Review of acpd-10-14513-2010 by Hodzic et al., entitled: "Can 3-D models explain the observed fractions of fossil and non-fossil carbon in and near Mexico City?"

Responses to Reviewers

The authors would like to thank both referees for their valuable comments, which have helped us improve the paper. Both referees acknowledged the scientific relevance of the paper and suggested its acceptance in ACP with minor revisions. We have addressed all of their concerns and suggestions as described below. The main modifications to the manuscript as requested by the reviewers were focused on bringing more clarity to the description of the results. For clarity, the reviewer comments are given in regular font below while our responses are italicized and in blue text.

Response to Dr. F. Dentener

R1.0) This is a nice piece of work, containing an careful analysis of a set of measurements and model results of C14 and other carbon relevant tracers over Mexico city, during Milagro 2006. While I have few comments to the scientific part of the manuscript, I found it relatively difficult to work way through the manuscript, due to the very compact writing with many abbreviations. Also I suggest to more clearly present the conclusions from this work, specifically addressing what exactly has been learned from combining model and measurements (compared to a stand-alone measurement analysis).

I also would like the authors to comment better in the revised manuscript on uncertainties of the measurements: on p. 14531 l. 16 there is something mentioned between the lines, but this needs a more prominent place, also in the light of the overall very few measurements available. Having only 4 measurements available on the PM10 filters: what is the risk that agreement is just by coincidence? I therefore recommend publication of this manuscript in ACP, with minor revisions. I mention here that my colleague Stefania Gilardoni also provided comments to the manuscript (... for free ... :)

A1.0) We thank Dr. Dentener and his colleague Dr. Gilardoni for highlighting the merits of the present paper and providing useful comments and suggestions. We have done our best to address all of the comments, as described below.

The uncertainties in the measurements have been addressed further in response to comments R1.16 and R2.3.

Specific comments

R1.1) The readability of the manuscript suffers from a surplus of abbreviations that are not intuitive (although it is good to have a list of abbreviations). At my first reading of the manuscript, I lost track of all things. My suggestion is that were abbreviations are not used more than ca. 5 times, substitute with the full wording. AIK dataset=>PM10; MAR dataset=>PM2.5 dataset. ROB and GRI simulations=> find something more appropriate. T0; T1=>urban- sub-urban.

A1.1) We agree that simplifying the abbreviations and limiting their usage help keep the reader more focused on the results of the paper. As suggested by the reviewers we made the following changes to the manuscript:

- *f_M and f_{NF} were used instead of fC_M and fC_{NF} for fractions of modern and non-fossil carbon.*
- *f^{OC}_{NF}, f^{EC}_{NF}, f^{TC}_{NF} were used instead of fC^{OC}_{NF}, fC^{EC}_{NF}, fC^{TC}_{NF} for fractions of non-fossil carbon contained in OC, EC and TC, respectively.*
- *the usage of T0 and T1 was limited and 'urban' and 'suburban' stations were used more frequently.*
- *biomass burning is now written explicitly instead of using "BB."*
- *we kept however the abbreviations for the ROB and GRI simulations in accordance with our previous paper (Hodzic et al., 2010, ACP).*
- *we have changed the abbreviations for the AIK and MAR datasets to "Swiss" and "US" datasets respectively, reflecting the nationality of the teams that performed the ^{14}C analyses in each case.*

R1.2) I would like the questions on page. 14518 (l. 22 to l. 24) explicitly answered in the abstract and conclusions.

A1.2) The questions on p.14518 read: "The goal of the paper is twofold: (i) to assess whether the current representation of OA in our model can explain the observed levels of non-fossil carbon in aerosols within Mexico City; and (ii) to use the model results to determine the relative contributions of urban sources, biomass burning (BB), and biogenic emissions to the observed levels of carbon in the vicinity of Mexico City."

These questions were already answered in the abstract of the ACPD version of the paper, which we have modified slightly for added clarity as follows:

"Model results show that the relatively high fraction of non-fossil carbon found in Mexico City seems to arise from the combination in about equal proportions of regional biogenic SOA, biomass burning POA and SOA, as well as non-fossil urban POA and SOA. Predicted spatial and temporal variations for f^{OC}_{NF} are similar to those in the measurements between the urban vs. suburban sites, and high-fire vs. low-fire periods. The absolute modeled values of f^{OC}_{NF} are consistent with the Swiss dataset but lower than the US dataset. Resolving the ^{14}C measurement discrepancies is necessary for further progress in model evaluation. The model simulations that included secondary organic aerosol (SOA) formation from semi-volatile and intermediate volatility (S/IVOC) vapors showed improved closure for the total OA mass compared to simulations which only included SOA from VOCs, providing a more realistic basis to evaluate the f^{OC}_{NF} predictions."

