Review of "An Improved Representation of Aerosol Mixing State for Air-Quality-Weather Interactions" by Stevens et al.

General comments:

This work implements a detailed representation of aerosol mixing state, which is categorized by hygroscopicity and BC mass fraction, into the GEM-MACH air quality and weather forecast model and examines the impact of the aerosol mixing state representations on the aerosol concentrations, radiative properties, and meteorology over the North American region. The authors show that the more-detailed representation of the aerosol hygroscopicity (three-category 1L2B and two-category HYGRO) increases aerosol concentrations, which increases cloud droplet number concentrations and cloud reflectivity in the model. They also show that resolving thinner coatings on BC (1L2B) slightly decreases absorption aerosol optical depths.

This study is interesting and the subject is of great interest to ACP. The manuscript is well written. However, the model descriptions of the aerosol mixing state are not satisfactory, which are cause for concern (see Major comments). Once these points are addressed satisfactory, the paper should in my opinion be suitable for publication in ACP.

Major comments:

1. Section 2:

The authors should explain the treatment of aerosols in the mixing state categories in the model in more details. It is unclear to me. Do the model trace both mass and number concentrations of aerosols in each bin? How to calculate diameter of the aerosols in each bin (e.g., stationary or moving structures)? How to treat the movement of aerosols between different categories (e.g., moving center approach, all particles (mass of species and number) in the bin are moved to a new bin)? Do the condensations of sulphate onto the lo-κ_hi-BC and lo-κ_lo-BC categories move the aerosols in these categories to the hi-κ category? Can the aerosols in the low hygroscopicity categories (lo-κ_hi-BC and lo-κ_lo-BC) be act as CCN and be activated to cloud droplets? What schemes of the gas phase chemistry and the aerosol thermodynamics the model use? How to calculate dust emissions in the model? Could the authors explain the calculation methods of the aerosol optical properties of BC-containing particles in more detail (please also see specific comments)?

2. Abstract:

The authors describe that "the more-detailed representation of the aerosol hygroscopicity in both 1L2B and HYGRO decreases wet deposition, which increases aerosol concentrations, particularly of less-hygroscopic species" and "We show that these increased aerosol concentrations increase cloud droplet number concentrations and cloud reflectivity". I cannot understand that the increases in less-hygroscopic aerosol concentrations increase cloud droplet number concentrations. The less-hygroscopic species are not able to act as CCN and are not removed from the atmosphere by in-cloud scavenging process, but can be activated to cloud droplets in the model? Please clarify this.

3. Mixing state of BC-containing particles:

I wonder if the authors have validated the BC mass fractions of BC-containing particles in the model calculations by the available surface or aircraft observations. Some measurements, such as SP2 (single-particle soot photometer), would provide the information of BC mixing state, which can be used for the model validation (e.g., Oshima et al., 2009b). Although there are small amounts of data available on the BC mixing state, the authors should mention that they have validated the BC mixing state or not in their previous studies and, if not, please mention that the comparison should be done in future works.

4. Horizontal resolution:

The authors used the 10 km horizontal resolution in the model calculations. In my opinion, the 10 km resolution is high for simulations with the detailed aerosol mixing state. Can the authors state some benefits in their results due to the high resolutions?

Specific comments:

Lines 4 and 459, "This is the first model …" Is it the first model, because some models treat more detailed aerosol mixing state representations (e.g., Jacobson, 2001).

Lines 9-14, the results shown in this study will depend on the regions, season, and scenarios (e.g., anthropogenic and biomass burning). The authors should clarify these conditions in the abstract.

Introduction, it would be better to compare the results with Stier et al. (2006), which showed the decrease or enhancement of absorption due to BC aging, which increases the CCN activity and light absorption of BC-containing particles.

Line 101, "our BC-free category is also hydrophobic". I cannot understand this. The authors mean that lo-κ_lo-BC category is hydrophobic even if there is no BC in the lo-κ_lo-BC category? Does the hi-κ category include BC? Are there BC-free categories in 1L2B representation? Please clarify this.

