Reply to Reviewer 2's Comments

General comments

This paper presents the implementation of two online dust emission parameterization schemes, the replacement of the thermodynamic module ISORROPIA with the newer version ISORROPIA II and also the inclusion of heterogeneous reactions of gaseous species at the surface of dust particles in the CMAQ model (US Environmental Protection Agency Community Multiscale Air Quality). The new system has been applied to simulate a dust storm event in Asia on April 2001, performing 9 simulations with different configurations, in order to reveal the impact of the new development to several atmospheric chemical species and the improvement succeeded by this development. A small section is also devoted to the influence of Asian pollution on the air quality in the US. The title of the manuscript reflects the contents of the paper and is consider sufficient.

In terms of scientific quality and significance, the purpose of this work is in line with the current state-of-the-art in the field of atmospheric chemistry modelling research. The inclusion of the complex interactions between gases and aerosols into a regional atmospheric chemistry model is a significant effort towards understanding the uncertainties and the behaviour of aerosols in the atmospheric system. I am in favour of such research efforts and my comments on the manuscript are written with this in mind.

The authors have made a large effort to analyze and discuss a lot of aspects of the new development in the CMAQ model, from meteorology to chemical reactions. This has resulted in a text rather difficult to read with several inconsistencies that I will describe in detail in the specific comments section. Several errors have been made in the description of the dust emission schemes, and the authors must carefully correct them as this part of the manuscript is the core part of the new development. The effects of the new processes in the CMAQ model are multidimensional and the message from this work must be clear, stressing out the solid conclusions and the remaining uncertainties. The specific comments will help to clarify some issues that have arisen during the review process and strengthen the quality of the paper through recommendations on the presentation and the discussion of the results.

Reply: We thank the reviewer for positive comments. We have addressed all the comments, corrected some typos and errors, and clarified some confusions. Please see below our point-by-point replies highlighted in bold.

Specific comments

Abstract

Page 13459, lines 14-15: The total dust emission amount indicated in the abstract comes from the Zender or the Westphal scheme? Please include this information here.

Reply: The total dust emission amount is from the Zender scheme and the information has been added to the abstract.

Page 13459, lines 25-26: The increase of gases and aerosols due to Asian pollution mainly affects the Western US as indicated in Section 5.4. Please include this information in the abstract.

Reply: This information has been added in the abstract.

Introduction

Page 13461, lines 3-5: I suggest you include the recent publication of Crowley et al. (2010) (see the full reference in the end of this document) where a full assessment of the heterogeneous processes on surfaces of solid particles is presented.

Reply: The reference has been added.

Page 13461, lines 7, 8: The modelling work of Cheng et al. (2008) and Huneeus et al. (2011) did not include any chemistry on dust particles. You should not include these publications in the same list denoting the studies of heterogeneous chemistry on dust particles (line 9). The references on Tang et al. (2004), Fairlie et al. 2010 (already in your ref list), Astitha et al. (2010) could be part of this list.

Reply: The citation has been corrected based on suggestions.

Section 2.1

Page 13464, lines 8-17: I would suggest that the authors do not refer to the second emission scheme as the "Zender scheme" when discussing the physical concept and equations. Zender et al. (2003) clearly state that “We generally follow the microphysical and micrometeorological approach to dust mobilization developed by Marticorena and Bergametti [1995]”. The original work on the horizontal dust flux is done by White (1979) and also Kawamura (1964) and it should be mentioned here.

Reply: As indicated clearly in the original manuscript, in the Zender scheme, Eq. (7) came from White (1979), Eqs. (8)-(9) came from Iversen and White (1982), Eqs. (10)-(11) came from Marticorena and Bergametti (1995). All these original references were cited for the physical concept and equations used in the Zender scheme in our manuscript. Since those equations and concepts were assembled and applied together by Zender et al. (2003), along with some other updates (e.g., the application of source factor S, the specific value for global tuning factor T, and the assumption of the optimal particle size) by Zender et al. (2003), we think that it is appropriate to name the second dust emission scheme to be “the Zender scheme”. To address the reviewer’s comment, we have also added the reference of Kawamura (1964).

Page 13464, line 26: Please briefly explain how the Heaviside function H is used in the context of the dust emission flux (the physical meaning).

Reply: The meaning for this function is that dust particles can only be emitted when $u_* - u_{*t} \geq 0$. A brief explanation has been provided in the revised manuscript.

Page 13465, line 2: “RF=1.0” will result in zero dust emission flux (Equation 1) for barren land? Please correct the values of RF used in Equation 1.

Reply: It was a typo and it should be RF=0.1.