And in the conclusion:

"i) The model results suggest that the relatively high fraction of non-fossil carbon found in Mexico City seems to arise from the combination of biogenic SOA sources, biomass burning POA and SOA, as well as non-fossil urban POA and SOA.

ii) Modeling results using the most complete SOA model (ROB simulation) show reasonable agreement with the PM_{10} Swiss dataset but are lower than the PM_1 US dataset. None of the simulations could explain the elevated values of f_{NF} reported by the US dataset, especially at the suburban site. If the Swiss dataset was the most accurate that would imply that our modeling of the organic aerosol mixture is reasonable especially during low biomass-burning periods. Conversely, if the US dataset was more accurate, that would indicate that the model predictions are too low for reasons that we have not been able to explain in this work."

R1.3) Also address more explicitly what was the added value of the model? I think one added value was that the PM1 and PM10 discrepancy could not be explained using model simulations.

A1.3) The added value of the model lies in the quantification of the relative contributions of urban sources, biomass burning, and biogenic emissions to the observed levels of non-fossil carbon inside and in the vicinity of Mexico City as explained for instance in page 14514 (ACPD version): "Model results show that the relatively high fraction of non-fossil carbon found in Mexico City seems to arise from the combination in about equal proportions of regional biogenic SOA, biomass burning POA and SOA, as well as non-fossil urban POA and SOA", and similar text on page 14536.

We also agree that another added value of the model is the fact that simulations could not account for measured differences between the Swiss and US filters suggesting possible measurement problems. This was already explained page 14536 (ACPD version): "The study shows, in agreement with previous assessments, that the difference between the two ^{14}C datasets could not be explained by the different size cuts between the two sets of filters, implying large uncertainties in field measurements of ^{14}C. Much smaller differences (<2%) between PM1 and PM10 f^{OC}_{NF} values is suggested from model results."

The model can also provide spatial and temporal resolution over the domain compared to the point measurements, as already shown in e.g. Figure 8 in the ACPD version. In addition, the model allows exploring different sensitivity cases, such as the use of different parameterizations for urban and biomass-burning SOA, as already discussed in the ACPD paper.

R1.4) p.14514 l. 10 source information on what?

A1.4) "Source information" here refers to the different corrections applied to each source to account for the excess non-fossil carbon due to the atmospheric nuclear bomb testing which depends on the type/age of the source. We modified the corresponding sentence to read:

"The non-fossil carbon fraction (f_{NF}), which is lower than the measured modern fraction (f_M) due to the elevated ^{14}C in the atmosphere caused by nuclear bomb testing, is estimated from the measured f_M and the source-dependent information on modern carbon enrichment."

R1.5) p.14514 l. 13 which known differences?

A1.5) The "known differences" refer here to the known and modeled differences in the impact of carbon sources to PM1 vs PM10, as already discussed on the ACPD version, on p. 14527.

R1.6) p.14514 l. 28 the S/IVOC param has newer insights; better skills, but correlation is as poor?

A1.6) We have changed "better skill" to "improved closure", to more accurately describe the findings of the paper. By improved closure, we mean that the predicted OC concentrations as well as the predicted f_{NF} are closer to the averages of the observations. We agree however that the degree of correlation between model and measurements does not improve, which indicates that substantial uncertainties remain in the measurements and modeling. However the correlation does improve when the effect of the underestimation of biomass burning POA is included in the model (Fig. 8 in the revised version), suggesting that the presence of additional sources of error is causing the lower correlation.

R1.7) p. 14515 l. 1 closing the gap: more quantitive. What was the problem?

A1.7) To improve the clarity of the text, we have changed "reducing or closing the gap between model and measurements" to "reducing or removing the difference in f_{NF} between model and measurements."

R1.8) p. 14516 l. 15 is anything know about the age of the burned material?

A1.8) We have added the following text to section 2.2 to clarify this point: "Assuming that the modern carbon content for biomass burning is similar to the factor of 1.16 for wood burning samples analyzed by Szidat et al. (2009) is reasonable as the wildfires that took place in pine forests in mountains and hills near the city are thought to dominate biomass burning OA in Mexico City during MILAGRO (e.g. Yokelson et al., 2007; Aiken et al., 2010)."

