

## Additional Response to Referee 1

We have addressed the referee's comments and suggestions (see also our previously uploaded response to the referee), and have heeled nearly all of the referee's suggestions and incorporated the changes in the revised manuscript. Below we provide additional details on how we have modified the manuscript according to two of the referee's comments in addition to what have been stated in our previous responses.

1. In response to the referee's comments, we have significantly expanded the model description section (Sect 2.1) in the revised manuscript. The modified discussion is as follows.

“Gas-phase chemistry is simulated with a version of the CBMZ mechanism that includes DMS chemistry (Zaveri and Peters, 1999). The MOdel for Simulating Aerosol Interactions and Chemistry (MOSAIC, Zaveri et al., 2008) provides an integrated treatment of aerosol chemical and microphysical processes. MOSAIC uses an 8-bin sectional approach to represent aerosol size distributions for both unactivated (i.e., interstitial) and activated (i.e. cloud-borne) aerosols, with both mass and number simulated for each bin. MOSAIC calculates condensation and evaporation of gas species (such as  $\text{H}_2\text{SO}_4$ , MSA,  $\text{HNO}_3$ , HCl, and  $\text{NH}_3$ ; Zaveri et al., 2008) using an accurate thermodynamic module that predicts particle deliquescence, water content and solid-liquid phase equilibrium in multicomponent aerosols (Zaveri et al., 2008). Aerosol coagulation, aerosol nucleation, and aqueous chemistry (in cloud droplets) are also treated. The aerosol activation scheme (Abdul-Razzak and Ghan, 2000) is based on the maximum supersaturation, which is diagnosed based on both the resolved vertical velocity and the parameterized turbulent motions. Particles are assumed internally mixed within each model size bin, and Köhler theory is used to relate the aerosol size distribution and composition to the number activated as a function of the maximum supersaturation (Abdul-Razzak and Ghan, 2000). For calculating number concentrations of CCN ( $N_{CCN}$ ) at a

supersaturation, a narrow lognormal size-distribution is assumed for the particles within a size bin. The aerosol composition of the bin gives a volume-weighted hygroscopicity, from which the dry-diameter for a specified critical supersaturation is calculated. The particles in the bin with dry-diameter exceeding this critical diameter contribute to  $N_{CCN}$ .

The cloud microphysics is represented with the Morrison double-moment scheme (Morrison et al., 2009) that was recently coupled with interactive aerosols (Q. Yang et al., 2011). In the Morrison double-moment microphysics, the cloud droplet number concentration and cloud water mixing ratio are used to determine a gamma distributed cloud droplet size spectrum, which affects various microphysical process including autoconversion. Changes of number concentrations and mass mixing ratios of cloud water are predicted in the scheme based on sources (primarily water vapor condensation) and sinks (e.g., autoconversion and accretion). Other hydrometeor species, including raindrops, are assumed to follow an exponential distribution (Morrison et al., 2009). The autoconversion scheme is based on Khairoutdinov and Kogan (2000) using an explicit autoconversion rate:

$$\frac{\partial q}{\partial t} = 1350 Q_c^{2.47} N_c^{-1.79}$$

where  $Q_c$  and  $N_c$  are cloud water mixing ratio and droplet number concentration, respectively. Unlike some other autoconversion parameterizations (e.g., Manton and Cotton, 1977), this formula does not include any threshold effective radius, but does implicitly depend on mean droplet radius given its dependence on both  $Q_c$  and  $N_c$  (i.e., mean droplet radius is proportional to  $(Q_c/N_c)^{1/3}$ ).

Aerosols impact clouds, and clouds also impact aerosols in the model. In-cloud wet scavenging is the main sink of submicron particles while cloud chemistry provides a major source of sulfate. Wet scavenging of aerosols refers to the removal of both cloud-borne/activated and interstitial/unactivated aerosols by precipitation, although the cloud-borne aerosol wet scavenging dominates. The cloud-borne aerosol wet-scavenging rate (first order rate,  $s^{-1}$ ) is assumed equal to the cloud water first-order removal rate, and includes

contributions from cloud water autoconversion and collection by rain in the Morrison microphysics. Evaporation of cloud droplets in dry air resuspends cloud-borne aerosol back to the interstitial state. The complete evaporation of a raindrop will also resuspend the rain-borne particulate matter as a single (and relatively large) particle, but this process was not treated in our WRF simulations. This resuspension by evaporating raindrops would have little impact on the simulated CCN number concentrations (at  $s = 0.1\%$ ), as a raindrop is typically composed of hundreds to thousands of cloud droplets (and activated CCN), but the evaporation releases a single large CCN. The aerosol impacts on clouds and shortwave radiation for the first indirect effect are implemented by linking the predicted cloud droplet number from the Morrison microphysics scheme with the Goddard shortwave radiative scheme. The effect of aerosols on precipitation is handled directly by the microphysics scheme for warm-rain processes, where the number of activated particles affects cloud droplet number, autoconversion rate, and precipitation.

The YSU scheme (Hong et al., 2006) is used to represent turbulent vertical mixing associated with the boundary layer. In the YSU scheme, the non-local mixing due to large eddy transport is considered for heat and momentum components, and an explicit treatment of entrainment is included in the heat and momentum flux profiles and the growth of the planetary boundary layer (PBL) height (Noh et al., 2003; Shin and Hong, 2011). This parameterized PBL entrainment in the YSU scheme is not equivalent to cloud-top entrainment (S. Hong, personal communication, 2012) since the YSU scheme assumes the PBL top as the minimum flux level as explicitly formulated, and the PBL height from YSU was found to be near the cloud base or mid-level of the stratocumulus clouds. The cloud-top entrainment discussed later likely results from parameterized local mixing, the resolved-scale motion, and complex feedback between microphysics and the MBL scheme.”

2. In Fig. 2, we have revised the caption significantly according to the referee’s comments. The revised caption is shown below, which also includes corrections

to a mistaken statement in our previous responses regarding how sigma was calculated. We realized later that the sigma shown only includes temporal variations.

“Fig. 2. Responses of aerosol and cloud properties to regional oceanic (OceanEmis, blue), anthropogenic (AnthroEmis, green), and enhanced anthropogenic (ScaledEmis, orange) emissions. The response, ‘ $\Delta$ ’, denotes differences in simulated quantities between two simulations with and without the specific regional emissions: REF – 0OCE, REF – 0ANT, and 5ANT – REF are used for assessing the response due to OceanEmis, AnthroEmis, and ScaledEmis, respectively. Panels a-f are for mean changes in accumulation mode aerosol ( $N_d$ , panel a) at 975 hPa, CCN ( $N_{ccn}$  at 0.1% supersaturation at 975 hPa, panel b), cloud-top cloud droplet number ( $N_d$ , panel c), aerosol optical depth (AOD, panel d), cloud top effective radius ( $r_e$ , panel e), and cloud optical depth (COD, panel f), respectively. The mean aerosol or cloud properties in the reference simulation are shown at the top of each panel for each region (regions P, I, and C) as  $\mu_{\text{ref}}$ . The aerosol-cloud sensitivity factor,  $\frac{\Delta \ln(Y)}{\Delta \ln(N_{ccn})}$ , are shown in red above or below each color bar, where Y is the cloud property (e.g.,  $N_d$ ,  $r_e$ , and COD). More details regarding the aerosol-cloud sensitivity factor are provided in the main text. The error bars show the 95% confidence intervals of the mean changes estimated based on the temporal variation of hourly spatially averaged responses in each region”.