Authors response to reviewer #1 comments:

We appreciate the valuable comments and suggestions made by anonymous reviewer 1 on the manuscript. All the comments are addressed in the revised manuscript and the point by point response to comments is listed bellow in blue color marked as "AC" (stands for Author Comments):

Major comments:

RC: One concern I have with the paper is the interpretation of the results in the context of the parameterizations employed. The model uses a horizontal grid spacing of 12 km and thus also use a cumulus parameterization to represent convective precipitation. However, aerosol effects on cloud droplets only occur within the resolved clouds (Lin microphysics) and aerosols do not affect the unresolved clouds. Therefore, the results presented represent an incomplete effect of aerosols on clouds in the atmosphere and it is not surprising that "Although, total non-convective rain is less than total convective rain in the domain, chemistry-induced effects on the former are more pronounced than those on the later" as stated in lines 12-13 in the abstract. For example, including aerosol effects on convective clouds could either enhance or reduce the overall affect of aerosols on clouds. While there are currently few convective parameterizations that include indirect effects (they are currently under development), changes in the resolved clouds do indirectly affect convective clouds in addition to the direct effects of aerosols on meteorology. The authors need to put their results into the proper context throughout the manuscript.

AC: We are in general agreement with this comment. We are aware that aerosol activation of cloud droplets is not included in the cumulus parametrization scheme, but perhaps we did not explain this clearly in the manuscript. We have removed from the abstract the somewhat confusing sentence quoted by the referee and have added more detail to the Model Description section of the manuscript.

We did, however, explain that the Grell 3-D option for convective cloud formation was used (because our 12 km resolution is not high enough to resolve convective clouds) and, as the referee points out, this option does not explicitly include aerosol effects. Also, in the second and third paragraphs of the Introduction, we discussed the indirect effects in some detail. We noted that resolved clouds influence cumulus formation through their effect on convection. It is also clear that convection affects resolved clouds by vertical transport of water vapour. In view of this interdependence, our use of the terms "thermal" and "microphysical" in describing convective and non-convective precipitation respectively is perhaps an oversimplification. We have looked again at the remainder of the manuscript to ensure that this does not introduce more confusion.

RC: Another concern I have is how the model has been evaluated and the lack of context regarding previous regional modeling studies of aerosol-cloud interactions. I appreciate the *evaluation done in Section 3, but it relies solely on surface observations and aerosol-cloud interactions occur aloft. It is well known that there can be large variations in PM in the vertical; therefore, the performance aloft is not necessarily the same as at the surface. It is not clear why this period in 2009 was chosen, when there have been campaigns over the past several years that have sampled aerosol concentrations in portions of the domain. The authors could compare simulated AOD with satellite measurements to get an idea of how well the column burden of aerosol is simulated. In addition, there is no evaluation of whether the aerosol-cloud interactions are reasonable. While the authors focus on previous papers that cite the performance of WRF-Chem in terms of air quality, the purpose of the paper is to study the effects of aerosol-cloud interactions on precipitation. At a minimum, the authors should discuss the performance of previous studies of WRF-Chem where aerosol-cloud interactions have been evaluated more rigorously (e.g. Yang et al. 2011; Saide et al. 2011; Shrivastava et al.2013). Another potential metric would be to compare the simulated cloud-top droplet number with MODIS measurements to determine the differences between WRF and WRF-Chem.*

AC: We agree that the inclusion of vertical information in the comparisons is appropriate. To this end, we have inserted a new Figure 3 that compares MODIS measurements with the WRF/Chem predictions for Aerosol Optical Depth (AOD) and Cloud Optical Thickness (COT). We use COT rather than cloud droplet number because it is a more direct satellite product (*i.e.* it does not require retrieval of droplet size distributions). We have discussed the comparison in the Model Description Section. Briefly, it highlights two points. For AOD, there is good general agreement in the south-east part of the domain, but in the north-west there is a significant underestimation. In this part of the domain, almost all emissions are biogenic and the absence of a secondary organic aerosol product in the model configuration used for this work causes this large discrepancy. The measured and simulated COT spatial distributions have the same general shape, but here again, the model underestimates the absolute value significantly. We have added the references mentioned by the referee, which are indeed relevant to the discussion. Please see our comments in response to referee #2 for additional discussion of these points.

RC: The paper neglects secondary organic aerosol formation. That could be an important factor for this domain and time period, which would subsequently affect aerosol cloud interactions. At a minimum, the authors need to discuss the implications of this omission in the model description, where PM is evaluated, and in the conclusions.

