Authors response to reviewer #2 comments:

We appreciate these valuable comments and suggestions. We concur with most of them and have addressed them below in the paragraphs marked "AC" (Author Comments):

Major comments:

RC: Major comments: One of my major concerns of this paper is about the simulation design to examine the effects of chemistry (majorly aerosols) on cloud then precipitation. The authors performed two sets of simulations using WRF/Chem: one with the full chemistry component and the other without chemistry and claimed that the differences between them would tell the impacts of chemistry (particularly aerosols) on precipitation. However, there's a major deficiency in the approach used here. In fact, the microphysics scheme (i.e., the Lin scheme in this work) of WRF/Chem will use the prescribed aerosols for cloud activation when there are no prognostic aerosols available from the chemistry module. Thus the differences between two simulations here can only tell the impacts of prognostic aerosols vs. prescribed aerosols instead of the "real aerosol" on precipitation. That's why it's not surprising that we see the overall increase of non-convective cloud precipitation over the domain which is conflicted with the 2nd aerosol indirect effect (more aerosols can lead to more and smaller CCNs and thus suppress the precipitation). So a more accurate way to simulate this effect is to do a 3rd simulation (which is what I would like to see in the revised paper) with chemistry but disable the aerosol emissions including precursors and aerosols related chemistry including cloud chemistry.

AC: Unfortunately, our description of the model setup was incomplete in this respect. We apologise for this omission, which led to the referee's concern. Prescribed aerosols were, in fact, excluded in the WRF calculation, so these did not influence the auto-conversion rate in the (single moment) Lin *et al*. microphysics scheme. This was done *via* parameters in the module_microphysics_driver.F, which calls the prescribe_aerosol_mixactivate code if chem_opt=0 and progn=1 in the namelist.input setting. In our calculations, we set progn=0, thus ensuring that we do not include the interaction of prescribed aerosols with cloud microphysics in the WRF simulations. (We also carried out some test runs for the case where the prescribed aerosol was included (progn=1) and – not surprisingly – we found a change in non-convective rain.) In our WRF calculation, the cloud droplet number concentration was prescribed at a constant value (100 cm^{-3}) in the microphysics scheme. Also, the auto-conversion calculation is based on a simple threshold scheme (Kessler, 1969), *i.e*. the trigger for collision/coalescence of cloud droplets into rain drops is only based on the amount of liquid water and any influence of cloud droplet number is excluded. The double-moment (again Lin *et al*.) microphysics scheme used in WRF/Chem, however, included the influence of aerosols on cloud and precipitation. Therefore, the difference between precipitation from WRF and WRF/Chem simulations in our study indeed represents the changes induced by chemistry and aerosols. We have clarified this point in the Model Description section of the manuscript

RC: Another concern is about the model evaluation as also pointed out by the other reviewer. The authors only conducted the surface evaluation, which in my opinion is a little weak itself as compared to many

other WRF/Chem studies. Only one site was chosen (the reason for choosing it is also not very clear to me) for the time series analysis. Wouldn't the evaluation be more robust if more representative sites (such as urban vs. rural and coastal vs. inland) are selected? Also why don't evaluate 1-hr or 8-hr max O3 and PM2.5 components which are routinely evaluated by most of air quality modelers? The model performance for higher altitudes (aloft) and for cloud-aerosol interactions is also missing. The satellite measurements such as MODIS can provide aerosol optical depth (AOD), cloud optical thickness (COT), and cloud condensation nulei (CCN) data and would be a great addition to the evaluation.

AC: We chose the single Toronto site for detailed comparisons because it is influenced by a variety of sources and thus provides a good test of the model's chemistry-related predictions. It is categorized as an "urban influenced" site because of the presence of a large freeway within a few hundred meters and an industrial area within 1-2 km. It is also 10 km from Lake Ontario, so (technically) it is on a land/water boundary because the model resolution is 12 km. Although it would be interesting to add detailed comparisons at more sites, we do not think this would add much more information about the capability of the model. Instead, this information is provided by the statistical comparisons in Table 2, which are based on the overall domain-wide comparison, including all sites. Also, we feel that the hourly results used in Figure 2 and Table 2 are more informative than averaged data.

We agree that higher altitude comparisons are important and have added comparisons of WRF/Chem AOD and COT predictions with MODIS products in a new Figure 3. Our response to referee #1 has a more complete discussion on this point.

