Reply to referee #1

First of all, we thank the reviewer for the careful reading of the manuscript and constructive comments. We have revised the manuscript, following the reviewer's suggestions.

We have also removed/added/changed the words, sentences, and figures in the manuscript. The changed and added parts are painted in a red color in the text.

General Comments:

This paper estimates the contribution of ammonium nitrate to AOD (aerosol optical depth) and ADRF (aerosol direct radiative forcing) over East Asia, based on CTM and radiative transfer simulations with data assimilation technique. Of novel interest, the paper specifically evaluates CTM performance for ammonium nitrate and firstly estimates the influences of NH4NO3 formation on AOD and ADRF. The authors use sound methods and the conclusions are supported by the evidence. In addition, the paper is well written and clear. Therefore, I recommend it for publication in ACP after minor revision. Detailed comments below:

Minor comments:

1. In this work, the authors evaluated CMAQ-estimated particulate concentrations by comparing with EANET-measured concentrations, especially for three particulate species (nitrate, sulfate, ammonium; Figs. 4-6). Why do authors only estimate the contribution of ammonium nitrate to AOD and ADRF? As the authors indicated in Eq. 4, the contributions of several particulate species, such as ammonium sulfate, elemental carbon, organic mass, to AOD and ADRF should be presented and discussed in the end of section 3.3. The concentration of ammonium sulfate and their radiative effects over E. Asia will be comparable. One figure or summary table will be enough.

Reply: In this study, we estimated the contribution of ammonium nitrate to AOD and DRF by aerosols in order to emphasize the important role of ammonium nitrate in regional climate over East Asia. Based on reviewer's comments, we added one paragraph at the end of Sect. 3.3 and one figure to describe the contributions of particulates species to AOD and DRF by aerosols. Please, check out pp. 23:6-23:13 and also newly-added Fig. 16.

2. The wavelength dependency of aerosol optical properties, such as AOD, SSA, and g, are properly considered in the radiative transfer calculations. But, in this paper, the authors only mentioned that AOD and SSA were estimated at a wavelength of 550 nm (Sec 2.3). Detailed descriptions should be added. In addition, how the authors considered the vertical distribution of aerosols in radiative transfer calculation?

Reply: Using our algorithm to convert CMAQ-simulated particulate concentrations into aerosol optical properties, the multi-wavelength aerosol optical properties were not calculated in this study. Instead, the angstrom exponent was used to consider the wavelength dependency of aerosol optical properties. Detailed descriptions were made in Sect. 2.6. Please, check out pp. 11:21-12:7.

3. Have the authors check the accuracy of ADAM-estimated dust concentrations? Dust aerosols in spring may largely contribute both scattering and absorption of aerosols.

Reply: Yes, it was also reported in our previous study (Park et al., 2011b) that dust aerosol in spring can significantly contribute to both the scattering and absorption of solar radiation, depending on the size and composition distribution of dust particle. In Park et al. (2011b), the accuracy of the ADAM-estimated dust concentrations was indirectly estimated via the comparison between AOD values calculated with and without the ADAM-estimated dust concentrations. Based on the comparison, the accuracy of AOD with the ADAM-estimated dust concentrations is higher than that without the ADAM-estimated dust concentration. Unfortunately, the accuracy of the ADAM-estimated dust concentration was not directly estimated, because of the absence of dust observation data over East Asia for the year of 2006.

4. In this study, the authors only show the ADRF at the surface, but atmospheric forcing and ADRF at the top of the atmosphere should be presented and discussed. In addition, it should be added how the AERONET ADRF were calculated (e.g., Fig. 12).
Reply: In this study, we showed the DRF by aerosols only at the top of atmosphere, mainly because the direct radiative forcing by ammonium nitrate at the surface is nearly equal to that at the top of atmosphere. In order to better show this, the surface and TOA forcings were analyzed in Fig. 16, following reviewer's comments. In this study, AERONET DRF by aerosols was downloaded from AERONET web site, as mentioned at pp. 10:13-10:15. In addition, we briefly described how AERONET DRF was calculated via GAME at pp. 10:15-10:22.

5. In this paper, it is hard to evaluate ASSIMILATED-AOD by comparing monthly averaged AERONET AOD. How the comparison given in Fig. 10 was made? Is assimilated AOD for only daytime is considered? Showing a comparison between assimilated AOD and AERONET AOD for simultaneous (or hourly mean for only day-time) is needed.
Reply: Our assimilation was conducted with the monthly-averaged CMAQ-estimated and MODIS-retrieved AODs. However, when we tried to assimilate daily MODIS-derived AOD data with CMAQ-calculated AOD, we obtained similar results. This was done and checked in our previous study (Park et al., 2011b). In addition, the daily comparison between assimilated and AERONET AODs was also partly conducted in our previous study (Park et al., 2011b).

6. P19202 L25: ". . ...the cloud bottom height was assumed to be 200 m above the surface following the. . ...". Under this assumption, where the aerosol layer is? Below the cloud layer or above the cloud layer? It is unclear how the authors calculate ADRF under all-sky conditions. Another issue that what's the accuracy of MODIS-derived cloud top height.
Reply: It is important whether the aerosol layer is present below cloud layer or above cloud layer. As it was mentioned in our manuscript, MODIS-retrieved cloud top height were used in this study. The cloud effect on DRF by aerosols should be calculated with the accurate data of cloud top height. The accuracy of MODIS-retrieved cloud top height was analyzed in the several previous studies (e.g., Genkova et al., 2007; Naud et al., 2007; Holz et al., 2008). Based on these analyses, MODIS-retrieved cloud top height has large uncertainty for cirrus cloud or overlapped clouds. We have recognized that the DRF by aerosols under all-sky conditions is highly uncertain with the uncertain MODIS-retrieved cloud top height. Even so, we intended to report that the high DRF by aerosols can be offset by clouds. It is expected that the accuracy of MODIS-retrieved cloud top height will be improved and the cloud effects on DRF by aerosols will also be more realistic in the future.

7. Minor technical corrections: P 19196 L 16, Title of Section 2.6, and so on: use "radiative transfer model" instead of "radiative transport model". SBDART is not a transport model.

Reply: Thank you for your kind correction. Based on your comments, the correction was made at pp. 4:19 and 10:23.

Additional references:

Genkova, I., Seiz, G., Zuidema, P., Zhao, G., and Girolamo, L. D.: Cloud top height comparisons from ASTER, MISR, and MODIS for trade wind cumuli, Remote Sens. Environ., 107, 211-222, 2007.

Naud, C. M., Baum, B. A., Pavolonis, M., Heidinger, A., Frey, R., and Zhang, H.: Comparison of MISR and MODIS cloud-top heights in the presence of cloud overlap, Remote Sens. Environ., 107, 200-211, 2007.

Holz, R. E., Ackerman, S. A., Nagle, F. W., Frey, R., Dutcher, S., Kuehn, R. E., Vaughan, M. A., and Baum B.: Global Moderate Resolution Imaging Spectroradiometer (MODIS) cloud detection and height evaluation using CALIOP, J. Geophys. Res., 113, D00A19, doi:10.1029/2008JD009837, 2008.