AC: As noted above, we agree that the accuracy of the results would be improved by the inclusion of a complete simulation of SOA cloud nucleation processes. In this context, we stated incorrectly that Version 3.4 of WRF/Chem was used; the version used for this study was 3.2, which does not have any SOA capability. (See also the response to referee #2 for further information about this unfortunate error.)

The erroneous model information has been corrected throughout the manuscript. In all future work in this area, we will use more recent versions of the models that have SOA capability. In any case, the implications of this omission have been noted above in the context of the satellite comparisons. We have stated explicitly in the manuscript that the study does not include SOA in order to avoid any misunderstanding. (see pg 27942, lines 14 ff)

Specific Comments:

RC: Page 27938, Line 16-17: It is interesting that the authors examine two particle ranges separately, but what is the motivation for doing so? Where they expecting to be differences in the correlation between the two size ranges?

AC: Yes, we expect the small and large particles to behave differently within the capability of the model. The size ranges chosen are the first and third sections of the 4-bin MOSAIC scheme and represent the nucleation and accumulation modes respectively. Panels (c) in Figures 8 and 9 show that there is a positive correlation between interstitial aerosols and cloud droplet number for small aerosols and a negative correlation for large aerosol particles. We interpret these correlations to indicate that small particles increase the number of cloud droplets, while larger particles are removed by the cloud droplets. These conclusions are reinforced by panels (d) in these Figures, which show that a higher cloud droplet number results in a lower number of interstitial aerosols. We have clarified the abstract to reflect this point.

RC: Page 27938, Lines 17-21: Here more results are presented, but the reader is left to figure out the importance of these statements. Would be useful to clarify what these findings mean.

AC: We have explained these points in the manuscript. In the interest of brevity, we did not think it appropriate to include these explanations in the Abstract.

RC: Page 27940, line 16: McKeen et al. (2007) did not use aerosol-cloud interactions in WRF-Chem. Please check the references for accuracy and/or whether they apply to this statement.

AC: The referee is correct; McKeen *et al* used WRF/Chem, but did not include cloud-aerosol chemistry. The reference has been deleted. The text has been revised to clarify this point.

RC: Page 27940, lines 19-20: The sentence follows WRF-Chem and implies these studies are WRF-Chem studies, but Rosenfeld et al. (2007) and Lynn et al. (2007) are not. I believe they just *use WRF and use prescribed aerosol numbers. Please re-phrase the text to be technically correct.*

AC: We agree. Also, Rosenfeld *et* al is not an appropriate reference for this discussion and we have deleted it. It is also correct that Lynn *et al* did not use WRF/Chem, but their study did simulate the effects of aerosols and so we feel that the reference should be included. We have modified the text to address the confusion about the use of WRF/Chem.

RC: Page 27940, line 23. This sentence seems to begin a new paragraph.

AC: Done

RC: Page 27941, line 17: I suggest replacing "agencies" with "organizations", since some of them are not "agencies". "NOAA, ESRL" should be NOAA/ESRL". The authors should write out what the acronyms are as well.

AC: Done

RC: Page 27942, line 1: The authors note at the end of the introduction that aerosol size is important in terms of aerosol-cloud interactions; however, 4 size bins are used for MOSAIC. How different would the aerosol-cloud interactions be if 8 size bins were used, which is also available in the public release of WRF-Chem?

AC: We agree that it would be interesting to use 8 bins, but we do not believe the extra accuracy would make a qualitative difference to the results. The work is intended to assess the effect of aerosols on precipitation using this particular modelling framework, which is being used in our group for the first time. In view of this, we believe it is adequate to use the smaller number of size bins. We plan future studies that will have more detail.

RC: Page 27942, lines 2-4: It appears that the authors are using a version of the code that does not include secondary organic aerosols (SOA). Given this is often a large fraction of the aerosol mass, especially during the summer, what impact will have that omission have on the present results? Including more organic mass could reduce the average hygroscopicity of the particles and inhibit aerosol activation.

AC: As noted above, we used WRF/Chem v3.2, which does not have any SOA capability. The comparisons with satellite measurements illustrates the consequences of this omission and emphasizes the importance of including SOA. This point is discussed in the text where the satellite measurements are introduced.

RC: Page 27942, lines 15-22: At this point it was not clear what the motivation was for this particular domain. The time period is mentioned in the abstract, but I cannot find any mention of time period in Section 2. Another question is why the summer of 2009 is chosen? If one is interested in how changing emissions affect precipitation, one would think all seasons would be important to investigate.

