Auxiliary Material for

Authors – Sholkovitz E. R., Sedwick P. N., Church T. M., Baker A. R. and Powell C.F.

Title - Fractional Solubility of Aerosol Iron: Synthesis of a Global-Scale Data Set

Tables S1 and S2 contain the compilation of data cited, discussed and plotted in this paper.

Table S1 and Figure 1

Table S1 is a compilation of *paired data* (total aerosol Fe load and fractional solubility of aerosol Fe) for the sites located on the maps in Figures 1 and 2 in the body of the main paper. Table S1 also includes information on collection dates, locations and sample types, as well as the protocols used to define 'soluble' aerosol iron ( ) in each of the twenty-six studies in Figure 1. Table S1 also contains two large sets of unpublished data, for samples collected from: (1) Bermuda and the adjacent Sargasso Sea during 2007 and 2008 (93 samples; T. Church, P. Sedwick and E. Sholkovitz, unpublished data); and (2) the North and South Atlantic Ocean during 2003-2008 (291 samples; A. Baker and C. Powell, unpublished data).

The data in Table S1 come from twenty-four published studies and two sets of previously unpublished data, noted above. The data are compiled in the form of Microsoft EXCEL files. References cited below are numbered to correspond to the numbers on the location map in Figure 1. The letters after the location numbers refer to the type of leaching methods used in each study – B for batch method, F_T for flow-through method and B/F_T for a combination batch and flow-through method. The second group of letters refer to type of leaching solutions used; they include Milli-Q (MQ) water, distilled water

(DI), formate pH 4.5 buffer (formate), ammonium acetate pH 4.7 buffer (acetate) and seawater (SW at pH ~ 8). The next and last set of numbers refers to the number of data point in each reference. For example, "**1-B-MQ-32**" means that site #1 has batch leach data for Milli-Q water for 32 samples.

[1-B-MQ-32] Kumur et al., 2010; [**2-B-MQ-49**] Kumar and Sarin, 2010; [**3-B-formate-26**] Johansen and Hoffmann, 2003; [**4-B-formate-16**] Siefert et al. 1999; [5**-B-MQ-31 and SW-19**] Chen et al. 2006; [**6-B-SW-28**] Hsu et al., 2005; [**7-B-MQ-40**] Hsu et al., 2009; [**8-B-MQ-26**] Chuang et al., 2005; [**9-B/F_T-MQ-8 and SW-9**] Aguilar-Islas et al, 2010; [**10-B/F_T-SW-15**] Wu et al. 2007; [**11-F_T-DI-54 and SW-54]** Buck et al., 2006. [**12-B-formate-59**] Chen, 2004; [**13-B-dilute HCl-18**] Zhuang et al. 1992; [**14-B-formate-23**] Siefert et al. 1996; [**15-F_T-DI-43 and SW-41**] Buck et al, 2010a; [**16-B-acetate-65**] Baker et al, 2006 a; [**17-B-acetate-36**] Baker et al. 2006 b. [**18-B-formate-29**] Chen and Siefert, 2004; [**19-B-formate-17**] Johansen et al., 2000; [**20-B-pH 2 NaCl-25**] Trapp et al., 2010; [**21-F_T-DI-18**] Sedwick et al. 2007**;** [**22-F_T-DI-93**] Church, Sedwick and Sholkovitz, unpublished data; [**23-B-pH 1 NaCl-25**] Zhu et a., 1997; [**24-B-pH 1 -12]** Witt et al., 2006; [25**-B-acetate-7 and pH 1 -7]** Witt et al. 2010; [**26-F_T-SW**-7] Bowie et al., 2009.

Full references for the studies cited in Table S1 and Figure 1 are as follows:
Reference number on map plus citation and location/date of sampling sites are included.

[1] Kumur A., Sarin, M. M., and Srinivas B. (2010) Aerosol iron solubility over Bay Bengal: Role of anthropogenic sources and chemical processing. *Mar. Chem*. 121, 167-175, doi:10.1016/j.marchem.2010.04.005.

