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Dust deposition to the East China Sea and its biogeochemical implications

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[1] Atmospheric Al concentrations published by Hsu et al. (2008) are used together with new information on mass particle-size distributions (MSDs) to calculate dust deposition to northern Taiwan and to evaluate the significance of the process for the East China Sea (ECS). Seasonal variations in aerosol Al in northern Taiwan show maxima in spring and winter and minima in summer and fall. The MSD of Al and Fe is monomodal, with a mean mass median diameter of 3.6 ± 1.2 μm, and the dry deposition velocity integrated over the entire MSD is 2.0 ± 1.5 cm s^{-1}. The estimated dust deposition to the ECS is $\sim$20 g m^{-2} a^{-1}, with wet deposition accounting for three fourths of the total. The quantity of dust deposited decreases exponentially with distance from the source regions. The annual dust input to the entire ECS is estimated to be $\sim$17 Mt, and when compared with riverine discharge, dust deposition appears to be a significant source of sediments and dissolved Fe. The large quantities of dust deposited also could have significant implications for the biogeochemical cycling of Fe (and Al), including effects on phytoplankton populations.

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1. Introduction

[2] Mineral aerosol particles play important roles in radiative forcing [*Levitus et al.*, 2001; *Bergin et al.*, 2001] and climate change [*Andreae and Crutzen*, 1997; *Kiehl*, 1999; *Yu et al.*, 2002]. Heterogeneous reactions with reactive gases, such as SO_2 and NO_x, can occur on the dust particles' surfaces [*Dentener et al.*, 1996; *Song and Carmichael*, 2001], thus influencing atmospheric acidity [*Wang et al.*, 2002]. Furthermore, windblown dust can influence marine ecosystems after the dust is deposited in the oceans and the available nutrients dissolve [*Duce et al.*, 1991; *Gao et al.*, 2001]. With reference to the latter, there has been particular interest in eolian Fe and some transition metals because they can stimulate the growth of marine algae [*Jickells et al.*, 2005, and references therein]; indeed numerous Fe-enrichment experiments have been conducted in the field and in the laboratory [*Boyd et al.*, 2007, and references therein].

[3] Dust storms occur annually in the arid/semi-arid areas and deserts in northern and northwestern China. While they are most frequent in spring, they also occur in late winter. There are at least three major regions for Asian dust (AD), including the Taklimakan Desert, the Gobi Desert, and the deserts in northern China; interestingly, the Loess Plateau is now considered to be a minor source for dust [*Zhang et al.*, 2003] (Figure 1a). Massive quantities of windblown dust from the Asian deserts can be entrained into the free troposphere and subsequently transported by the prevailing westerly winds [*Merrill*, 1989; *Zhang et al.*, 1997]. Recently, *Laurent et al.* [2006] have estimated the emission of dust from the Chinese and Mongolian deserts to be between 100 and 460 Mt a^{-1}. Indeed, air parcels carrying yellow sand can travel thousands of kilometers or more from the source regions [*Duce et al.*, 1980; *Blank et al.*, 1985]. Areas influenced by the deposition of Asian dust include the adjacent Chinese marginal seas (e.g., the Yellow and East China Seas and the Sea of Japan) [*Gao et al.*, 1992; 1997]; Korea, Japan and other parts of eastern Asia [*Choi et al.*, 2001]; the North Pacific, and even North America [*Husar et al.*, 2001].

[4] Estimates of AD emission and fluxes show considerable variability. One assessment suggests that the quantity of dust annually emitted from China is $\sim$800 Mt, and roughly 50% of that is transported to the North Pacific Ocean and beyond [*Zhang et al.*, 1997]. In an earlier study, *Duce et al.* [1991] estimated the annual dust deposition to the entire North Pacific as up to 480 Mt. From a modeling study, *Gao et al.* [2001] obtained a dust deposition to the North Pacific of 86 Mt a^{-1} (here Fe deposition was

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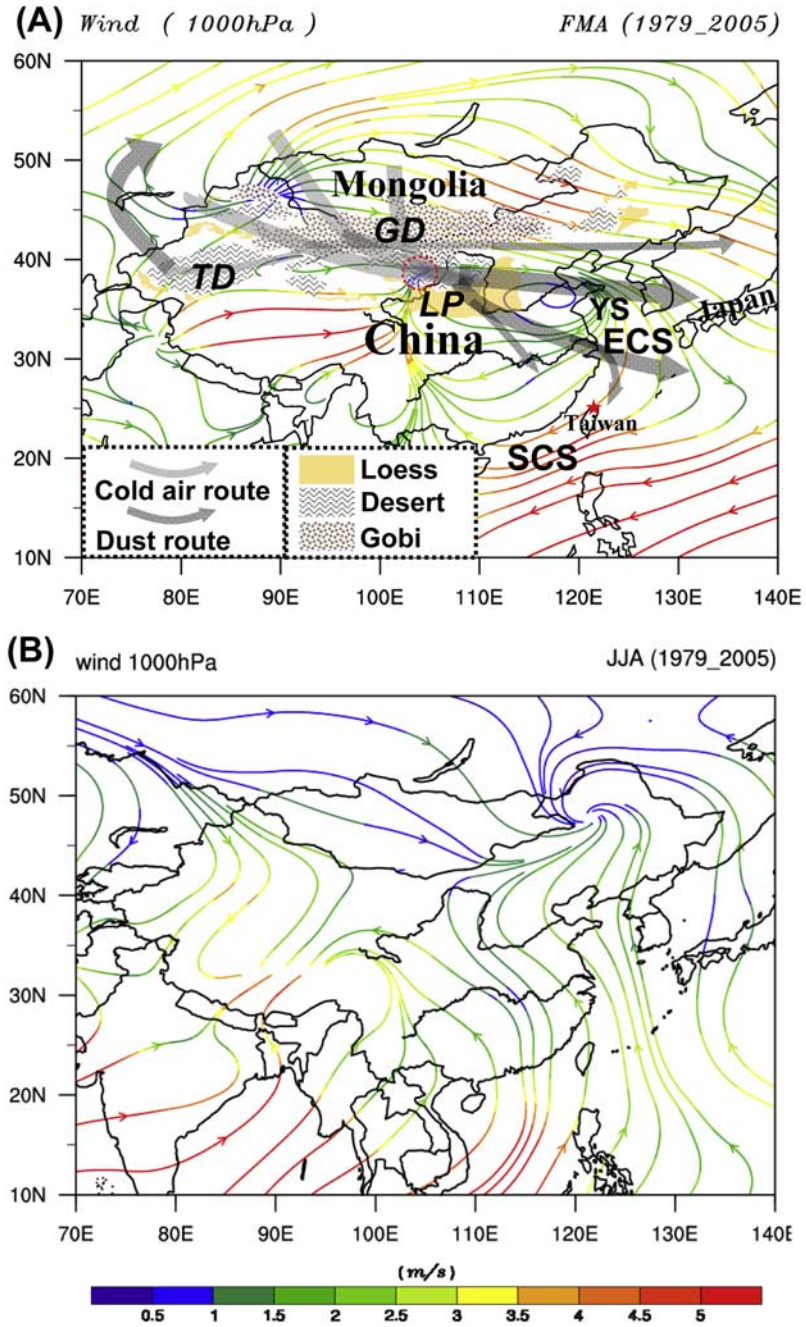

Figure 1. (a) A synoptic illustration showing the three major arid/semiarid areas: Takalimakan Desert (TD) in northwestern China, Gobi Desert (GD) in the boundary of inner and outer Mongolia, and Loess Plateau (LP) in eastern China. Also displayed are the three transport routes for cold air (light shaded arrows) and windblown soil dusts (dark shaded arrows) out of the continent. Note that a minor branch separates from the southern path, leading to the southern East China Sea and Taiwan. Transport paths are modified from *X. Y. Zhang et al.* [1993], *Zhang et al.* [1997], *Xuan* [1999], and *Sun et al.* [2001]. The dashed red circle approximates the location of Badain-Juran Desert, a major dust source region. The sampling site for the present study was Taipei, Taiwan (stars). The base map shows typical 1000 hPa streamlines during winter/spring (February, March, and April) for 1979–2005. Arrows indicate prevailing wind directions, and the color scale indicates wind speed (m s^{-1}). The northeasterly monsoon is obvious south of 30°N. (b) 1000 hPa streamlines in summer (represented by the months of June, July, and August).

converted to dust deposition by assuming that the Fe content in dust is 3.5%). *Uematsu et al.* [2003] estimated the annual atmospheric deposition of AD to the western North Pacific to be 64 Mt; but earlier *Prospero et al.* [1989] had estimated the dust input to the western North Pacific (25°N to 50°N, west of 150°E) to be about 300 Mt a^{-1}. For the Chinese marginal seas, *Gao et al.* [1997] calculated annual dust deposition rates of 31 and 36 Mt for the East China Sea (including the Bohai and Yellow Seas) and the South China Sea, respectively, but those estimates were based on a very limited data set.

