

Dear authors, thank you for the careful revision of your manuscript.

Please perform the following corrections (lines refer to the track changes document)

Summary Response: Thank you for your careful review of the manuscript. We have addressed all the comments and suggestions as outlined below. Please find our detailed responses and corresponding revisions (lines refer to the track changes document).

(1) - Line 110: “mol/m-3 (air)” please replace “/” by “.” or empty space

Response: Thank you for your suggestion. We have revised “mol/m⁻³ (air)” to “mol m⁻³ (air)” in line 110 as recommended.

(2) - Lines 112-114: please clarify in the text whether the concentrations used for pH calculations are monthly mean values or something else.

Response: Thank you for your suggestion. We have clarified the methodology for pH calculations as follows:

Line 114: “The aerosol pH is calculated based on H⁺ concentrations for each aerosol mode at each time step.”

(3) - Lines 137-138: please replace “According to the utilization of” by “Using”

Response: Thank you for your suggestion. We have replaced “According to the utilization of” with “Using” in line 138.

(4) - Lines 139-140: please replace the sentence starting by “Compared to the default setting of 3.5%,” by “The total iron content in dust aerosol in our model in the main dust sources, including North Africa, Middle East and Central Asia, is higher than the default setting of 3.5% (Fig. S1a)” and modify accordingly figure S1 caption.

Response: Thank you for your suggestion. We have revised the sentence in the manuscript and supplement as follows:

Line 140: “The total iron content in dust aerosol in our model in the main dust sources, including North Africa, Middle East and Central Asia, is higher than the default setting of 3.5% (Fig. S1a).”

Line 14 in the supplement: “Changes in dust total iron surface concentrations from the developed model compared to the setting of 3.5%, averaged over the 2001-2017 springs.”

(5) - Line 142: the observed DUST iron content

Response: Thank you for your suggestion. We have added “dust” to clarify the context. The revised phrase now reads:

Line 143: “the observed dust iron content.”

(6) - Line 144: suggest THAT

Response: Thank you for your suggestion. We have replaced “which” with “that” in

line 144 as recommended.

(7) - Line 154: is this eq 5a you refer to?

Response: Thank you for your suggestion. We have corrected the reference to “Eq. 5a” instead of “Eq. 5” in line 155.

(8) - Line 160: replace “have been shown” by “are shown”

Response: Thank you for your suggestion. We have replaced “have been shown” by “are shown” in line 161.

(9) - Line 161: replace “Through the annually averaged comparison of” by “The comparison of annually averaged...”

Response: Thank you for your suggestion. We have restructured the sentence as follows:

Line 162: “The comparison of annually averaged accumulation mode aerosols’ pH with observations collected by Pye et al. (2020) shows that...”

(10) - Line 162: add after the reference “that”

Response: Thank you for your suggestion. We have restructured the sentence as follows:

Line 162: “The comparison of annually averaged accumulation mode aerosols” pH with observations collected by Pye et al. (2020) shows that...”

(11) - Line 164: please replace “by” by “to”

Response: Thank you for your suggestion. We have replaced “by” by “to” in line 165 as recommended.

(12) - Lines 181,182: please specify if it is upper or lower limit threshold

Response: Thank you for your suggestion. We have specified the upper threshold in line 181.

(13) - Lines 185,186: “The locations and months are consistent between observations and the model.” Are monthly mean modelled values are used? What about the year of the observations is the same with the simulated values or you consider this as climatological mean? Please describe precisely what you have done.

Response: Thank you for your suggestion. We have clarified that monthly mean modelled values were used, and the observations were treated as climatological means over the period 2001-2017. The revised text reads:

Line 186: “The locations and months are consistent between observations and the model. We utilized monthly mean modelled values, averaged climatologically over the period 2001-2017.”

(14) - Line 272- 274: The observations of iron used here ARE the total iron which INCLUDES dust ... between simulation and observations IS shown ...Our model only CALCULATES IRON SOLUBILITIES BETWEEN 0 and 10%

Response: Thank you for your suggestion. We have revised the sentence as follows:

Line 271: The observations of iron used here are the total iron which includes dust and pyrogenic iron. ... What's more, the comparison about iron solubility between simulation and observations is shown in Figure S7. Our model only calculates iron solubility between 0 and 10%.

