Reply to Anonymous Referee #3

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.

This paper analyzes the importance of meteorological uncertainties on aerosol simulations for Mexico City using the WRF-Chem model. The study is methodical and the results are relevant to the evaluation of aerosol simulations. Publication is therefore recommended in ACP, subject to a few minor revisions outlined below. (Note only the last 2 digits of page numbers are used)

General Comments:

1. Section 6 ("… other initialization method") seems to be included as an afterthought, and the conclusions drawn from it (pg07, ln10) are possibly an over interpretation. I would recommend including the description of the second method with the initial method (Section 2, pg98). Figures 2 and 12 could be merged which would make it easier to see the difference. I suspect that the conclusion is that there is not much difference between the methods. Concluding that therefore large-scale features dominate the uncertainties (Pg07, ln9-12) seems like a stretch.

Response: We have moved the description of the second method to Section 2 following the initial method. Figures 2 and 12 have been merged as suggested. We have also revised the conclusion in Section 7: "*We have also demonstrated the uncertainties in SOA simulations using another ensemble initialization method, namely "climatology method". These two initialization methods yield very similar results, indicating that there is not much difference between these two methods*."

2. Pg06, ln6-9 mentions very briefly other sources of uncertainty. I would recommend merging this with (Pg7, ln9-12) and expanding the discussion a bit, especially with more references if possible. The study has not evaluated the uncertainties due to model parameterization and input fields other than the meteorological fields. Evaluating these may belong to a future study, but there should be more mention of them here.

Response: We have merged Pg06 ln6-9 with Pg7 ln9-12 and also expanded the discussion in Section 7: "*However, the ensemble mean is not yet consistent with the measurements in both urban and suburban area of Mexico City, showing that meteorological initial uncertainties can only partially explain the uncertainties in the SOA simulations. Other uncertainties, such as those from meteorological model, e.g., PBL schemes (Bei et al., 2010), emission (Li et al., 2011a), SOA formation mechanism (Li et al., 2011a), and photochemical models, should be considered also in the ensemble simulation system. Future studies need to be performed to further investigate the impacts*

of these uncertainties on SOA simulations."

3. In addition to comment 1 above, I would like to see some more detail of the ensemble initialization methods rather than relying entirely on the references. Would it not make sense to show average met fields in Fig 2 to be able to compare with the spread used for the ensembles?

Response: We have added a detailed description of the ensemble initialization methods in Section 2:

For the WRF-3DVAR method: "*A set of random control vectors with a normal distribution was generated. A control increment vector is then transformed back to model space via an empirical orthogonal functions (EOF) transform, a recursive filter, and physical transformation via balance equation. The perturbed variables include the horizontal wind components, potential temperature, perturbation pressure, and mixing ratio of water vapor, whose error statistics are defined by the domain specific climatological background error covariance that are derived from one-month simulations in the same domain using the NMC method (Parrish and Derber, 1992). Other prognostic variables such as vertical velocity (w) and mixing ratios of cloud water (qc), rainwater (q_r), snow (q_s) and graupel (q_g) are not perturbed*."

For the climatological method: "*Deviations of the initial and boundary condition data for each member from the climatological mean for the entire period are then scaled down to 20% to reduce the ensemble spread to below typical observation error magnitudes (Kalnay, 2003) and added to the unperturbed initial and boundary conditions derived directly from the NCEP-FNL analyses valid at 0000 UTC 29 March, which are used for the 12-km domain ensemble simulation."*

We have added standard deviation into the ensemble spread in Figure 2.

Minor Comments:

1. Pg94, ln19-20: What does this mean? Do you mean "the same ratio" instead of "significance"? There are only 2 episodes, so care should be taken before making such a general claim – from Figs 4, 5, 10 and 11 it seems that the ratio of ensemble spread to mean does vary quite a bit.

Response: Yes, we mean the same ratio. We have reworded the sentence in the abstract: "*The magnitude of the ensemble spreads may vary with different meteorological episodes but the ratio of the ensemble spread to mean does not change significantly.*"

2. Pg99, ln13: I found the use of brackets to explain 2 things at once detrimental to the flow of the paper. I would strongly recommend eliminating this to be clearer – even if it means that the paragraph will be longer and there will be some repetition. This method is applied multiple times in the paper (eg. Pg02, ln5-12) and I would recommend removing

it everywhere, including when "[SOA] ([POA]) " is used. (Although in some instances, it seems that "[SOA] ([POA])" is used when you just mean "[SOA] and [POA]".)

Response: We have removed the brackets that are used to explain 2 things at once in the whole manuscript.

Technicalities:

1. Fig 1: Could you mention the terrain heights / intervals in the caption?

Response: We have added the intervals of the terrain heights in the caption of Figure 1: "*Figure 1. (a) WRF domain (red box is the WRF-CHEM domain) and (b) WRF-CHEM domain (red box indicated in Fig. 1a) and the observation sites for aerosol measurements in MCMA (red dot: T0, blue dot: T1). Inner box indicates the domain shown in Figures 6-8. Contours in both panels represent the terrain height with the intervals of 500 m (top) and 200 m (bottom), respectively*."

2. Fig 4: Is the caption wrong? It looks like reference is the blue line and best member is the orange line.

Response: We have corrected the caption of Figure 4: "*Figure 4. The temporal evolution of (a) the surface POA and (b) SOA concentration from each ensemble member (thin green lines), ensemble mean (bold black line), reference deterministic forecast (bold blue line), and the best member (compare to observations, bold red line) of the CTRL ensemble simulations (29 March 2006) and observations (red dots) at T0. (c) and (d) denote the ratio of the ensemble spread and the ensemble mean (brown line) for SOA and POA, respectively.*"

3. Pg02, ln03: "attribute" is not used correctly.

Response: We have corrected the sentence in Section 4: "*The large variations in [SOA] distributions in Mexico City basin are principally attributed to the striking discrepancies in the surface winds between these two extreme members."*

4. Pg06, ln07: "midnight" instead of "the midnight" – please check manuscript for other incorrect uses of "the".

Response: We have changed "the midnight" to "midnight" and also corrected the misuse of "the" in the manuscript.

5. Pg07, ln13: "It is worth mentioning" or something like that instead of "It is worthy to note".

Response: We have removed "It is worthy to note" in Section 7.