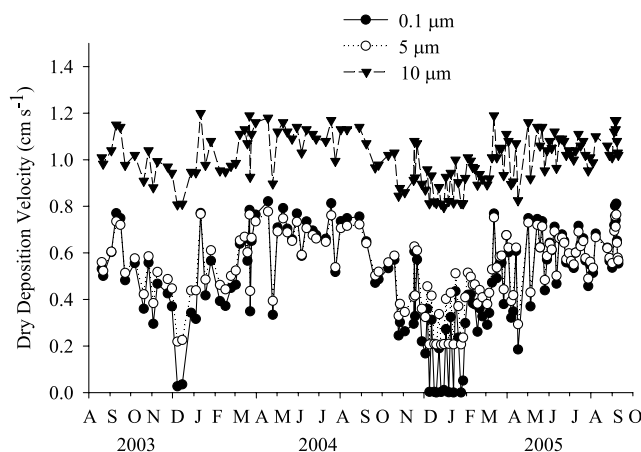
**Figure 8.** Atmospheric concentrations ( $\text{nmol m}^{-3}$ ) of water-soluble Fe measured between 20 August 2003 and 10 September 2005 at the northwest coast of the Gulf of Aqaba ( $29^{\circ}31'N$ ,  $34^{\circ}55'E$ ); the two open circles represent the soluble Fe concentrations below the detection limits (close to zero).

determined the particle size for many of the samples using SEM imaging of the filters, 1/50 of the original filter was cut randomly from the filter samples and analyzed under the SEM using ImageJ software. Accordingly, particle size distribution of each aerosol sample was determined and used to calculate the dry deposition velocity of total aerosol mass for each sampling date.

[21] The sample deposition velocity was largely dictated by the large particles with an aerodynamic diameter greater than  $5 \mu\text{m}$  which comprised the majority of particles in our samples (Figure 9). Deposition velocity for a  $5 \mu\text{m}$  particle for example is  $0.7 \text{ cm s}^{-1}$  while a  $20 \mu\text{m}$  particle from the same sample will fall at  $3 \text{ cm s}^{-1}$ . Very similar velocities (difference around 10%, Figure 9) were found for all particles smaller than  $5 \mu\text{m}$  collected in the same date. The small aerosol particles however presented orders of magnitude higher temporal variability in their deposition velocities compared to the large ones. For instance, the



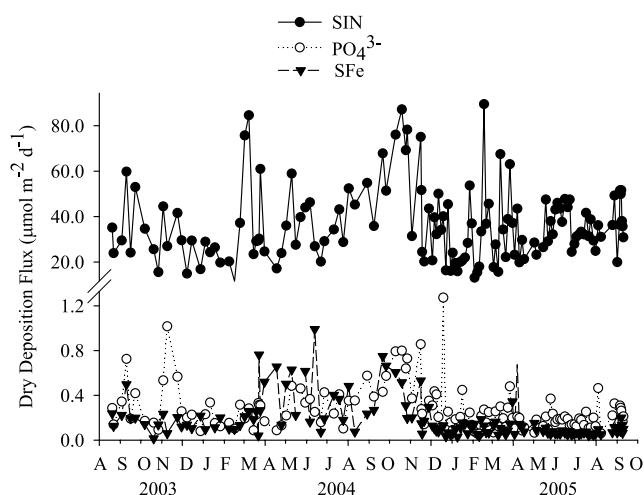
**Figure 9.** An example of the model output for the aerosols sampled on 20 August 2003 including the sedimentation, dry deposition and total dry deposition velocities ( $\text{cm s}^{-1}$ ) varied with different sizes of aerosol particles.



**Figure 10.** Temporal variation of the dry deposition velocities calculated for the three sizes ( $0.1$ ,  $5$  and  $10 \mu\text{m}$  diameter) of aerosol particles during the sampling period of 20 August 2003 to 10 September 2005.

deposition velocities of  $0.1$  and  $10 \mu\text{m}$ -diameter particles in our 2-year sampling period (20 August 2003 to 10 September 2005) ranged between  $0.0002$  and  $0.8 \text{ cm s}^{-1}$  (for  $0.1 \mu\text{m}$ ) and between  $0.8$  and  $1.2 \text{ cm s}^{-1}$  (for  $10 \mu\text{m}$ ) respectively (Figure 10). Accordingly, total-mass deposition velocity would show less temporal variability over the sampling period if the aerosols were dominated by large particles ( $>5 \mu\text{m}$ ) than if constituted by only small ones ( $<5 \mu\text{m}$ , Figure 9).

