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Title: Sources, transport and deposition of iron in the global atmosphere

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Referee 1 (C. Pérez García-Pando)

Comment 1

The paper proposes a new emission inventory of Fe from combustion sources, and estimates and evaluates the global size-resolved Fe emission, concentration and deposition, including the contribution from natural dust sources using a recently published mineralogical soil database. In my opinion, this work is impressive and represents a significant step forward towards constraining the deposition of soluble iron into ocean waters. I only have some minor comments that intend to clarify a few aspects of the paper and allow the reproducibility of the method by other modelers.

Response

Thank you very much for the reviewer's positive comments. Please see a point-to-point response to specific comments in the following.

Comment 2

Section 2.1: With respect to equation 1 and its description, please provide in the form of table(s) in the Supplementary material the values and ranges of all the parameters included in the equation that are used to calculate the emissions. Some of these values and ranges appear in the text, but others are missing (for example A_y). Please provide a comprehensive table.

Response

Thank you for the good comment. According to the comment, we added in the revised manuscript a new Table, **Table 1** that lists all parameters used in estimating the emissions. A_y is computed by country and time using an empirical method. We describe the method in **Table 1**, but the parameters used in the functions are referred to a Table in the published paper (*Table S1* of Wang et al., 2014) to avoid repeating.

Table 1. Parameters used in the estimation of Fe emissions from combustion sources.

Parameter	Description	Values or data sources
a	Fuel consumption	The fuel data was taken from a global 0.1°×0.1° fuel data set which is used to construct a global CO_2 emission inventory (Wang et al., 2013; available at http://inventory.pku.edu.cn/home.html).
b	Completeness of combustion	■ coal (98%); ■ petroleum (98%); ■ wood in stoves (88%); ■ wood in fireplaces (79%); ■ crop residues (92%); ■ biomass burning (considered in van der Werf, 2010).
c	Fe content of the fuel	■ coal: based on Fe contents in coal produced by country (**Table S2**) and an international coal-trading matrix (Chen et al., 2014); ■ wood (a geometric mean of 0.036% and range in **Fig. S1**); ■ crop residues (a geometric mean of 0.060% and range in **Fig. S1**); ■ grass (a geometric mean of 0.027% and range in **Fig. S1**); ■ dung cakes (0.13±0.09 %); ■ biodiesel (0.00024±0.00023 %); ■ heavy fuel oil (32±2 ppm); ■ diesel (13±7 ppm); ■ gasoline (3.3±2.6 ppm);

		■ liquefied petroleum gas (4.9±3.3 ppm).
f	Fraction of Fe retained in residue ash relative to the amount of Fe in the burnt fuel	■ coal used in industry and power plants (30-45%); ■ petroleum used in industry and power plants (43-58%); ■ solid biofuels used in industry and power plants (60-70%); ■ petroleum consumed by motor vehicles (2-12%); ■ anthracite coal used in the residential sector (99.2-99.8%); ■ bituminous coal used in the residential sector (91-97%); ■ straw used in the residential sector (79-95%); ■ wood used in the residential sector (89-99%); ■ forest fires (49-98%); ■ savanna fires (24-79%); ■ deforestation (43-50%); ■ woodland fires / peat fires (41-56%).
J_x	Fraction of Fe emitted in a particle size	■ coal fly ash (0.1-0.3% in PM_1; 10-30% in PM_{1-10}; the remainder in $PM_{>10}$); ■ oil fly ash (80-95% in PM_1; the remainder in PM_{1-10}); ■ biomass fly ash (1-3% in PM_1; 50-60% in PM_{1-10}; the remainder in $PM_{>10}$).
A_y	Fraction of a specific control device	A_y is computed for each country and each year using a function by Grubler et al. (1999) and Bond et al. (2007): $A_y = (F_0 - F_f) \exp[-(t-t_0)2/2s^2] + F_f$, where F_0 and F_f are the initial and final fractions of the technology, t_0 is transition beginning time, and s is transition rate. Parameters were determined for developing or developed countries and listed in Wang et al. (2014a).
$R_{x,y}$	Removal efficiency for each particle size by different control device (Zhao et al., 2008)	■ cyclone (10% for PM_1; 70% for PM_{1-10}; 90% for $PM_{>10}$); ■ scrubber (50% for PM_1; 90% for PM_{1-10}; 99% for $PM_{>10}$); ■ electrostatic precipitator (93.62% for PM_1; 97.61% for PM_{1-10}; 99.25% for $PM_{>10}$).

References:

Wang, R., Tao, S., Shen, H. Z., Huang, Y., Chen, H., Balkanski, Y., Boucher, O., Ciais, P., Shen, G. F., Li, W., Zhang, Y. Y., Chen, Y. C., Lin, N., Su, S., Li, B. G., Liu, J. F., and Liu, W. X.: Trend in global black carbon emissions from 1960 to 2007, Environ. Sci. Technol., 48, 6780–6787, doi:10.1021/Es5021422, 2014.

Comment 3

Section 2.4: The gridded fuel data (PK-FUEL) doesn't seem to be available from the web page announced in the text. If is announced as available, please make it available.

Response

Sorry for the misleading information. Unfortunately, we cannot release original fuel data in our web page at present due to policies using some data products developed by other groups, but the fuel data can be converted from the CO_2 emissions in the PKU-CO_2 inventory with CO_2 emission factors. The sentence on line 22 page 7653 was revised as: "*The annual emissions of Fe were estimated based on the 0.1° gridded fuel data which is used to construct a global CO_2 emission inventory (Wang et al., 2013; available at http://inventory.pku.edu.cn/home.html).*".

Comment 4

Section 2.6: Emissions of Fe from mineral sources. Please provide in the text the assumed Fe content for each mineral.

Response

Thank you for the comment. The content of Fe in each mineral is listed in the revised manuscript, and the following sentence was added on line 22, page 7654: "*Then, a global 0.5° × 0.5° map of Fe content in clay fraction was obtained (Fig. S2) with the Fe content of each mineral (4.3% for illite, 2.6% for smectite, 0.23% for kaolinite, 12.5% for chlorite, 6.7% for vermiculite, 0.34% for feldspars, 62.8% for goethite and*

69.9% for hematite) measured in Journet et al. (2008) and compiled in Journet et al. (2013).”.

Comment 5

In contrast to combustion sources, the uncertainty calculated for Fe from dust only accounts for the uncertainty in the emission. I would suggest (at least) acknowledging that the elemental composition (and therefore the iron content) in each of the minerals can be regionally variable in nature, which adds additional uncertainty.

Response

Thank you for the comment. Due to lack of a global distribution of elemental composition in minerals, we acknowledge this uncertainty in the revised text. Accordingly, the following sentence was added in Section 2.6: “*Note that we only account for the variation of dust emissions when assessing the uncertainty in Fe emissions from dust. However, there is also a variation of elemental composition of minerals in nature. For example, the Fe content can vary from 0.8 to 8.4% in illite depending on the environmental condition (Murad and Wagner, 1994), and from 0.02 to 0.81% in kaolinite (Mestdagh et al., 1980). This uncertainty is not accounted for in our study due to lack of a global distribution of elemental composition in minerals.*”.

Comment 6

Section 2.7: Please clarify and detail the following issues: What is the assumption behind your treatment of sedimentation, dry and wet deposition for Fe in PM1-10 and PM>10 as if it was dust? Are you assuming the same density as dust? Are you treating the combustion Fe as hydrophobic in those size ranges? Is there heterogeneous chemistry for dust (and therefore they can be in-could scavenged)? Please provide further details and justification.