AC: We chose the Eastern Canada and North Eastern US domain because it is important to our research interests and to those of our funding agencies. We agree that it is desirable to simulate all seasons, but one of the main purposes of this manuscript (in addition to reporting aerosol effects on precipitation) is to report the performance of the model configuration and determine whether it is suitable for application in this region. We are currently working on additional studies in which we consider longer time scales and different seasons. These follow-up studies will be based on the configuration reported here, but will correct deficiencies such as the absence of SOA. We chose the summer period because it is the most important season to study convective *vs* non-convective rain, which is an important focus of the work. We agree that it would be important to include the winter period, but this will also introduce additional difficulties. For example, the coupling of prognostic aerosols to ice nuclei is not properly included in the current WRF/Chem microphysics scheme; this will require correction, especially for use in our domain.

RC: Page 27942, line 25: A 3-day period is rather long one to use without data assimilation. Why not use a 2-day period? However, the relatively small domain may limit large errors in the synoptic fields through the boundary conditions. Some discussion regarding this aspect is warranted.

AC: At the beginning of the project, we did studies of various simulation periods before deciding on 3 days. These showed that negligible changes resulted if we used 2 days re-initialization: the performance was not improved by using the shorter time period. We have added this information to the indicated location.

RC: Page 27944, line 24: Figure 1a has a contour of 4 degrees, which is pretty large and will make the model look better than it really is. I do not doubt the model performs reasonably well, especially when examining time series such as in Figure 2. However, I think at a minimum a 2 degree contour interval should be used in Figure 1b.

AC: We agree. The figures have been re-plotted according to the referee's recommendation.

RC: Page 27945, line 10: The color contour in Figure 1b is biased towards the large values over the ocean where there are no observations. I suggest reducing the to 0.5 or 1 mm up to 5 or 6 mm so that differences between observed and simulated values can be seen more clearly.

AC: Agreed. The figure has been re-plotted.

RC: Page 27945, line 20: The authors mention that some stations have larger errors that may be due to grid spacing. What is leading the author to this conclusion? Are the stations located near land/water boundaries for example?

AC: We have rewritten this paragraph to make it clearer. Our main point is that the statistics of the comparison are poor. Measurements of short, intense, localized rain events by sparsely distributed measurement stations might underestimate the spatially averaged rainfall by missing some events.

RC: Page 27945, line 26-27: The authors should change "WRF-Chem convective scheme" to "Grell 3D convective scheme" to be more specific. They have not shown the performance of other convective schemes in WRF-Chem, so their sentence is implying there is a problem with all the schemes. Also, the errors may also be due to other parameterizations (land use, PBL, radiation, microphysics) as well that will influence the meteorology and affect precipitation. It is not clear why just the convection scheme is blamed here.

AC: We have reworded this to identify the Grell 3D convective scheme.

RC: Page 27945, line 29: It would be useful to include a small panel on the right of each plot showing the diurnal average of the observed and simulated quantities.

AC: We agree that this would be informative, but the plots are already crowded and we hesitate to add more detail. Also, qualitative information about the diurnal differences between measured and simulated values can be seen in the existing plots and we do not think significantly more value would be added by plotting the diurnal averages. We have, however, included the daily mean values, which give part of this information.

RC: Page 27946, line 1: I assume that in addition to the urban land use category, the fact that it is likely located close to a land/water boundary is another factor for making predictions there more challenging?

AC: The site is 10 km from Lake Ontario, but the model resolution is 12 km, so it is technically on a land/water boundary. We think this is of lesser importance, however, than the other urban influences, such as the presence of a large freeway within a few hundred meters and an industrial area within 1-2 km.

RC: Page 27946, line 6: I cannot see the under-prediction in wind speed at night in this figure. Perhaps if a diurnal average were shown, this point would be more clear to the reader. I am not following why the PBL errors would contribute to the wind speed errors. Please be more specific.'

AC: See our response to referee#2. There were typos in the designation of under- and over-estimations. Also, the differences are very small. The reference to PBL errors was speculation and has been removed.

RC: Page 27946, line 27: I am quite skeptical of the authors reasoning with errors in the "treatment of radiative transfer (or photochemistry)". It is equally possible that the simulated cloud cover (which have not been evaluated) is the problem, which would subsequently affect photochemistry. Have the authors allowed the convective clouds to affect radiation? If not, that could be a reason contributing to photochemistry errors. That is a point that should be included in the model description.