R1.9) p. 14517 l. 2 residence time is days to ca. 1 week.

A1.9) The lifetime of EC is typically estimated with global models as ~1-2 weeks, and this is now more precisely indicated in the paper: "EC is exclusively generated by combustion of fossil fuels and biomass and is effectively chemically inert on atmospheric residence times of 1-2 weeks."

R1.10) p. 14517 in discussing all these measurements: are they fully comparable, e.g. have there been intercomparisons?

A1.10) Most of the ^{14}C data discussed p. 14517 were collected by independent groups and have not been compared until this study, to our knowledge.

R1.11) p. 14518 the outcome of these two main goals should be more clearly mentioned in abstract and conclusions.

A1.11) Done - see response to A1.2 above.

R1.12) p. 14520 it would be useful to know why the large time intervals have chosen like this (sensitivity issue). In your recs you say that this would have to improve. Is this possible?

A1.12) The main reason for collecting 1 or 2 samples per day during the MILAGRO field experiment is the elevated cost of the ^{14}C sample analysis and their limited use until now in the modeling community. E.g. 3h samples can be collected and should provide sufficient material for analysis for future field campaigns, at least in polluted regions.

R1.13) p. 14520 l. 8 explain why fNF for EC at T0 is assumed equal to 0.05 and which is the uncertainty derived from this assumption.

A1.13) This assumption is based on the measurements of f_{NF} for EC at T0, as discussed in Aiken et al. (ACP 2010). Here the average value for filters reported during low biomass burning periods is used i.e. 0.05.

R1.14) p. 14520 l. 14 At T0 the EC/BC ratio varies between 1.8 and 9.3. The ratios indicate that EC and BC do not "agree reasonably well" at this site. It is not clear if the estimate of EC at T0 is based on this agreement; in such a case, please explain comment the uncertainty.

A1.14) It appears that the reviewer may be misunderstanding the quantities shown in Table 3. EC/BC stands for the concentrations of EC (elemental carbon) or BC (black carbon) as we are using both measurements equivalently in this paper, and not for the ratio of the EC and BC measurements. Depending on the measurement technique, both "EC" and "BC" are used in the paper, as appropriate. This is justified given the good agreement between the EC and BC datasets during MILAGRO, see e.g. Paredes-Miranda et al. (2009) and Supp. Info Figure S-1. To avoid confusion, we have changed the heading of Table 3 as "EC (BC)".

R1.15) p. 14520 l. 15 low TIME resolution

A1.15) We gather that the reviewer is objecting to the dash in "low-time" and we have changed this text to "low time".

R1.16) p. 14520 l.20 not only fcm(OC) exceeds also the f (Cnf,oc)>1. You only discuss this later, but it should be here.

A1.16) This is a good point, which was discussed later in the ACPD paper (P14531), as addressed by comment R.1.20 below from this reviewer. We have moved the text from P14531 to the end of P14520 (ACPD version) and also expanded it to address comment R2.3 from reviewer #2, to address the possible sources of uncertainty earlier:

"Finally, for some samples the estimated values of f_{OC}^{NF}>1.0. This may be due to measurement noise, and suggest that the contribution of noise to scatter in the US data is likely to be at least 0.10. Alternatively, perhaps the assumptions for converting f_M into f_{NF} are too conservative (too low assumed average f_M/f_{NF}=1.10) for the whole dataset or

for some samples with high biomass burning impact. A third option is that the assumed value of f_{EC}^{NF} may be too low for some samples. As the EC concentration is only ~1/4 of TC, the uncertainty range of 0.13 - 0.04 = 0.09 for f_{EC}^{NF} (from Table 3) will only cause an uncertainty of 0.03 in the estimated f_{OC}^{NF}. Given the much larger uncertainties in the measurements and model, we have not considered this effect directly in the rest of the manuscript."

R1.17) p. 14520 is there a reason for using a constant factor 1.1? Are the results sensitive?

