Dear Reviewer,

Thank you for the comments to help improve the quality of the paper. We have revised the manuscript to address your comments and a detailed response to each comment is provided in this file.

The comments are in regular font, the responses are in red, and the changes in the manuscript are in blue.

Anonymous Referee #3

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General Comments: This manuscript presents results of concentrations and sources Ultrafine Organic Aerosols from 9 years chemical transport model simulations, which is important for health effects studies. The presentation quality is excellent and no major scientific problems with the presentation. I only have some minor concerns, which are listed below.

Specific comments:

1. In model evaluation, the authors claim that their calculated monthly MFB and MFE meet Boylan and Russel (2006) standard. It is better to point out the standard in manuscript.

Responses: The PM model performance criteria of MFB and MFE, suggested by Boylan and Russell (2006), are a function of PM concentration as follows,

MFB (%) $\leqslant \pm 140e^{-(Co+Cm)} + 60$

MFE (%) $\leqslant 125e^{-2(Co+Cm)/3} + 75$

where Co and Cm represent the observed and predicted PM concentrations, respectively.

We added above criteria equations in the Supplemental Materials.

2. Emission inventory is a key part in air quality modeling, but the authors do not write much about emission inventory (based on which year, what the sectors are, and etc.). Besides, the simulation period is as long as 9 years. It is better if the authors can consider the changes in emissions over this period.

Responses: The details of emission inventory were provided in the Part I paper, so we didn't repeat in this paper. This is clarified on lines 144-146 of the revised manuscript. A few additional details are provided below to help the reviewer understand what was done.

The emission inventory base year was 2000. We did consider the changes in emissions over the modeling period when information is available. The EMFAC 2007 model (CARB 2008) was used to scale the mobile emissions using predicted temperature and relative humidity fields through the entire nine-year modeling episode. Biogenic emissions were generated using the Biogenic Emissions Inventory System v3.14 (BEIS3.14), which includes a 1-km resolution land cover database with

230 different vegetation types (Vukovich and Pierce 2002). Sea-salt emissions were generated on-line based on the formulation described by de Leeuw et al. (de Leeuw et al. 2000) for the surf zone and the formulation described by Gong (Gong 2003) for the open ocean. Emissions from wildfires and open burning at 1 km × 1 km resolution were obtained from the Fire INventory from NCAR (FINN) (Hodzic et al. 2007; Wiedinmyer et al. 2011). These emissions included the changes over the model period. However, for other emissions, for example, stationary sources and residential wood burning emissions were kept unchanged due to unavailable information to scale the emissions.

3. The authors compared ratio from model to CMB derived ratio. Are the CMB derived ratios 100% reliable? How is its uncertainty? And it is also better to introduce the CMB method in the manuscript.
Responses: The uncertainties of CMB derived SOA range from 1% - 22% (Daher et al. 2012). The information was added in the caption of Figure 3.

We used the CMB results to evaluate the model performance of SOA and POA predicted by the UCD/CIT model. Because the CMB method is not the focus of this study, we think it is not necessary to introduce the CMB method in the manuscript. The references on the CMB method were provided in the original manuscript.

4. The authors discuss the impact of vapor wall losses on SOA concentrations. How will it affect model evaluation? In Figure 1, model overpredicts OA at some sites. After vapor wall losses correction, model may mismatch with observations.
Responses: The vapor wall losses correction didn't significantly affect the model evaluation results for total OA. The predicted SOA/TOA fractions in the base model is very low (generally < 10%) at the observation sites located in urban areas (Figure 4 and Figure S1). Even though the vapor wall loss correction led to substantial increase of SOA concentrations, the total OA concentrations didn't increase much.

A sentence was added in the manuscript in the lines 363-366:
"Due to low SOA/TOA fractions (< 10%) at the observation sites located in urban areas (Figure 4 and Figure S1), the substantial increase of SOA by the vapor wall loss corrections does not strongly change the total OA concentrations and therefore does not significantly affect the model evaluation results shown in Figure 1."

References:

CARB, 2008: Calculating emission inventories for vehicles in California. *User's Guide EMFAC 2007 version 2.30*, **Accessed in 2010**.

Daher, N., and Coauthors, 2012: Characterization, sources and redox activity of fine and coarse particulate matter in Milan, Italy. *Atmos Environ*, **49,** 130-141.

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Gong, S. L., 2003: A parameterization of sea-salt aerosol source function for sub- and super-micron

particles. *Global Biogeochemical Cycles*, **17**.
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Vukovich, J. M., and T. Pierce, 2002: The Implementation of BEIS3 within the SMOKE modeling framework. MCNC-Environmental Modeling Center, Research Triangle Park and National Oceanic and Atmospheric Administration.
Wiedinmyer, C., S. K. Akagi, R. J. Yokelson, L. K. Emmons, J. A. Al-Saadi, J. J. Orlando, and A. J. Soja, 2011: The Fire INventory from NCAR (FINN): a high resolution global model to estimate the emissions from open burning. *Geosci Model Dev*, **4,** 625-641.