RC: The authors explicitly mentioned in a few occasions that their results showed the impacts of anthropogenic aerosols on the precipitation. Does it mean that the simulation didn't include any dust and seasalt emissions? I failed to see any information regarding this. Please clarify.

AC: Our focus was on the role of anthropogenic aerosols and clearly we overemphasized that in the manuscript, thereby giving this incorrect impression. Certainly, other species including dust, biogenic aerosols and sea salt were also included in the calculation. We have revised the manuscript to make this clear. We have also inserted additional information about the sources of the non-anthropogenic aerosols in sub-section 2-2 (in which the previous sections 2-2-1 and 2-2-2 are merged) and added information about the emission rates for dust and sea salt at the end of that section.

Specific Comments:

RC: Page 27938, line 12-13: it's not surprising because aerosol-cloud feedback was not treated for convective cloud in the current version of WRF/Chem.

AC: We agree. We have rephrased the sentence.

RC: Page 27941, 1st paragraph: This paragraph is redundant and should be removed. The description should be put into the related subsections. For example, the part about emission should go to Sect. 2.2 if it's not covered there. The ones about the model setup should go to the corresponding section.

AC: We have rephrased and reduced this paragraph somewhat to clarify its intended purpose, which is to summarize our approach and note our intention to use the surrogates created here in future scenario studies.

RC: Page 27941, line 13-14: redundant and should be removed.

AC: We have shortened the text, but we believe it provides a useful guide to readers, so we have not removed it completely.

RC: Page 27941, line 19: This version of model was released in 2012 and the references are very old ones. Please cite the more recent papers.

AC: We are very grateful to the referee for this comment. While investigating the reasons for the older references, we found that the version of WRF/Chem used for the calculations was v3.2 and not v3.4 as we had previously thought. This very unfortunate mistake was the result of a change in personnel early in the project. The person who installed the model suddenly left the research group immediately thereafter, leaving notes stating that he had installed v3.4. There was a gap of about two months during which no one worked on the project. When work was recommenced, the model was running properly, so no one checked this aspect. We discovered the error only while responding to this comment. We immediately reviewed all other model components and ensured there were no other discrepancies of this kind. We have corrected this point throughout the manuscript and express our gratitude again to the referee for the careful review that helped us to correct this error.

RC: Page 27942, line 2-3: I don't think MOSAIC simulates methanesulfonate, carbonate, and calcium.

AC: These species are included in MOSAIC (see Zaveri et al., 2008). They are not included in our WRF/Chem output, however, so we have removed this text.

RC: Page 27942, line 7-8: what are the schemes for thermodynamic equilibrium and aqueous-phase chemistry?

AC: The bulk aqueous-phase chemistry of Fahey and Pandis (2001) is used, which includes 50 aqueous-phase species and 147 aqueous-phase processes (21 dissolution

equilibria, 17 dissociation equilibria, and 109 reactions). We have added this information in the Model Description section of the manuscript.

RC: Page 27943, 1st paragraph: this paragraph is mainly for SMOKE and should be moved to Sect. 2.2.1.

AC: Done. Also, Section 2.2.1 has been merged into a new Section 2.2

RC: Page 27945, line 3: +1.27 C (40%). This bias is quite large for temperature and what could be the reason?

AC: The reason for the large percentage bias is simply the fact that the mean temperature for the colder months (1.37 degrees C) is a small number. A small absolute model error, therefore, results in a large percentage error. For the same reason, the percentage bias decreases to 3.5% for the warmer months. While the percentage error is large, we believe that the absolute value of this bias is not excessive.

RC: Page 27945, line 9: what do you mean by the stable? Stable boundary layer condition, which I don't think should be the case? Please clarify.

AC: We agree that this is speculation and possibly incorrect. We have removed the clause "which is more variable during the spring." and revised the next sentence in order to state the information more clearly.

RC: Page 27945, line 12: "especially during the summer months". I don't think it can be told by Fig. 1b.

AC: We agree. The statement was based on other data from the study that were not included in the manuscript. The sentence has been revised.

RC: Page 27945, line 12-13: The reason doesn't seem to be right, since apparently model predicts more rain instead of less rain.