A1.17) This factor is uncertain, both for the individual sources and also due to the contributions of different sources of modern carbon such as wood burning vs. biogenic SOA vs. cooking. As discussed in P14520, the factor of 1.1 was chosen "under the assumption that modern carbon comes in equal proportions from sources with similar ages as wood and recently photosynthesized biogenic material", which is consistent with previous source estimates from many previous publications from the MILAGRO study, as summarized in the recent overview / review of MILAGRO by Molina et al. (ACP, 2010). We have also added the following text to the paper to characterize the sensitivity to this parameter: "Since the extreme values for this parameter are thought to be 1.16 for wood burning and 1.065 for biogenic SOA, we estimate the sensitivity of the results to the choice of this parameter as a few percent."

R1.18) p. 14521 why so few measurement. And can the correspondence just be coincidence?

A1.18) The small number of measurements was due to both logistical issues with sampling and transporting filters from Mexico City, as well as the high cost of 14C analysis when carbon fractions are analyzed (EC, OC, WSOC). Note that compared with the US dataset, the Swiss dataset has fewer samples but the samples cover longer times and have more "depth" (f_M of several components of TC, rather than of TC only). I.e. the Swiss dataset is based on 12 individual analyses of ^{14}C (4 samples x 3 fractions), which is comparable to the number of individual analyses for the US dataset. The number of samples is sufficient to show a substantial difference between the Swiss and US datasets (see e.g. Fig. 6a), which is currently unexplained, as discussed in the paper.

R1.19) p. 14530 few high BB events not captured by model. Can be corrected for this?

A1.19) The main reason for the underestimation of some BB plumes is related to the timing of smoldering emissions in the late evening and night, and their transport in the shallow nighttime boundary layer (Aiken et al., 2010). The FLEXPART Lagrangian dispersion model, that better represents point sources than WRF, was able to capture most of the variability of certain plumes and show agreement with the AMS BBOA observations (Aiken et al., 2010).

Based on those previous results, a possible way to estimate the effect of the underestimation of BBOA plumes on the ^{14}C budget is to replace the model primary BBOA with the measurements, whenever the measurements are larger than the model. However we prefer to do this only as a sensitivity analysis in one location of the paper, and to not do this for the whole paper, as this hybrid approach introduces other uncertainties and complications. To show the sensitivity to this assumption, we have

added an additional figure (Figure 8a and b in the revised version) where we explore the sensitivity to this correction. The conclusion of this sensitivity analysis is that effect increases the modeled ^{14}C by ~5-10% during the sampling periods and reduces the differences with the measurements.

This is now explained in Section 4.4:
*"**Sensitivity analysis to biomass burning emissions***
The predicted amount of primary organic material originated from biomass burning is another uncertain parameter that could influence the modeled levels of non-fossil carbon. As shown in Hodzic et al. (2010) the biomass burning contribution is underpredicted at the urban site during MILAGRO, especially for nighttime and early morning intense plumes, which could result in an underprediction of the non-fossil carbon fraction. The likely reasons for the underestimation of biomass burning plumes is related to the model resolution (5x5km2) that cannot accurately represent subgrid fire plumes, the representation of smoldering emissions in the late evening and night (Aiken et al., 2010), and the limited accuracy of the wildfire emission inventories that do not account for fires not detectable in a 1km satellite pixel. To estimate the impact of the underestimation of BBOA plumes on the ^{14}C budget, the modeled BBOA was replaced with the measured BBOA at the urban and suburban sites, whenever the measured values were larger than the modeled ones. The sensitivity to this correction is shown on Figure 8. This effect increases the modeled ^{14}C by ~5-10% during some ^{14}C sampling periods and reduces the differences with the measurements (Figure 8a). The improved agreement between model and measurements is particularly noticeable for the two Swiss filters that were taken during periods influenced by increased fire activity and affected by early morning BBOA plumes (i.e. March 21-22). The hourly comparison between the model results obtained with and without this correction during the entire MILAGRO campaign (Figure 8b) shows that only 10% of the modeled f_{NF} values are very sensitive to the representation of the biomass burning in the model. The most significant difference in f_{NF} (0.4-0.5) is obtained for the mornings of March 11 and 21, during which the BBOA plumes of ~20 μg/m^3 was not captured by the model."

And in the Conclusion:
"Correcting for modeling errors for the biomass burning aerosols increased the modeled ^{14}C by ~5-10% during the sampling periods and reduces the differences with the measurements, but could not explain the discrepancies with the PM1 US dataset."

R1.20) p. 14531 0.57-1.07 =>measurement noise: should be discussed earlier

A1.20) This has been done, as discussed in response A1.16 above.

R1.21) p. 14533 what is the general conclusion of 4.4.1: levels better ; correlation not?