[22] To estimate the dry deposition fluxes of nutrients to the Gulf, we assumed that the soluble nutrient species ( $\text{NO}_2^-$ ,  $\text{NO}_3^-$ ,  $\text{NH}_4^+$ ,  $\text{PO}_4^{3-}$  and Fe) were evenly distributed on various sizes of aerosol particles. This assumption may bring an inherent uncertainty of up to a factor of 7 to the estimated nutrient fluxes. This uncertainty was determined by taking into account the wide range of possible deposition velocities based on the particle size analysis and using variable size-dependent distribution of nutrients in our samples (e.g., assuming that all of the content of a certain nutrient is associated with one size fraction or another). Nonetheless, this maximum uncertainty is relevant only for  $\text{NH}_4^+$  which is mainly associated with the more variable deposition of the fine mode aerosols ( $<1 \mu\text{m}$ ) of anthropogenic origin [Huebert *et al.*, 1998; Spokes *et al.*, 2000; Jickells *et al.*, 2003]. Other nutrient species including  $\text{NO}_3^-$  [Savoie and Prospero, 1982; Spokes *et al.*, 2000],  $\text{PO}_4^{3-}$  and Fe [Duce *et al.*, 1991; Prospero *et al.*, 1996] generally occur in the coarse aerosol mode. The coarse mode includes the large particles ( $>5 \mu\text{m}$ ) that basically determines the dry deposition velocity of the total aerosol mass (Figure 9), thus using the total mass deposition velocity for components associated with the coarse fraction (e.g.,  $\text{NO}_3^-$ ,  $\text{PO}_4^{3-}$  and SFe) does not involve a large error. To determine the best estimate for the dry deposition fluxes of  $\text{NH}_4^+$ , we applied the dry deposition velocities of a fine mode particle ( $D = 0.75 \mu\text{m}$ ) calculated using the particle deposition model for each sample since all the fine mode particles had relatively similar deposition velocities at any given date (although these differed between dates).



**Figure 11.** Estimates of the dry deposition fluxes ( $\mu\text{mol m}^{-2} \text{d}^{-1}$ ) of water-soluble inorganic N, P and Fe to the Gulf of Aqaba during the sampling period of 20 August 2003 to 10 September 2005.

[23] The calculated fluxes of water-soluble inorganic N, P and Fe to the Gulf showed high temporal variability, and ranged between 12–87, 0.08–1 and 0.01–1  $\mu\text{mol m}^{-2} \text{d}^{-1}$  during the intermittent sampling period (20 August 2003 to 28 November 2004) and between 13–90, 0.06–1 and 0.03–0.7  $\mu\text{mol m}^{-2} \text{d}^{-1}$  during the continuous sampling period (28 November 2004 to 10 September 2005), respectively. The mean fluxes of water-soluble  $\text{PO}_4^{3-}$  and Fe (0.2 and 0.1  $\mu\text{mol m}^{-2} \text{d}^{-1}$ ) derived from the continuous sampling of aerosols during 2004–2005 were found to be significantly lower than those (0.3 and 0.3  $\mu\text{mol m}^{-2} \text{d}^{-1}$ ,  $P < 0.01$ ) derived from the aerosols sampled one day per week during 2003–2004. However, no statistically significant difference was observed for the mean SIN fluxes (39 and 34  $\mu\text{mol m}^{-2} \text{d}^{-1}$ ,  $P > 0.1$ ) between the two sampling periods. The generally higher fluxes of soluble nutrients estimated between August 2003 and November 2004 could be a result of the intermittent sampling method applied that may have captured episodic aerosol events over the Gulf which skewed the statistics. No clear seasonal pattern was identified for the SIN flux, and the maximum input of SIN over the Gulf occurred in October 2004 as well as around the beginning of March in both years (Figure 11). Unlike SIN, soluble  $\text{PO}_4^{3-}$  fluxes presented a discernable seasonal pattern with winter and fall fluxes (mean 0.4  $\mu\text{mol m}^{-2} \text{d}^{-1}$  in September to December) approximately a factor of 2 higher than those in other seasons (mean 0.2  $\mu\text{mol m}^{-2} \text{d}^{-1}$ , Figure 11), consistent with the weak seasonal trend of the soluble  $\text{PO}_4^{3-}$  concentration in the aerosols. The more pronounced seasonal pattern in the  $\text{PO}_4^{3-}$  fluxes was due to the particle size distribution with larger particles and thus faster deposition during the winter. The higher fraction of larger particles may be attributed to more frequent breakouts of Sahara dust storms that carry larger particles over the eastern Mediterranean during the winter [Kocak et al., 2004] than in other seasons.