AC: The description here is too brief and we agree that it is unclear. We mean to say that the comparison is made more difficult by the nature of the convective storms that are prevalent in this area during the summer. These storms are relatively brief and localized, so they are not well represented by the (fairly sparse) point measurements that are available. The model resolution is too coarse to reproduce such measurements accurately and the comparison, therefore, suffers. We have deleted the sentence "This is mostly due to difficulty in predicting intense convective rain, which is prevalent in this region" and expanded the following text to clarify the meaning.

RC: Page 27945, line 17-19: The plots for time series of precipitation should be added.

AC: We think it excessive to include the 5 additional monthly plots to the publication. Perhaps this could be added as supplementary material?

RC: Page 27946, line 2-3: It's confusing to me that the overprediction is due to the under prediction of nighttime T?

AC: Correct. This is a typo. "under-prediction" has been changed to "over-prediction".

RC: Page 27946, line 5-6: Again this statement is confusing how the positive bias is due to underprediction at night?

AC: The referee is correct here as well; we apologise for this carelessness. We have removed the statement and revised the text.

RC: Page 27947, 1st paragraph: This paragraph is isolated and doesn't fit there. Please either remove it or move it to the more suitable section.

AC: We have merged this paragraph with the last one in the section.

RC: Page 27947, line 9-13: Figure 2 shows disagreement or large "spikes" instead of just well reproduced time dependence of PM2.5. The statement needs revision.

AC: The "spikes" are the result of using hourly results rather than the more customary time averaged data. We have revised the text.

RC: Page 27947, line 22-25: The content within the parenthesis is well known and should be removed.

AC: We hesitate to remove this because it might not be well known to members of the community who focus on chemistry as opposed to meteorology. It is also relevant to some of the information about cloud-aerosol interactions that we added in response to referee #1.

RC: Page 27949, line 11-13: How about the large areas with warming? What's the causes for it?

AC: This is an important point and should have been discussed more fully. Similar warming is also reported in work by Zhang *et al*. (2010). Our simulations of the vertical aerosol distributions show more light-absorbing aerosols such as black carbon at higher altitudes (*e.g.* 8 km) in the north than in the south. The paragraph

has been expanded to explain this. (See also our response to referee #1 on this point.)

RC: Page 27950, line 14-16: How is aqueous phase (cloud) chemistry treated in this study? Some background information should be provided.

AC: Agreed. The bulk aqueous-phase chemistry of Fahey and Pandis (2001) is used, which includes 50 aqueous-phase species and 147 aqueous-phase processes (21 dissolution equilibria, 17 dissociation equilibria, and 109 reactions). Oxidation of dissolved S(IV) by hydrogen peroxide, ozone, trace metals, and radical species are explicitly treated, as are the non-reactive uptake of nitric acid, hydrochloric acid, ammonia, and other trace gases. Aqueous chemistry processes can lead to the transfer of aerosol particles between size bins due to increased mass from cloud-borne sulfate, nitrate, ammonium, and other ions. We have modified this paragraph to include this information.

RC: Page 27952, line 10: The model performance is reasonably good, but I won't say it's very good based on the statistics table.

AC: We have changed "very" to "reasonably"

RC: Table 2: It might be interesting to show the performance for the WRF run without chemistry as well to see how the feedback could affect the model performance.

AC: This comparison can be seen in the (newly-plotted) Figure 5.

RC: Fig. 1: The stride of scales was too large for both T2 (every 4 degree) and preciptiation (3mm/day) and makes the results looking much better that they should be. Please use smaller ones.

AC: These have been re-plotted in the revised manuscript.

RC: Fig. 5b: The integrated mass in ug m-3? Shouldn't it be ug m-2?

AC: Yes. This has been corrected in the revised manuscript.

technical notes:

AC: Thank you for noting these details. All have been corrected in the revised manuscript

Page 27940, line 14: such an interactive
Page 27940, line 26: and with
Page 27941, line 17: NOAA/ESRL
Page 27942, line 2 and 4: "including" to "include"; delete "certain"; delete "are all included"

Page 27942, line 9: It should be "by Chapman et al. (2009)". There are similar typos throughout the paper and should be fixed as well
Page 27942, line 26: allowing for
Page 27944, line 17: the US.
Page 27944, line 23: "measured results" to "measurements" or "observations"
Page 27945, line 28: ":" to ","
Page 27945, line 29: #60430?
Page 27948, line 8-9: "probable" to "possible"
Page 27948, line 14: The highest
Page 27952, line 21, the prediction of convective precipitation