[5] The northeasterly (NE) monsoon is a major influence on the atmospheric circulation in eastern Asia from late September to early May, and it is critical for the southeastward transport of AD beyond 30°N. In a prior study, we found that AD plumes arrived in northern Taiwan two to seven times per year and that during the high-dust season, dust concentrations were enhanced by ~75% compared with those in summer [*Hsu et al.*, 2008]. Therefore, one might reasonably expect that the quantity of AD deposited into the East China Sea would be large enough to significantly influence marine biogeochemical cycles. However, none of the estimates of dust deposition to the East China Sea that have been done to date have been based on large data sets, and none have explicitly considered the potential effects on marine biogeochemistry.

[6] *Hsu et al.* [2008] identified major AD events in northern Taiwan based a 5-year data set and proposed a criterion (geometric mean concentration times twice the geometric standard deviation) to define significant major AD events. Here, we determine the size distribution of Al and Fe from a yearlong measurement program, and we combined those results with the existing 5-year dust data set to calculate dry deposition rates. We then estimate an integrated wet plus dry deposition of mineral dust and dust-derived Fe for the entire ECS, and finally, we evaluate the significance of the AD deposition with reference to ECS biogeochemistry, sediment budget, and eolian Fe deposition. This study is the first to link the atmospheric deposition of AD with various aspects of marine biogeochemistry over the Chinese marginal seas.

2. Synoptic Atmospheric Circulation

[7] Investigations of AD outbreaks revealed three principal transport pathways in the zone from 30°N to 45°N [*Sun et al.*, 2001; *Uematsu et al.*, 2003; *Gao et al.*, 2003]. Figures 1a and 1b illustrate the mean 1000 hPa streamlines over East Asia during the NE monsoon (represented by the three months of February, March, and April) and summer (SUM) monsoon (represented by the three months of June, July, and August). During the NE monsoon, strong surface winds generated by anticyclones over the Yellow River watershed (around 35°N, 115°E) can reach a mean wind speed of ~4 m s^{-1} or more over Taiwan, and this can lead to the transport of AD southward (Figure 1a). *Lin* [2001] suggested that the transport of AD from northern China to Taiwan requires 2 to 3 days and that the dust was confined primarily to an altitude of 500 to 1500 m, which is considerably lower than what has been observed over Japan (2–6 km; see *Iwasaka et al.* [1988]).

[8] During the SUM monsoon, the wind speeds and directions differ markedly from those in the winter, and the southeasterly flow in summer normally does not bring much dust to the Taiwan region. Figure 1b shows that the prevailing southeasterly winds in summer are driven by subtropical- or cross-equatorial flows. Note that during summer, the wind speed is only about 1 m s^{-1} and thus much lower compared with that in winter. These seasonal differences in wind patterns are a major influence on the seasonality in dust concentrations as illustrated in Figure 2.

3. Materials and Methods

3.1. Sampling

[9] Size-resolved aerosol particle samples were collected with the use of a cascade impactor which was located on the rooftop of the Institute of Earth Sciences, Academia Sinica in Taipei City. This site is ~10 km northeast of the Central Weather Bureau of Taiwan where the PM_{10} aerosol samples were collected for the prior studies of atmospheric Al [*Hsu et al.*, 2008]. Taipei itself is located about 20 km from the northern tip of Taiwan (Figure 1). The study area is characterized by abundant rainfall (2325 mm a^{-1}; http://www.cwb.gov.tw/V4/index.htm), and it is affected by the transport pathways described above.

[10] A 10-stage micro-orifice uniform deposit impactor (MOUDI, Model 110, MSP Corporation, and Minneapolis, Minnesota, USA) was used to collect nine sets of size-segregated aerosol samples between April and December 2003. Of these, five sets were obtained during the NE monsoon and the other four during the SUM monsoon. Basically, there was one set collected each month, except for November. The actual sampling dates were: 4/27–29; 5/26–30; 6/27–30; 7/30–8/1; 8/27–29; 9/24–26; 10/29–31; 12/19–22; 12/26–29. From our analyses of the size-resolved samples, it appears that the collection of some giant, locally generated, suspended-dust particles is unavoidable, but as explained below this did not affect our main results.

[11] The MOUDI impactor has 10 size-fractionating stages (50% cutoff diameters: 10, 5.6, 2.5, 1.8, 1.0, 0.56, 0.32, 0.18, 0.10, and 0.056 μm) with inlet (nominal cut-size 18 μm) and backup (<0.018 μm) filters. Membrane filters (PTFE, 47 mm in diameter and 1.0 μm pore size, Pall Gelman, East Hills, NY, USA) were used for all stages with the exception of the backup stage which used a quartz filter as a substrate; those final filters were not analyzed owing to high levels of impurities. Here, the inclusion of the inlet stage allowed the collection of particles in eleven size fractions between 0.056 and >18 μm.

[12] The sampling interval for each set of size-resolved samples was approximately three days, and the sampler operated at a flow rate of 30 l min^{-1}. Sample filters were conditioned for at least 48 h and then were weighed using a microbalance (MX 5, Mettler-Toledo Inc., Greifensee, Switzerland; detection limit 1 μg) in a weighing room at a controlled relative humidity of 35 ± 5% before and after collection.

3.2. Chemical Analyses

[13] The aerosol-laden filters were digested with a mixture of strong acids (HNO_3, HF and $HClO_4$); details of the sample preparation procedures are given by *Hsu et al.*

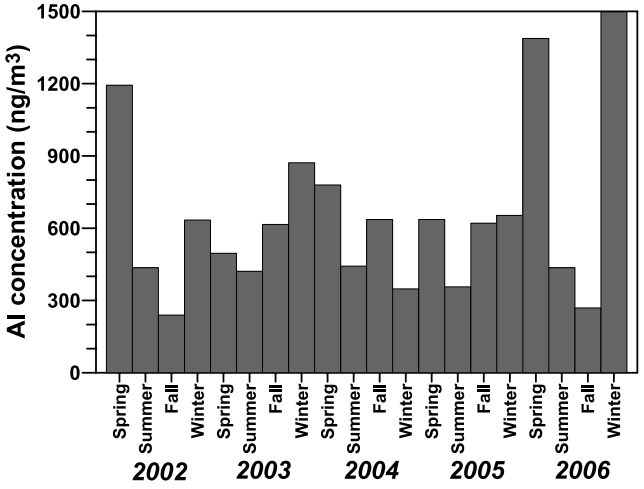

Figure 2. Seasonal variations of aerosol Al concentrations (ng m^{-3}) from February 2002 to February 2007; this data set is from *Hsu et al.* [2008]. For each season the mean value of the data reported in the work of *Hsu et al.* [2008] is presented here.

[2008]. Metal concentrations were determined with the use of a quadrupole-based, inductively coupled plasma mass spectrometer (ICP-MS, Elan 6100, PerkinElmer, Waltham, Massachusetts). For each run, a blank reagent and three membrane filter blanks were prepared and were subjected to the same procedures as the samples. In our study, the method detection limit (MDL; using units of ng/m^3) for Al and Fe was assessed by using three times the standard deviation calculated from ten blanks divided by a 48-h average flow volume. The MDLs of Al and Fe were similarly ~0.5 ng m^{-3} for the size-resolved samples, which is better than that (2 ng m^{-3}) for the PM_{10} samples; this difference is because the filtered air volumes for size-resolved samples were much higher. Quality assurance and quality control (QA/QC) of the chemical analyses were validated by analyzing a standard reference material (SRM), NIST SRM 1648 (urban particulate matter from NIST, National Institute of Standards and Technology, Gaithersburg, Maryland). The recovered values for Al and Fe fell within 5% of their certified values. Further details of the analytical methods may be found in the work of *Hsu et al.* [2004a, 2005a, 2008].