[24] Dry deposition fluxes of soluble nutrients to the Gulf were also estimated using the mean deposition velocities previously reported for aerosol  $\text{NO}_3^-$ ,  $\text{NH}_4^+$ ,  $\text{PO}_4^{3-}$  (1, 0.1,

2  $\text{cm s}^{-1}$  [Duce et al., 1991]) and Fe (1  $\text{cm s}^{-1}$  [Jickells and Spokes, 2001]). Using these fixed mean velocities the uncertainties in flux were estimated to be about a factor of 3 [Duce et al., 1991]. Using the mean deposition velocities above may give a better estimate (smaller uncertainties) of the mean fluxes of nutrients to the Gulf than the values obtained from averaging the deposition for individual samples (which may have an uncertainty of up to a factor of 7 based on propagation of maximum error in model inputs); however using the mean deposition velocity temporal or seasonal variation of nutrient fluxes caused by the change of daily meteorological conditions and particle size distributions is not fully captured. The calculated mean fluxes of soluble inorganic N, P and Fe using the mean velocities for the whole sampling period were 36, 0.7 and 0.2  $\mu\text{mol m}^{-2} \text{d}^{-1}$ , respectively. Interestingly, these values are very similar to the mean SIN and SFe fluxes (36 and 0.2  $\mu\text{mol m}^{-2} \text{d}^{-1}$ ) derived from the average of individual sample estimates, and the only difference between the two calculations is in the soluble  $\text{PO}_4^{3-}$  fluxes (0.7 and 0.3  $\mu\text{mol m}^{-2} \text{d}^{-1}$ ), however, even these estimates are within the uncertainty ranges reported for both methods.

[25] Our estimate of the mean SIN flux (36  $\mu\text{mol m}^{-2} \text{d}^{-1}$ ) is comparable to that estimated in the eastern Mediterranean at Crete (32  $\mu\text{mol m}^{-2} \text{d}^{-1}$  [Kouvarakis et al., 2001; Markaki et al., 2003]), but is about twice as much as that reported from the Israeli Mediterranean coast (18  $\mu\text{mol m}^{-2} \text{d}^{-1}$  [Herut et al., 1999] (see Table 3)). The SIN dry flux over the Israeli coast was estimated indirectly assuming that it was 1/3 of the wet flux [Herut et al., 1999], and thus may not be accurate. The mean SIN flux over the Gulf is at least an order of magnitude higher than the flux estimates for the open Pacific (0.6–3  $\mu\text{mol m}^{-2} \text{d}^{-1}$ ) and Atlantic oceans (1–5  $\mu\text{mol m}^{-2} \text{d}^{-1}$  [Chen, 2004] (see Table 3)). This is expected because the long distance from land to the open ocean sites results in a relatively low aerosol flux over the open ocean. The mean  $\text{PO}_4^{3-}$  flux over the Gulf (0.3  $\mu\text{mol m}^{-2} \text{d}^{-1}$ ) was about a factor of 3 lower than that reported from the Israeli Mediterranean coast (0.9  $\mu\text{mol m}^{-2} \text{d}^{-1}$  [Herut et al., 1999]), but comparable to the higher ends of the flux ranges over the open Pacific (0.08–0.4  $\mu\text{mol m}^{-2} \text{d}^{-1}$ ) and Atlantic oceans (0.1–0.3  $\mu\text{mol m}^{-2} \text{d}^{-1}$  [Chen, 2004] (see Table 3)). Our estimate was also in a good agreement with the seawater soluble  $\text{PO}_4^{3-}$  fluxes reported by Markaki et al. [2003] to the eastern Mediterranean (Table 3). The mean SFe flux over the Gulf was about 1–2 orders of magnitude and 1–3 times higher than flux estimates for the central North Pacific [Chen, 2004] and the tropical North Atlantic [Chen and Siefert, 2004] (see Table 3), respectively.