A1.21) This is correct, as already discussed in the manuscript, with the following text: "The comparison indicates a strong sensitivity to this parameter, with a near doubling of the modeled f^{OC}_{NF} values when increasing the contribution of urban emissions from 0 to 0.40." and "The scatter in the comparison with the US data does not improve for most of the points." However we note that the correlation does improve when accounting for the underestimation of biomass burning POA in the model, as discussed in the next section of the revised paper and in response A1.19 above.

R1.22) p. 14536 l 10: larger than expected uncertainties. I didn't know what to expect.

A1.22) The sentence has been updated to read: "The study shows, in agreement with previous assessments, that the differences between the two ^{14}C datasets could not be explained by the different size cuts between the two sets of filters, implying large uncertainties in field measurements of ^{14}C."

R1.23) p.14547 Table 3 EC/BC is dimensionless; 0.05(3) should be 0.05(d); for consistency in the footnotes "...EC is assumed to be 5%".

A1.23) This topic has already been addressed in response to comment A1.14 above. Briefly, EC/BC in the ACPD paper stands for the concentrations of EC (or BC) and not for their ratio therefore this quantity is not dimensionless. This confusing notation "EC/BC" has now been replaced by "EC (BC)". The footnotes have also been corrected.

Response to Referee#2

R2.0) This is an interesting manuscript that for the first time directly compares modeled and measured fossil fractions of carbonaceous aerosol. The use of a model to more closely interpret the 14C measurements results is a very promising method and leads to an improved understanding of the sources of OC and EC. The authors do a quite good job of highlighting the remaining uncertainties of measurements, models and emission inventories, which will stimulate further research. The manuscript is, however, relatively difficult to read, due to many abbreviations (sometimes up to 5 or six per sentence), some of which are unnecessary or unnecessarily complicated. This will be addressed in the specific comments. I recommend publication in ACP with some revisions detailed below.

A2.0) We thank the referee for his/her kind comments and suggestions on how to clarify the paper. We addressed all of the comments as described below. We have substantially reduced the use of acronyms in the manuscript, as detailed in response to comment A1.1 of Referee #1.

General Comments

R2.1) Use of fM vs. fNF (Nb. I encourage the authors to just use f instead of fC to denote 'fraction of carbon', for reasons detailed in the specific comments). Despite the advantages of using fNF detailed in section 2.1 I would argue that the use of fM would be scientifically 'cleaner'. This parameter is directly measured. The conversion of the measured fM to fNF with a constant factor of 1.1 for the whole measurement period is less accurate than a conversion of the modeled fNF values to fM. The latter automatically takes into account the day-to-day variability of sources and the speculations on page 14531, line 18-21, whether a factor of 1.1 is also applicable for biomass burning periods could be avoided. However, I respect the author's decision of using fNF, and I anyway expect that the difference between the two approaches is not large. It would be good to see a short estimate based on the model, how this conversion parameter could vary for different scenarios (based on the f_M/f_{NF} ratios from page 14520 and the modeled aerosol composition).

A2.1) We have changed the notation from "fC" to "f" for simplicity as discussed above.

Regarding the choice of f_M vs f_{NF}, there are arguments in favor of using each metric, as already discussed on section 2.1 of the ACPD paper. The overriding argument for us is that f_{NF} is the physically meaningful quantity, while f_M tends to cause much confusion as many researchers automatically interpret f_M as if it was f_{NF}. We estimate the uncertainty in the conversion factor to be only a few percent, based on the time variation of the sources, and the fact that pure biomass burning OA has f_{NF} = 1.16 and pure biogenic SOA has f_{NF} = 1.065 while urban non-fossil OA will likely have a value in between, and all three sources are active for most of the campaign. As our paper clearly shows, at present there are uncertainties in the measurements and models which are much larger than the uncertainties in estimating f_{NF} from f_M. For these reasons we have chosen to retain the use of f_{NF} in the revised manuscript.