3.3. Data Sources

[14] Data for aerosol Al concentrations in samples collected at the Central Weather Bureau of Taiwan from February 2002 and February 2007 were published in the work of *Hsu et al.* [2008]. In those samples, Al concentrations varied from below the detection limit (2 ng m^{-3}) to over 30,000 ng m^{-3}. A total of thirty Asian dust events were identified on the basis of spikes in Al concentrations and air mass trajectory analyses. Many of the events occurred in 2002 and 2006; in contrast, 2003, 2004, and 2005 can be characterized as low-dust years.

[15] Seasonal variations in atmospheric Al (Figure 2) were combined with the mass-particle size information obtained with the cascade impactors in the present study to calculate dust deposition. Note that the two types of samples (i.e., PM_{10} and size-resolved aerosols) were not sampled at the same place, not over the same time period, and not with the same sampling protocols. Nonetheless, these differences would not affect our conclusions in a fundamental way. In particular during the NE monsoon, when the AD outbreaks occur, most or all of the Taipei Basin is under influence of ADs, and the size distribution of dust particles varies little.

[16] Our deposition calculations were further based on the assumption that the Al content of mineral dust was the same as that of Chinese desert dust and loess, that is, 7% Al by weight [*Zhang et al.*, 2003]. Note that this value is different from that (i.e., 8% Al by weight in the upper continental crust [*Taylor*, 1964]) used in the work of *Hsu et al.* [2008] for calculating the mineral dust concentration. Data for monthly precipitation during the same period were acquired from the Central Weather Bureau of Taiwan, and that information was used to calculate the wet deposition as described below.

4. Results and Discussion

4.1. Temporal Variations of Atmospheric Al

[17] The seasonal variation in Al concentrations from the prior 5-year study is shown in Figure 2, with prominent maxima in spring and winter and minima in summer evident. Two years, 2002 and 2006, were characterized as high-dust years while in contrast, 2003–2005 were considered low- to moderate-dust years. These spikes in Al of up to several thousands of ng m^{-3} were quite comparable with those measured onboard over the East China Sea [*Hsu et al.*, 2009], and in nearby areas [*Zhang and Iwasaka*, 1999; *Fang et al.*, 1999; *Zhang et al.*, 2001; *Choi et al.*, 2001]. The interannual mean Al concentration of 638 ng m^{-3} equates to a dust concentration of 9 μg m^{-3}, which is also comparable with those measured over 18 years at a Japanese network (several hundreds of ng of Al m^{-3}; see *Var et al.* [2000]). For comparison, we compiled data from several East Asian cities and the western Pacific (Table 1), and it can be seen that the dust concentrations during the 5-year study at our site are quite comparable with what has been measured at other East Asian cities. It is noteworthy that Al minima usually occur during periods of rain [*Hsu et al.*, 2004a], indicating that frequent rainfall can effectively wash dust particles from the atmosphere [*Kim et al.*, 2007; *Türküm et al.*, 2008]. Also, this implies that wet deposition may be important for biogeochemical cycles in the study region where the annual rainfall is 2000 mm (see further discussion in section 3.6).

[18] The present results combined with previous observations of high springtime and wintertime concentrations at Onna (26°30′N, 127°50′E), Japan [*Tsunogai et al.*, 1985] demonstrate the extensive influence of AD on the regions extending southward of 30°N. That is, the area influenced by AD covers the entire southern part of the ECS during the NE monsoon. These results also show that the regional background of dust loading in air is enhanced not only in spring but also in winter or throughout the entire NE monsoon period when the dust load on average is ~75% higher than in SUM monsoon [*Hsu et al.*, 2008]. The Al mean concentration (1126 ng m^{-3}) in spring 2002 was only slightly lower than what has been reported for the northern

Table 1. Comparison of Dust-Derived Al Concentrations Over East Asian Cities and the North Pacific Ocean

Study Location	Study Time	Al (ng/m^3)	Reference
Northern Taiwan (25.07°N, 121.52°E)	5 years	638 (PM_{10})	this work
Cheju, Korea (33.52°N, 126.48°E)	11 months	1651 (TSP)	*Prospero* [1996]
Okinawa, Japan (26.92°N, 128.25°E)	18 months	1119 (TSP)	*Prospero* [1996]
Hong Kong (22.55°N, 114.30°E)	18 months	967 (TSP)	*Prospero* [1996]
South Taiwan (21.87°N, 120.87°E)	2 months	317 (TSP)	*Prospero* [1996]
Shemya Island (52.92°N, 174.06°E)	11 years	107.3 (TSP)	*Prospero* [1996]
Midway Island (28.22°N, 177.35°E)	12 years	62.6 (TSP)	*Prospero* [1996]
Oahu Island (21.33°N, 157.70°E)	12 years	54.6 (TSP)	*Prospero* [1996]
Qingdao, China (~35°N, 122°E)	2 months of each spring and summer	1600 (TSP)	*Liu et al.* [2002]
Xiamen, China (24°N, 118°E)	5 months	1500 (TSP)	*Gao et al.* [1997]
Hong Kong (22.5°N, 114.3°E)	4 months	~630 (PM_{10})	*Ho et al.* [2003]
Hong Kong (22.5°N, 114.3°E)	5 years	275 (PM_{10})	*Qin et al.* [1997]
Seoul, Korea (37.53°N, 127.07°E)	1.5 spring months	2490 (PM_{10})	*Kim et al.* [2003]

ECS [*Gao et al.*, 1997] and in or around Japan [*Tsunogai et al.*, 1985; *Hashimoto et al.*, 1994; *Var et al.*, 2000].

[19] The magnitude of the atmospheric dust loads presumably is affected by the strength and frequencies of AD events as well as by processes that occur during transport and by the scavenging effects of wet deposition. These results just cited indicate that there is a relatively uniform geographical distribution (or small gradients) of airborne dust over the ECS, at least during parts of the NE monsoon season. They also would indicate that differences in the dust concentrations and properties between the two sites used for the cascade impactor and PM_{10} sampling are likely to be small.

4.2. Mass-Particle Size Distributions of Airborne Particulate Al and Fe

[20] The mean mass-particle size distributions (MSD) of Al and Fe for nine sets of size-segregated aerosol samples are displayed in Figure 3. Obviously, the two crustal elements had similar size distribution patterns along with strong correlations in their atmospheric concentrations as presented later; this is due to the shared mineral dust origin. Both Al and Fe concentrations peak at cascade impactor stage 4 (i.e., 50% cutoff aerodynamic diameter 2.5 to 5.6 μm), and the overall shape of the distributions is very consistent with previous results from nearby East Asian regions and some remote sites in the Western Pacific [*Park et al.*, 2004; *Fu et al.*, 2004]. When compared with observations in Chinese dust source areas [*Zhang et al.*, 1998] or at Beijing [*Gao et al.*, 1997], the size bin with maximum concentrations at Taiwan appears to be finer.