Bay of Bengal, 27 Dec. 2008 to 28 Jan. 2009

[2] Kumar A., and Sarin M.M. (2010) Aerosol iron solubility in a semi-arid region: temporal trend and impact of anthropogenic sources. *Tellus*, 62B. 125-132, doi:10.1111/j.1600-0889.2009.x

Mount Abu, India (1680 m ASL, 22.7 N & 74.6 E), Jan. – Dec. 2007

[3] Johansen A. M., and Hoffmann M. R. (2003) Chemical characterization of ambient aerosol collected during the northeast monsoon season over the Arabian Sea: Labile-Fe(II) and other trace metals. *J. Geophys. Res*, 108, D14, doi:10.1029/2002JD00380.

Arabian Sea, March 1997

[4] Siefert, R.L., Johansen, A.M., Hoffmann, M.R. (1999) Chemical characterization of ambient aerosol collected during the southwest monsoon and intermonsoon seasons over the Arabian Sea: Labile-Fe(II) and other trace metals. *J. Geophys. Res.* 104 (D3), 3511–3526, doi: 10.1029/1998JD100067.

Arabian Sea, May 1995

[5] Chen Y., Street J. and Paytan, A. (2006) Comparison between pure-water- and seawater-soluble nutrient concentrations of aerosols from the Gulf of Aqaba. *Mar. Chem*. 101, 141-152, doi: 10.1016/j.marchem.2006.02.002.

Gulf of Aqaba, 20 Aug. 2003 to 21 Nov. 2004

[6] Hsu S-C., F-J. Li, and W-L. Jeng (2005) Seawater solubility of natural and anthropogenic metals within aerosols collected from Taiwan coastal sites, *Atmos. Environ.*, 39, 3989-4001, doi: 10.1016/j.atmosenv.2005.03.033.

Taiwan coastal sites, 1992

[7] Hsu, S-C., Wong G.T. F., Gong, G-C., Shiah F-K., Huang Y-T., Kao S-J., Tsai F., Lung S-C. C, Lin F-J., Lin I-I., Hung C-C. and Tseng C-M. (2009) Sources, solubility, and dry deposition of aerosol trace elements over the East China Sea. *Mar. Chem.*, 120, 116-127, doi:10.101016/j.marchem.2008.10.003.

East China Sea, north, north-east of Taiwan, 2005 – 2007

[8] Chuang, P.Y., Duvall, R.M., Shafer, M.M., Schaur, J.J. (2005) The origin of water soluble particulate iron in the Asian atmospheric outflow. *Geophys. Res. Lett*. 32, L07813, doi:10.1029/2004GL021946.

Kosan, coastal site in South Korea, 31 March – 2 May, 2001

[9] Aguilar-Islas, A. M., J. Wu, R. Rember, A. M. Johansen, and L. M. Shank (2010) Dissolution of aerosol-derived iron in seawater: Leach solution chemistry, aerosol type, and colloidal iron fraction, *Mar. Chem.*, 120, 25-33, doi:10.1016/j.marchem.2009.01.011.

Alaska, 2007, ocean and land sites
Tropical and Equatorial central North Pacific, Oct. 2006

[10] Wu, J., Rember, R. and Cahill, C. (2007) Dissolution of aerosol iron in surface waters of the North Pacific oceans as determined by a semicontinuous flow-through method. *Global Biogeochem. Cycles*, 21, GB4010, doi:10.0129/2006GB002851.

Subtropical eastern North Pacific, April 2006
Sargasso Sea, subtropical western North Atlantic

[11] Buck C.S., Landing W.M., Resing J.A., Lebon G.T. (2006) Aerosol iron and aluminum solubility in the northwest Pacific Ocean: Results from the 2002 IOC cruise. *Geochem. Geophys. Geosyst.* 7, Q04M07, doi:10.1029/2005GC000977.