R2.2) I do not necessarily agree with the interpretation of Figure 2: If, for clarity, only the urban data from the urban station T0 were plotted in the first two panels it be quite obvious that the REF model does a better job of reproducing the observations for T0 than the ROB model, especially the variability.
Even for the PM2.5 (MAR) data set the measured f_{NF} values range from 0.3-0.7 and the REF f_{NF} also range from 0.3-0.7, whereas the ROB f_{NF} values fall into the narrow range of 0.3-0.45. It seems that for the urban location the inclusion of S/IVOC increases the modeled carbon amounts, which leads to a better agreement with the observed carbon amounts, but at a cost of an underestimation of f_{NF}. This is also in line with the overestimation fossil SOA by the MAR model that can be seen in Figure 9b. Might this be an indication that S/IVOC are a bit overrepresented in the model? Or that they might derive less from fossil sources than assumed? A more detailed discussion would be helpful at his point. Also, since aerosol processes and sources are quite different for the urban and suburban stations, it would be better if the data from both stations would be plotted separately in 6 panels.

For the suburban station the range of modeled f_{NF} values does not change drastically between the models. Why is that?

A2.2) We disagree with the reviewer's interpretation of Figure 2. It is true that there is more variability on the model results with the REF model and less with the ROB model. However in the REF case there is still no correlation between the model and the observations, so the higher variability may be present on that simulation for the wrong reasons. For example for the REF simulation f^{TC}_{NF} is slightly ANTI-correlated with the observations for either dataset, and this effect is lessened with the ROB or GRI simulations. And as discussed on response A1.19 above and in a new section in the revised paper (and the new Figure 8), the correlation between model and observations at the urban site appears to be more affected by the underestimation of biomass burning POA during some early morning plumes at T0.

In addition the REF simulation greatly underestimates total SOA while doing a better job in predicting urban POA / HOA (Hodzic et al., ACP 2010). In particular, the biogenic SOA fraction seems to be well captured by the REF model, while we have shown a large underprediction of the fossil contribution. Therefore we do know that the REF simulations should not be trusted for the determination of f_{NF}. This was already explained in the ACPD manuscript, and has been slightly reworded for clarity as: "As discussed in Hodzic et al. (2009, see Figure 11) the traditional approach can predict biogenic SOA fairly well when compared to specific tracers, which is consistent with studies at other

locations. However SOA formation associated with urban emissions is severely underpredicted by the REF simulation leading to a larger relative fraction of non-fossil TC within the city for the wrong reasons."

It is true however that the ROB simulation does not improve the degree of correlation between the measurements and observations, and this does point at many remaining uncertainties in the simulations and measurements, as discussed in the paper. One such uncertainty is the likely underestimation of primary BBOA in the model during some early morning periods, as addressed in a new section of the paper and response A1.19 above. When this effect is taken into account, the correlation does improve at the urban site. In addition, the observed f_{NF} may be too high for the US dataset as explained in section 4.4 which might be one of the reasons for the model underprediction of f_{NF}. Regarding fossil SOA in figure 9, this quantity is calculated by difference and is affected by uncertainties in the other fractions, which precludes drawing strong conclusions from that comparison.

Also, we do not see the need for plotting the results for T0 and T1 as separate panels and increasing the number of panels from 3 to 6, as the difference between the two stations is clearly visible: results for T0 are plotted in red while the ones for T1 are indicated in blue.

R2.3) In the manuscript a lot of emphasis lies on comparing measured and modeled f_{NF}^{OC}, however this quantity is not measured directly, except in 4 cases. For the PM2.5 MAR data set f_{NF}^{OC} is calculated using two major assumptions: first that f_{NF}^{EC}=0.05, whereas it could probably vary from 0.04 to 0.15 or even higher; second that the EC/TC ratio is known, which is notoriously difficult to determine and quite method dependent. Even if the methods agree reasonably well, the uncertainty of this ratio is considerable. What typical uncertainties do these assumptions introduce for f_{NF}^{OC}? I think this should be taken into account for the intercomparison with the model and for f_{NF}^{OC} values >1.

A2.3) It is true that f^{EC}_{NF} is uncertain, however the effect of this uncertainty is small. We have added the following text to address this point in section 2.2: "As the EC concentration is only ~1/4 of TC, the uncertainty range of 0.13 - 0.04 = 0.09 for f^{EC}_{NF} (from Table 3) will only cause an uncertainty of 0.03 in the estimated f^{OC}_{NF}. Given the much larger uncertainties in the measurements and model, we have not considered this effect directly in the rest of the manuscript."