AC: This has been re-worded to improve clarity. By "radiative transfer" we meant the effects of simulated cloud cover and the latter, of course, affects photochemistry. Also, the cumulus_radiation_feedback option was turned on in the namelist, so convective clouds did influence radiation

RC: Page 27947, lines 1-6: I do not follow the logic regarding the emission processing and the performance of the model in this paragraph. Having emissions using the simulated meteorology is obviously better than pre-defined emissions, but they have not done another simulation with pre-processed emissions to shown any change in performance. They seem to be suggesting that the current statistics for chemistry is similar to previous studies that use pre-processed inventories, so that at least they did not make the results worse.

AC: We agree it is obvious that SMOKE-processsed inventories give more accurate emissions than interpolated pre-processed data. We have revised this paragraph to remove this confusion.

RC: Page 27947, lines 7-12: The model does not include SOA, so the inclusion of SOA would likely make the bias even higher (21-31% is not that bad, however) for April, May, and June and could improve the results for the other 2 months. The authors need to comment on how missing SOA affects these results. It is hard to know that the PM simulation results are sufficiently "small" as stated in the next paragraph when SOA is excluded.

AC: The paragraph was re-worded to remove some confusion between the measurements of one station, shown in Figure 2, and the averages from 65 stations, shown in Table 2. The new figure showing the comparison between satellite measurements and model predictions of AOD adds further information on the effect of missing SOA.

RC: Page 27947 lines 26-27: The thermal effects (which the authors here imply are from the differences in parameterized convective rain) here are due to changes resulting from both the direct effect and the aerosol effects on the resolved clouds

AC: Agreed. We have dealt with this point in our response to the first comment and also addressed it in the manuscript.

RC: Page 27948, line 1: In Figure 4, what does the total in a single column mean? I suggest changing Figure 4 to have 3 columns per month. One column for observed precipitation and the other two columns for simulated precipitation (WRF and WRF-Chem) – divided into convective and non-convective precipitation.

AC: Agreed. The plot has been changed and the text has been clarified.

RC: Page 27949, line 6: The direct effect will affect both convective and non-convective precipitation.

AC: We have addressed this point earlier in the manuscript and have removed this sentence, which is a repetition of our previous statements.

RC: Page 27949, line 13: Are the units for the column integrated PM correct? Should it not be ug/m2?

AC: Correct. We have fixed the units.

RC: Page 27949, line 15: It is clear that aerosols lead to cooling over the southern part of the domain, but how do aerosols lead to warming over the northern US and Canada?

AC: Our simulations of the vertical distributions of aerosols show there are more light-absorbing aerosols such as black carbon at high altitudes (*e.g.* 8 km) in the north than in the south. A similar point is also made in Zhang *et al*. (2010). The paragraph has been expanded to explain this.

RC: Page 27949, line 26: Should “increase” be “decease”?

AC: the word “increase” here is correct. Although the cloud-resolved rain in the southern part of the domain is less than in the north, the chemistry-induced increase is larger there than that in the north.

RC: Page 27949, lines 27-28: I am not following the logic. Yes there is more non-convective rain in the northeastern US, but the decrease in convective rain in that region is not as strong as in the southern part of the domain.

AC: Here we focus on the difference between the WRF/Chem and WRF results. We are making the point that higher aerosol concentrations in the southern part of the domain cause decreased convective precipitation because of cooling associated with cloud nucleation. Higher aerosol concentrations along the eastern seaboard cause increased precipitation due to cloud nucleation in a region of elevated humidity, coupled with temperature stabilization by the ocean. We have rewritten the paragraph to clarify this.

RC: Page 27950, line 9: Is the primary wind direction really east-north-east? For the whole five-month period? I would have guessed the primary wind direction is from the west to southwest. To they mean that transport is primarily towards the northeast?

AC: We regret the confusion in nomenclature. We plotted the time-averaged wind vectors and their direction was predominantly from southwest toward northeast. We have modified the sentence to read “toward the east-north-east”.

RC: Page 27951, line 9: Suggest dropping “correctly”. Yes the model is producing the aerosol indirect effect, but to say it is correct requires further observations (e.g. observed cloud droplet number) that the authors have not shown.

AC: We have removed the word "correctly".

RC: Page 27951, line 18: This statement could actually be proved by saving the aerosols (by size) removed by precipitation and analyzing those results.

AC: Support was also provided by doing short runs with wet scavenging turned off. The results are stated in the revised manuscript.

RC: Page 27951, line 26: Change “reproduce” to “produce”. To reproduce means the model was compared against some observations which is has not in this case.

AC: Done.