Reply to Anonymous Referee #1

We thank the reviewer for the careful reading of the manuscript and helpful comments. We have revised the manuscript following the suggestion, as described below.

The paper investigates the uncertainties in SOA simulation by focusing on the uncertainty in the meteorology. The main strength of the paper lies in a very detailed analysis of two specific days using a large number of ensemble simulations, perhaps this is also a potential weakness of the paper, as analysis of just two days makes it hard to generalize the results. The number of analyzed days is probably limited by the long run time of the chemistry model. Nevertheless, the paper clearly shows the importance of meteorology when SOA is simulated in the Mexico City basin for the specific days included in this paper. A more detailed description of the two observation sites is needed to show how representative these locations are when compared with the model results.

Response: We have selected "Convection-North" and "Convection-South" days in ensemble simulations, which can be classified into "O_3-South" and "O_3-North" episode types, respectively, except that the convective activity prevents the formation of a clean convergence zone sweeping through the basin in the late afternoon. During the MILAGRO-2006 field campaign, more than 90% of days can be defined as "O_3-South" or "O_3-North", indicating that the two days we simulated represent most of the meteorological situations during the campaign. Therefore we have clarified in Section 3: "*According to the flow type, "Convection-North" and "Convection-South" days can be classified into "O_3-South" and "O_3-North" episode types, respectively, except that the convective activity prevents the formation of a clean convergence zone sweeping through the basin in late afternoon (de Foy et al., 2005; 2008). Considering that "O_3-South" and "O_3-North" episodes dominate the MILAGRO-2006 field campaign period (de Foy et al, 2008), the two days we have simulated represent most of the meteorological conditions during the campaign.*"

We have also added a paragraph in Section 2 to provide a detail description of the two observation sites: "*The ensemble simulation results are compared to the Aerosol Mass Spectrometry (AMS) observations analyzed using the Positive Matrix Factorization (PMF) technique at an urban background site (T0) and a suburban background site (T1) in Mexico City. The T0 monitoring station is located in the northwestern part of the basin of Mexico City, influenced by road traffic emissions (300 m from four major roads surrounding it), domestic and residential emissions, and also potentially influenced by local industrial emissions and from the Tula industrial area (60 km to the north-northwest, in the Hidalgo State). T1 supersite is located around 50 km to the north of Mexico City, in an area isolated from major urban agglomerations but close to small populated agglomerations, and around 500 m from the closest road.*"

Specific comments:

P16297, line 18: Is the WRF model initialized at 00:00 UTC at the beginning of 24 and 29?

Response: Yes, the WRF model is initialized at 00:00 UTC on both 24 and 26. We have clarified in Section 2: "*The WRF model is initialized at 00:00 UTC and integrated for 30h for all the selected days.*"

P16297, line 26: Can the author briefly justify the choice of WRF parameterizations used here? Are these the most up to date and appropriate use of parameterizations schemes for air quality applications? This would be useful information for air quality modelers.

Response: We have clarified in Section 2: "*The physical process parameterizations, particularly the PBL parameterization, play an important role in the air quality simulation. We have performed sensitivity studies to investigate the impact of different PBL schemes on ozone and aerosol simulations and found that the MYJ TKE PBL scheme yields more reasonable results than the other PBL schemes in the WRF model compared to the observations. However, it is worth mentioning that the MYJ TKE PBL scheme is appropriate in the simulations in Mexico City, but might not work well in other megacities due to different meteorological situations, topography, land use, etc.*"

P16297, line13: Can the author briefly expand on what a "…flexible gas phase…" means and why this choice has been made?

Response: We have clarified in Section 2:
"*In the present study, a specific version of the WRF-CHEM model (Grell et al., 2005) is used for photochemical ensemble simulations. The version of the WRF-CHEM model is developed by Li et al. (2010; 2011a, b; 2012) at the Molina Center for Energy and the Environment, with a new flexible gas phase chemical module which can be utilized in different chemical mechanisms, including CBIV, RADM2, and SAPRC. The gas-phase chemistry differential equations are solved by an Eulerian backward Gauss-Seidel iterative technique. The short-lived species, such as OH and* $O(^1D)$*, are assumed to be in steady state. The solution is iterated until all species are within 0.1% of their previous iterative values.*"
The flexible gas phase module can avoid changing the code in the WRF-Chem model when the different chemical mechanism is used.