Page 13465, lines 2-9: The parameter EF, which is assumed to characterize the fraction of erodible land, is constant everywhere in the domain? Does it change depending on the soil properties in each arid area (roughness elements etc)? If not, then this parameter is mainly a tuning factor rather than a measure of the erodibility of the land.

Reply: Yes, EF is assumed to be constant everywhere. It mainly serves as a tuning factor, which can be improved to incorporate spatial variability of the erodibility of the land in

future work. This has been pointed out as one of the limitations of this work in the conclusion section.

Page 13465, line 10: I strongly disagree that a constant threshold velocity can be valid for any study on dust emissions, either regional or global. The dust emission process occurs on very small spatial scales and is directly related to the soil properties (mineralogy, roughness elements, smooth surface) and particle sizes (diameter of the particles capable to initiate saltation).

Reply: "might be valid for regional scale studies" has been removed.

Page 13465, Equation 2: The value 121 should be 1.21.

Reply: In the original equation of Fecan et al. (1999), w and w' represent the percentage gravimetric soil moisture (i.e., in a unit of %). However, in our formula and also computer codes, we used a unit of kg kg^{-1} for gravimetric soil moisture. So 121 should be the correct value for the units used in our study.

Page 13466, Equation 7: This equation must be wrong. The last terms "H(u*-u*t)" are not included in the dust flux formulation described in Marticorena and Bergametti (1995), Zender et al. (2003) and elsewhere. Furthermore, why did you use the parameter EF in this equation of the horizontal flux? In the calculation of the vertical flux (Equation 11) you already include the "source erodibility factor S". I don't see any physical meaning in adding one more constant factor in Eq.7 rather than a possible double-counting of the same concept.

Reply: $H(u_*-u_{*t})$ should be read as a function H(x) with u_*-u_{*t} as x (i.e., the independent variable). To avoid confusion, the term (u_*-u_{*t}) has been removed from the equation. Instead, the dependence of H on this term is described in the text. The parameter EF acts as a tuning factor in this study, rather than duplicating the function of source erodibility factor S.

Page 13466, line 21: There is no u*tl in the previous equation of the dust horizontal flux. The authors use this parameter also in Eqs. 8 and 9, which should be replaced by u*t.

Reply: u_{*tl} was defined in line 23 of page 13465.

Page 13467, Eqs 8 and 9: In Iversen and White (1982) some of the values used in the empirical relationship are different from the ones shown here (instead of 0.1666681 the value is 0.129 etc). Please explain why you changed these values or refer to the appropriate publication. In addition, there is an error in Equation 9, at the last term of the right side (gD2.5 belongs to the denominator). Please correct it accordingly.

Reply: 0.1666681 was a typo, it should be 0.01666681. In the original reference, the value of 0.1291 was placed out of the square root function. In our paper, we moved this value inside of square root function, which results in a value of 0.01666681; the term "$gD^{2.5}$" has been moved to denominator.

Page 13467, Eq 10: The Reynolds number calculated in equation 10 is a constant value since the diameter of the particle is fixed to 75_m. This means that the threshold friction velocity is also a fixed number and there is no need for both Eqs 8 and 9. The use of Eq.10 could be employed only at the 1st time-step when there is no initial value for u*t, and then Re*t= u*t D/v providing a small change in the calculation of the threshold friction velocity.

Reply: The reviewer is correct about Re_{*t}, which is a fixed value in this study. However, Eqs. 8 and 9 only provide an initial values of u_{*t} and the final value of u_{*t} will also be corrected for the effect of soil moisture by applying Eqs. (2)-(6). There was a typo in line 12 of page 13467. It should be Eqs.(2)-(6) instead of Eqs. (3)-(8). So the final u_{*t} is varied spatially. To address the reviewer's comment, we have clarified this in the revised manuscript.

Page 13467, line 12: Maybe you mean that u*t is calculated from Eq2 with u*tl from Eqs 8 or 9 and then used in Eq.7? Of course if the previous comment is true, there is no need for both equations 8 and 9. If my comment is precise then the dust flux calculated by Zender's approach is subject to correction for soil moisture as done for the Westphal. If so, please indicate that explicitly in the text.

Reply: Reviewer is correct about the correction for soil moisture for the Zender's approach, which has been indicated in this revised manuscript.

Page 13467, line 17: How do you calculate the “source erodibility factor” S (from an input table, a database, online calculation)? Add an explanation in the text.

Reply: It’s from a database provided by Ginoux et al. (2001), which was cited in the original manuscript and the text has been revised to indicate this.

