

Reply to Reviewer 1's Comments

General comments: The authors implemented an updated thermodynamic equilibrium module, two wind-blown dust emissions schemes, and 9 dust related heterogeneous reactions into CMAQv4.7. They applied the model to simulate air quality for a dust storm in April 2001, conducted nine different simulations, and described the results. The article is well prepared and merits publication. However, several issues need to be addressed before publication.

Reply: We thank the reviewer for positive comments. We have addressed all the comments. Please see below our point-by-point replies highlighted in bold.

Specific comments are given below: Specific Comments

Comment #1 Page 13466, first paragraph, line 1: Variable “w” - the threshold gravimetric soil moisture has a prime in the equation but not in the explanatory text.

Reply: A prime sign was missing for the second “w”. We have fixed it.

Comment #2 Page 13468, second paragraph, line 10: Eqs. (1), (2), and (14) have been referred for dust flux generation. Please check these equations; they are not the correct equations for dust flux.

Reply: It was a typo and should be “Eqs. (1) and (11)”.

Comment #3 Page 13473, equation for scattering coefficient: $[SO_4]$, $[NO_3]$, etc, are not defined.

Reply: Those variables are now defined in the updated manuscript.

Comment #4 Page 13476-13477, section 4.2.1: Based on the results presented here, can the authors make any suggestion on the dust emissions scheme to be used in regional air quality models?

Reply: Both schemes predicted a similar spatial pattern and also total dust emissions. However, the Zender scheme is more physically-based than the Westphal scheme and also gives a better model performance (see new evaluation results in terms of surface dust concentrations and other species concentrations between simulations with Zender and Westphal schemes in Sect. 4.2.2 in the revised manuscript), we would recommend the

Zender scheme for dust emission estimate in air quality models. The above points have also been added in the second paragraph of Sect. 6.

Comment #5 Page 13477, second paragraph, line 14-17: Dust is emitted from surface. Concentrations are expected to be higher near the source. However, it indicates the total concentrations of dust at 5 km are higher than the surface. What is causing the total concentration aloft to be higher than the source region?

Reply: Reviewer is correct about that the concentrations of dust at surface are higher than those aloft near the source region. However, over the downwind and remote areas the dust concentrations aloft are higher than those at surface. This is because that most dust particles at or near surface have been deposited at and near source regions and cannot be transported for a long distance. On the other hand, dust particles with diameters $< 20 \mu\text{m}$ may be uplifted and transported for a long distance, leading to higher concentrations aloft than at surface over the remote areas far away from the source regions, as shown in Figure 1 (c)-(f).

Comment #6 Page 13479, 13480: The authors describe the importance of crustal species here but comparison of the predicted crustal materials with any observed data is not provided. Some crustal materials are measured in the US (IMPROVE and STN sites). The section would have benefitted from such a comparison. I am not suggesting the authors to provide such a comparison. But an acknowledgement that such a comparison has not been done is needed.

Reply: This is a good point. We have indicated a need for this comparison in the future work.

Comment #7 Page 13479, second paragraph, line 15-20: One sentence indicates that the two simulations show very small differences for non-volatile species like sulfate (line 15). While the next sentence indicates that the CRUST_ONLY predicts relatively lower sulfate over the East Asia. Need to reconcile these statements.

Reply: “non-volatile species such as SO_4^{2-} ” has been deleted.

Comment #7 The prediction of lower sulfate for CRUST_ONLY is explained by less oxidation of SO₂ into sulfate. The lower oxidation can occur via lower OH, H₂O₂ or other oxidants that converts SO₂ into sulfate. In the CRUST_ONLY simulation, the crustal materials affect the model results via the updated ISOROPIA. It is appropriate to discuss how the updated ISPROPIA affects these oxidants and subsequently the SO₂ oxidation.

Reply: More discussions on this have been added in Section 5.1.

Comment #8 Page 13480, first paragraph, line 15: Here, the mixing ratio of gas-phase NO₃⁻ is referred; should it be NO₃?

Reply: It was a typo and should be NH₃.

Comment #9 Page 13503, Table 6: It is not clear which columns are for Beijing and which columns are for Japan. Need to distinguish the columns for Beijing and Japan.

Reply: A footnote has been added to Table 6 to distinguish the data over Beijing and Japan.

Comment #10 Figure 4, 6, 7, 8, 9: Legends and numbers are difficult to read; bolder and larger fonts are helpful.

Reply: Those plots were generated by the PAVE software. It is not possible for the user to change the font types and size, since they are default font size. The legends and numbers can be readable by zooming in for the electronic version or for the printed version.

Comment #11 Page 13510, Figure 6: The spatial distribution of the differences between DUST and CRUST_ONLY for NO_x (second panel on the figure) shows that the heterogeneous reactions mostly increase NO_x. However, the spatial distribution of the differences between DUST_HIGH_UPTAKE and CRUST_ONLY for NO_x shows that they increase NO_x in some areas while decreasing in other areas. Please explain the reason for such behavior.

Reply: The plot for NO_x from DUST_HIGH_UPTAKE was incorrect and we have replaced it with the correct one. Both plots show a similar trend now.

The spatial distribution of the differences between DUST and CRUST_ONLY for H₂O₂ (third panel on the figure) shows that the heterogeneous reactions increase H₂O₂. However, the spatial

distribution of the differences between DUST_HIGH_UPTAKE and CRUST_ONLY for H_2O_2 shows that they decrease H_2O_2 . Please explain the reason for such behavior.

Reply: As indicated in Sect. 5.2, the increase of H_2O_2 was due to the conversion of HO_2 to H_2O_2 on dust particles. Although H_2O_2 is also absorbed by dust particles, the uptake coefficient is rather small. In contrast, in the simulation DUST_HIGH_UPTAKE, the H_2O_2 level decreases. This is because of the use of an uptake coefficient that is much higher for H_2O_2 than for HO_2 as compared with those used in the CRUST_ONLY simulation (it is increased by a factor of 20 for H_2O_2 but only by a factor of 2 for HO_2). Therefore, the rate of loss of H_2O_2 via the uptake process to the surface of dust particles is much larger than the rate of reproduction of H_2O_2 from the conversion of HO_2 on dust particles, leading to a new decrease in the H_2O_2 level as shown in Fig. 6. This has been indicated in lines 24-25 in page 13481.

The spatial distribution of the differences between DUST and CRUST_ONLY for NO_3^- (fourth panel on the figure) shows that the heterogeneous reactions decrease NO_3^- . However, the spatial distribution of the differences between DUST_HIGH_UPTAKE and CRUST_ONLY for NO_3^- shows that they increase NO_3^- . Please explain the reason for such behavior.

Reply: The small reduction of NO_3^- has been explained in lines 15-21 in page 13481. An increase in NO_3^- , which is expected, is due to the much higher uptake of NO_3^- precursors such as NO_2 , NO_3 , N_2O_5 , and HNO_3 on dust particles. This has been indicated in line 17 in page 13482.