Response

Sorry for missing these informations. According to the comment, hygroscopic growth, density of particles, and hygroscopic properties of Fe are described in the revised manuscript. Accordingly, the third paragraph in **Section 2.7** was re-written as: “*The emitted Fe from combustion sources were partitioned into three particulate modes with the following characteristics: Fe in* PM_1 *as a fine mode (MMD = 0.34* μm*, geometric* $\sigma = 1.59$*); Fe in* PM_{1-10} *as a coarse mode (MMD = 3.4* μm*, geometric* $\sigma = 2.0$*); Fe in* $PM_{>10}$ *as a super coarse mode (MMD = 34.0* μm*, geometric* $\sigma = 2.0$*) (Mamane et al., 1986; Querol et al., 1995; Valmari et al., 1999). Hygroscopic growth, sedimentation, dry and wet deposition accounted for Fe in* PM_{1-10} *and* $PM_{>10}$*, as for dust, and Fe in* PM_1 *as for BC (Balkanski et al., 2004, 2010, 2011). Hygroscopic growth of particles in the model is treated as a function of ambient relative humidity and the composition of soluble aerosol components based on Gerber's experiment work (Gerber, 1988). The uptake of water on aerosols increases the particle size of Fe, while the loss of water on aerosols decreases the particle size of Fe. For the particle density, the fraction of low density mass in coal fly ash is found to increase with decreasing particle size (Furuya et al., 1987). The major fraction for particles with a diameter less than 10* μm *is composed by mass with a density of 2.4-2.8 g* cm^{-3}*, and by mass with a density of 1.6-2.4 g* cm^{-3} *for particles with a diameter from 10 to 100* μm*. Therefore, we applied a density of 2.6 and 2.0 g* cm^{-3} *for Fe transported in* PM_{1-10} *and* $PM_{>10}$ *respectively in the model. For Fe in* PM_1*, we assumed that the density is the same as BC (1.5 g* cm^{-3}*). For the hygroscopic properties of Fe, it is found that Fe in large-size coal ash is dominated in aluminosilicate glass, similar to that in dust (Chen et al., 2012), and thus we assume that the Fe in* PM_{1-10} *and* $PM_{>10}$ *can be treated as insoluble dust, which is removed by sedimentation, dry deposition and below-cloud scavenging. For the Fe in* PM_1*, it is found that approximately 25% of Fe in fine particle (diameter<0.61* μm*) is bound to organic matter and thus insoluble (Espinosa et al., 2002). Thus, we assumed that 25% of Fe in* PM_1 *was hydrophobic, which is removed by sedimentation, dry deposition and below-cloud scavenging, but not by in-cloud scavenging. The remainder Fe in* PM_1 *was hydrophilic, which is removed by sedimentation, dry deposition, below-cloud scavenging, and in-cloud scavenging. Due* *to limited understanding of the heterogeneous chemistry of Fe in the cloud, we did not account for the conversion of Fe from hydrophobic to hydrophilic in the atmospheric transport, and the ratio between the two phases varies due to their different removal rates in the atmosphere.*".

Comment 7

Section 3.5: I am confused by the last sentence of the section. Wouldn't it be desirable to have dust and plant material in the Fe emission estimates from biomass burning? Why would be Luo et al. overestimating? Perhaps just note that their estimates are larger than yours partly because their estimates implicitly include additional sources of Fe.

Response

Thank you very much for the good comment. Now, we realized that our lower estimates of the biomass burning emissions of Fe than previous studies is likely caused by the fact that previous estimates include the part by pyro-convection of Fe from soil and plant materials. In contrast, since we estimated the emissions based on the difference of Fe mass initially present in the fuel and that in the post-burn residues, rather than on the atmospheric concentration of Fe, we could miss the emissions of Fe by pyro-convection. Although our estimate provides a more explicit source attribution of Fe in the atmosphere, it could underestimate the total source. Accordingly, the sentences on line 20-24, page 7661 were revised as: "*Note that the dust and plant material entrained in fires can contribute to the Fe concentrations in the atmosphere, as noticed by Luo et al. (2008). As a result, their estimates include the pyro-convection of Fe from soils and plant materials. In contrast, our estimate is based on the mass balance of Fe from the burnt fuel. This might explain partly why our estimate of the biomass burning emission of Fe is lower than that in previous studies (Luo et al., 2008; Ito, 2013). Although our estimate provides an explicit source attribution of Fe, which is useful for modelling the Fe solubility, it underestimates the total sources. We propose that the emissions of Fe by pyro-convection in the fires should be estimated separately in the future.*".

Comment 8

Section 4.2 Please note that the overestimation of iron from dust may not come (at least not all of it) from the assumption that the composition of dust resembles the composition of clay. Clay minerals form aggregates of larger sizes and the mineralogy database is mostly based on wet sieving that breaks the aggregates into small clay-sized particles.

Response

Thank you for the good comment. Accordingly, the following sentences were added on line 6, page 7664: "*In addition, when compiling data in the mineralogy database, Journet et al. (2014) noticed that wet sieving is used to determine soil texture, leading to loss of soluble minerals (e.g., calcite or gypsum) and a possible overestimation of the content of minerals rich in Fe such as hematite and goethite. This impact might also contribute to an overestimation of Fe content in dust.*".

Comment 9

Section 4.6. This section needs further detail (this is connected to my previous comment on sedimentation, dry and wet deposition). The authors analyze the wet MMD. It would be very helpful to understand how this is treated in this specific model.

Response

Thank you for the good comment. The following paragraph was added on line 21-23, page 7667 to explain the mechanisms changing the size of Fe: "*According to Schulz et al. (2007), after the particles containing Fe are emitted into the atmosphere, there are three major processes that change the size distributions in the model. First, formation of sulphate increases the aerosol mass in the accumulation mode and, since the particle number is kept constant, the aerosol diameter increases for Fe in* PM_1*. Second, removing processes such as sedimentation removes the larger particles more efficiently, shifting the mode diameter to a smaller one for Fe in* PM_1, PM_{10} *and* $PM_{>10}$*. At last, the hygroscopic growth creates instaneous changes in the size of particles as a function of ambient relative humidity (Schulz et al., 2007), and the uptake of water on aerosols increases the size, while the loss of water on aerosols decreases the size. Therefore, the change of the size of Fe is dependent on the relative importance of the mechanisms increasing / decreasing the size.*

For example, the size of Fe in PM_1 *emitted from coal combustion increased from 0.3 µm to >2 µm after being transported away from the source regions, because the hygroscopic growth by uptake of water on aerosol particles are more important. In contrast, the size of Fe in* $PM_{>10}$ *emitted from coal combustion in East Asia decreases over the southern Pacific Ocean, because sedimentation is the dominating process. The size of Fe in* $PM_{>10}$ *from coal combustion would decrease from 33 µm in the source regions to <10 µm over the oceans.*". Please also refer to our response to **Comment 6** for a description of the treatment of Fe in the model.

Comment 10

Page 7667, line 28: replace "of two mechanisms" by "of the two mechanisms" (this refers to mechanisms presented before and the omission of "the" creates confusion when reading.)

Response

Thanks for correcting the confusion, and "*of two mechanisms*" was revised as "*of the mechanisms increasing / decreasing the size*".

Comment 11

Page 7670, line 5: "orders of magnitude" instead of "magnitudes"

Response

Revised accordingly.

Comment 12

Caption of Figure 1: the Fe emission is log-transformed but the x-axis in not on log-scale

Response

Sorry for the problem. Figure 1 is plotted on a log-scale x-axis (as below).

