The authors would like to thank both of the reviewers for their interest in the paper and the obvious effort that they put into reviewing it. We have tried to implement as much as we could of their suggestions, even though it increases somewhat the size of the final, revised manuscript.

Below we list the reviewers' comments in bold and our response in italics.

Response to Reviewer 1

Why did the authors compare the hourly diurnal averages instead of comparing the hourly data directly, as is more often done? The authors should justify this choice in the manuscript. I am especially concerned with the E and SW days, because only two days were averaged. Is it really possible to obtain statistically relevant data averaging only two data points?

Actually, the WNW and SW cases had two days each and the E case had six days. As the reviewer suggests, we had considered doing the hourly comparisons, but since one of the objectives was to follow the same lines of evaluation as we did in our 2009 paper, i.e. to compare the properties of the trace gases and aerosols as a function of the air mass origin, we did the same diurnal averaging as we did in the 2009 paper. Our evaluation is not to check if the model reproduces exactly each detail in the time series but to assess the model performance with respect to following the trends and locating the maxima and minima. The statistical relevance is a valid concern if the primary goal was to show how different the environmental properties are with respect to the three cases of air mass origin. We have already addressed this, however, in the 2009 paper and the current manuscript is more directed to understanding these differences with the use of the model.

As recommended by the reviewer, we have expanded upon our choice of analysis strategy, following what we have just described above.

The manuscript should include a description on the measurements error, daily variability and standard deviations of the averages being compared. Also, all these parameters should be taken into account during the description and discussion of the comparisons.

As recommended by the reviewer, we have added to table 3 the measured hourly average standard deviations to compare with the average differences between the predicted and measured values of the parameters. The natural variability is now part of the discussion section.

Furthermore, the Pearson0s r analysis is, in general, not a good choice when comparing data with significant standard deviations associated, as in this case. I suggest to include, scatter plots of the data shown in figures 3 and 4, showing the standard deviations and best linear fits (which should be calculated with a robust method, in order to take into account uncertainties in x and y variables). This would make it easier to visually demonstrate the quality of the correlations. These figures could be included as supplementary material.

The Pearson's correlation coefficient is defined as the ratio of the covariance between two variables to the product of the individual variances. Intrinsically its use is not limited by the magnitude of the variances, as suggested by the reviewer. We could have found higher correlations if we had instead used non-parametric statistics like the Spearman, but we decided to select a more rigorous statistic. Adding dispersion plots for the trace gases and meteorological parameters is a good idea, in addition to the table. We have put these dispersion plots in the supplementary material

As it is mentioned in the introduction, the air masses affecting Altzomoni might have other sources of pollutants in addition to Mexico City, such as the major population centers of Puebla, Cuernavaca, and Cuautla (P9816, L25-28). Also, a recently published paper (Salcedo, D., et al. Sci. Tot. Environ. 414, 417, 2012) showed that pollutant concentrations in the neighboring state of Morelos (where Cuernavaca and Cuautla are located) might be quite high; however, the emission inventory for Morelos is not very detailed. In P9820, L3-6, it is mentioned that the emission inventory described by Fast el al. (2009) was used. However, this emission inventory was developed for the Mexico City Metropolitan Area. What about the other urban areas (Puebla, Cuernavaca, Cuautla)? What was used for them? In this respect, I am concerned with the statement in P9820, L10-12: "Baumgardner et al. (2009) reported that in the area to the east of Mexico City there are no major sources of anthropogenic pollutants, although the small villages in this region may have some minor impact". On one hand, the authors are contradicting themselves, given that in the introduction of the manuscript Cuernavaca, Cuautla and Puebla are described as major population centers along with Mexico City (P9816, L25-27). On the other hand, the statement is probably not accurate given the size of the population and the relatively large industrial activity that exists in the states of Puebla and Morelos. Because of all of the above, the emission inventories of the region being simulated must be clearly described and supported in the manuscript. Also, a discussion is missing of the effects of the error in the emission inventory on the differences observed between measurements and simulations.