[26] The above estimates of atmospheric nutrient fluxes are based on the soluble fractions of aerosol nutrients extracted in pure water. We report these values for comparison with other studies most of which use solubilities based on pure water extractions. However, this may overestimate the atmospheric deposition of certain nutrients to the Gulf (e.g.,  $\text{PO}_4^{3-}$  and Fe) as a consequence of their lower solubilities in seawater than in pure water [Chen et al., 2006]. Solubility experiments for the aerosols in the Gulf showed that aerosol inorganic N (mainly  $\text{NO}_3^-$  and  $\text{NH}_4^+$ ) had similar solubilities in pure water and in seawater, while seawater-soluble  $\text{PO}_4^{3-}$  and Fe fractions were respectively

**Table 3.** Dry Deposition Fluxes (Mean  $\pm$  SD) of Soluble Inorganic N (SIN Including  $\text{NO}_2^-$ ,  $\text{NO}_3^-$  and  $\text{NH}_4^+$ ),  $\text{PO}_4^{3-}$  and Fe (SFe) to the Gulf of Aqaba<sup>a</sup>

Location	SIN	$\text{PO}_4^{3-}$	SFe	Reference
Eilat, Israel	$36 \pm 16$	$0.3 \pm 0.2$	$0.2 \pm 0.2$	this work
Eilat, Israel	$38 \pm 17^b$	$0.2 \pm 0.2^b$	$0.02 \pm 0.02^b$	this work
Israeli Mediterranean coast	$18^{b,c}$	$0.9^b$		Herut et al. [1999]
Crete Island	32			Kouvarakis et al. [2001]
Erdemli	$120^b$	$0.5^b$		Markaki et al. [2003]
Crete Island	$32^b$	$0.3^b$		Markaki et al. [2003]
Central North Pacific	0.6–3	0.08–0.4	0.004–0.02	Chen [2004]
Tropical North Atlantic	1–5	0.1–0.3	0.06–0.2	Chen and Siefert [2004]

<sup>a</sup>Unit is  $\mu\text{mol m}^{-2} \text{d}^{-1}$ . Compare to the estimates reported in the literature for the eastern Mediterranean area and open oceans.

<sup>b</sup>Seawater soluble inorganic nutrients from dry fallout.

<sup>c</sup>Calculated from wet deposition (dry = 1/3 of wet).

20% and an order of magnitude lower than those in pure water. On the basis of these experiments, pure-water SIN and  $\text{PO}_4^{3-}$  measurements can be converted to their respective seawater solubilities through equations (4) and (5), and seawater-soluble aerosol Fe can be estimated as one tenth of its pure-water concentrations [Chen et al., 2006].

$$SW(SIN) = 1.02 \times PW(SIN) + 0.51 \quad (4)$$

$$SW(\text{PO}_4^{3-}) = 0.86 \times PW(\text{PO}_4^{3-}) + 0.012 \quad (5)$$

where  $SW(SIN)$  and  $PW(SIN)$  represent atmospheric SIN concentrations ( $\text{nmol m}^{-3}$ ) derived from seawater and pure-water extractions, respectively. Similar abbreviations are used for  $\text{PO}_4^{3-}$ . Using the relationships of nutrient solubilities between pure water and seawater, the mean atmospheric deposition of seawater-soluble inorganic N, P and Fe to the Gulf can be calculated as 38, 0.2 and  $0.02 \mu\text{mol m}^{-2} \text{d}^{-1}$  respectively (Table 3). We use these values rather than calculations based on pure water solubilities to evaluate the ecological effects of atmospheric deposition of nutrients to the Gulf.

### 3.4. Ecological Effects

[27] The ratios of the SIN to soluble  $\text{PO}_4^{3-}$  fluxes from the atmosphere to the Gulf varied from 32 to 541 with a mean ratio of 196, well above the Redfield ratio (N: P = 16: 1). The average 196 times higher input of SIN than  $\text{PO}_4^{3-}$  from the atmosphere suggests that the atmospheric aerosol deposition could increase the likelihood of P limitation in the Gulf ecosystem. Obviously the actual limiting nutrient will depend on the N:P ratios of all nutrient sources to the euphotic zone of the Gulf coupled with the specific demands by organisms. It was found that the deep Gulf water, another major nutrient source to the euphotic zone, also had an N:P ratio slightly higher than 16 (A. Paytan, unpublished data, 2005), which further suggests that the nutrient ratio of the atmospheric input will be a critical factor controlling the nutrient balance (N/P ratio) in the Gulf.