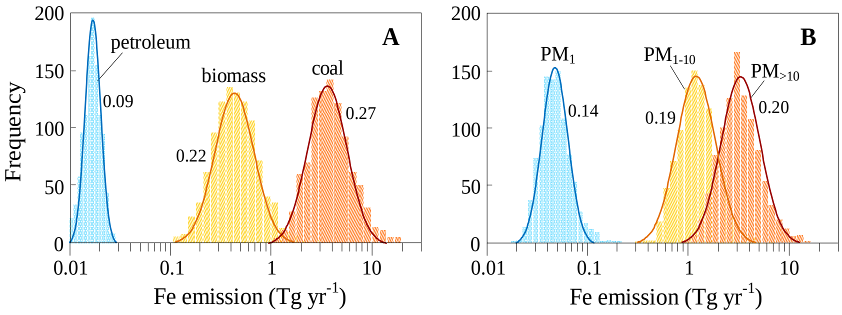

Figure 1. Frequency distributions of Fe emissions from different fuel types (**A**) and particle sizes (**B**). The distributions are derived from 1000 Monte Carlo simulations. The standard deviation of $\log_{10}$-transformed Fe emissions is shown for each distribution. The x-axis is plotted on a log scale.

Comment 13

Caption of Figure 7: measurement instead of measuring

Response

Thank you for the comment. Revised accordingly.

Comment 14

Figure 9: One cannot distinguish the colors for the dust and combustion contributions

Response

Sorry for the unclear figure. In the revised version, Figure 9 was plotted with contrasting colors for the dust and combustion contributions (as below).

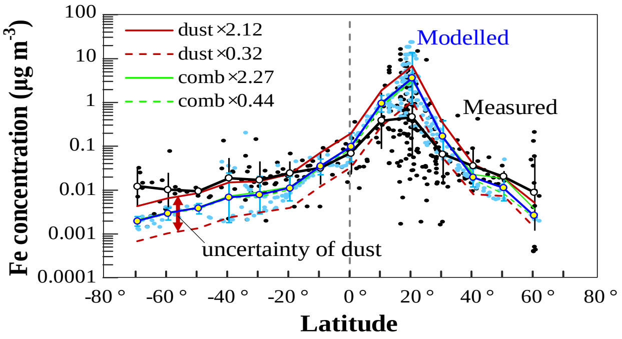

Figure 9. Zonal distribution of modelled (cyan dots) and measured (black dots) Fe concentrations attached to aerosols in surface air over the Atlantic Ocean from 70 °S to 60 °N. The solid lines with circles show the modelled (blue) and measured (black) Fe concentrations as geometric means in each band with error bars for the geometric standard deviations. As sensitivity tests, Fe concentrations from mineral sources were scaled by factors of 0.32 and 2.12 (solid and dashed red lines) as 90% uncertainties in dust emissions (Huneeus et al., 2011) and Fe concentrations from combustion sources were scaled by factors of 0.44 and 2.27 (solid and dashed green lines) as 90% uncertainties in Fe emissions from combustion.

Comment 15

Figure 10: Why do you show the median of the observations? You also show the median of the model or the mean? Please describe

Response

Thank you for the comment. We were using the median concentrations in the measurements, because we were trying to minimize the impact of very high or low concentrations on a specific day measured during 1988-1994. In the previous version, the modelled concentration was computed as the monthly mean. To be consistent, we computed medians in the model in the revised version (the mean and median is very close in the model). To make it clear, the following sentence was revised in the capture of Figure 10: "*Modelled Fe concentrations are derived from all sources (Fe_total) and from mineral sources only (Fe_dust) as medians of all days for each month in 2005.*".

Anonymous Referee #2

Comment 1

The amount of iron emissions is the source of the large uncertainties in the calculation of iron deposition. The authors presented a comprehensive emission inventory for iron. The estimated source strength results in higher iron deposition from coal combustion than previous estimates. The work conducted in this paper may contribute to improve our understanding of iron cycle, although more works will be required to improve the treatment of the iron solubility. Here, my concern is the method to estimate the biomass burring emissions.

Response

We thank you very much for the reviewer's positive comments and useful discussion on the methodology. Please see a point-to-point response to all specific comments in the following. Regarding a particular concern on the Fe emissions from biomass burring, we updated the methods according to the reviewer's comments, and all results were updated (please refer to our responses to **Comment 9**, **11**, **15** and **21**).

Comment 2

Abstract p. 7647, l. 3: Krishnamurthy et al. (2009) have already accounted for the combustion sources of Fe and their deposition in their biogeochemical model. Please rephrase the sentence.

Response

Thank you for pointing out the work of these authors. Accordingly, the sentence was rewritten as: "*In particular, the combustion sources of Fe and the subsequent deposition over oceans have been accounted for in only few ocean biogeochemical models of the carbon cycle.*". In addition, the following sentence was added on line 3, page 7647 in the **Introduction**: "*However, few global models have accounted for the impact of Fe from combustion on the open-ocean biogeochemistry (Krishnamurthy et al., 2009; Okin et al., 2011), due to large uncertainties in the sources and chemical forms of Fe from combustion.*".
References:

Krishnamurthy, A. et al. (2009), Impacts of increasing anthropogenic soluble iron and nitrogen deposition on ocean biogeochemistry, Global Biogeochem. Cycles, 23, GB3016, doi:10.1029/2008GB003440.
Okin, G. S., et al. (2011), Impacts of atmospheric nutrient deposition on marine productivity: Roles of nitrogen, phosphorus, and iron, Global Biogeochem. Cycles, 25, GB2022, doi:10.1029/2010GB003858.

Comment 3

p. 7647, l. 13: Please show the size range of mineral dust for the comparison.

Response

Thank you for the comment. Accordingly, the sentence was revised as: "*Of these emissions, 2, 33 and 65% were emitted in particles < 1 µm (PM_1), 1-10 µm (PM_{1-10}), and > 10 µm ($PM_{>10}$), respectively, compared to a total Fe emission from mineral dust of 41.0 Tg yr^{-1} in a log-normal distribution with a mass median diameter of 2.5 µm and a geometric standard deviation of 2.*".

Comment 4

p. 7647, l. 15: Please show Fe emissions from motor vehicles.

Response

Thank you for the comment. Accordingly, the sentence was revised as: "*For combustion sources, different temporal trends were found in fine and medium-to-coarse particles, with a notable increase in Fe emissions in PM_1 since 2000 due to an increase in Fe emission from motor vehicles (8.0 and 10.3 Gg yr^{-1} for 2000 and 2007, respectively).*".

Comment 5

p. 7647, l. 20: Please state "monthly" mean concentrations and "daily" measurements for the comparison.

Response

According to the comment, the sentence was revised as: "*The modelled Fe concentrations as monthly means were compared with the monthly (57 sites) or daily (768 sites) measured concentrations at a total of 825 sampling stations.*".

Comment 6

p.7648, l.2: Please state "coal" combustion, because Fe emissions from biomass burning and oil combustion were not higher than previous estimates. Please also indicate the uncertainty and clarify if the differences from previous estimates are statistically significant.

Response

Thank you for the comment. Unfortunately, we cannot perform a statistical analysis to compare our estimates with previous ones, because the uncertainties were not provided in previous studies. So we compare the central estimate with that in previous studies while judging if previous estimates are within our uncertainty range or not. Accordingly, the sentence was revised: "*Our central estimates of Fe emissions from fossil fuel combustion (mainly from coal) are generally higher than those in previous studies, although they are within the uncertainty range of our estimates. In particular, the higher than previously estimated Fe emission from coal combustion implies…*". Please also refer to our response to **Comment 18** for more discussion.

Comment 7

1 Introduction
p. 7649, l.6: Additional Fe emission from ships should not change the total Fe emission so much. The total Fe emission should be corrected to 1.2 Tg/yr in 2001 (see below). Do you mean "by prescribing high iron solubility"? Please consider to rephrase the sentence.