The reviewer is, of course, correct in identifying the emission's inventory as the primary culprit when it comes to uncertainties associated with differences between measurements and models. This is one of the principal reasons that we were more interested in documenting the differences in trends rather than absolute magnitude differences, although these are also important to understand. We do describe the inventory that we use. The base year for this inventory is 1999 and was the first emissions inventory developed for the whole country. Hence the state of Morelos is included, as well. Emissions were estimated for each state and municipality for six pollutants: nitrogen oxides (NOx), sulfur oxides (SOx), volatile organic compounds (VOC), carbon monoxide, particulate matter less than 10 and 2.5 micrometers (PM10/PM2 .5) and ammonia. Estimates of emissions of these compounds were made taking into account fixed and area sources, motor vehicles, mobile sources that do not circulate on roads and natural sources (INE-SEMARNAT, 2006). In this inventory Morelos CO emissions only account for c.a. 1% nationally, unlike emissions from Puebla where in some cases such as PM10 brings more than 50% of national emissions. The national emissions inventory of Mexico for 1999, disagree significantly with those reported by Salcedo et al. (2012), since the 1999 inventory reports rates per year much higher for almost all pollutants but CO. Furthermore, the values presented here are similar to those presented in the inventory reported by Baumgardner et al. (2009). We now include this additional information describing the emissions inventory in the manuscript.

OTHER IMPORTANT ISSUES:

P9816, L17. "It is the primary objective of the study described herein, to extend the evaluation by Baumgardner et al. (2009) who looked at the physical and chemical properties of the regional mixed layer of Mexico's Megapolis during the MILAGRO project." The objective of the study is to extend the evaluation of what? With what purpose? The objectives of the manuscript are not clearly described, nor justified, in this or any other statement in the manuscript.

The introduction has been expanded to make clearer the objectives and jutification of the follow-up study described in the current manuscript. The principal objective of this paper is to expand the analysis looking in greater detail at the diurnal trends in the particle size distributions and to use the WRF-Chem model to better understand the behavior of the regional mixed layer and evolution of the trace gas and particle properties that were measured at the elevated monitoring site.

Section 2.1 and Table 1. The time resolution of all measurements should also be indicated. Only for the AMS is time resolution mentioned.

These are now added to the table of instruments.

P9825, L3-7. Water mixing ratio for E and SW days is described, but no for WNW days.
Why?

Mixing ratio for water vapor is included in the figures but inadvertently left out of the discussion. We have now included the WNW micing ratios in the discussion.

P2825, L9-27 and Fig. 4a. The description of the CO trends is confusing and not accurate.

There was an unfortunate mixup when the paper was submitted for discussion. The figures that were in the pdf of the manuscript were correct and the description in the text of the CO trends reflected what the figure shows. We were asked to submit the figures also separately as eps files. Somehow the figure 4 that was converted to an eps was a very old version and bears little resemblance to the correct figure. We ask that the reviewers look at the Figure 4 that is in the revised, annotated document and decide if our somewhat modified description is indeed accurate.

I suggest to include a figure (as a supplemental material if necessary) directly comparing CN and CO traces in order to visually show the similarities.

We have added this figure in the supplementary material as suggested, but only for air origins from the east, not all three directions as this creates a very cluttered graph.

P 9826, L10-24. "The O3 is underestimated by the model throughout the 24 h period regardless of the air mass origin (Fig. 4c). When the model results are shifted two hours later with respect to the measurements, the correlations are all positive and significant at the P < 0.01 level (Table 3), indicating good agreement in trends even though the absolute values are on average more than 0.02 ppm less than observed." According to Table 3, only SW days have a positive significant correlation coefficient (0.598); WNW and E days have a correlation coefficient equal to 0.136 (not in bold, i.e. not statistically significant) and -0.601 (negative), respectively. Although the correlation coefficient for E days is statistically significant, a negative correlation cannot be considered "good agreement". Hence, Table 3 does not show a good agreement in O3 trends for all days as is stated in the

manuscript. In fact, good agreement in O3 between measurements and model is mentioned several times in the manuscript (for example P9830, L19-21 and P9831, L9-11), and is used as a justification to reach some of the conclusions of the study. Given the arguments above, authors should revise the manuscript removing the references to the "O3 good agreement", and check if their conclusions still hold.

