Supplementary information

Text S1

Simulation of Asian dust

When dust travels over the Asian continent through the atmosphere, it can experience mixing and deposition, as well as undergo chemical reactions (Formenti et al., 2011). The Asian continent suffers from air pollution to varying extents, with dynamically changing emissions of anthropogenic pollutants such as NO_x, SO₂, and NH₃ (Kim et al., 2014). The aging processes, i.e., the reactions of dust aerosols with anthropogenic pollutants, result in the Asian dust carrying a large amount of nutrients and bioavailable trace metals, prior to its deposition in the oceans. In this study, we followed the method of Guieu (2010) to mix the dust and cloud water (100 g L⁻¹) to simulate the aging process. The chemical compositions of the reference eastern Asian rains and cloud water used in this study are summarized in Table S1. As the uptake of organic acidic gases during transport is complicated for Asian dust, we did not add oxalic acid, which was used for simulating the Saharan dust by Guieu et al. (2010), to simplify the reaction of dust surface and emphasize the importance of inorganic acids (H₂SO₄ and HNO₃) (Fan et

15 al., 2006; Formenti et al., 2011; Shi et al., 2012).

References

Fan, S. M., Moxim, W. J., and Levy, H.: Aeolian input of bioavailable iron to the ocean. Geophysical research letters, 33, 359-377, doi: 10.1029/2005GL024852, 2006.

Formenti, P., Schütz, L., Balkanski, Y., Desboeufs, K., Ebert, M., Kandler, A. Petzold, D. Scheuvens,

20 S. Weinbruch, and Zhang, D.: Recent progress in understanding physical and chemical properties of African and Asian mineral dust. Atmospheric Chemistry and Physics, 11, 8231-8256, doi: 10.5194/acp-11-8231-2011, 2011.

Kim, I.-N., Lee, K., Gruber, N., Karl, D. M., Bullister, J. L., Yang, S., and Kim, T.-W.: Increasing anthropogenic nitrogen in the North Pacific Ocean, Science, 346, 1102-1106, doi:

25 10.1126/science.1258396, 2014.

Meskhidze, N., Chameides, W. L., Nenes, A., and Chen, G.: Iron mobilization in mineral dust: Can anthropogenic SO2 emissions affect ocean productivity?. Geophysical Research Letters, 30, 267-283, doi: 10.1029/2003GL018035, 2003.

Sakihama, H., Ishiki, M., and Tokuyama, A.: Chemical characteristics of precipitation in Okinawa Island,

- Japan. Atmospheric Environment, 42, 2320-2335, doi: 10.1016/j.atmosenv.2007.12.026, 2008.
 Sasakawa, M., and Uematsu, M.: Chemical composition of aerosol, sea fog, and rainwater in the marine boundary layer of the northwestern North Pacific and its marginal seas. Journal of Geophysical Research: Atmospheres, 107(24): ACH 17-1–ACH 17-9, doi: 10.1029/2001JD001004, 2002.
 Shi, J., Gao, H., Zhang, J., Tan, S., Ren, J., Liu, C., Liu, Y., and Yao, X.: Examination of causative link
- between a spring bloom and dry/wet deposition of Asian dust in the Yellow Sea, China, Journal of Geophysical Research: Atmospheres, 117, D17304, doi:10.1029/2012JD017983, 2012.
 Wang, Z., Akimoto, H., and Uno, I.: Neutralization of soil aerosol and its impact on the distribution of acid rain over east Asia: Observations and model results. Journal of Geophysical Research: Atmospheres, 107(D19): ACH 6-1–ACH 6-12, doi: 10.1029/2001JD001040, 2002.
- 40 Watanabe, K., Ishizaka, Y., and Takenaka, C.: Chemical characteristics of cloud water over the Japan Sea and the Northwestern Pacific Ocean near the central part of Japan: airborne measurements. Atmospheric Environment, 35, 645-655, doi: 10.1016/S1352-2310(00)00358-7, 2001.

45

Zhang, J., Zhang, G. S., Bi, Y. F., and Liu, S. M.: Nitrogen species in rainwater and aerosols of the Yellow and East China seas: Effects of the East Asian monsoon and anthropogenic emissions and relevance for the NW Pacific Ocean. Global Biogeochemical Cycles, 25, 113-120, doi: 10.1029/2010GB003896, 2011.

	nU	NO ₃ -	SO4 ²⁻
	pm	(M)	(M)
Reference eastern Asian rains*	3.89–7.61	10 ⁻⁵	10-5
Simulated cloud water	1**	10-1	10-1

Table S1. Primary chemical composition of the rains in the eastern Asian region and the simulated eastern Asian cloud water.

*Sasakawa and Uematsu, 2002; Watanabe et al. 2001; Zhang et al. 2011; Sakihama et al. 2008; Wang et al. 2002.

** Meskhidze et al., 2003.

Metal	Detection limit ($\mu g L^{-1}$)*	Recovery (%)	RSD (%)**
Zn	0.012	90.6	3.17
Cu	0.226	95.2	2.09
Cd	0.016	88.5	0.87
Pb	0.019	93.2	2.93
Co	0.017	97.9	0.24
Fe	3.738	95.4	3.88
Mn	0.056	90.9	4.48

Table S2. Recovery yield, accuracy, and detection limit for trace metal analysis

* Detection limit was calculated as three times the standard deviation of the blank.

** RSD means 'Relative Standard Deviation'.



Figure S1. Changes in Chl *a* during the incubation experiments at each station. The successive increase during the incubation period in this study is identified by the dotted line.



Figure S2. The relationship between the consumed N:P ratio $(C_{N:P})$ and supply N:P ratio $(S_{N:P})$ in the control and the various nutrient treatments during the successive increase in the incubation period at each station