Western and central North Pacific, May – June, 2002

[12] Chen, Y. (2004) Sources and fate of atmospheric nutrients over the remote oceans and their role on controlling marine diazotrophic microorganisms. Ph.D. Thesis, University of Maryland.

Four cruises to central subtropical Pacific: April, 2001, July, 2002, Sept. & Oct, 2002 and Aug. 2003

[13] Zhuang, G.S., Yi, Z., Duce, R.A., Brown, P.R. (1992) Link between iron and sulfur cycles suggested by detection of Fe(II) in remote marine aerosols. *Nature* 355, 537-539, doi: 10.1038/355537a0.

Island of Barbados, 1986
Islands of Midway, Oahu, Enewetak & Fanning

[14] Siefert, R. L., Webb, S. M., and Hoffmann, M. R. (1996) Determination of photochemically available iron in ambient aerosols. *J. Geophys. Res*. 101, D9, 14,441-14,449, doi: 10.1029/96JD00857.

Land-based sites in USA: Whiteface Mt.(1992) , New York, Pasadena, Calif. (1993), Yosemite, Calif. (1993) and San Nicolas Island, Calif. (1993)

[15] Buck, C. S., W. M. Landing, J. A. Resing, and C. I. Measures (2010) The solubility and deposition of aerosol Fe and other trace elements in the North Atlantic Ocean: Observations from the A16N CLIVAR/CO2 repeat hydrography section, *Mar. Chem*., 120, 57-70, doi: 10.1016/j.marchem.2008.08.003.

A16N CLIVAR line in the North Atlantic, 20 June – 7 Aug. 2003

[16] Baker, A.R., Jickells, T., Witt, M, Linge, K.L. (2006a) Trends in the solubility of iron, aluminium, manganese and phosphorus in aerosol collected over the Atlantic Ocean. *Mar. Chem.* 98**,** 43-58, doi: 10.1016/j.marchem.2005.06.004.

JCR cruise, 2001; ANT18 cruise, 2000 and IRONAGES II cruise, 2002, North Atlantic

[17] Baker, A.R., French, M., Linge, K.L. (2006b) Trends in aerosol nutrient solubility along a west-east transect of the Saharan dust plume. *Geophys. Res. Lett.* 33, L07805, doi:10.1029/2005GL024764

M55 cruise, Oct. & Nov., 2002, North Atlantic

[18] Chen, Y., Siefert, R.L., 2004. Seasonal and spatial distributions and dry deposition fluxes of atmospheric total and labile iron over the tropical and subtropical North Atlantic Ocean, *J. Geophys. Res.* 109, D09305, doi:10.1029/2003JD003958

Two cruises to subtropical North Atlantic: Jan. & Feb., 2001; June – Aug., 2001

[19] Johansen, A. M., Siefert, R. L., and Hoffmann, M. R. (2000) Chemical composition of aerosols collected over the tropical North Atlantic Ocean. *J. Geophys. Res*., 105, D12, 15277-15312, doi: 10.1029/2000JD900024.

Cruise from Barbados to Cape Verde and back, April 1996

[20] Trapp J. M., F. J. Millero, and J. M. Prospero (2010) Trends in the solubility of iron in dust-dominated aerosols in the equatorial Atlantic trade winds: Importance of iron speciation and sources, *Geochem Geophys Geosyst*., 11: Q03014, doi:10.1029/2009 GC002651.

Island of Barbados, Aug. & Sept., 2007

[21] Sedwick, P.N., Sholkovitz, E.R., Church, T.M. (2007) Impact of anthropogenic combustion emissions on the fractional solubility of aerosol iron: Evidence from the Sargasso Sea. *Geochem. Geophys. Geosysts*. 8, Q10Q06, doi:10.1029/2007GC001586

Sargasso Sea, subtropical western North Atlantic, 2003 and 2004, shipboard samples

[22] Sedwick, Church and Sholkovitz [unpublished data]

2007 & 2008 – samples from the island of Bermuda and multiple cruises to the Sargasso Sea,

[23] Zhu, X. R., J. M. Prospero, and F. J. Millero (1997), Diel variability of soluble Fe(II) and soluble Fe in North African dust in the trade winds at Barbados, *J. Geophys. Res.*, 102, 21,297-21,305, doi: 10.1029/97JD01313.