[28] The water column of the Gulf between April and October is stratified with a mixing depth much less than 100 m based on the temperature and nutrient profiles of the water column [Lindell and Post, 1995]. During these seven months, atmospheric input of nutrients would lead to an increase of 80, 0.5 and  $0.04 \text{ nM}$  for soluble inorganic N, P and Fe respectively in the up 100 m of seawater (the depth

above which most of the phytoplankton biomass exists). Obviously, this might not be reflected in the measured nutrient concentrations because the nutrients are not conservative and will be lost as a result of biological uptake and scavenging. Nonetheless, such atmospheric input of nutrients particularly SIN is important ( $\sim 35\%$ ) when compared to the averaged concentrations of dissolved inorganic N and P (230 and 20 nM (A. Paytan, unpublished data, 2005)) and may also impact the Fe content (6 nM [Chase et al., 2006]) in the euphotic zone during the stratification period.

[29] If all of the atmospheric soluble input of  $\text{PO}_4^{3-}$  (mean  $0.2 \mu\text{mol m}^{-2} \text{d}^{-1}$ ) was effectively utilized by marine phytoplankton in the Gulf, assuming a Redfield uptake ratio, this source could support close to  $0.3 \text{ mg C m}^{-2} \text{d}^{-1}$  uptake by phytoplankton. This corresponds to about 0.1% of the total production in the Gulf ( $219 \text{ mg C m}^{-2} \text{d}^{-1}$  [Levanon-Spanier et al., 1979; Lindell and Post, 1995]). A similar calculation for SIN using the C:N Redfield ratio and the estimated SIN deposition (mean  $38 \mu\text{mol m}^{-2} \text{d}^{-1}$ ) could support  $3 \text{ mg C m}^{-2} \text{d}^{-1}$  of primary production which is about 1.4% of the total production in the Gulf. It is important to note that although the atmospheric nutrient contribution to the total annual C production seems small (0.1–1.4%) this is a source of “new” nutrients to the ecosystem, and assuming typical  $f$  ratios in oligotrophic systems (0.05–0.1 [Eppley and Peterson, 1979]) this source may constitute a substantial fraction of the new production. Moreover, as discussed above this is a minimum estimate since the nitrogen input in gas form is not considered here (see discussion above). Importantly, the atmospheric deposition may be particularly essential for supporting photosynthetic C uptake during the summer when the water column is stratified. Levanon-Spanier et al. [1979] measured a surface C production of about  $29 \text{ mg C m}^{-2} \text{d}^{-1}$  during the summer (July and August), we measured similar rates in the summer of 2003 and 2004 (unpublished), of this summer production about 10% could be supported by the atmospheric input of SIN using the average flux of  $35 \mu\text{mol m}^{-2} \text{d}^{-1}$  in July and August (Figure 11). Thus, for the stratified summer period assuming an  $f$  ratio of 0.1, atmospheric N deposition may support all of the new production. Indeed, incubation experiments with dust additions to the natural phytoplankton assemblage of the Gulf showed that aerosols could induce significant increase in chlorophyll a and cell numbers, which are measures for



primary production (K. Mackey et al., manuscript in preparation, 2007).

[30] The total C production driven by the atmospheric deposition of SIN ( $3 \text{ mg C m}^{-2} \text{ d}^{-1}$  or  $249 \text{ } \mu\text{mol m}^{-2} \text{ d}^{-1}$ ) would require at most  $0.009 \text{ } \mu\text{mol m}^{-2} \text{ d}^{-1}$  of bioavailable Fe using the intracellular Fe:C ratios for coastal species ( $25\text{--}38 \text{ } \mu\text{mol mol}^{-1}$ ) at specific growth rates greater than  $1.0 \text{ d}^{-1}$  [Raven, 1988; Sunda and Huntsman, 1995]. Assuming that the seawater-soluble aerosol Fe is bioavailable, atmospheric deposition of SFe ( $0.02 \text{ } \mu\text{mol m}^{-2} \text{ d}^{-1}$ ) would be in large excess compared to that required for the phytoplankton growth driven by the SIN deposition [see also Chase et al., 2006].