Response

Thank you very much for the comment. The total Fe emission was corrected to 1.2 Tg yr^{-1} in 2001 for Ito, 2013, and the sentence on line 6, page 7649 was rephrased as: "*By applying a high emission factor of Fe from ships and accounting for a large Fe solubility from oil fly ash, Ito (2013) later derived the same total Fe emission of 1.2 Tg* yr^{-1}*, but with a significant contribution by shipping to soluble Fe deposition over the northern Pacific Ocean and the East China Sea.*".

Comment 8

Table 1: Please correct the Fe emissions from biomass burning in Ito (2013) to 0.57 (= 102/0.18/1000) and 0.14 (= 25/0.18/1000) (see Table 4 and Table 5 in Ito, 2013).

Response

Thank you very much for pointing out the mistake. We included these corrections in the revised manuscript. We mistook the Fe solubility in Ito, 2013 to be 11%.

Comment 9

2.1 Emissions of Fe from combustion sources.
p. 7650, equation (1): The definition of the combustion rate (b) is not clear. What is the difference between the fraction of fuel not burned (1 – b) and that retained in residue ash? If (1 – b) includes the compound retained in residue ash, why did you multiply (1–f) by b?

Response

Sorry for the confusion. In equation (1), the combustion rate (*b*) is the fraction of burnt fuel relative to total fuel, and the rest (1- *b*) is unburnt. The fraction of Fe in residue ash (*f*) is the fraction of Fe retained in residue ash relative to the mass of Fe in burnt fuel, and the rest (1- *f*) is going to fly ash or cyclone ash. To make it clear, the sentence on line 18, page 7650 was revised as: "*b is the completeness of combustion (defined as the fraction of fuel burnt in the fires), …, f is the fraction of Fe retained in residue ash relative to the amount of Fe in burnt fuel, …*".
Thank you for the good comment. We realized that, when collecting *f* from the literature, if Fe in unburnt fuel was included in residue ashes, it will lead to an overestimation of *f*. For example, when Raison et al. (1985) reported that 25-43% of the manganese was transferred to the atmosphere, it is a percentage relative

to the manganese present in initial fuels and part of them was unburnt. So, in the revised manuscript, we used the completeness of combustion in the literature to convert the percentage relative to the element in initial fuels to that relative to the element in the burnt fuel. If the *f* is estimated based on the distribution of the element in bottom ash, cyclone ash and fly ash (Chalot et al., 2012), we use the original data. As the completeness of combustion is close to 100% for fossil fuels and the *f* is the percentage to the amount in burnt fuels for biofuels, we only updated the *f* for the biomass burning used in our paper (Raison et al., 1985; Pivello and Coutinho, 1992; Mackensen et al., 1996; Holscher et al., 1997). Accordingly, the last paragraph in Section 2.3 was revised as below: "*Many studies measured the budget of elements other than Fe in the burning of biomass. We collected the budget measured for elements whose physical and chemical properties are similar to those for Fe (e.g. low volatility). In the literature, the percentage of the element transfer to the atmosphere based on the element present in initial fuels was converted to that based on that in burnt fuels using the completeness of combustion (Raison et al., 1985; Pivello and Coutinho, 1992; Mackensen et al., 1996; Holscher et al., 1997). Raison et al. (1985) reported that 44-59% of the manganese in burnt fuel was transferred to the atmosphere in three prescribed vegetation fires (f = 41-56%). Pivello and Coutinho (1992) reported that 63% of the potassium, 76% of the calcium and 61% of the magnesium in burnt fuel were transferred to the atmosphere in a Brazilian savanna fire (f = 24-39%). Mackensen et al. (1996) reported that 18-51% of the potassium in burnt fuel was transferred to the atmosphere for two plots of forest fires in eastern Amazonia (f = 49-82%). Holscher et al. (1997) reported that 50% of the calcium and 57% of the magnesium in burnt fuel was transferred to the atmosphere during a deforestation fire in Brazil (f = 43-50%). Laclau et al. (2002) reported that 61% of the potassium, 79% of the calcium and 72% of the magnesium were bound in residue ash in the complete combustion of leaf litter from the littoral savannas of Congo (f = 61-79%). Chalot et al. (2012) reported that 70% of the copper and 55% of the zinc in all combustion products were bound in residue ash in the combustion of phytoremediated wood (f = 55-70%). In summary, we assumed that the partitioning of Fe is similar to these analogue elements, and applied a fraction of Fe in residue ash in burnt fuel (f) of 49-82% for forest fires (Mackensen et al., 1996; Chalot et al., 2012), 24-79% for savanna fires (Pivello and Coutinho,1992; Laclau et al., 2002), 43-50% for deforestation (Holscher et al., 1997), and 41-56% for woodland and peat fires (Raison et al., 1985)*.".

Reference:

Chalot, M., Blaudez, D., Rogaume, Y., Provent, A. S., and Pascual, C.: Fate of trace elements during the combustion of phytoremediation wood, Environ. Sci. Technol., 46, 13361–13369, doi:10.1021/Es3017478, 2012.

Holscher, D., Moller, R. F., Denich, M., and Folster, H.: Nutrient input-output budget of shifting agriculture in Eastern Amazonia, Nutr. Cycl. Agroecosys., 47, 49–57, 1997.

Mackensen, J., Holscher, D., Klinge, R., and Folster, H.: Nutrient transfer to the atmosphere by burning of debris in eastern Amazonia, Forest Ecol. Manag., 86, 121–128, doi:10.1016/S0378-1127(96)03790-5, 1996.

Pivello, V. R. and Coutinho, L. M.: Transfer of macro-nutrients to the atmosphere during experimental burnings in an open cerrado (Brazilian Savanna), J. Trop. Ecol., 8, 487–497, doi:10.1017/S0266467400006829, 1992.

Raison, R. J., Khanna, P. K., and Woods, P. V.: Mechanisms of element transfer to the atmosphere during vegetation fires, Can. J. Forest Res., 15, 132–140, doi:10.1139/X85-022, 1985.

Comment 10

p. 7650, l.21: What is the range of the fraction of Fe emitted in particle size (J_X)? Please show the values of JX, which were used to estimate Fe emissions in Table 3. p. 7651, l.5: What is the range of b? Please show the values of b, which were used to estimate Fe emissions in Table 3.

Response

According to a suggestion by another reviewer, all parameters (including J_x and *b*) used in estimating the Fe emissions are listed in **Table 1** in the revised manuscript.

Table 1. Parameters used in the estimation of Fe emissions from combustion sources.

Parameter	Description	Values or data sources
a	Fuel consumption	The fuel data was taken from a global 0.1°×0.1° fuel data set which is used to construct a global CO_2 emission inventory (Wang et al., 2013; available at http://inventory.pku.edu.cn/home.html).
b	Completeness of combustion	■ coal (98%); ■ petroleum (98%); ■ wood in stoves (88%); ■ wood in fireplaces (79%); ■ crop residues (92%);