Barbados, September, 1992

[24] Witt, M., A. R. Baker, and T. D. Jickells (2006), Atmospheric trace metals over the Atlantic and South Indian Oceans: Investigation of metal concentrations and lead isotope ratios in coastal and remote marine aerosols, *Atm. Environ*., 40, 5435-5451, doi: 10.1016/j.atmosenv.2006.04.041.

Data from a cruise between South Africa and Australia in Indian Ocean, March/April 2002

[25] Witt, M. L. I., T. A., Mather, A. R. Baker, J. C. M. De Hogg, and D. M Pyle (2010), Atmospheric trace metals over the south-west Indian Ocean: Total gaseous mercury, aerosol trace metal concentrations and lead isotope ratios, *Mar. Chem*., 121, 2-16, doi:10.1016/j.marchem.2010.02.005

Data from a November 2007 cruise in South Indian Ocean between Seychelles and Mauritius

[26] Bowie, A. R, D. Lannuzel, T. A. Remenyi, T. Wagener, P. J. Lam, P. W. Boyd, C. Guieu, A. T. Townsend, and T W. Trull, (2011), Biogeochemical iron budget of the Southern Ocean south of Australia: decoupling of iron and nutrient cycles in the sub-antarctic zone by the summertime supply, *Global Biogeochem. Cycles*, 23, GB4034, doi:10.1029/2009GB003500

Seven aerosol samples between Tasmania and Antarctica, February 2007

Table S1 and Figure 2

Table S1 also contains Baker and Powell's unpublished data for 291 samples from twelve cruises in the Atlantic Ocean between 2003 and 2008 –see Figure 2 for site location map. Batch leaching with pH 4.7 ammonium acetate buffer was employed – see Baker et al. (2006a). Total Fe for these samples was determined using neutron activation analysis – see Baker et al., "Estimation of atmospheric nutrient inputs to the Atlantic Ocean from 50°N to 50°S based on large-scale field sampling: Iron and other dust-associated elements", in preparation for *Global Biogeochemical Cycles*, 2011. The majority of sites come from eastern North Atlantic; some data comes from the South Atlantic Ocean.

Table S1 – Notes on data in the compilation in Table S1

The majority of data on total aerosol loading and % Fe solubility were taken directly from tables in cited papers – exceptions are noted below. In some cases the data are presented in the original paper in the forms of plots but not in tables. In these cases, the cited authors have sent us [us = we = the authors = Sholkovitz et al.] the data or we have read data off published plots in the cited papers. Site #'s refer to location map in Figure 1 of main paper.

Site #1, Kumur et al. [2010] – Total Fe and % soluble Fe read off their figure 5
Site # 2, Kumur and Sarin [2010] - Total Fe and % soluble Fe data sent to us
Site #3, Johansen and Hoffman [2003] - Total Fe and % soluble Fe data sent to us
Site #5, Chen et al. [2006] – Total soluble Fe read off their figure 6, total Fe load from their table 1.
Site #6, Hsu et al. [2005] – Total Fe loading data sent to us; % soluble Fe data came from their paper
Site #7, Hsu et al. [2009] – Total Fe load and % soluble Fe data sent to us
Site #8, Chung et al [2005] - Total Fe load and % soluble Fe data read off their figure 2
Site #16, Baker et al [2006a] – Total Fe load and % soluble Fe data sent to us
Site #17, Baker et al [2006b] – Total Fe load and % soluble Fe data sent to us
Site # 19, Johansen et al. [2000] - Total Fe load and % soluble Fe data sent to us