P16297, line15: As above, expand briefly on the "…Non-traditional SOA…"

Response: We have briefly expanded the description about the non-traditional SOA module in Section 2:
"*The secondary organic aerosol (SOA) formation is simulated using a non-traditional*

SOA model including the volatility basis-set modeling method in which primary organic components are assumed to be semi-volatile and photochemically reactive and are distributed in logarithmically spaced volatility bins (Li et al., 2011a). The partitioning of semi-volatile organic species is calculated using the algorithm suggested by Koo et al. (2003), in which the bulk gas and particle phases are in equilibrium and all condensable organics form a pseudo-ideal solution (Odum et al., 1996). Nine surrogate species with saturation concentrations from 10^{-2} to 10^{6} μg m^{-3} at room temperature are used for the primary organic aerosol (POA) components following the approach of Shrivastava et al. (2008). The SOA formation from each anthropogenic or biogenic precursor is predicted using four semi-volatile organic compounds whose effective saturation concentrations at 298 K are 1, 10, 100, and 1000 μg m^{-3}, respectively. The NO_x-dependent SOA yields from anthropogenic and biogenic precursors are included (Lane et al., 2008), and the oxidation hypothesis of semivolatile and intermediate volatile organic compounds by Grieshop et al. (2009) is used. The contributions of glyoxal and methylglyoxal are also considered in the study. Detailed description about the volatility basis-set approach can be found in Li et al. (2011a)."

P16299, line 1: The authors state that boundary condition and emissions inventory are kept unchanged for all ensembles. Does this apply to the biogenic emissions too? It may not be important, but has the author considered this?

Response: No, the biogenic emissions are calculated using the MEGAN model (Model of Emissions of Gases and Aerosols from Nature) developed by Guenther et al. (2006, 2007). We have clarified in Section 2:
"*The emission inventory used in this study is developed at the Molina Center by Lei et al. (2012), which includes fossil fuel combustion (mobile, area and point sources) and open burning of biomass and trash. The biogenic emissions are calculated using the on-line MEGAN model (Model of Emissions of Gases and Aerosols from Nature) developed by Guenther et al. (2006), in order to consider the variations of biogenic emissions due to the temperature change in the ensemble simulations.*"

P16299, line 6: The Authors have chosen the 29th, but why?

Response: The large-scale meteorological situations on 24 and 29 March are similar at 500 hPa and 700 hPa, but the strength of southerly winds at the surface on these two days are different, leading to the different movement of plumes formed in Mexico City. Considering the similarity and difference of meteorological conditions on these two days, we have provided a more detailed analysis on the simulations on 29 March and attempted to highlight the difference of simulation results on 24 March compared to those on March 29 to avoid the repetition of the analysis method.

P16300, section 4.1: The authors often state that ensemble means better performance compared to the reference deterministic forecast. However, a quantitative index may be a

useful addition. Something like a Taylor plot showing correlation, bias and perhaps mean gross error for the ensemble mean, min, max and best member may help to visualize each selected member performance.

Response: We have added a Taylor diagram (Taylor, 2001) (Figure 5) to present the variance, bias and correlation of the modeled [SOA] and [POA] against observations at T0 on 29 March for the ensemble mean, best, minimal, and maximal member, and the reference deterministic forecast. We have clarified in Section 4:
"*Figure 5 is a Taylor diagram (Taylor, 2001) to present the variance, bias and correlation of the modeled [SOA] and [POA] against observations at T0 on 29 March for the ensemble mean, best, minimal, and maximal member, and the reference deterministic forecast. As shown in Figure 5, overall, the ensemble mean and best member exhibit better performance than the reference deterministic forecast.*"

Minor comments:

P16295, line 17: the sentence "They found that the …" is not clear, please reword.

Response: We have rewritten the sentence in Section 1: "*They found that the largest unpredictability in O_3 simulations was attributed to the increasing uncertainties in meteorological fields during peak O_3 period, and the impacts of wind speeds and PBL height on O_3 simulations are more straightforward.*"

P16299, line16: In Figure 3 it is difficult to distinguish the wind barb (at least in my copy). I suggest making the wind barb larger and maybe showing only half of them. Also the labels of the geo-potential height are too small. In the caption the GFS-FNL is referred in the text as NCEP-FNL.

Response: We have updated Figure 3 as suggested. We have also changed "GFS-FNL" in the caption to "NCEP-FNL" as in the text.

Reference:

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