		■ biomass burning (considered in van der Werf, 2010).
c	Fe content of the fuel	■ coal: based on Fe contents in coal produced by country (**Table S2**) and an international coal-trading matrix (Chen et al., 2014); ■ wood (a geometric mean of 0.036% and range in **Fig. S1**); ■ crop residues (a geometric mean of 0.060% and range in **Fig. S1**); ■ grass (a geometric mean of 0.027% and range in **Fig. S1**); ■ dung cakes (0.13 ±0.09 %); ■ biodiesel (0.00024 ±0.00023 %); ■ heavy fuel oil (32 ±2 ppm); ■ diesel (13 ±7 ppm); ■ gasoline (3.3 ±2.6 ppm); ■ liquefied petroleum gas (4.9 ±3.3 ppm).
f	Fraction of Fe retained in residue ash relative to the amount of Fe in the burnt fuel	■ coal used in industry and power plants (30-45%); ■ petroleum used in industry and power plants (43-58%); ■ solid biofuels used in industry and power plants (60-70%); ■ petroleum consumed by motor vehicles (2-12%); ■ anthracite coal used in the residential sector (99.2-99.8%); ■ bituminous coal used in the residential sector (91-97%); ■ straw used in the residential sector (79-95%); ■ wood used in the residential sector (89-99%); ■ forest fires (49-98%); ■ savanna fires (24-79%); ■ deforestation (43-50%); ■ woodland fires / peat fires (41-56%).
J_x	Fraction of Fe emitted in a particle size	■ coal fly ash (0.1-0.3% in PM$_1$; 10-30% in PM$_{1-10}$; the remainder in PM$_{>10}$); ■ oil fly ash (80-95% in PM$_1$; the remainder in PM$_{1-10}$); ■ biomass fly ash (1-3% in PM$_1$; 50-60% in PM$_{1-10}$; the remainder in PM$_{>10}$).
A_y	Fraction of a specific control device	A_y is computed for each country and each year using a function by Grubler et al. (1999) and Bond et al. (2007): $A_y = (F_0-F_f)$ exp $[-(t-t_0)2/2s^2] + F_f$, where F_0 and F_f are the initial and final fractions of the technology, t_0 is transition beginning time, and s is transition rate. Parameters were determined for developing or developed countries and listed in Wang et al. (2014a).
$\mathbf{R}_{\mathbf{x,y}}$	Removal efficiency for each particle size by different control device (Zhao et al., 2008)	■ cyclone (10% for PM$_1$; 70% for PM$_{1-10}$; 90% for PM$_{>10}$); ■ scrubber (50% for PM$_1$; 90% for PM$_{1-10}$; 99% for PM$_{>10}$); ■ electrostatic precipitator (93.62% for PM$_1$; 97.61% for PM$_{1-10}$; 99.25% for PM$_{>10}$).

Comment 11

The combustion factor (b) is included in the calculation of fuel consumption for biomass burning. Why did you multiply fuel consumption by b for biomass burning?

Response

We thank the reviewer very much for the comment. We realized that GFED3.1 (used in developing our fuel inventory) has applied the completeness of combustion, which is equivalent to b in equation 1. Accordingly, the sentence on line 6 were revised as: "*Fixed completeness of combustion (b) were assigned to coal (98%), petroleum (98%), wood in stoves (88%), wood in fireplaces (79%) and crop residues (92%) (Johnson et al., 2008; Lee et al., 2005; Zhang, et al. 2008). As the fuel consumptions for biomass burning have already accounted for the completeness of combustion based on the type of fires (van der Werf et al., 2011), we applied a combustion completeness of 100% for them.*". Fe emissions from biomass burning and related simulations were all updated.

Reference:

van der Werf, G. R., et al.: Global fire emissions and the contribution of deforestation, savanna, forest, agricultural, and peat fires (1997–2009), Atmos. Chem. Phys., 10, 11707–11735, 2010.

Comment 12

2.2 Fe contents in fuel. p. 7651, l.23: Crop residues are not shown in Fig. S1. Please correct it.

Response

Sorry for the confusion. In the previous version, we show the grass and crop residues in one group. In the new version, grasses and crop residues are plotted separately in **Fig. S1**.

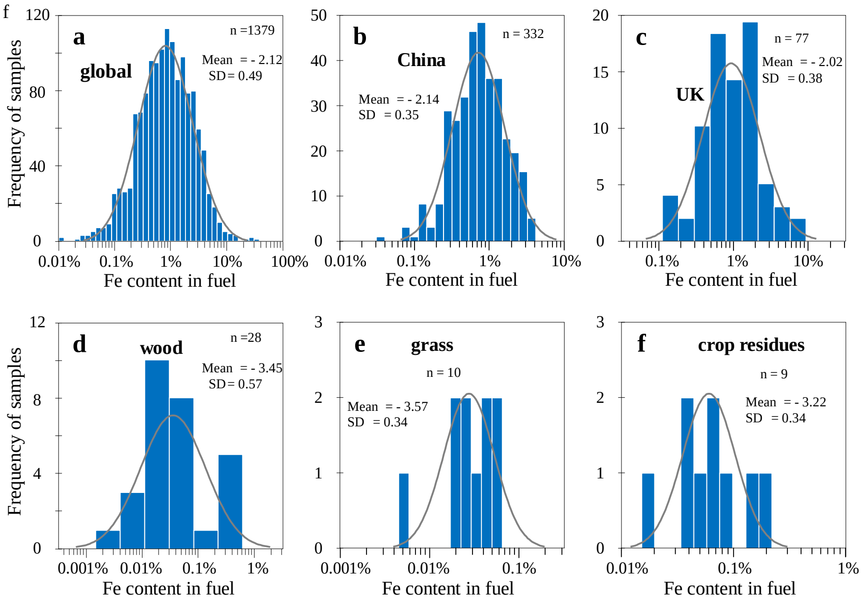

Figure S1. Frequency distribution of Fe contents in coal (a-c), wood (d), grass (e) and crop residue (f). The number of samples (n) and the mean and standard deviation (SD) of log$_{10}$-transformed Fe content are shown in each panel. The x-axis is plotted on a log scale.

Comment 13

Please show the mean and standard deviation of Fe content used for biomass burning.

Response

Sorry for missing the information. We applied the Fe content in grass for the savanna and grassland fires, and the Fe content in wood for the deforestation, forest, woodland and peat fires. The following sentence was added on line 24, page 7651: "*We applied the Fe content in grass (-3.57±0.34 for log*$_{10}$*-transformed Fe content) for the savanna and grassland fires and the Fe content in wood (-3.45±0.57) for the deforestation, forest, woodland and peat fires.*".

Comment 14

The values of Fe contents derived for woods and grasses are close to the average value of 3.5% in upper crustal materials. How did you deal with the contamination?

Response

Sorry for that we had not made x-axis of **Fig. S1** very clear in the previous version. The x-axis is showing the log$_{10}$-transformed Fe content as a percentage. Thus, the geometric mean Fe content is 0.036% for wood and 0.027% for grass (much lower than that in crustal materials). To make it clear, we re-set the x-axis without percentage in **Fig. S1** (please see a new **Fig. S1** in our response to **Comment 12**).

Comment 15

2.3 Partitioning of Fe in combustion. p. 7653, l.14: The percentages of Fe emitted into the atmosphere from the burning of straw (3%) and wood (6%) in residential stoves or fireplaces are much lower than those from open fires. It is likely that the majority of Fe in plant materials is retained in residue ash. How did you separate Fe in soil minerals entrained by pyro-convection from that in fly ash for biomass burning?

Response

Thank you for the good comment (the percentage of Fe transferred to the atmosphere from burning of straw is 13%, rather than 3%). First, for biomass burning, our estimate is based on the difference of Fe initially present in the fuel and that in the post-burn residues (Raison et al., 1985; Pivello and Coutinho, 1992; Mackensen et al., 1996; Holscher et al., 1997), rather than on the content of Fe in the atmosphere, so it is not influenced by pyro-convection of Fe from soil minerals to the atmosphere. Second, regarding the lower percentage of Fe transferred to the atmosphere in the residential stoves than that for biomass burning in the field, we propose that wind is a possible reason. According to Pivello and Coutinho (1992), wind can uplift ashes in the wildfires to the atmosphere, which is a key factor to explain the much higher percentage of element transferred to the atmosphere in the burning of biomass in the field than that measured in a furnace. Accordingly, the following sentence was added on line 18, page 7653 in the text: "*Here, the percentage of Fe transferred to the atmosphere for biomass burning in the field is larger than that in the residential stoves (see values above), and this is likely due to the wind, which can uplift more ashes into the air in the case of wildfires (Pivello and Coutinho, 1992).*".