[21] Assuming all Al MSDs were lognormally distributed, the entire MSDs can thus be fitted by a least squares linear regression method [*Arimoto et al.*, 1985, 1997; *Arimoto and Duce*, 1986; *Dulac et al.*, 1989; *Gao et al.*, 1997] as follows:

$$Y = A + B \ln D, \quad (1)$$

where Y is a standard normal deviate of the cumulative percentage of the total mass of Al on each impactor stage, ln D is the natural logarithm of the particle diameter, and A and B are the intercept and slope, respectively. For these nine sets of MSD data, the calculated correlation coefficients squared (R^2) for lognormal fitting ranged from 0.931 to 0.998. The mass median diameters (MMDs) and

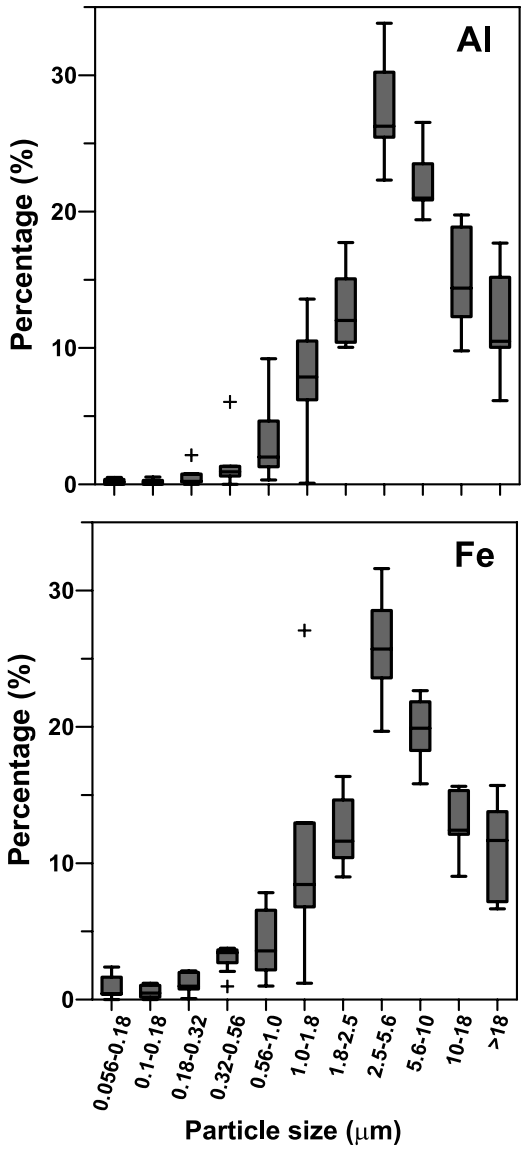

Figure 3. Mass-particle size distributions of atmospheric Al and Fe for nine sets of size-resolved aerosol samples; the peak size bin was 2.5–5.6 μm.

Table 2. Comparisons of Mass Median Diameters and the Corresponding Dry Deposition Velocity for Airborne Al (Dust) From Our Measurements in Northern Taiwan With Other Literature Data

Study Area	Mass Median Diameters (μm)	Mean Dry Deposition Velocity (cm s^{-1})	Reference
Taipei, Taiwan (Southern ECS)	3.6 ± 1.2	2.0 ± 1.5	this work
Northern ECS	3.2	1.9	*Gao et al.* [1997]
North Pacific	3.0 ± 1.8	1.1 ± 1.5	*Arimoto et al.* [1997]
Tropical North Pacific	2.0 ± 4.8	...	*Arimoto et al.* [1985]
Remote North Atlantic	~2.0	0.3 ~ 1.0	*Arimoto et al.* [1997]
Western Mediterranean	2.8	1.8	*Dulac et al.* [1989]
Eastern equatorial Pacific	3.7	...	*Maenhaut et al.* [1983]
Nearshore and coastal environments	...	2	*GESAMP* [1989]; *Duce et al.* [1991]
North Pacific	...	2	*Uematsu et al.* [1985]
Chinese deserts	...	4.4–6.8	*Zhang et al.* [1998]
Xian, China	3.1	3.2	*X. Y. Zhang et al.* [1993]
Qingdao, China	...	4.7	*Qi et al.* [2005]
Chicago, USA	...	2.5	*Yi et al.* [2001]

geometric standard deviation (σ), of the fitted lognormal distribution were obtained from the regression equation (1), as follows:

$$A = 1/\sigma \tag{2}$$

$$B = -\ln(MMD)/\sigma. \tag{3}$$

[22] The MMDs were then computed from the fitted lognormal distribution, yielding a range of 2.1 to 5.6 μm and an average of 3.6 ± 1.2 μm. Interestingly, no seasonality in the MMDs was found even though the size-resolved sampling covered seasons with both high and low dust loadings. Thus the results suggest that the MSD of atmospheric Al (or mineral dust) was not dependent on season and/or atmospheric dust loadings [*Buat-Ménard et al.*, 1983; *Arimoto et al.*, 1997]. For comparison, data from other studies as compiled in Table 2 show that the MMDs of airborne Al (dust) obtained here are very comparable with those from diverse areas in the Pacific Ocean [*Maenhaut et al.*, 1983; *Arimoto et al.*, 1997; *Gao et al.*, 1997]. *Arimoto et al.* [1997] found that the MMDs measured in the North Pacific were larger and more variable than those measured in the North Atlantic. They suggested that such difference could be attributed to the wet conditions and particle aggregation over the North Pacific. It is worth noting in this context that *Porter and Clarke* [1997] found that the dust size distribution at the Mauna Loa Observatory, Hawaii, always peaked at a particle size greater than 3.0 μm regardless of atmospheric dust loadings. However, during atmospheric transport, larger particles theoretically should settle out of suspension faster than smaller ones and this should lead to a change in the mass size distribution with time. Apparently, this was not the case in our studies, and a similar paradox has also been pointed out in studies conducted over the Atlantic Ocean by *Maring et al.* [2003].

4.3. Estimating Eolian Dust Deposition

4.3.1. Dry Deposition

[23] Both dry and wet deposition contribute to the air-to-sea flux of mineral dust. Here they were calculated separately on the basis of long-term data sets, including both the aerosol and meteorological data. The dry deposition flux (F_d) of a given substance was calculated as the product of atmospheric concentration (C) and dry deposition velocity (V):

$$F_d = C \times V. \tag{4}$$

[24] Traditionally, dry deposition was calculated using only an assumed dry deposition velocity corresponding to the MMD for certain chemical species or components of interest [*Duce et al.*, 1991]. Numerous investigations have demonstrated that dust dry deposition fluxes can be dominated by a relatively small fraction of aerodynamically giant particles [*Arimoto and Duce*, 1986; *Dulac et al.*, 1989; *X. Y. Zhang et al.*, 1993; *Zhang et al.*, 1998; *Arimoto et al.*, 1997; *Gao et al.*, 1997]. Accordingly, it is essential to consider the complete MSD in the calculations of dry deposition velocity [*Arimoto et al.*, 1985; *Arimoto and Duce*, 1986; *Dulac et al.*, 1989]. Hence, in our study, the dry deposition rate of atmospheric Al was estimated by considering the entire MSDs.

[25] For the numerical method we used, which we call the "100-step" method, the entire MSD is separated into 100 individual particle-size bins, with each bin corresponding to 1% of the total mass and characterized by particle diameter at its center ($D_{0.5\%}$, $D_{1.5\%}$, $\cdots$, $D_{99.5\%}$) [*Arimoto et al.*, 1985; *Arimoto and Duce*, 1986; *Dulac et al.*, 1989]. Thus, in this way, the modeled MSD was used together with dry deposition models to estimate the dry deposition flux (F_{d-100}) of eolian dust particles as in the following equation:

$$F_{d-100} = (C/100)\Sigma\, V_d\left(D_{(i-0.5)\%}\right), \tag{5}$$

where $D_{(i-0.5)\%}$ is the diameter at the center of a size interval i and C is the mass of dust or other aerosol substances of interest.

[26] Here we used the two-layer model of *Slinn and Slinn* [1980] to calculate the dry deposition velocities (DDVs) for each of the particle size bins. In the computation, the data for wind speed (10 m s^{-1}) and relative humidity (85%), as provided by the CWB of Taiwan, were based on long-term observations at an offshore island (Pengchiayu) in the southern ECS, while the other parameters applied in this study were similar to those adopted by *Dulac et al.* [1989].

