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Interactive comment on "Improvement and further development in CESM/CAM5: gas-phase chemistry and inorganic aerosol treatments" *by* J. He and Y. Zhang

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Reply to Reviewer 4's Comments

This work describes several major model development and improvements in a recently-released Earth system model, i.e., NCAR's CESM/CAM5.0. The paper focuses on model development effort and initial assessment of improved model treatments in representing the atmosphere. The model development includes a comprehensive gas-phase chemical mechanism based on CB05 and detailed inorganic aerosol treatments that combine several nucleation parameterizations including advanced ion-mediated

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nucleation module of Yu (2010) and that simulate aerosol thermodynamics based on ISORROPIA II. A comprehensive model evaluation was performed for climatic/radiative variables and chemical concentrations and column mass abundances on global and several regional domains including US, Europe, and Asia using observations from global surface networks (including NCDC, GPCP, BSRN, and NOAA/CDC) and satellite datasets (including MODIS, CERES, TOMS/SBUV, MOPITT,GOME) as well as regional observational networks (including CASTNET, IMPROVE, STN over CONUS; the EMEP, BDQA, and AirBase over Europe; MEP of China, NIES of Japan, and TAQMN over East Asia). The simulation results were analyzed in detail and the improved model performance was linked clearly to specific model representations that were improved in this work, which helps the readers to better understand the benefits of such model improvements. It was found that CB05_GE with new and modified inorganic aerosol treatments in MAM7 is more accurate than simple gas chemistry coupled with default MAM7 and can predict many more gaseous species, and give improved performance for predictions of organic carbon and PM2.5 over CONUS, NH3 and SO42ôĂĂĂ over Europe, SO2 and PM10 over East Asia, and cloud properties such as CF, CDNC, and SWCF. This work represents a significant contribution to the global air quality, climate, and Earth system communities and addresses several major model deficiencies and limitations in chemistry and aerosol treatments in current climate/Earth system models. It will thus potentially reduce the uncertainties of climate/Earth system predictions that are associated with those treatments in global climate and Earth system models. Although NCAR's CAM v5.0 represents the latest advancement in atmospheric model component in the contemporary Earth system model, it still uses simple gas-phase chemistry (only 8 chemical reactions among 6 gas-phase species) and highlysimplified thermodynamics that treats sulfate and ammonium only. In particular, it does not simulate nitrate and chloride which are important on a global scale. Modeling sulfate microphysics without considering these components introduces errors and uncertainties because it neglects microphysical interactions between components such as the interplay among sulfate, nitrate and ammonium (Yu et al., 2005) as well as sulfuric acid

condensing onto sea-salt particles (O'Dowd et al., 1997). This work addresses the major model deficiencies in simulating chemistry and aerosols in CAM5.0 by incorporating the state of the science CB05_GE gas-phase chemical mechanism with 273 reactions among 93 gas-phase species and the latest version of ISORROPIA II. Incorporating CB05_GE enables an explicit simulation of many gas-phase species such as CO, O3, and HNO3 and also improves model performance for secondary PM species and PM precursors such as SO2 and NH3 in several regions. CB05_GE has been coupled with the most detailed aerosol module based on MAM7 in CAM5.0. To my best knowledge, CAM5.0 with the coupled CB05_GE/MAM7 represents the most advanced chemistry-aerosol treatments in current generation of Earth system models. In a similar effort done by NCAR, MOZART version 4 (MOZART-4) (Emmons et al., 2010), an extended version of gas-phase chemistry used in Lamarque et al. (2012) has been incorporated into an offline version of CAM version 4.0 (older than CAM v5.0) driven by GEOS5 meteorological analyses. MOZART-4 was coupled to the simplified bulk aerosol module that cannot simulate aerosol size distribution. While MOZART_4E contains more explicit anthropogenic VOCs species than CB05_GE, it lumps monoterpenes as one species (i.e., _-pinene). By contrast, CB05_GE contains explicit treatments for more biogenic VOCs such as _-pinene, _-pinene, limonene, terpinene, and ocimine, thus enabling a more accurate treatment for secondary organic aerosol (SOA). It would be interesting to compare the two comprehensive gas-phase chemical mechanisms in the same CAM model to investigate their capabilities in simulating gas-phase chemistry and resulting secondary aerosol, in particular, SOA, on a global scale. The incorporation of ISORROPIA II into CAM5.1 significantly advances aerosol treatments in current global and Earth system models. Although an older version of ISORROPIA was incorporated in several global models, most of which are offline-coupled meteorology-chemistry models such as GEOS-Chem (Bey et al., 2001) and the GISS Caltech (Liao et al., 2003); very few models are online-coupled models (e.g., the GUWRF/ Chem (Zhang et al., 2012b)); and none of them are Earth system models that are much more complex than global or regional offline or online-coupled air quality models. Differ-