Reference:

Chalot, M., Blaudez, D., Rogaume, Y., Provent, A. S., and Pascual, C.: Fate of trace elements during the combustion of phytoremediation wood, Environ. Sci. Technol., 46, 13361–13369, doi:10.1021/Es3017478, 2012.

Holscher, D., Moller, R. F., Denich, M., and Folster, H.: Nutrient input-output budget of shifting agriculture in Eastern Amazonia, Nutr. Cycl. Agroecosys., 47, 49–57, 1997.

Mackensen, J., Holscher, D., Klinge, R., and Folster, H.: Nutrient transfer to the atmosphere by burning of debris in eastern Amazonia, Forest Ecol. Manag., 86, 121–128, doi:10.1016/S0378-1127(96)03790-5, 1996.

Pivello, V. R. and Coutinho, L. M.: Transfer of macro-nutrients to the atmosphere during experimental burnings in an open cerrado (Brazilian Savanna), J. Trop. Ecol., 8, 487–497, doi:10.1017/S0266467400006829, 1992.

Raison, R. J., Khanna, P. K., and Woods, P. V.: Mechanisms of element transfer to the atmosphere during vegetation fires, Can. J. Forest Res., 15, 132–140, doi:10.1139/X85-022, 1985.

Comment 16

2.7 Modelling the atmospheric transport and deposition of Fe aerosols. p.7656, l. 19: Please indicate the uncertainty and clarify if the differences in national emissions between different years are statistically significant. How did you assign the concentration over the ocean to specific country? If you can run the model for the 2000-2011 dust emissions, why do you need this scaling?

Response

Thank you for the comment. In our study, we have scaled Fe concentrations from combustion at land sites based on the Fe emissions from combustion in the corresponding country, and at oceans sites based on the global total Fe emissions from combustion. According to the comment, we show the results without this scaling (see our response to **Comment 17**). There is very minor change in the comparison of modelled and observed Fe concentrations, indicating that the variation of Fe concentrations at the measuring sites is dominated by the variation of Fe in space.

In addition, when modelling the Fe concentrations from dust, we have not accounted for the impact of land use change on the dust emissions at present stage, and thus we used the average Fe concentrations over 2000-2011 when comparing against observations.

To clarify the methods and uncertainties, the last paragraph in Section 2.7 was revised as below (the variations of annual Fe emissions from combustion among different years were provided for major regions): "*Running the model for the whole period 1990-2007 was too heavy computational. Therefore, we run the model for one representative year. We plan to run the simulations for more years for a future study. In the present study, simulations were run for a typical year (2005) for the Fe emitted from the combustion of coal (three size classes), petroleum (two size classes) and biomass (three size classes). The Fe emitted from combustion as monthly means averaged over 1990-2007 were used as an input to the model, which produces the distribution of Fe concentrations attached to aerosols in the surface layer of the atmosphere contributed by combustion sources. When evaluating the modelled Fe concentrations by observations, we added the Fe concentrations contributed by combustion sources and dust together. However, there is a notable temporal variation of the combustion-related emissions over this period. The coefficient of variation (defined as the standard deviation relative to the mean) of annual Fe emissions from combustion* *over 1990-2007 is 46, 28, 17, 22, 26 and 22% for Europe, North America, South America, Africa, Asia and Oceania, respectively. To account for the impact of the changing emissions, when comparing the model with observations, we scaled the modelled Fe concentration from combustion at each land site by the ratio of the national Fe emission in the year to the 1990-2007 average in the country, and then compare it with the measured concentrations. For sites in the oceans, we scaled the concentrations following the same method using the global total emissions. In addition, since the change of land use during the period has not been accounted for when estimating the dust emissions in the model, we used the average Fe concentration by dust over 2000-2011 when comparing against observations and estimating the average contribution to Fe concentrations by different sources. Therefore, uncertainties induced by the nonlinearity of Fe concentrations to emissions and the interannual variation of dust emissions have not been accounted for in our study, which should be notified when comparing the model against the observations.*".

Comment 17

Please show the results without this scaling.

Response

According to the comment, **Fig. 12** without this scaling was shown in **Fig. S5** with the following sentences added on line 9, page 7666 in the revised main text: "*Due to heavy computational load, we modelled the Fe concentrations from combustion in a typical year using the average Fe emissions during 1990-2007, and compared them with measurements during 1990-2007 by scaling the modelled Fe concentrations from combustion to a specific year with the temporal change of emissions at each site (Section 2.7). To investigate the influence of this scaling process, we compared the modelled Fe concentrations without scaling among the four groups of sites (see results in **Fig. S5**). As a result, without this scaling, there is very minor change in the comparison between the modelled and observed Fe concentrations with* R^2 *change from 0.73 to 0.72. It indicates that the variation of Fe concentrations among the measuring sites is dominated by the variation of Fe concentration in space.*".

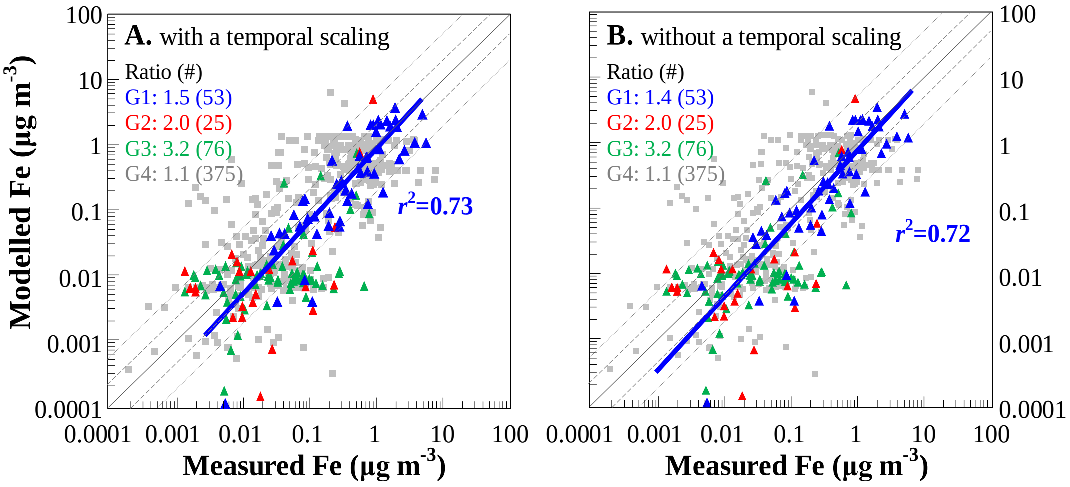

Figure S5. Comparisons of modelled and measured Fe concentrations attached to aerosols in surface air with (**A**) or without (**B**) applying a temporal scaling for the Fe concentrations from combustion based on the temporal change of the emissions. The four groups (G1, G2, G3 and G4) are the same as that shown in Fig. 12 in the main text based on the contribution of combustion to Fe concentrations: G1, contribution ⩾ 50% (blue triangles); G2, 30%⩽ contribution <50% (red triangles); G3: 15%⩽ contribution <30% (green triangles); G4, contribution <15% (grey squares). The ratios between measured and modelled concentrations as geometric means are listed with the number of stations in the brackets for each group. The fitted curves for the G1 stations are shown as blue lines with coefficients of determination (r^2).

Comment 18

3.5 Comparison of Fe emissions with previous studies. Table 1: Correct PM1-10 to PM1 for 0.035.

Response

Thank you very much for pointing out the mistake. The PM_{1-10} is corrected to PM_1 accordingly.