Page 13467, Eq 12: This equation must be wrong. It is the same as used in Zender et al. (2003) but a careful comparison with the original equation in Marticorena and Bergametti (1995) will reveal the differences: The original equation is log(F/G)=0.134(%clay)-6 coming from linear fitting of data in Marticorena and Bergametti (1995). This means that _=F/G=10ˆ(0.134(%clay)-6) and for 10% of clay the sandblasting efficiency will be _2.18E-05 cm-1 (shown in Fig.4 of Marticorena and Bergametti (1995)). For Mclay=0.1, equation 12 gives _=2.18E-03 cm-1, two orders of magnitude different. If the authors have used this equation in their calculations they should carefully check the calculated sandblasting coefficient _ with published values and the original work.

Reply: In Marticorena and Bergametti (1995), α=F/G and has a unit of cm^{-1}. In this study, we use the SI units for all the formula and variables. In Eq. 12, α is actually in a unit of m^{-1}. So the calculation results are consistent between the original work and ours. To avoid confusion, we have added the unit for α in the text.

Page 13468, line 10: There is no Eq.14 in the text.

Reply: It was a typo and has been fixed in the revised manuscript.

Section 2.2 (Heterogeneous chemistry)

A recent publication by Crowley et al. (2010) is dedicated to the heterogeneous processes on surfaces of solid particles present in the atmosphere, for which uptake coefficients and adsorption parameters have been presented on the IUPAC (website in 2009). In this publication, data of uptake coefficients is evaluated and a recommendation is made for each reaction, based on several arguments presented in the paper. A reference to this work must be included in the text, as it is a recent study based on experimental data and it is closely related to this publication. Some of the uptake coefficients suggested by Crowley et al. (2010) for the uptake on mineral dust particles are quite different from the values used herein. A brief discussion on the consequences from using lower or higher values for the gamma coefficients compared to the ones in Table 2 would be very informative in the text.

Reply: The reference has been cited in revised manuscript. A brief discussion regarding the differences of uptake coefficients in Crowley et al. (2010) and this study was added in Sect. 2.2. Additional simulations may be performed using different sets of uptake coefficients such as those recommended by Crowly et al. (2010), which has been mentioned in the future work of Sect. 6

Section 2.3 (Crustal species-ISORROPIA II)

The mineralogy of the soil that prescribes the chemical composition of atmospheric dust particles is very important and still not available for all the arid areas of the world. The inclusion of ISORROPIA II is a very good step towards improving the thermodynamics of the aerosols in the model.

1. How do you treat the products from the heterogeneous reactions in the model, i.e. the sulfates produced on dust are added to the sulfates produced from gas to particle conversion or they are kept separately in the thermodynamics and the other aerosol processes in the model?

Reply: The aerosol products such as sulfates from heterogeneous reactions are added to sulfates produced from other gas-to-particle conversion, to be consistent with the internal mixing assumption for all aerosols used in CMAQ.

2. The ratios between crustal species and dust (page 13470, line 17) is assigned to the dust emitted flux or the concentration?

Reply: The ratios are assigned to the emitted flux. This has been indicated in the revised text.

Section 3 (Model configuration)

Page 13471, line 24: Please be more specific on the meteorological setup chosen for the 9 simulations described in Table 3. Is it WRF with FNL and nudging?

Reply: It is WRF with FNL and nudging for 10 simulations described in Table 3. This information has been added in lines 24 and 25 of page 13471.

Page 13471, line 25: The WRF and the CMAQ models use the same geographical projection or a necessary recalculation is performed?

Reply: They use the same projection.

Page 13472, line 11: Which dust emission scheme did the authors use for the simulations, Zender or Westphal? This is not stated anywhere in the next sections and the description of both schemes in Section 1 is not justified by the performed simulations or a discussion in the text.

Reply: For all the simulations with dust treatments in Table 3, except for DUST_W, the Zender scheme was used. This information has been clarified in the paragraph 4 of Sect. 3. To address the reviewer's comment, we have conducted a new simulation with the Westphal scheme called DUST_W. We also evaluated the performance of the simulations with both the Zender and Westphal schemes (see Sect. 4.2.2). The Zender scheme gives an overall better performance for all the aerosol related variables such as dust, $PM_{2.5}$, PM_{10}, SPM, and AOD, so we chose to use the Zender scheme to investigate the impacts of dust treatments and performed additional simulations with this scheme as described in Sect. 5.