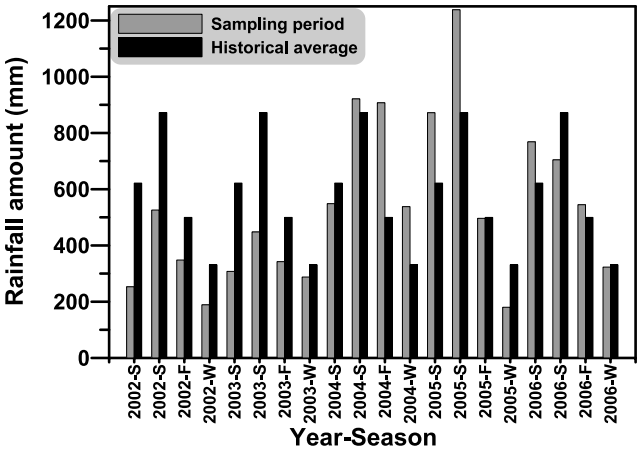

Figure 4. Seasonal precipitation (mm) during the observational period (2002 and 2006) in Taipei, Taiwan. Data were taken from the Central Weather of Bureau of Taiwan.

[27] The integrated dry deposition velocities averaged over the method-derived 100-bin sizes for the nine samples with MSDs fell into the range of 0.7 to 5.4 cm s^{-1} and averaged 2.0 ± 1.5 cm s^{-1} (n = 9). In comparison, the DDVs corresponding to the MMDs of the samples were only ~0.2 cm s^{-1}, that is, much lower than what was obtained from the 100-step method. The fact that the DDV obtained by considering the entire MSD was much larger than that for the MMD shows that giant particles can contribute a major portion of an aerosol substances' dry deposition.

[28] Comparisons of the DDV between our results and previous studies conducted in various other areas are summarized in Table 2. Our value was quite consistent with that (average 1.9 cm s^{-1} and range 0.9 ~ 4.0 cm s^{-1}) in the northern ECS as determined by *Gao et al.* [1997], although their investigation was very limited in both sample numbers and time. In addition, our value also agreed well with the common value of 2 cm s^{-1} for coastal or marginal seas as recommended by *Duce et al.* [1991]. This result is consistent with the results of the MMD comparisons presented above, and it suggests a similarity between the southern and northern parts of the ECS. Therefore, it likely implies again that on a yearly basis, the whole ECS is under a rather uniform influence of airborne dust and dust-derived constituents, and therefore the atmospheric loadings and MSDs of dust do not vary as much as in areas closer to the sources.

4.3.2. Wet Deposition

[29] With reference to the wet deposition flux of atmospheric dust, we estimated it indirectly by using the following equation [*Duce et al.*, 1991]:

$$F_w = P \times S \times C_a/\rho, \tag{6}$$

where P is the precipitation rate (mm a^{-1}), S is the scavenging ratio (dimensionless) for mineral dust, C_a is the concentration of dust in the air (μg m^{-3}), and ρ is the density of air (~1200 g m^{-3}). The scavenging ratio, which is defined as the concentration ratio of certain species in rain to in air, is a function of numerous factors including meteorological conditions (e.g., cloud type, phase contact time, precipitation intensity, etc.), microphysical conditions (e.g., size and hygroscopy of particles, number of condensation nuclei, etc.; see *Slinn* [1983]), and solubility of elements in rainwater [*Chester et al.*, 1997]. A range of scavenging ratio between 500 and 2000 was suggested to be an appropriate value for dust aerosol in coastal and marginal seas [*Arimoto et al.*, 1985; *Uematsu et al.*, 1985; *Group of Experts on Scientific Aspects of Marine Pollution* (*GESAMP*), 1989; *Duce et al.*, 1991; *Gao et al.*, 1997]; here the value for S we used for computing the wet deposition of eolian dust is 1000, and the resulting uncertainty would be a factor of 2.

[30] Seasonal rainfall as recorded during the sampling period by the CWB of Taiwan (http://www.cwb.gov.tw/V4/index.htm) was used for the wet deposition calculations. However, comparing the observed rainfall in the sampling period with the historical average (i.e., 1971 to 2000) (Figure 4), it was clear that in 2002 and 2003, when draft conditions existed, precipitation was reduced and fewer rainy days occurred (not shown). Although comparing the two cases (observed versus climatological precipitation) would be a means of better understanding the seasonality of dust deposition, the resulting differences in the interannual mean wet deposition fluxes had relatively small uncertainties (<10%). Therefore, we reported the wet deposition only from the observed rainfall data. The frequent and abundant rainfall in the study area implies that wet removal is likely to be especially important for dust deposition, and its importance for total deposition is discussed in detail below.

4.3.3. Combined Fluxes

[31] The dry and wet deposition fluxes of eolian Al were calculated season by season from equations (5) and (6) using the Al concentrations shown in Figure 2. Assuming that the Al abundance of the airborne mineral dust is similar to that of the Chinese desert dust and loess, that is, 7% by weight [*Zhang et al.*, 2003], we converted Al fluxes to mineral dust fluxes by dividing the former by 0.07, as shown in Figure 5. Even though the characteristics of the dust (e.g., chemical composition, grain size, and mineral assemblages, etc.) would be altered during transport, these influences are likely to be minor and not considered further here. Indeed, taking into account all the main factors together that lead to uncertainties in the estimated deposition flux (e.g., dry deposition velocity, scavenging factor, and precipitation), the errors in our estimates of airborne dust fluxes should be roughly a factor of 3 or less [*GESAMP*, 1989].

[32] Dry deposition ranged from 0.54 to 3.38 g m^{-2} per season while wet deposition was higher, from 0.99 to 12.6 g m^{-2} per season. The annual mean total deposition was 21.4 g m^{-2} a^{-1} (Figure 5). Particles larger than 10 μm in diameter accounted for ~25% of the dry deposition of dust and because our estimation of dust deposition was based on Al data for PM_{10} samples, the derived value is probably underestimated somewhat. In this regard, it is worth pointing out (1) that an unknown fraction of the dust is from local sources but one would expect the greatest impact to be on the largest particles, (2) there is no accepted way of distinguishing particles from local versus distant sources, and (3) that the effect is probably not large given all of the other assumptions in the deposition calculations. In fact, the estimates of annual dust deposition in the study area were comparable to those (~3.6 g m^{-2} mo^{-1})

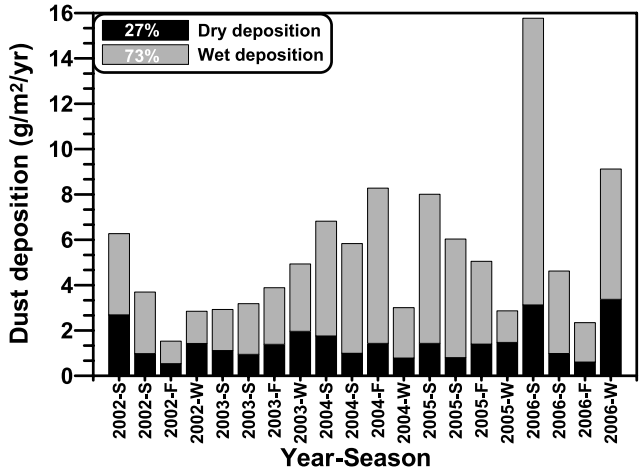

Figure 5. Seasonal variations in atmospheric deposition fluxes (g m^{-2} a^{-1}) of eolian dust through dry and wet deposition to northern Taiwan between 2002 and 2006. The percentages of dry and wet deposition accounting for the total deposition are shown.

measured during a 5-month high-dust period at a coastal site in southeastern China (Xiamen), which is approximately 300 km southwest of our site [*Gao et al.*, 1997]. In addition, the calculated dust deposition in our study also was consistent with that estimated for coastal areas of China (21 g m^{-2} a^{-1} [*Uematsu et al.*, 2003]).

[33] The seasonal pattern of eolian dust fluxes (Figure 5) generally mirrored the concentrations of atmospheric Al (Figure 2), that is, maxima in spring and minima in summer or fall. However, the seasonality in dust deposition was not so evident in 2003 because during that year, only two moderate AD events occurred, and rainfall and hence wet deposition was low. In sum, the quantities and temporal variability of the dust fluxes are a function of precipitation patterns as well as the strength and frequency of AD events.