(15) - Lines 328 - 332: here it seems that you compare regional (NWP) with global budgets. Please clarify. This is particularly important because your model also performs well for OXL concentrations over East Asia but not elsewhere as shown in your figure S4b.

Response: Thank you for your suggestion. The primary role of oxalate is illustrated by our model both in the NWP and globally (Table 2 and line 287-293). We have clarified the comparison as follows:

Line 324: “The contribution of the oxalate-promoted processing was about twice that of proton-promoted processing in the NWP. This finding is consistent with global results (Table 2) and aligns with previous global modelling (Johnson and Meskhidze, 2013; Scanza et al., 2018) and East Asian observational research (Shi et al., 2022). Differently, As the oxalate concentrations appear to be underestimated in their model and the simulated mainland coarse-mode aerosol acidity in our model (Figure S2) is obviously lower than those. Furthermore, regional differences can also play a role, as our model performs well for oxalate concentrations over East Asia but not elsewhere (Figure S4). Future studies should ...”

(16) - Line 347-348: Please specify to which period the rate corresponds. Is this a decrease over the studied period (then it is not a rate).

Response: Thank you for your suggestion. It is the variation in 2017 compared to 2001 spring. We have revised the sentence as follows:

Line 345: “However, the amount of soluble iron deposition produced from atmospheric processing showed a much lower decrease (18%) in 2017 compared to 2001 spring (Figure S9). The coarse-mode proton-promoted soluble iron deposition even increased by 7% as shown in Fig. S9d.”

(17) - In the supplement, please change “oxalate-ligand” to “oxalate” everywhere

Response: Thank you for your suggestion. We have changed “oxalate-ligand” to “oxalate” throughout the supplement.

(18) - Figure S2: explicitly write in the caption that 1) the spatial distribution shown is near-surface, 2) the relationship shown in (b) is for fine aerosol, 3) how the aerosol pH was calculated (using monthly mean concentrations ?)

Response: Thank you for your suggestion. We have updated the caption to explicitly state the details as follows:

Line 18 in supplement: “Figure S2. (a) Spatial distribution of surface aerosol pH in

accumulation mode in 2013 and observationally estimated ground-level fine-aerosol pH (dots) from Pye et al. (2020). (b) The linear relationship between simulated surface aerosol pH in accumulation mode and observationally estimated ground-level fine-aerosol pH. (c) Spatial distribution of surface aerosol pH in coarse mode in 2013. The aerosol pH is calculated based on H⁺ concentrations for each aerosol mode at each time step.”

(19) - Explain colored symbols in Figure S4 caption

Response: Thank you for your suggestion. We have added an explanation of the colored symbols.

Line 36 in the supplement: “(b) The comparison between estimated oxalate concentration in cloud water and observations. Red circles represent locations in East Asia (EA), and green circles represent locations elsewhere.”

(20) - Figure S10 caption: Interannual variation of the percent contribution of each iron solubilization process to the total soluble iron deposition

Response: Thank you for your suggestion. We have revised the caption as follows:

Line 63 in the supplement: “Figure S9. Interannual variations of dust soluble iron deposition from proton-promoted (a, d), oxalate-promoted (b, e) and emissions (c, f) in coarse and fine mode (atiken + accumulation) to the Northwest Pacific averaged of 2001-2017 springs.”

Line 68 in the supplement: “Figure S10. Interannual variations of contribution of each iron solubilization process including proton-promoted (a, d), oxalate-promoted (b, e) and emissions (c, f) in coarse and fine mode (atiken + accumulation) to the dust total iron deposition to the Northwest Pacific averaged of 2001-2017 springs.”

(21) - Figure S13 caption: “high production rate of ... area (...)” do you mean “area of high contribution of...”

Response: Thank you for your suggestion. We have updated the sentence as follows:

Line 83: “over the area of high production rate of proton-promoted soluble iron”

(22) - Figure S14 caption: CHANGES IN THE spatial distribution

Response: Thank you for your suggestion. We have added the “Changes in the” to the caption in line 87.