Lines 111 and 114, "add the mass to the mixing-state category that matches the new particle's properties" and "the mass is moved to the mixing-state category that matches the hygroscopicity and BC mass fraction of the aerosol mass". How to treat number concentrations of aerosols?

Lines 137-138, "For the radiation calculations, seaspray and dust are always assumed to exist as pure particles, externally mixed from the other components". How to estimate number concentrations and diameters of seaspray and dust particles from the internally mixed aerosol representations?

Line 140, "The absorption enhancement of the BC cores is calculated following Bond et al. (2006)." Do the authors implement the Mie theory for concentrically coated spheres (core-shell treatment) and use complex refractive indices of individual chemical species? Or do the authors simply use the absorption enhancement factors for BC-containing particles in hi-κ and lo-κ_lo-BC categories? If in the latter case, how to calculate extinction (or scattering) coefficients of BC-containing particles and how to treat enhancement due to RH?

Line 147, "we perform simulations only from June 15th to July 31st, and." It is better to clarify the year (2016) here.

Lines 221 and 255, "all aerosol in the low-κ categories are excluded from wet deposition processes". Is this "in-cloud scavenging processes" rather than "wet deposition processes"? Coarse particles in the low-κ categories can removed from the atmosphere by below-cloud scavenging.

Figure 2 and lines 233-246, the authors show the temporal means of the wet deposition fluxes normalized by the surface aerosol concentrations. Do the authors show the wet deposition fluxes normalized by the aerosol column, if possible, because the in-cloud scavenging processes play efficiently at higher altitudes?

Lines 253 and 217, "After normalizing by the surface concentrations of aerosol, wet deposition rates of BC, POA, SOA, and dust were reduced in the HYGRO and 1L2B simulations by 27%, 40%, 12%, and 10%" and "These differences are due mostly to increases in less-hygroscopic species, with concentrations of BC, POA, SOA, and dust being increased in the HYGRO and 1L2B simulations by 16%, 16%, 21%, and 93%." Why the wet deposition rate of dust (10%) is smaller, although the surface concentrations of dust are largely increased?

Line 269, "We convert observed organic carbon to organic matter assuming a mass-to-carbon ratio of 1.8". Could the authors show references (ratio of 1.8) here?

Lines 315-322 and Figure 4, it is difficult to understand the BC transport and evaporation from Figure 4. Could the authors explain more clearly? How about the changes in BC column over the source (forest fires) and downwind regions?

Lines 374-379, why AAOD decrease in the Gulf of Mexico and just south the Great Lakes region? Are these regions close to the anthropogenic source regions? Is the resolving the coating thickness of BC important for freshly emitted anthropogenic BC particles? Could you add explanations?

It is helpful to show the emission map of BC (both anthropogenic and forest fires) during the model simulation period in Supplemental figures to clarify the source and remote regions.

Technical comments:

Table 3, I could not find the word "Table 3" in the text.

Lines 92-93, "0-2.5 um and 2.5-10 um". Are these diameters?

Line 101, "in that we our BC-free", remove "we", typo?

Line 310, "BC concentrations at the surface". BC mixing ratio?

Figure S1, please add units, like Figure 1.

References:

Jacobson, M. Z. (2001), Strong radiative heating due to the mixing state of black carbon in atmospheric aerosols, Nature, 409, 695–697.

Oshima, N., M. Koike, Y. Zhang, Y. Kondo, N. Moteki, N. Takegawa, and Y. Miyazaki (2009b), Aging of black carbon in outflow from anthropogenic sources using a mixing state resolved model: Model development and evaluation, J. Geophys. Res., 114, D06210, doi:10.1029/2008JD010680.

Stier, P., J. H. Seinfeld, S. Kinne, J. Feichter, and O. Boucher (2006), Impact of nonabsorbing anthropogenic aerosols on clear-sky atmospheric absorption, J. Geophys. Res., 111, D18201, doi:10.1029/2006JD007147.