4.4. Comparison of the Eolian Dust Fluxes With Those in Nearby Regions

[34] As the observed Al concentrations and calculated dust deposition to northern Taiwan were generally comparable to those previously measured over the ECS, we suggest that the distribution of AD over the ECS around 25°N–30°N is relatively homogeneous, at least on an annual scale. This working hypothesis enables us to apply our estimates of dust deposition for northern Taiwan to the southern ESC and then to evaluate the biogeochemical significance of the deposition process. In addition, for further comparisons with our results, we compiled literature data on AD deposition to diverse areas from source regions to areas downwind of the ECS, which could be representative of the southern ECS.

[35] Literature data on atmospheric dust fluxes to the Chinese marginal seas and the North Pacific are given in Table 3. On the basis of a limited data set, *Gao et al.* [1992] obtained for the first time a rough estimate of the total dust deposition to three marginal seas of China, showing a slight decreasing tendency southward from the Bohai Sea (30 to 196 g m^{-2} a^{-1}) to the Yellow Sea (9 to 79 g m^{-2} a^{-1}), and to the ECS (7 to 113 g m^{-2} a^{-1}). A few years later, *Gao et al.* [1997] revised the estimation of eolian dust input fluxes into the northern ECS by obtaining dry and wet deposition fluxes of 13 g m^{-2} a^{-1} and 14 g m^{-2} a^{-1}, respectively. *J. Zhang et al.* [1993] measured the fallout fluxes into the Yellow Sea through dry and wet deposition and reported values of 37 and 17 g m^{-2} a^{-1}, respectively.

[36] Combining these published dust fluxes with our results, we find a pattern of decreasing dry deposition with latitude, from the Yellow Sea (37 g m^{-2} a^{-1}) to the northern ECS (13 g m^{-2} a^{-1}) and then to the southern ECS (5.8 g m^{-2} a^{-1}). In contrast, the wet deposition fluxes were nearly identical (~15 g m^{-2} a^{-1}) among these Chinese marginal seas. Such a geographic pattern is a coupled function of the spatial distributions of dust loadings and precipitation.

[37] With reference to precipitation, although the amount of rainfall is a critical parameter in determining the wet deposition of aerosols, the rain frequency, that is, the number of rainy days in a given period, is likely to be another key factor in determining deposition rates. On the basis of the historical data set of the CWB, there are at least 15 rainy days over the offshore sea north of Taiwan every month during the NE monsoon, and the total precipitation during that part of the year is ~1000 mm. In comparison,

Table 3. Comparisons of Total, Dry, and Wet Deposition Fluxes of Eolian Mineral Particles Into the Southern ECS With Those Measured in the Chinese Marginal Seas and the North Pacific Ocean

Study Area	Deposition Fluxes (g m^{-2} a^{-1})			Reference
	Total	Dry	Wet	
Southern East China Sea	21.4	5.8	15.6	this work
Northern East China Sea	27	13	14	*Gao et al.* [1997]
Yellow Sea	53.7	36.8	16.9	*J. Zhang et al.* [1993]
Qingdao (Yellow Sea)	51.3	...	...	*Kai and Gao* [2007]
Northern East China Sea	7.5–113	...	...	*Gao et al.* [1992]
Yellow Sea	8.6–79	...	...	*Gao et al.* [1992]
Bohai Sea	30–196	...	...	*Gao et al.* [1992]
Japan Sea	5–43	...	...	*Irino and Tada* [2000, and references therein]
North Pacific Ocean	5.3	1.5	3.8	*GESAMP* [1989]; *Duce et al.* [1991]
Remote islands over the Pacific	0.12–0.64	...	...	*Prospero et al.* [1989]
Tropical North Pacific Ocean	0.15	0.02	0.13	*Arimoto et al.* [1985]
Chinese coastal sea	21[a]	~12.6	~8.4	*Uematsu et al.* [2003]
Global ocean	2.5[a]	1.0[a]	1.5[a]	*Duce et al.* [1991]

[a]Modeling result.

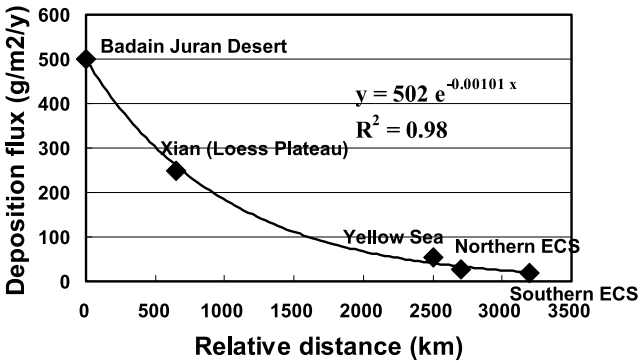

Figure 6. Variation of dust deposition rates (g m^{-2} a^{-1}) with distance from the Badain-Juran Desert, a representative dust region [*Zhang et al.*, 1998], to the downwind areas including Xi'an (located in Loess Plateau; see *X. Y. Zhang et al.* [1993]), the Yellow Sea [*Gao et al.*, 1997], the northern East China Sea [*Gao et al.*, 1997], and the southern East China Sea (this study). The results of regression fitting showed an exponential decrease, with $R^2 = 0.98$.

during the SUM monsoon there is 1300 mm of precipitation and 12 rainy days every month. This abundant rainfall can explain our finding that wet deposition exceeds dry deposition, and it would support the idea that much of the AD in the air transported southward would be removed by rainfall along with dry deposition. However, comparing our results with those compiled for Chinese marginal seas and from regional models, the modeled dust deposition over the Chinese marginal seas (Bohai, Yellow and East China Seas) appears to be underestimated by a factor of 2–4 [*Uematsu et al.*, 2003] to as much as an order of magnitude [*Zhao et al.*, 2003, 2006]. This highlights the fact that more measurements of dust deposition are needed to better constrain the dust fluxes.

[38] Combining estimates of dust deposition over source regions, as represented by the Badain-Juran Desert [*Zhang et al.*, 1998] with those for the downwind marginal seas, including our estimate for the southern ECS, we find an exponentially decreasing tendency with distance from the source region (Figure 6). From this decreasing trend, the half-life distance, which is defined as the distance in km required for the AD flux to decrease by a factor of 2 is estimated to be ~700 km. This is consistent with a value of 500 to 600 km, which was derived from the relationship of atmospheric Al concentrations (not deposition) at several remote islands in the northwestern Pacific by *Tsunogai et al.* [1985]. This relationship is very useful in that it allows one to estimate the dust deposition at a given downwind location. When the equation given in Figure 6 is used to predict the eolian dust inputs to several remote islands in the North Pacific (e.g., Midway, Oahu, Enewetak, and Fanning Island), one obtains a value of approximately 0.5 g m^{-2} a^{-1}, and this is very comparable to what has been measured at those sites [*Prospero et al.*, 1989].

4.5. Relative Contributions of Dry and Wet Deposition

[39] On average, dry and wet deposition account for 27% and 73% of the total dust deposition, respectively. These proportions are very consistent with those (31% versus 69%) measured at the coastal site of Xiamen (24°N, 118°E), which is located southeastern China and is similarly characterized by ample rainfall [*Gao et al.*, 1997]. The proportion of wet deposition was very slightly lower than what has been measured at the remote islands of Midway and Oahu in the North Pacific (~80%; see *Uematsu et al.* [1985]).

[40] On the basis of a variety of techniques including numerical modeling, direct observations, and indirect calculations by using atmospheric parameters, various authors have presented their findings on the relative contributions of dry and wet deposition of eolian dust or dust-related species (e.g., Fe). For example, *Gao et al.* [2003] suggested that according to numerical modeling results, the wet deposition of airborne Fe contributes a mean proportion of ~40% over the coastal oceans and ~60% over the open oceans. Therefore, the results we obtained here appear to be more similar to what they found for the open ocean than for the coastal ocean. However, *Uematsu et al.* [2003] concluded that dry deposition into the western North Pacific Ocean was slightly more important than wet deposition, accounting for 60% of the total. In summary, the proportion of dust wet deposition seems to increase seaward and southward, but again the data are limited.

[41] While our results showed that more dust was removed by wet than dry deposition even during the springtime dust season (Figure 5), this may be explained, at least in part, by the differences in latitude, and in turn, the synoptic meteorological conditions for the various studies. Along these lines, *Zhao et al.* [2003] has concluded that although dry deposition is the dominant dust removal process close to dust source regions, the scavenging of dust particles via precipitation is the principal removal process over the Pacific, where wet deposition greatly exceeds dry deposition (the latter accounting for less than 10% of the total dust flux).