Comment 19

Please present the uncertainty and clarify if the differences from previous estimates are statistically significant.

Response

Thank you for the good comment. Accordingly, the 90% confidence interval was provided for our estimate in **Table 2**. However, we cannot perform a statistical analysis to compare our estimates with previous ones, because the uncertainty range was not estimated in previous studies. So, we compared our central estimates with those in previous studies while stating if their estimates are within our uncertainty range or not. Accordingly, the sentence on line 18-21, page 7660 was revised as: "*The estimate by Bertine and Goldberg (1971) for the same year (1967) is within the uncertainty range of our estimate (1.2-7.2 Tg yr*$^{-1}$ *as 90% confidence), but half of our central estimate (3.0 Tg yr*$^{-1}$*) after accounting for different removal efficiencies by particle size and control device.*"; the sentence on line 24, page 7660 was revised as: "*Their estimates of the total Fe emissions (1.7 Tg yr*$^{-1}$ *for 1996 and 2001) are close to our central estimates (1.6 Tg yr*$^{-1}$ *for 1996 and 1.3 Tg yr*$^{-1}$ *for 2001 with a 90% confidence range of 0.7-3.8 and 0.6-3.1, respectively).*"; the sentence on line 27, page 7660 was revised as: "*Their estimates of fossil fuel emissions (0.51 Tg yr*$^{-1}$ *for 1996 to 0.66 Tg yr*$^{-1}$ *for 2001) are within the uncertainty range of our estimate (0.56-2.46 Tg yr*$^{-1}$ *as 90% confidence for 1996 and 0.42-1.76 Tg yr*$^{-1}$ *for 2001), but lower than our central estimates (1.18 and 0.86 Tg yr*$^{-1}$ *for the two years, respectively) for the same size class ...*"; the sentence on line 25-28, page 7661 was revised as: "*In a recent study focused on East Asia (Lin et al., 2015), the emission of Fe from combustion sources in East Asia in 2007 was estimated to be 7.2 Tg yr*$^{-1}$*, far higher than all other studies (Luo et al., 2008; Ito, 2013) and the central estimate in our study (1.6 Tg yr*$^{-1}$*, with a 90% confidence of 0.66-3.84).*".

Comment 20

p. 7660, l.22: The total Fe emission should be corrected to 1.2 Tg/yr in 2001 (see above).

Response

Thank you very much for correcting the number. The right number is cited in the revised manuscript.

Comment 21

p. 7661, l.24: How did you confirm that dust material entrained in fires causes a likely overestimation? It is more likely that a lack of the dust material entrained by pyro-convection causes an underestimation. Please clarify this point.

Response

Thank you for this good comment. Now, we realized that our lower estimates of the biomass burning emissions of Fe than previous studies is likely due to the fact that previous estimates include the part by pyro-convection of Fe from soil and plant materials. In contrast, since we estimated the emissions based on the difference of Fe mass initially present in the fuel and that in the post-burn residues, rather than on the atmospheric concentration of Fe, we could miss the emissions of Fe by pyro-convection. Although our estimate provides a more explicit source attribution of Fe in the atmosphere, it could underestimate the total source. Accordingly, the sentences on line 20-24, page 7661 were revised as: "*Note that the dust and plant material entrained in fires can contribute to the Fe concentrations in the atmosphere, as noticed by Luo et al. (2008). As a result, their estimates include the pyro-convection of Fe from soils and plant materials. In contrast, our estimate is based on the mass balance of Fe from the burnt fuel. This might explain partly why our estimate of the biomass burning emission of Fe is lower than that in previous studies (Luo et al., 2008; Ito, 2013). Although our estimate provides an explicit source attribution of Fe, which is useful for modelling the Fe solubility, it underestimates the total sources. We propose that the emissions of Fe by pyro-convection in the fires should be estimated separately in the future.*".

Comment 22

4.2 Evaluation of Fe concentrations in surface air. p.7664, l.6: It is not clear whether the overestimation of Fe concentration occurs only at stations near continents due to high Fe content or high dust concentration. Please show the comparisons of Fe content and dust concentration separately to clarify this point.

Response

Since the Fe contents in dust and the concentrations of dust were not provided along with the measured Fe concentrations over the Atlantic ocean close to the Sahara in **Fig. 6** (Baker et al., 2006; Buck et al., 2010; Chen and Siefert, 2004), we cannot compare the Fe content and dust concentrations separately based on these data. In fact, there are very limited measurements of Fe content in dust. However, we compared the modelled Fe content in dust over the African desert with a measurement reporting the Fe content of dust in this region (Formenti et al., 2014). According to Formenti et al., at a site with a distance of ~2000 km to the Sahara, the measured Fe content in dust is 2.5-2.7% when the dust is originated from local erosion and 4.3% when the dust is originated from the Sahara. In comparison, the Fe content in our model is 4.5-5.5% over the desert, higher than 2.5-2.7% in the measurement, and 4-5% over the region distant from the desert, close to the measured 4.3%. Accordingly, **Fig. S4** (as below) was added to Supplementary Material, and the following sentence was added on line 6, page 7664 in the text: "*To illustrate this impact, the global distribution of Fe content in dust simulated by assuming that Fe content of emitted dust is equal to that in the clay fraction of soil is shown in **Fig. S4**. We can see that the Fe content in dust over the Sahara desert is 4.5-5.5%, which decreases with the distance to the Sahara. According to a measurement at a site with a distance of 2000 km to the Sahara desert, the Fe content in dust is 2.5-2.7% when the dust is originated from local erosion and 4.3% when dust is originated from the Sahara (Formenti et al., 2014). In the model, the Fe content is 4.5-5.5% over the Sahara desert, which is higher than the measured 2.5-2.7%, and 4-5% over the regions distant from the Sahara desert, close to the measured 4.3% in dust after a long-range transport from Sahara (Formenti et al., 2014).*".

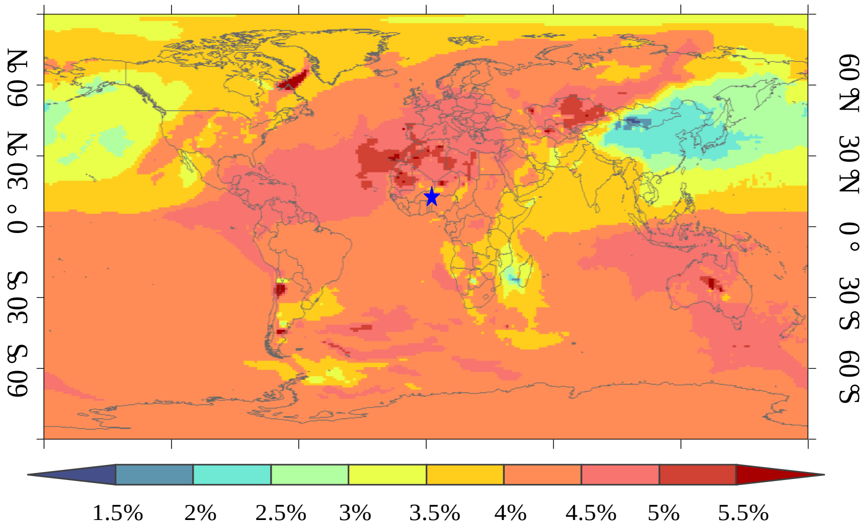

Figure S4. Global distribution of Fe content in dust simulated by assuming that Fe content of emitted dust is equal to that in the clay fraction of soil. The Banizoumbou ground-based site where Formenti et al. (2014) measured the composition of dust is shown as a blue pentagram.

Reference:

Baker, A. R., Jickells, T. D., Witt, M., and Linge, K. L.: 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, 2006.