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Table S2

Table S2 contains data on the fractional solubility of Fe in aerosols and soils as presented in the following papers: *Bonnet and Guieu* [2004]; *Breslin and Duedall* [1987]; *Buck et al*., [2010b]; *Chester et al.* [1993]; *Crecelius* [1980]; *Cwiertny et al*. [2008]; *Desboeufs et al*., [2005]; *Duvall et al*., [2008]; *Hardy and Crecelius* [1981]; *Hodge et al*., [1978]; *Hsu* *et al.*, [2010]; *Jickells* [1999]; *Jickells and Spokes* [2001]; *Journet et al*., [2008]; *Mackie et al*., [2006]; *Mendez et al*., [2010]; *Mori et al*., [2011]; *Oakes et al*. [2010]; *Ooki et al*., [2009]; *Paris et al*., [2010]; *Schroth et al*., [2009]; *Seguret et al.*, (2011); *Shi et al*., [2011]; *Spokes et al*., [1994]; *Spokes and Jickells* [1996]; *Theodosi et al*., [2010]; *Wagener et al*., [2008]; *Upadhyay et al*., [2011]; *Zhu et al*., [1993].

Unlike the paired-data in Table S1, the majority of papers in Table S2 only report data on % Fe solubility and not on total aerosol Fe loading. Table S2 also provides information on collection dates, locations, and sample types, as well as the protocols that are used to define soluble aerosol iron. A full reference list for papers cited in Table S2 follows:

Bonnet, S., and C. Guieu (2004), Dissolution of atmospheric iron in seawater, *Geophys. Res. Lett*., L03303, doi:10.1029/2003GL018423.

Breslin, V. T., and I. W. Duedall (1987), Metal release from particulate oil ash in seawater, *Mar. Chem*., 22, 31-42, doi:10.1016/0304-4203(87)90046-6.

Chester, R. K., K. J. T. Murphy, F. J. Lin, A. S. Berry, G. A. Bradshaw, and P. A. Corcoran (1993), Factors controlling the solubilities of trace metals from non-remote aerosols to the sea surface by the 'dry' deposition mode, *Mar. Chem.*, 42, 107-126, doi:10.1016/0304-4203(93)90241-F.

Crecelius, E. (1980), The solubility of coal fly ash and marine aerosols in seawater, *Mar. Chem*., 8, 245-350, doi:10.1016/0304-4203(80)90013-4.

Cwiertny, D. M., J. Baltrusaitus, G. J. Hunter, A. Laskin, M. M. Scherer, and V. H. Grassian (2008), Characterization and acid-monbilization study of iron-containing mineral dust source material, *J. Geophy. Res*., 113, D05202, doi:10.1029/2007JD009332.

Desboeufs, K.V., A. Sofikitis, R. Losno, J. L. Colin, and P. Ausset (2005), Dissolution and solubility of trace metals from natural and anthropogenic aerosol particulate matter, *Chemosphere* 58**,** 195-203, doi:10.1016/j.chemosphere.2004.02.025.

Duvall, R. M., B. J. Majestic, M. M. Shafer, P. Y. Chuang , B. R. T. Simoneit, and J. J. Schauer (2008), The water-soluble fraction of carbon, sulfur, and crustal elements in Asian aerosols and Asian soils, *Atmos. Environ*., 42, 5872-5884, doi: 10.1016/j.atmosenv.2008.03.028.

Guieu, C., S. Bonnet, and T. Wagener (2005). Biomass burning as a source of dissolved iron to the open ocean, *Geophys. Res. Lett*., 32, L19608, doi:10.1029/2005GL022962.

Hardy, J. T., and E. A. Crecelius (1981), Is atmospheric particulate matter inhibiting marine primary productivity?, *Environ. Sci. Technol*., 15, 1103-1105, doi: 10.1021/es00091a013.