4.6. Implications for Biogeochemistry in the East China Sea

4.6.1. Significance of Dust Depositions on the Local Marine Sediment Budget

[42] The spatial distribution of sediments on the shelf of the ECS is characterized by high sedimentation in the narrow inner shelf (particularly in the area near the Changjiang estuary) and little or no contemporary sedimentation on the outer shelf, the latter being almost exclusively composed of relict sediments from the Holocene. Although the Changjiang (Yangtze River) delivers massive quantities of sediments into the ECS (approximately 500 Mt a^{-1}), only a minor portion (10% or less) of that material is dispersed away from the Chinese coast [*Liu et al.*, 2007].

[43] Apart from the inner shelf, the Okinawa Trough (OT), which is on the eastern margin of the ECS, is the most important receptacle for terrigenous sediments flowing out from the Chinese coast [*Huh et al.*, 2006]. This deep basin has a mean sedimentation rate of 0.01 to 0.05 cm a^{-1}, but at its southern extreme sedimentation is higher, $\geq$0.1 cm a^{-1} [*Su and Huh*, 2002; *Huh et al.*, 2006]. The corresponding mean bulk sediment mass accumulation rate is 100 to 500 g m^{-2} a^{-1} [*Huh and Su*, 1999]. As the lithogenic component composes approximately 80% of the bulk sediments [*Hsu et al.*, 2004b], the accumulation

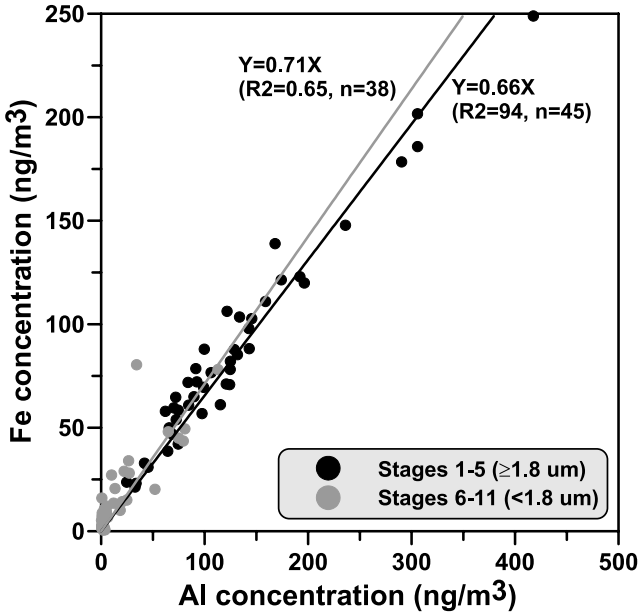

Figure 7. The correlation of atmospheric Al and Fe in size-resolved aerosol samples. The data were treated as two groups: coarse (size $\geq$ 1.8 μm, indicated by solid circles) and fine (size < 1.8 μm, indicated by gray circles) modes. Also shown are the regression lines through the origins (solid and gray lines for coarse and fine mode samples, respectively) as well as the regression equations and R^2 values.

rate of terrigenous (aluminosilicate) sediments is 80 to 400 g m^{-2} a^{-1}.

[44] For comparison, the average deposition of AD we estimate for the adjacent oceans (24 g m^{-2} a^{-1}) is equal to ~6 to 30% of the sedimentation of terrigenous material in the OT. Interestingly, this proportion agrees with that (10–20%) estimated for the Mediterranean Sea with dust transported from the Sahara Desert [*Guerzoni et al.*, 1997]. Further, we estimate the total dust deposition into the entire ECS, that is, geographically covering the ECS shelf and slope and the Okinawa Trough located at the shoreward side of the Ryukyu Islands, to be ~18 Mt a^{-1}. Compared with the terrigenous sediment budget (i.e., ~80 Mt a^{-1}) the dust input to the ECS may be seen to account for ~20% of the OT terrigenous sediment budget.

[45] In sum, eolian dust evidently serves as an important contributor to the OT sediments; this contention has been strongly supported by recent observations on settling particles captured sediment traps [*Katayama and Watanabe*, 2003; *Otosaka et al.*, 2004; *Li et al.*, 2004; *Yuan and Zhang*, 2006]. In addition, the dust and elemental fluxes derived here may be viewed as a reference for contemporary eolian dust deposition and so they may be useful for assessing the changes in glacial/interglacial climate that have been recorded in sediment cores [*Rea*, 1994; *Duce*, 1995; *Lunt and Valdes*, 2002].

4.6.2. Eolian Fe Deposition

[46] We estimated the Fe deposition to the ECS by multiplying the calculated Al deposition rates by a representative aerosol Fe/Al ratio. As shown in Figure 7, the Fe/Al ratios in size-segregated aerosols varied slightly as a function of particle size. The average observed Fe/Al mass ratios were 0.66 and 0.77 for the particles whose aerodynamic diameters were $\geq$1.8 μm, that is, the five larger size bins and <1.8μm, that is, the six smaller size bins, respectively. As the difference in Fe/Al ratios was not large, we used a concentration-weighted mean mass ratio of ~0.68 for calculating the atmospheric deposition flux of total Fe. This ratio agrees with the results of AD measurements in dust regions over China [*Zhang et al.*, 1997, 1998] and at locations downwind [*Arimoto et al.*, 2004; *Sun et al.*, 2005].

[47] The total Fe deposition flux calculated in this manner was 1.14 g m^{-2} a^{-1} (20 mmol m^{-2} a^{-1}). We further estimated the deposition of dissolvable Fe (DFe) because only Fe in dissolved form is bioavailable even though some recent studies have suggested that particulate/colloidal Fe sometimes can be utilized by marine organisms [e.g., *Nodwell and Price*, 2001; *Yoshida et al.*, 2006]. The dissolvable (also termed soluble) portions of aerosol Fe in seawater have been found to be highly variable, ranging from <1% to $\gg$10%, depending upon various factors [*Hand et al.*, 2004; *Chen et al.*, 2006; *Hsu et al.*, 2005b]. Given this uncertainty, seawater solubilites for aerosol Fe ranging from 1 to 10% were used in our study following the approach of *Baker and Croot* [2009]. The resulting DFe deposition was 11 to 114 mg m^{-2} a^{-1} (0.2 to 2 mmol m^{-2} a^{-1}), and the total DFe deposition to the whole ECS was 0.01 to 0.1 Mt a^{-1}. These values may be compared with the estimates (0.14 to 0.96 Mt a^{-1}) for the entire North Pacific [*Gao et al.*, 2003], recognizing that the area of the ECS is equal to ~10% of that of the North Pacific.

[48] Two large Chinese rivers, the Changjiang and Yellow River, deliver massive quantities of DFe into the ECS, and this undoubtedly dominates the biogeochemical cycling and budget of DFe. Nonetheless, a DFe budget for this region has not been constructed to date. Thus, we compared the atmospheric Fe input with the fluvial Fe input to the ECS. By adopting a DFe concentration range of 72 to 550 nM for the Changjiang [*Zhang*, 1995, and references therein; *Koshikawa et al.*, 2007] and a mean concentration of 240 nM for the Yellow River [*Zhang*, 1995, and references therein] the respective discharge rates of 0.004 to 0.03 Mt a^{-1} and <0.001 Mt a^{-1} were derived for the two rivers. Furthermore, if one assumes that 50% of the riverine DFe is scavenged during estuarine mixing [*Wang and Liu*, 2003], the two rivers would supply ~0.02 Mt of DFe each year to the ECS.