Buck, C. S., Landing, W. M., Resing, J. A., and Measures, C. I.: 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, Marine Chemistry, 120(1), 57-70, 2010.

Chen, Y. and Siefert, R. L.: 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.-Atmos., 109, D09305, doi:10.1029/2003jd003958, 2004.

Formenti, P., Caquineau, S., Desboeufs, K., Klaver, A., Chevaillier, S., Journet, E., and Rajot, J. L.: Mapping the physico-chemical properties of mineral dust in western Africa: mineralogical composition, Atmos. Chem. Phys., 14, 10663–10686, doi:10.5194/acp-14-10663-2014, 2014.

Comment 23

4.6 Size distributions of Fe-containing particles. p.7668, l.1: It is not clear why the size of Fe was larger over the tropical ocean, due to more precipitation over the tropical regions. How does the precipitation increase the size of Fe?

Response

Sorry for the confusion due to a typo in the sentence. We meant to write that the size of Fe was smaller over the tropical oceans due to more precipitation over the tropical regions, because below-cloud scavenging removes large particles (diameter >10 μm) more efficiently than that for small particles (diameter of 0.1-1 μm). It is corrected in the revised manuscript.

Comment 24

6. Global atmospheric Fe budget. p.7669, l.17: Please directly compare the emission and deposition from combustion sources (see the supporting information of Ito, 2015).

Response

Thank you for the good comment. The sentences on line 15-19, page 7669 were revised as: "*We estimate that the annual deposition of total Fe over global oceans is 8.4 Tg yr^{-1} over the studied period (1990-2007). Among the total Fe deposition over the oceans, 93.1, 5.4, 1.4 and 0.13% was originated from dust (7.82 Tg yr^{-1}), coal combustion (0.455 Tg yr^{-1}), biomass (0.122 Tg yr^{-1}) and oil combustion (0.011 Tg yr^{-1}). In a recent global study modelling the Fe solubility, Ito (2015) estimated a larger deposition of Fe from mineral dust (13 Tg yr^{-1}), biomass burning (0.14 Tg yr^{-1}) and oil combustion (0.02 Tg yr^{-1}), but a lower deposition of Fe from coal combustion (0.16 Tg yr^{-1}), mainly due to the difference in the estimation for the sources (**Table 2**).*"

Comment 25

7 Influence of different Fe solubilities from different sources. Please show the comparison of soluble Fe and Fe solubility with measurements.

Response

An explicit modelling of the Fe solubilities also requires a fine consideration of the atmospheric processing of Fe in the atmosphere, which is beyond the target of our present study. So we only emphasized the importance of considering the contribution from combustion on the soluble Fe concentrations. According to the comment, we have compared the modelled daily soluble Fe concentrations with observations over the Atlantic and Pacific Oceans (Baker et al., 2007; Sedwick et al., 2007; Buck et al., 2006; Buck et al., 2010) with or without accounting for the contribution of Fe from combustion sources (see the figure as below).

Accordingly, a new **Fig. 19** (as below) was added in the revised manuscript and the following sentence was added on line 14, page 7670: "*As illustrated in **Fig. 19**, consideration of Fe from combustion sources, with assumed constant Fe solubilities for different sources, can largely improve the modelled soluble Fe concentrations when comparing against observations at 176 sites over the Atlantic and Pacific oceans (Baker et al., 2007; Sedwick et al., 2007; Buck et al., 2006; Buck et al., 2010). Further improvement of the modelled soluble Fe concentrations requires an explicit modelling of the atmospheric processing of Fe emitted from mineral dust and combustion sources, which is beyond the target of present study.*"

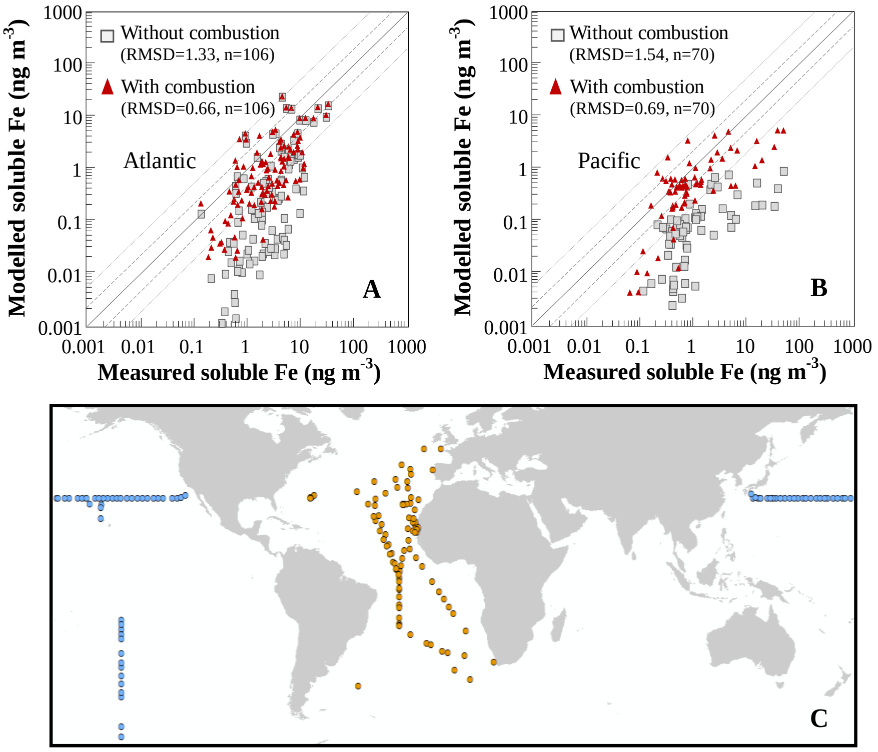

Figure 19. Comparison of modelled and observed soluble Fe concentrations over the Atlantic (A) and Pacific (B) Oceans. Model simulations were run for 2004 and 2005 for Fe emissions from dust, but with (red triangles) or without (grey squares) accounting for emissions from combustion. Fixed Fe solubilities were applied for Fe from mineral dust (0.44%) and combustion of coal (22.5%), oil (79%) and biomass (18%). The measured daily soluble Fe concentrations in 2004 and 2005 were compiled from the literature (Baker et al., 2007; Sedwick et al., 2007; Buck et al., 2006; Buck et al., 2010). The modelled and observed soluble Fe concentrations were compared on the same days. To evaluate the model performance, the root mean square deviations (RMSD) are computed for $\log_{10}$-transformed concentrations. The 1:1 (solid), 1:2 and 2:1 (dashed), and 1:5 and 5:1 (dotted) lines are shown. The locations of the measurement sites over the Atlantic (brown cirlces) and Pacific (blue circles) Oceans are shown in (**C**).

Reference:

Baker, A. R., Weston, K., Kelly, S. D., Voss, M., Streu, P. and Cape, J.N.: Dry and wet deposition of nutrients from the tropical Atlantic atmosphere: Links to primary productivity and nitrogen fixation, Deep-Sea Res. Pt. I, 54, 1704-1720, 2007.

Sedwick, P. N., Sholkovitz, E. R., and Church, T. M.: Impact of anthropogenic combustion emissions on the fractional solubility of aerosol iron: evidence from the Sargasso Sea, Geochem. Geophy. Geosy., 8, Q10Q06, doi:10.1029/2007gc001586, 2007.

Buck, C. S., Landing, W. M., Resing, J. A., and Lebon, G.T.: Aerosol iron and aluminum solubility in the northwest Pacific Ocean: Results from the 2002 IOC cruise, Geochem., Geophy., Geosy., 7, Q04M07, doi:10.1029/2005GC000977, 2006.

Buck, C. S., Landing, W. M., Resing, J. A., and Measures, C. I.: 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, 2010.