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ent from ISORROPIA that has been used in most regional air quality models such as CMAQ and CAMx and several global models, ISORROPIA II considers the effects of crustal species (e.g., magnesium, potassium, calcium) on inorganic aerosol thermo-dynamics that have been neglected in most regional air quality models and in nearly all global models. Not until recently, ISORROPIA II has been incorporated into CMAQ 5.0 (Appel et al., 2013). To my best knowledge, aerosol thermodynamics involving coarse particles has not been treated in nearly all regional and global models, not until recently, such effects were considered in CMAQ 4.7 and newer versions. Although this work only considers aerosol thermodynamics involving crustal species for fine-mode particles, it lays a foundation to further account for aerosol thermodynamics involving coarse particles and the effects of crustal species on coarse particles. While climate models are typically run for 30-year or longer, very few work focusing on chemistry and aerosols can afford to simulate a 30-year period, due primarily to a large compu-tational burden of long-term simulations using such air quality models. In this work, seven 1-year simulations were performed with various model treatments to illustrate the benefits of each improved model representation. I believe such 1-year model sim-ulations are well sufficient for model development and initial assessments. It would be a natural extension for the authors to apply such a model with advanced chemistry and aerosol treatments for decadal simulations to demonstrate enhanced model capabili-ties in their future work. In sum, this work represents a significant model development effort that deserves publication on ACP. The results are very interesting and promising. The paper is very well written and provides a very good documentation of their model development effort. The subject is appropriate to ACP. Therefore, I would strongly rec-ommend its acceptance for publication on ACP after some minor modification. Several comments for improv- ing the information content and presentation of the paper are listed below in specific comments.

Reply:

We thank the reviewer for positive comments.

Specific Comments 1. P27765-27767 for Tables 3-5: It will be better if the authors can add the model evaluation results for RMSE and NME for each variable.

Reply:

RMSE and NME were indeed calculated for all simulations but were not included in Tables 3-4 due to limited space. To address the reviewer's comments, we have added RMSE and NME in the Tables 6-7 from the three new 5-yr simulations.

2. P27777 for Figure 9, It will be better if the authors can compare the results for cloud and radiative variables for the summer time instead of whole year because the indirect aerosol forcing is more important during the summer.

Reply:

The results for cloud and radiative variables during Northern Hemisphere (N.H.) summer (JJA) are overall similar to those during the whole year. Although the magnitude of changes in CDNC, COT, and SWCF over specific regions is larger in N.H. summer-mean than annual-mean, the global mean changes in CDNC and SWCF are larger in annual mean than in N.H. summer-mean.

To address the reviewer's comment, we have added comparison for chemical, cloud and radiative variables for JJA mean in the revised paper, see Figures A2 and A3 in the supplementary material. We also evaluated the model performance during JJA and added this info in Tables A2 and A3 in the supplementary material.

Interactive comment on Atmos. Chem. Phys. Discuss., 13, 27717, 2013.