[49] A comparison of the inputs (i.e., 0.01 to 0.1 Mt a^{-1} for atmospheric deposition, versus ~0.02 Mt a^{-1} for the riverine input) implies that eolian DFe is a significant source of DFe for the ECS, especially the offshore areas. Moreover, *Obata et al.* [1997] has published a profile of seawater Fe in dissolved and leachable forms measured at an offshore site (28°00′, 127°30′) in the ECS, which showed a surface enrichment and high concentrations at depths below 500 m. This explicitly implicates that atmospheric inputs are the chief source of the surface DFe and sediments (or sediment resuspension) are a main source of DFe in the deep waters [*Obata et al.*, 1997]. Nevertheless, because the DFe concentrations at depths between 100 and 300 m are constantly low, the upward benthic contribution to the DFe-rich surface appears to be small; this is consistent with the low upwell-

Table 4. Estimates of Residence Times for Dissolved Al and Fe in the Upper 100 m of the East China Sea[a]

	Atmospheric Input (mmol m^{-2} a^{-1})	Upwelling Flux (mmol m^{-2} a^{-1})	Concentrations (nmol)	Inventory (mmol m^{-2})	Residence Time (years)
DFe	0.20–2.04	0.015	0.63–7.3[b]	0.20	0.10–1.1
DAl	0.62–6.22	0.13	9–24.5[c]	1.45	0.24–3.0

[a]Also given here are the estimated atmospheric inputs, the estimated upwelling fluxes, the reported water concentrations, and the calculated inventories for the two species.
[b]From *Obata et al.* [1997].
[c]From *Minakawa and Watanabe* [1998].

ing flux of DFe from just below the surface mixed layer (discussed below). However, we cannot accurately assess the relative contribution of sedimentary sources to the budget of DFe throughout the ECS water column because of the limited data on seawater DFe.

4.6.3. Implications for the Biogeochemical Cycling of Dissolved Al and Fe in the Mixed Layer and Biological Stimulation

[50] In order to make the first-order approximation of the biogeochemical effects of dust and dust-bound Fe deposition to the ECS, these deposition rates are considered in the context of the biogeochemical cycling of dissolved Al (DAl) and DFe and potential biological stimulation in the study area. Relevant published data are quite limited, but according to *Obata et al.* [1997] and *Minakawa and Watanabe* [1998] both DFe and DAl are enriched in the surface mixed layer of the offshore area, and the upper 100 m of the water column is strongly influenced by atmospheric input. Following *Jickells* [1999], we estimated the respective upwelling fluxes of DAl and DFe from the depth between 100 and 200 m to be 0.13 and 0.015 mM m^{-2} a^{-1}, in which the vertical velocity (w) of 0.04 m d^{-1}, similar to the maximum used by *Jickells* [1999], and the vertical eddy diffusivity (K_z) of 10^{-5} m^2 s^{-1} [*Matsuno et al.*, 2005] were used.

[51] When compared with atmospheric inputs of 0.62–6.22 mM m^{-2} a^{-1} for DAl and 0.20–2.04 mM m^{-2} a^{-1} for DFe, in which the seawater solubility of Al and Fe in eolian dust has been assumed similarly to be 1–10% [*Baker and Croot*, 2009; *Measures et al.*, 2009], the upwelling sources could be either significant (i.e., <10% for DFe, and ~20% for DAl) or insignificant (i.e., <2% for both), depending upon the assumed seawater solubility. Nevertheless, atmospheric inputs seem to be an important source for DAl and DFe in the mixed layer [*Obata et al.*, 1997; *Minakawa and Watanabe*, 1998]. Furthermore, we estimated the residence times of DAl and DFe in the upper 100 m mixed layer to be 0.24 to 3.0 and 0.10 to 1.1 years, respectively, in which the relevant parameters used are given in Table 4. Note that the uncertainties in the estimates of upwelling fluxes and residence times for DAl and DFe are large; this is because relevant information for key parameters used in the calculations is scarce.

[52] It is impossible at this point to determine whether the DFe from eolian Fe deposition would be bioavailable. That would depend on the inorganic and organic speciation of Fe, redox chemistry, size fractionation of the Fe species (colloids) on the one hand, and the Fe demand and which form of Fe is preferred by the phytoplankton community on the other hand. In this respect, the question arises whether autotrophs are Fe limited in the ECS. The concentrations of 1.5 nM in the upper 100 m [*Obata et al.*, 1997] are not that low, but Fe could become limiting during phytoplankton blooms especially if the blooming species had a high Fe demand.

[53] Elevated biogenic opal fluxes associated with sharp increases in Al flux were observed in the Japan Sea during the spring of 2002 in studies using sediment traps [*Otosaka et al.*, 2004], and the increased Al fluxes in the settling particles captured in those studies were attributed to atmospheric deposition of Asian dust. In the work of *Hsu et al.* [2008], frequent strong Asian dust events were in fact observed in spring 2002. Interestingly, the concurrence between high Al fluxes and biogenic fluxes (e.g., opal and organic matter) have also been observed by other sediment traps deployed in around the ECS [*Katayama and Watanabe*, 2003; *Li et al.*, 2004; *Yuan and Zhang*, 2006]. These investigators have suggested that the supply of dust-associated bioavailable Fe and other nutrients was the main reason for the enhanced fluxes of biogenic materials. Apparently, primary production and/or nitrogen fixation can be stimulated by eolian dust deposition especially in offshore areas of the ECS where the influence of riverine inputs is small. On the basis of the results of novel genetic techniques, C. C. Chung et al. (The influences of Asian dust storms on cyanobacterial picoplankton succession and nutrient status in subtropical Kuroshio Current, submitted to *Microbial Ecology*, 2009) recently showed that the activity of *Synechococcus* spp. in the ECS was stimulated during an Asian dust storm; this was taken as evidence that DFe can indeed be a limiting nutrient.

5. Summary

[54] Data for Al in size-resolved aerosol samples from northern Taiwan in 2003 were combined with information on temporal variations of aerosol Al from our companion paper [*Hsu et al.*, 2008] to estimate wet and dry dust deposition rates. To evaluate the significance of the derived dust flux for marine biogeochemistry we assumed it to be representative of the ECS south of 30° N. The major conclusions of the study are as follows:

[55] 1. The mass particle-size distribution of atmospheric Al was monomodal, peaking at 2.5 to 5.6 μm, with MMDs for the dust particles averaging 3.6 ± 1.2 μm. The dry deposition velocity integrated over the entire MSD was 2.0 ± 1.5 cm s^{-1}; this is the same value that is commonly applied in studies of coastal and marine regions.

[56] 2. Total dust deposition rates calculated from Al were estimated to be ~20 g m^{-2} a^{-1}, with dry and wet deposition accounting for approximately one-fourth and three-fourths of the deposition, respectively. The frequency and strength

of AD storms and rain events are the major determinants of the quantity of dust deposited.

[57] 3. The deposition of AD decreases approximately exponentially with distance from the source, here represented by the Badain-Juran Desert. Interestingly, the wet deposition rates across the Yellow Sea, the northern ECS, and the southern ECS (from north to south) are almost identical; this shows that the relative significance of wet deposition gradually increases southward owing to the more abundant precipitation in the south. In contrast, both dry and total dust deposition gradually decrease southward owing to the decreasing atmospheric dust loads.

[58] 4. On the basis of the similarity of dust loadings over the Taiwan/ECS region, we applied our estimate of dust deposition to northern Taiwan to the entire ECS to assess its biogeochemical significance. Dust input into the ECS thus estimated was $\sim$18 Mt a^{-1}, which is $\sim$20% of the terrigenous sediment load delivered to the OT. Therefore, AD appears to be a significant source of sediments even for a marginal sea basin that receives massive amounts of fluvial sediment.

[59] 5. From the Al deposition, we estimated the total Fe and DFe and an assumed seawater solubility of 1 to 10% for aerosol Fe. The resulting deposition rates of total eolian Fe and DFe were 1.14 g m^{-2} a^{-1} and 11 to 114 mg m^{-2} a^{-1}, respectively. Even when compared with the discharge rates of DFe from the Changjiang and the Yellow River, the results suggest that eolian dust may be a vital source of DFe in the ECS; this has largely been ignored in marginal seas and coastal waters.

[60] 6. When compared to dust deposition rates on the basis of regional atmospheric chemistry models, our estimates and those values reported for the adjacent seas (i.e., $\sim$20 g m^{-2} a^{-1}) appear to be somewhat higher; this merits further evaluation. Our study also highlights the importance of the southeastward transport of AD south of 30N, which is neglected in general.

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