[31] The discussion above is based on annual average atmospheric fluxes of nutrients to the Gulf. It is important to note that the effect on primary production and C sequestration based on the averaged fluxes is much smaller than that triggered by individual dust events on a daily basis, as the response time of the phytoplankton is fast. For example, the estimated atmospheric flux of SIN to the Gulf was  $52 \text{ } \mu\text{mol m}^{-2} \text{ d}^{-1}$  on 29 August 2004 (Figure 11), which can support 14% of summer surface total C production ( $29 \text{ mg C m}^{-2} \text{ d}^{-1}$ , see the paragraph above) on a daily basis. This is a significant fraction of total production and may be the most important component of new production during these specific events. This effect is comparable to and even higher than those reported for other oligotrophic regions such as the Mediterranean Sea and the North Atlantic. The atmospheric N depositions (wet and dry) amount to approximately 6–10%, 12% and 4–6% of the new N need on an annual basis, in the Gulf of Lions, the oligotrophic zones of the eastern Mediterranean and the Canary Islands region, respectively [Guerzoni et al., 1999; Neuer et al., 2004]; while the atmospheric dry deposition of N supports about 10% of the new production at the  $30^\circ\text{N}$  eastern Atlantic site [Neuer et al., 2004].

#### 4. Conclusions

[32] The air masses over the Gulf of Aqaba are strongly impacted by mineral dust transported from the surrounding deserts. Accordingly, water-soluble aerosol  $\text{NH}_4^+$  and  $\text{PO}_4^{3-}$  concentrations are relatively low over the Gulf compared to adjacent more industrialized areas because of the lower contribution of anthropogenic aerosol sources. The main SIN compounds in the water-soluble fraction of aerosols over the Gulf are nitrate (60%) and ammonium (38%). The SIN accounts for about 86% of TSN, while both inorganic  $\text{PO}_4^{3-}$  (69%) and organic P (31%) are important in contributing to TSP in the Gulf area. Our estimates of soluble nutrient fluxes from the atmosphere to the Gulf show a high temporal variability. Aerosol soluble P fluxes present a seasonal pattern with higher fluxes during the winter months than during other periods, consistent with the higher frequency of Saharan dust outbreaks during the winter. The mean fluxes of atmospheric nutrients to the Gulf are of the same order of magnitude as values reported from the neighboring area.

[33] We evaluate the ecological effects of atmospheric deposition of nutrients on the Gulf ecosystem based on seawater-soluble mean nutrient fluxes. The atmospheric deposition ratio of macronutrient species ( $\text{SIN} : \text{PO}_4^{3-} =$

196: 1) tends to increase the likelihood of P limitation in the Gulf. Atmospheric deposition could contribute a substantial fraction (35%) of dissolved inorganic N to the euphotic zone during the stratification period (April to October); however, this estimate may have a relatively large uncertainty due to a maximum uncertainty of a factor of 7 of the deposition flux calculations. This SIN input could support about 10% of surface primary production during the summer months, and may constitute a substantial fraction of surface new production of the Gulf. Such nutrient contribution is similar or even higher than those reported for other oligotrophic regions such as the Gulf of Lions, the eastern Mediterranean Sea and the eastern subtropical North Atlantic. Atmospheric deposition of SFe to the Gulf is in large excess compared to that required for the phytoplankton growth driven by atmospheric SIN input in this region.

[34] Data reported here is the first attempt to quantify nutrient fluxes from the atmosphere to the Gulf of Aqaba which is important for understanding the nutrient dynamics of this arid (desert) environment with no nutrient inputs from either river runoff or atmospheric wet deposition. Moreover, this external source of nutrients may contribute to the balance of the net loss of nutrients from the system which results from the circulation pattern of the Gulf (incoming nutrient poor surface water and outgoing subsurface water higher in nutrients). The Gulf which represents an oligotrophic region with high dust deposition may be representative of more wide spread future conditions of increased aridity and dust fluxes [Teegen et al., 2004; Woodward et al., 2005] and thus data presented here are important for understanding and modeling future anthropogenic aerosol impacts.

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