Response to Reviewer 1

We thank the reviewer for his/her careful evaluation of the manuscript. We especially appreciate the reviewer's comments providing references to recent work on uptake coefficients and mineralogy information. We think adding discussion on these topics has greatly improved our paper. Below we give a detailed response to each of the concerns raised by the reviewer. Reviewer's comments are in regular font and our replies are in bold font characters.

General comments

This study examines the effects of dust particles on tropospheric chemistry through the inclusion of heterogeneous reactions at the particle surface and the radiative effects on the photolysis rates. Several additions have been made to WRF-Chem to accommodate the new chemical reactions and the responsiveness of the photolysis rates at the presence of atmospheric dust particles. The paper is in line with the current state-of-the-science in the field of atmospheric chemistry. Even though a lot of similar applications have appeared in the literature, the area under investigation in India provides a new element that adds to the scientific knowledge on the aerosol processes in that specific region. The title of the manuscript reflects the content of the paper and is considered sufficient. I am in favor of publishing the paper with Atmospheric Chemistry and Physics, after carefully addressing the specific comments that follow.

Specific comments

The results from this study are highly related to one specific dust event (model simulation for 15 days in April 2010) and cannot be generalized for all dust events in the region. This should be acknowledged in the abstract and summary of the manuscript.

Dust storms in northern India are characterized by large increase in AOD (>50%) and decrease in α (>70%) (Dey et al., 2004; Prasad and Singh, 2007). Both these features were observed during this dust storm indicating that this was a typical pre-monsoon season dust storm (see Kumar et al., 2014 for further details) and can be considered as representative of dust storms in northern India. We already mentioned this dust storm as "typical" in both the abstract and the summary, and this is now acknowledged in section 4.1 also. However, following the reviewer's suggestion, we have added the following line in summary. "Although this study analyzed a typical dust storm in northern India but more such studies should be conducted in future to lend further confidence in these results".

How are the products from the heterogeneous reactions treated in the model? For example, 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? This is important to understand the results from the simulations.

The gases reacting with dust particles are assumed to be lost irreversibly on the particle surface but do not contribute to aerosol mass in the model configuration used here. This is

because we use the GOCART aerosol module, which does not include an aerosol thermodynamic module in WRF-Chem. We agree that heterogeneous uptake of trace gases by aerosols can potentially affect aerosol mass, composition and size distribution. However, the main objective of this study was to understand the impact of a dust storm on the levels of trace gases only. The GOCART model is suited to meet this objective. We have now included this information in section 2.2.

The discussion of aerosol thermodynamics in WRF-Chem is absent in the manuscript and should be included in the text. The product of the heterogeneous reactions at the surface of dust particles is a new particle, as described in section 2.3 (atmospheric aging). It can be sulphate or nitrate covering the dust particle, after the adsorption of the related gaseous compound. That new particle takes part in all aerosol processes (thermodynamics, deposition, advection etc.)? This should be clearly stated in the text.

As stated in our reply above, the GOCART aerosols scheme does not consider aerosol thermodynamics. In Section 2.3, we discuss the simplified treatment of atmospheric aging of dust aerosols in which aged dust particles are treated similar to the original dust particles and take part in advection and deposition processes but not in aerosol thermodynamics. The limitation of our study due to use of the GOCART model and need for a detailed aerosol module for future studies are already discussed in the second last paragraph of summary.

The 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 at the IUPAC (International Union of Pure and Applied Chemistry) website in 2009 (http://iupac.pole-ether.fr/). 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 work. A brief discussion on the consequences from using lower or higher values for the gamma coefficients compared to the ones in Table 1 should be included in the text. Sensitivity runs with different gamma coefficients would be a more appropriate way of assessing the uncertainties that are related with the adoption of one value.

We have included this reference in the revised manuscript. We also conducted two sensitivity simulations using lower and upper bounds of γ values reported in the literature. The results from these sensitivity simulations are now discussed in a new section (4.6) called "Effects of uncertainty in reactive uptake coefficient (γ)".

Another important aspect of the heterogeneous formation on the surface of dust particles is the mineralogy of the dust itself. The chemical composition of dust is essential for the realization of several reactions and for the behavior of the particle in the atmosphere. Especially for the HNO3 uptake, it is important to know how much CaCO3 is available as the reaction of HNO3 with CaCO3 has a different path than that with the rest of the minerals (Grassian 2002). There is no discussion on this aspect anywhere in the text and I strongly suggest to devote a part in this discussion. The mineralogy of dust particles is very difficult to measure worldwide and only recently we have some compilation of mineralogy databases from scientists in Europe and the US. These can be informative for assuming a percentage of the dust particle as CaCO3.

We thank the reviewer for bringing this to our attention. At least, two pathways of HNO_3 reaction on mineral dust particles are observed depending on the mineral with which HNO_3 is reacting (Grassian, 2002). Surface-limited reactions are observed when HNO_3 molecules react with Al_2O_3, Fe_2O_3, or TiO_2, whereas both surface and bulk reactivity are observed for HNO_3 reacting with $CaCO_3$. The first pathway leads to adsorption of HNO_3 molecules to the surface of dust particles thus making it available for renoxification processes while the second pathway leads to irreversible removal of HNO_3 molecules. Peterson (1968) estimated that dust particles in India contain about 20% of $CaCO_3$. If we assume that 20% of the total HNO_3 molecules reacting with dust particles are lost irreversibly and that the remaining 80% adsorb to the surface, then the reaction of one HNO_3 molecule with a dust particle should be associated with the production of 0.4 molecules of $NO_{x.}$ This is assuming a yield of 50% for NO_x production from HNO_3 uptake as observed by Chen et al. (2011). In our simulations, we did not consider these different pathways but assumed that HNO_3 reacting with all minerals adsorbs to the surface, and consequently assumed a production of 0.5 molecules of NO_x from HNO_3 uptake (Dust_JH configuration). However, this assumption should not affect the results presented in this study. The comparison between the NO_x distributions as simulated by Dust_JH and Dust_JH_NoReNOx, showed that the renoxification process leads to a very small (0.1-0.2 ppbv) increase in NOx values. The use of a yield of 0.4 molecules of NO_x from HNO_3 uptake should lead to an even smaller increase. We have now included this information in section 2.2.

For the model configuration my comment is on the coarse resolution of the gridded domain. I would expect to see a higher resolution simulation, i.e. 10km or 5km, to capture the fine scale of the chemical reactions. Have the authors tried to use a finer spatial resolution? How did they come up with the specific choice?

The use of a finer resolution will not only affect the scale of chemical reactions but might also have influence on modeled dust emissions because the wind speed might vary somewhat with grid spacing. However, we did not use a finer spatial resolution because the anthropogenic emissions used here (MACCity emission inventory; Granier et al., 2011) are only available at a resolution of 0.5^ox0.5^o. Emission inventories at higher spatial resolution (e.g. HTAP emission inventory at 0.1^ox0.1^o) have become available only recently and can be used in future simulations.

I am not in favor of referring to accompany papers for parts of the manuscript that ought to be included in the text. Especially when the accompany paper is still under review. This comment

refers explicitly to the initialization of the WRF model as it relates to the meteorology and thus the dust emissions generated by the model, which are essential part of this work.
The accompanying paper has now been published in ACP. We have also included a more detailed description of the model configuration in the revised Section 2.

Supplement: In some occasions the wording in the supplement is not appropriate: i.e. “Both the methods use the SSA…” should be “Both methods use the SSA….” A careful review of the text is necessary.
We have carefully revised the text in both the manuscript and the supplementary material.