Hodge, V., S. R. Johnson, and E. D. Goldberg (1978), Influence of atmospherically transported aerosols on surface ocean water composition, *Geochem. J*., 12, 7-20.

Hsu, S-C. et al. (2010), Effects of acidic processing, transport history, and dust and sea salt loading on iron from Asia dust, *J. Geophys. Res*., 115, D19313, doi:10.1029/2009JD013442.

Jickells, T. D. (1999), The inputs of dust derived elements to the Sargasso Sea, *Mar. Chem.*, 68, 5-14, doi: 10.1016/S0304-4203(99)00061-4.

Jickells, T. D., and L. Spokes (2001), Atmospheric Iron Inputs to the Oceans, in *The Biogoechemistry of Iron in Seawater*, (ed., K. Hunter and D. Turner), pp. 85-122, John Wiley, New York.

Journet, E., K. V. Desboeufs, S. Caquineau, and J-L. Colin (2008), Mineralogy as a critical factor of dust iron solubility, *Geophys. Res. Lett*., 35, L07805, doi:10.1029/2007GL031589.

Mackie, D. S., J. M. Peat, G. H. McTainsh, P. W. Boyd, and K. A. Hunter (2006), Soil abrasion and eolian dust production: Implications for iron partitioning and solubility, *Geochem. Geophys. Geosysts*., 7, Q12Q03, doi:10.1029/2006GC001404.

Mendez, J., C. Guieu, and J. Adkins (2010), Atmospheric input of manganese and iron to the ocean: Seawater dissolution experiments with Saharan and North American dusts, *Mar. Chem*., 120, doi:10.1016/j.marchem.2008.08.006.

Mori I., M. Nishikawa, A. Shimizu, M. Hayasaki, and T. Takasuga (2011), Solubility of Iron in aerosol collected during Kosa (Asian dust) events in Japan, *SOLA*, 7A, 5-8, doi:10.2151/sola.7A-002.

Oakes, M., N. Rastogi, B. J. Majestic, M. Shafer, J. J. Schauer, E. S. Edgerton, and R. J. Weber (2010), Characterization of soluble iron in urban aerosols using near-real time data, *J. Geophys. Res*., 115, D15302, doi:10.1029/2009JD012532.

Ooki, A, J. Nishioka, T. Ono, and S. Noriki (2009), Size dependence of iron solubility of Asian dust particles, *J. Geophys. Res*., 114, D03202, doi:10.1029JD010804.

Paris, R., K. V. Desboeufs, P. Formenti, and C. Chou C (2010), Chemical characterisation of iron in dust and biomass burning aerosols during AMMA-SOP0/DABEX: implication for iron solubility, *Atmos. Chem. Phys*., 10, 4273-4282, doi: 10.5194/acp-10-4273-2010.

Schroth, A. W., J. Crusius, E. R. Sholkovitz, and B. C. Bostick (2009), Iron Solubility driven by speciation in dust sources to the ocean, *Nature Geoscience*, 2, 337-340, doi:10.1038/NGEO0501.
Seguret, M. J. M., M. Koçak, C. Theodosi, S.J. Ussher, P.J. Worsfold, B. Herut, N. Mihalopoulos, N. Kubilay and M. Nimmo (2011), Iron solubility in crustal and anthropogenic aerosols: The Eastern Mediterranean as a case study, *Mar. Chem.*, 126, 229-238, doi:10.1016/j.marchem.2011.05.007.

Shi, Z. B., M. T. Woodhouse, K. S. Carslaw, M. D. Krom, G. W. Mann, A. R. Baker, I. Savov, G. Fones, B. Brooks, T. D. Jickells, and L. G. Benning, (2011), Minor effect of physical size sorting on iron solubility of transported mineral dust, *Atmos. Chem. Phys. Discuss.*, 11, 14309-14338, doi: 10.5194/acpd-11-14309-2011.