The negative correlation between O3 in the model and the measurements was an unfortunate typographical error. We apologize that this provided so much confusion.

P9830, L19-24. "The temperature and water vapor mixing rations were well correlated regardless of air mass origin, as was the ozone. The CO was very well correlated prior to the arrival of the RML but the significant underestimation by the model after that time causes the correlation coefficients to decrease." As mentioned above, the statements related to O3 and CO are not justified.

Please see above our explanation of this mix-up in figure 4.

P9837, L25. "The fidelity of the model, validated by the measurements, is generally quite good. . ." This statement is too general, and not quantitative or descriptive at all. Also, it contradicts most of the discussion, which describes all the differences in CO, O3, AP and PSDs observed.

We have changed this statement to be more quantitative, pointing out that whereas the correlation coefficients are statistically significant at confidence levels less 0.01, the absolute difference are still large. We also have expanded the abstract and summary to be more quantitative when describing the results.

MINOR CORRECTIONS AND SUGGESTIONS:
Fig 1. Because the urban areas of Cuernavaca and Puebla are also mentioned in the text, they could also be included in the map.

The new Fig. 1 includes these cities.

P9816, L26. Should be "Cuernavaca"

Corrected

P9817, L5-20. "In the following presentation we describe observations of diurnal trends in the mass size distributions of sulfate, nitrate, ammonium and organic matter and compare these observations with simulations from the WRF-Chem model." This description of the manuscript does not agree

with the title, nor with the content of the manuscript, which also include modeled trace gases.

Corrected to read "In the following presentation we describe observations of diurnal trends in trace gases and the mass size distributions of sulfate, nitrate, ammonium and organic matter and compare these observations with simulations from the WRF-Chem model."

P9817, L13-18. I suggest to make reference to Fig. 1, where the position of the site is marked in a map. Also, it might be helpful to have the relative altitude with respect to the MCMA, rather that only meters above sea level.

Edited as recommended.

P9817, L23. "AGL" should be "a.g.l." in order to be consistent with the rest of the document, where the latter is used.

Edited as recommended.

Fig 2. The wording of the label and the text describing the figure is very confusing.

As the reviewer notes, the label and text is confusing, probably do to the complexity of the graph. We have rewritten the text to try and clarify what the figure is showing.

Table 3. What do the dashes in the error column mean?

This appears to be a problem when converting to the PDF. Where there are just dashes in the table cell, the word document has numbers. We will double check this in the final version of the manuscript.

P9824, L25. Should be ". . .temperature and water mixing ratios".

Edited as recommended.

P9825, L21. It should be "4a" (?)

Edited as recommended.

P9828, L14. "a decrease" would be more accurate than "a shift".

Edited as recommended.

Table 3. Use WMR, instead of MR for water mixing ratio. Also, define MR (or WMR) in the text and/or the table caption/footnote.

Edited as recommended.

P9830, L20. Should be ratios instead of "rations".

Edited as recommended.

Fig 9 label. "RMA" should be "RML" (?)

Edited as recommended.

Response to Reviewer 2

What is the observed and calculated partitioning between compounds at Altzomoni?

We have now added a figure, and a description in the text, that compares the partitioning between the model and measurements.

P 9816, line 23-25. "... mixed layer also known as the convective boundary layer is the upper portion of the boundary layer .." Typo? Is the lower portion of the boundary layer not convective and not part of the mixed layer?

The reviewer is correct and we have now modified this section of the text, explaining that the mixed layer and convective boundary layer are synonymous and are those layers that form after sunrise as the warmed ground heats the air, creating shallow thermals that rise and cause intense mixing (Wallace and Hobbs, 2006)

Regional mixed layer. I was not familiar with this term. The top 20 or so Google entries are from papers written by the same group as the ACPD article under review. It seems to be a worthwhile concept, and one that you get an intuitive feeling for as the article progresses. However, it could use a fuller definition when it first appears. A comparison to the boundary layer observation of Herndon et al GRL (2008) at Pico de Tres Padres might be useful?

We have expanded our description of the RML, noting that this is a terminology that we introduced in our 2009 paper in order to explain the air that arrives at the research site. Herndon et al. (2008) refer to the Shaw et al. (2007) paper in the same way that we do. We compared the time of day when the RML reached the measurement site in the our 2009 paper, as well asin the current manuscript to the Shaw et al. results.