Section 4 (Model evaluation)

Page 13476: The evaluation of the dust emission scheme used for the 9 simulations is missing from this section. Figure 2 with the monthly AOD plots is not adequate to form a solid evaluation of the dust scheme. This is very crucial as the main developments of this study rely on the dust emission online scheme embedded in the model. I would strongly suggest two ways the authors can deal with that: 1) use the data of dust concentration measurements in Asian sites published in Cheng et al. (2008). Data for April 2001 is included in that publication (10 stations out of the 16 give values for April 2001). 2) Use the daily AOD values from the AERONET Level 2 products (_13 AERONET sites in Asia have AOD values for April 2001). Selecting stations close to the desert areas will exclude the influence from anthropogenic sources and give a clear picture of the model performance. The AOD statistics presented in Table 7, (a spatial and temporal average on the domain), include all species with the new development and cannot account as an evaluation of the dust scheme.

Reply: These are very good suggestions. We have included more evaluation of dust schemes in Sect. 4.2.2 in the revised manuscript by using the data from Cheng et al. (2008)

(see Table 4) and also daily-average AOD data from AERONET (see Figure 3). The assessments show overall good results from CMAQ_Dust.

Section 5.2

Figure 6: The spatial differences between DUST and CRUST_ONLY show increase of H2O2 but the differences between DUST and DUST_HIGH_UPTAKE show decrease in H2O2. The same occurs for NOx and the opposite for NO3. This means that changing the uptake coefficients into higher values can have the opposite effect in some species, indicating the high uncertainty of the values. The authors should mention this in the text and try to elaborate on the reasons for this behavior.

Reply: The plot for NO_x from DUST_HIGH_UPTAKE was incorrect and we have corrected it. The new one shows a similar pattern. For H_2O_2, 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, 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. For NO_3^-, 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.

Conclusions

It is not clear if the 9 simulations were performed with the Zender or the Westphal scheme. Which of the two schemes performed better in terms of efficiently producing the dust distribution in the selected domain and which scheme would the authors recommend to the modellers using CMAQ/Dust in the future?

Reply: As we stated earlier, all simulations with dust treatments in Table 3 were performed with the Zender scheme except for DUST_W. As indicated in Sect. 2.1, the major difference between the two schemes is that the Zender scheme splits the dust flux into two components, horizontally saltating mass flux of large particles and vertical mass flux of dust, whereas the Westphal scheme calculates vertical fluxes directly. We incorporated both approaches in CMAQ-Dust to assess the sensitivity of dust emissions and their impacts to different dust flux parameterizations. However, the results show similar spatial patterns and very comparable total dust emissions between two schemes. The evaluation results also show that the Zender scheme gives an overall better performance than the Westphal scheme. We therefore chose the Zender scheme for the rest of simulations. Based on the above facts, we would recommend the Zender scheme for dust-related applications. This has been indicated in Sect. 6.

Technical corrections

Page 13459, line 18: "aerosol optical depth than the default CMAQ..."

Reply: It's been revised.

Page 13460, line 22: "treatments for dust blowing up or uplifting processes..." should be rephrased to "parameterizations of dust emission processes...".

Reply: It is rephrased as suggested by the reviewer.

Page 13461, line 11: "For example,...".

Reply: It's been revised.

Page 13465, line 22: "In the current version of the emission scheme, only the ...'

Reply: It's been revised.

Page 13466, line 1: There is a missing prime symbol in w (threshold gravimetric soil moisture).

Reply: It's been fixed.

Page 13468, line 1: "the mass faction" should be "mass fraction".

Reply: It's been fixed.

Page 13483, line 28: "...altitudes is much larger..."

Reply: It's been fixed.

Tables 4-7: The values "Data Number" and "Mean Observed" must be clearly stated and independent of the model configuration in the first column. In addition, in tables 4-6 the different data networks must be clearly separated. These actions will facilitate the proper reading of the tables.

Reply: Values for "Data Number" and "Mean Observed" have been clearly separated from other model configuration in the first column for those Tables. We have also added more footnotes to those tables to clearly separate different data networks.

Figure 2: Indicate the satellite that gave the monthly AOD plot (first row). Also the title of each plot does not correspond to the respective simulation (1st plot: should be MODIS AOD, if MODIS data is used etc).

Reply: The caption of Fig. 2 has been revised. The specific titles for each plot are added.

Figures 3-9: It would benefit the reader if a brief sentence on the scope of each figure was included in the legend. For example: In Figure 6 the sentence "Effects of heterogeneous chemistry" could be added.

Reply: All figure captions have been revised based on the suggested changes.

Figures in the supplement: The figures in the supplement need refinement as some titles are not shown and the borders of the plots are not uniform.

Reply: This has been fixed.