Spokes, L. J., T. D. Jickells, and B. Lim (1994) Solubilization of aerosol trace metals by cloud processing: A laboratory study, *Geochim. Cosmochim. Acta*, 58, 3281-3287, doi: 10.1016/0016-7037(94)90056-6.

Spokes, L. J., and T. D. Jickells (1996), Factors controlling the solubility of aerosol trace metals in the atmosphere and on mixing with seawater, *Aquatic Geochem.*, 1, 355-374, doi: 10.1007/BF00702739.

Theodosi, C., N. Markaki, and N. Mihalopoulos (2010), Iron speciation, solubility and temporal variability in wet and dry deposition in the Eastern Mediterranean, *Mar. Chem.*, 120, 100-107. doi:10.1016/j.marchem.2008.05.004.

Upadhyay N., B. J. Majestic, and P. Herckes (2011), Solubility and speciation of atmospheric iron in buffer systems simulating cloud conditions, *Atmos. Environ.*, 45, 1858-1866, doi:10.1016/j.atmosenv.2011.01.010.

Wagener, T., E. Pulido-Villena, and C. Guieu (2008), Dust iron dissolution in seawater: Results from a one-year time-series in the Mediterranean Sea, *Geophys. Res. Letts.*, 35, L16601, doi:10.1029/2008GL034581.

Zhen, Y., G. Zhuang, P. R. Brown, and R. A. Duce (1992), High-performance liquid chromatographic method for the determination of ultratrace amounts of iron(II) in aerosols, rainwater, and seawater, *Anal. Chem.*, 64, 2826-2830, doi: 10.1021/ac00046a028.

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Table S3

This table contains a Microsoft Excel spreadsheet that is associated with the two end-member mixing model presented in equations # 1 to 3.

Specifics of the model, as per the main paper, are as follows.

The pervasive curvilinear trend in % vs. clearly suggests a mixing hyperbola produced by the conservative mixing of two aerosol populations. To demonstrate that this is the case for the *global data set*, we have formalized the simple conservative mixing of two aerosol end member populations, A and B, with each having distinct atmospheric loadings of soluble aerosol Fe (and , respectively) and total aerosol Fe (and , respectively). This yields the following expressions for the soluble aerosol Fe loading ( ) and total aerosol Fe loading ( ) of the mixture (Faure and Mensing, 2005)*.

= + (1 -) (1)

= + (1 -) (2)

where and are the fractions of aerosols A and B, respectively, in the aerosol mixture. The percent fractional solubility for the aerosol iron in the mixture (bulk aerosol) is then given by:

% = 100(/) = 100(+ (1 -))/(+ (1 -)) (3)

We have estimated % as a function of for various combinations of plausible end-member aerosol iron loadings (and) and corresponding aerosol iron solubilities. To that end, Figure 10C shows the mixing hyperbolae for eight case studies which cover the range of end-member properties that have been observed in global data set (Figures 3-9). Table S3 presents eight case studies [I-VIII], each with different values of Fe solubility and Fe loading for the two end-members. Also presented in this table are the individual plots of vs. % for each of the eight case studies. These same plots appear in Figure 10 in the main paper.

Cases I –VIII have the following end-member values of (ng m^{-3} air) for the Fe loading of non-soil- dust and soil-dust end-members respectively: I (50, 1000), II (50, 500), III

(10, 2000), IV (50, 2000), V (100, 2000), VI (50, 2000), VII (50, 2000) and VIII (50, 500).

Cases I –VIII have the following end-member values for the fractional solubility of Fe (%FeS): for the non-soil- dust and soil dust end-members respectively: I (50, 1), II (50, 1), III (50, 1), IV (50, 1), V (50, 1), VI (60, 1), VII (60, 0.5 and VIII (30, 0.5).

* Faure G, and Mensing T. M. (2005) *Isotopes: Principles and Applications*. 3rd Edition, John Wiley and Sons, Hoboken, N. J.