P 9820, line 12-13 no major sources to the east of Mexico City Are you restricting domain by mountains to the east? Else there is Puebla in next valley to the east.

This sentence has been modified to read "In addition to the cities of Puebla to the east and Cuautla and Cuernavaca to the SW, there are two important natural sources of trace gases and particles: biogenic emissions (from vegetation and biomass burning) and volcanic exhalations."

P 9823, line 15. “aerosol layer that forms at the top of the RML” Do you mean aerosol layer found at the top of the RML? Aerosol at top could have been formed at any depth.

We have modified this to clarify that this is a layer that forms from aerosols, not that the aerosols form, as recommended by the reviewer.

P9824, lines 19-24 It would be helpful to reader to note that the timing of CN will be returned to in discussions of time sequences of other pollutants.

A sentence has been added to this effect.

P 9825 “rations” Typo. Should be “ratios”.

Corrected

P9825, line 11-13 Is agreement for E and SW better than for WNW. Observation – calculation difference for black line appears to be about as good as for red line.

There was an unfortunate mixup when the paper was submitted for discussion. The figures that were in the pdf of the manuscript were correct and the description in the text of the CO trends reflected what the figure shows. We were asked to submit the figures also separately as eps files. Somehow the figure 4 that was converted to an eps was a very old version and bears little resemblance to the correct figure. We ask that the reviewers look at the Figure 4 that is in the revised, annotated document and decide if our somewhat modified description is indeed accurate.

P 9826, line 11 model results are shifted by two hours SW data shifted by one hour.

This paragraph has been rewritten to correctly describe the shifts and correlations for each of the air mass origins.

Section 3.4 Discussion of size distributions presented in Figs 5 - 8 is hard to follow. There are 12 panels, each one of which contains 6 curves. Each curve has 3 attributes, shape, Dp at peak, and amplitude. That is a lot of information to keep in short term memory cache. It might help to consolidate material on Aitken mode, rather than strictly following a species by species description.

This section has been rewritten for better clarity and to decrease the stress on memory caches.

P9832 line 10-13 13:1 dilution for CO Could you present number from which this dilution factor is calculated.

The source of this dilution factor is now described in the text.

P9836, line 21-22 "there were no clouds" How does this impact earlier statement that oxidation of SO2 is enhanced by higher humidity? Is model creating clouds?

Days were selected when visually there were no clouds and although the model will predicted higher relative humidity at night, consistent with the measures, they never exceeded approximately 80%. This is high enough to enhance SO2 oxidation without the need for actual liquid water content from clouds.

P 9833, line 6-7, ozone is a factor of 3 lower Concentrations and figure numbers would be useful.

Modified as recommended.

P 9833, line 14 "measured O3" where and when?

Modified as follows "The measured O_3 under conditions of southwesterly winds, is 0.15 ppm, off the scale in this figure, suggesting that there is much more O3 actually being produced than predicted. The same is seen in the comparisons in easterly air masses where the measured maximum O3 at Altzomoni is 0.11 ppm, twice what is predicted.

Fig. 4a and c. Measured CO and O3 for WNW flow stop at 15:00. Text refers to measured CO at 18:00 (Page 9832, line 3-4)

This has been clarified now in the text.

Fig 7. Observed concentrations of NH4+ are too high in comparison to SO4= and NO3. I will neglect changes in peak width. Consider WNW flow in Figs, 5a, 6a, 7a. The equivalence ratio, NH4+/(2 SO4= + NO3-) is _ [1.2]/18 / (2 * [0.5]/96 + [1.2]/62) = 2.2. Fully neutralized H2SO4 plus HNO3 has an equivalence ratio of 1. The equivalence ratio from the calculations is 0.73. The difference between this number and one may be real or may be due to approximating concentration by peak height.

We are unable to understand if there is a question or concern to be addressed here.

Fig. 9 caption I am having trouble with horizontal bars. It would be helpful to indicate the figures in which the concentrations for the bars are presented. A mark on that figure would be valuable. Or if not in any figure, indicate that.

Edited as recommended