1) Referee #1
Comment 1: The partitioning between the gas and condensed phase are treated in a similar way, despite being fundamentally different in nature. For a solid surface, the adsorption and desorption processes do follow a different formalism, typically through a Langmuir-Hinshelwood formalism which takes into account a given number of adsorption sites. The consequences is that adsorption decreases with time or increasing concentration, while here it is simulated in a constant way with time. how can you justify such an assumption? Also products such as sulfate are probably staying on the surface, thereby also using adsorption sites i.e., poisoning the surface. How would your model change if you implement such time/concentration dependence?

Response: We assumed that the gas-particle partitioning onto dust is operated by an absorption process (Eq. 7) by several reasons (see section 3.2.1). First, unlike pure metal oxide which is governed by the adsorptive partitioning, the composition of authentic mineral dust such as Arizona Test Dust (ATD) is complex. The fresh ATD contains inorganic salts that are hygroscopic and form the water film above efflorescence relative humidity or deliquescence relative humidity. Second, the partitioning process is dynamic due to the formation of various hygroscopic salts of sulfate and nitrate due to the reaction of alkaline carbonates and metal oxides with inorganic acids (sulfuric acid and nitric acid). Third, the sulfate formation in our study increased as increasing humidity due to the dissolution of tracers into the water layer (see section 3.2.1). If partitioning is processed by the adsorptive mode, water molecules compete for the site with tracers and reduce partitioning of tracers (Cwiertny et al., 2008). The amount of the surface water on dust particles, which was measured using FTIR (submitted in the other journal), was multi-layered.

Editor: It seems to me that the only change in response to this comment was that you replaced all ‘adsorption’ by ‘absorption’ in the manuscript. However, this raises more questions than it adds to clarification. You argue that the hygroscopic fraction of the dust forms an aqueous phase where then the chemical processing in the ‘dust phase’ occurs. Thus, the extent of this processing will depend on the mass (volume) of the hygroscopic dust material. It is not clear to me why parameters such as the absorption rate constants (e.g. R5 and Equation 5) can be used that relate to a particle surface (m-2) and not to a (partial) particle volume.

This should be clarified throughout the manuscript. The text should be carefully checked for consistency (e.g. p. 15, l. 10 ‘adsorption-desorption’) and conclusions should be refined (e.g. why is BET surface area needed (p. 20, l. 26) if the hygroscopic material determines the reactions medium?)

2) Referee #1
Comment 4: Too many rate constants are estimated without any justification. Please justify and explain your estimations.

Response: Most of the rate constants shown in Table 3 were estimated using the indoor chamber data obtained in the previous study (Park and Jang, 2016). The rate constants of R10 (electron-hole production) and R11 (recombination of electron-hole) in the manuscript is estimated using Eq. 10 (photoactivation rate, J_{ATD}) in the manuscript (Section 3.2.3). The rate constant of R13 (reaction of SO_2 with dust-phase OH radicals) is set to the same reaction rate constant for the reaction of SO_2 with

OH radicals in gas phase. Without sunlight, autoxidation of SO_2 (R9) is dominant in dust phase and its rate constant was obtained from indoor chamber data under various humidity conditions (Exp. D1-D3 in Table 1). With sunlight, the photochemical reaction is the major source for sulfate production. Using the same approach with autoxidation, the rate constant of R12 was estimated under different humidity conditions. Also, the rate constants of R14 (heterogeneous autoxidation of SO_2 in the presence of ozone) and R15 (heterogeneous oxidation of O_3) were estimated using experiments D4 and L5 in Table 1, respectively. The rate constants of R18 (heterogeneous autoxidation of NO_2) and R19 (heterogeneous photocatalytic oxidation of NO_2) were estimated using experiments D5 and L7 in Table 1, respectively.

Editor: In my opinion, your response is neither a justification nor an explanation why so many rate constants were estimated. Please add a more detailed discussion of uncertainties and background on these constants in order to fulfill the reviewer's inquiry.

Also, I noticed that e.g. Kd,SO2 is used for dry particles – how can this be justified?

3) Referee #3:

Comment 2: In addition to react with SO_2 and NO_2, OH radicals produced on the surface of particles under UV conditions can undergo heterogeneous reaction with particles as well as self-reactions, resulting in the significant decrease of OH radicals participate in the oxidation of SO_2 and NO_2, and subsequently overestimating sulfate and nitrate formation in the model. Furthermore, in addition to compete OH radicals with SO_2, the presence of NO_2 can also react with SO_2 on the surface of particles to promote sulfate formation at high RHs as like in aqueous phase. However, these mechanisms were not considered in dust phase in the model (Table S1).

Response: In our model, the apparent rate constant of the formation of the dust-phase OH radicals is estimated using indoor chamber data. The synergistic effect of NO_2 on sulfate formation under UV light is explained by the HONO production through the reaction of NO_2 with electrons or holes in dust phase (R16). HONO will then be decomposed via photolysis to form OH radicals (R17).

Editor: The reviewer's concern of missing reactions in your mechanism is well justified. How would the recombination of OH or their loss on particle surfaces affect your results? Please add a discussion about what it is known about such processes and to what extent they may compete with other processes in your system.

4) Difference between dust chemistry and aqueous phase chemistry

Editor: Related to the comment above, I do not understand the fundamental difference between the aqueous phase chemistry (Section 3.1.3) and dust chemistry (Section 3.2). Both are reactions that occur in a bulk aqueous phase and thus mechanistically they should be treated equally (even though different chemical reactions occur). Please justify the differentiation into two types of processes.

It should be stated throughout the manuscript that the ‘dust phase’ is also technically an aqueous phase

5) Use of Henry’s law constants

a) What is the ionic strength and/or acidity of the aqueous phases? Is the application of Henry’s law constants (for ideal solutions) justified? If not, how does this affect the results and the possibility of extrapolation of the derived rate constants to other conditions?

b) Is the pH sufficiently low that the uptake of SO2 can be indeed solely described by the physical Henry’s law constant KH,SO2? This can be only applied if the solution is sufficiently acidic and no dissociation occurs; otherwise the effective Henry’s law constant including dissociation should be included. Please justify.

6) Language

The whole manuscript should be carefully checked for proper use of English language. I list some rather unusual or unclear expressions below (line numbers refer to the marked-up manuscript that was attached to the response of the reviews)

p. 6, l. 10: ‘calculated to mass absorbance’ – is there a word missing (e.g. ‘obtain’)?

p. 9, l. 3: What is a ‘carry over for sulfate’

p. 11, l. 11: ‘numeric number’ is redundant

Figure 2, y-axis should be ‘Uptake coefficient’