Specific Comments:

R2.4) Use of abbreviations: The many abbreviations make this paper very difficult to read. I strongly suggest changing fC_{NF} to f_{NF} etc. It is not so easy to distinguish MfC_{NF}^{OC} from fC_{NF}^{EC} or from fC_{F}^{EC} at first glance. Constantly pausing for a second look very much distracts from reading the paper and taking in the content. One letter less in these complicated expressions really helps quite a bit and 'fM' is anyway a quite common expression for 'fraction of modern carbon'. Other abbreviations could be made more intuitive following suggestions from Reviewer 1. "BB" could always be written as biomass burning, since there is no immediate need to make the manuscript as short as possible.

A2.4) We agree that the use of abbreviations can make the paper difficult to follow, and therefore we have included the simplifications suggested by both reviewers as already explained in our response A1.1 above.

R2.5) Abstract, p14514, line 7-9: "which is… testing" this explanation could be omitted in the abstract. People familiar with 14C analysis know this and for people not familiar this is confusing here.

A2.5) We prefer to keep this sentence as this is not understood by most people in the organic aerosol community, who are for the most part not familiar with 14C analysis, and otherwise this can cause much confusion. E.g. the Marley et al. (2009) paper reported f_M with no mention of the difference between f_M and f_{NF}, and their reported f_M values have been quoted in multiple presentations and papers as if they were f_{NF} values. For this reason we believe that there is a need to clarify this point, especially for people interested in organic aerosols in Mexico City and other megacities.

R2.6) Abstract, p14514, line 25-30: "… showed better skill in explaining fC^{OC}_{NF}…" I cannot find any comparison of the Ref model with f^{OC}_{NF} in the manuscript and for f^{TC}_{NF} it is not clear to me that the ROB model has a better skill (see general comment 2).

A2.6) As discussed in A2.2. above, we believe that the large underprediction of OA by the REF model is troublesome and the fact that the fractions of non-fossil carbon are similar is most likely a coincidence due to cancellation by chance of several large errors. However, we have rephrased this sentence to better reflect the information which is directly shown in the manuscript, with the following text: "The model simulations that included secondary organic aerosol (SOA) formation from semi-volatile and intermediate volatility (S/IVOC) vapors showed improved closure for the total OA mass compared to simulations which only included SOA from VOCs, providing a more realistic basis to evaluate the f_{NF} predictions."

R2.7) p14531, line 17: I don't think this can be concluded, since f^{OC}_{NF} was not directly measured and the values larger than 1 are more likely due to the assumptions in deriving f^{OC}_{NF}.

A2.7) This point was also raised by reviewer #1 in R1.16 and R1.20. We addressed the two points separately in the ACPD paper, which was confusing. A third source of uncertainty are the value of f_{NF}^{EC} =0.05 used to estimate f_{NF}^{OC} from f_{NF}^{TC} for the US dataset may be too low. We have modified the following text at the end of P14520 (ACPD version), to address these points earlier and in one place:

"Finally, for some samples the estimated values of f_{NF}^{OC} >1.0 which may be due to measurement noise, and suggest that the contribution of noise to scatter in the US data is likely to be at least 0.10. Alternatively, perhaps the assumptions for converting f_M into f_{NF} are too conservative (too low assumed average f_M/f_{NF}=1.10) for the whole dataset or for some samples with high biomass burning impact. A third option is that the assumed value of f_{NF}^{EC} may be too low for some samples. As the EC concentration is only ~1/4 of TC, the uncertainty range of 0.13 - 0.04 = 0.09 for f_{NF}^{EC} (from Table 3) will only cause an uncertainty of 0.03 in the estimated f_{NF}^{OC}. Given the much larger uncertainties in the

measurements and model, we have not considered this effect directly in the rest of the manuscript."

R2.8) p14532, line 25: should it not read '…lower by …' instead of '…higher by …'?

A2.8) Yes, this was an error in the manuscript and we thank the referee for catching it. We have modified this text to read "At T1, where only the US dataset is available, the reported non-fossil OC is higher by 5–8 µgC/m3 compared to the model on most of the days."

R2.9) Table 1: AM and PM commonly refer to the time periods of 00:00 – 12:00 hrs and 12:00 -24:00 hrs are therefore confusing here. Why not use 'day' and 'night'? A 3 superscript is missing in the units for EC.

A2.9) These comments refer to Table 3. We agree that using 'day' and 'night' filters instead of AM and PM is easier to process for the reader, and have made the corresponding modifications in the manuscript. The footnote of Table 3 was modified to read: "(a) Sampling time period corresponds to 6:00-18:00LT for DAY filters, to 18:00-6:00 for NIGHT filters, and to 9:00 to 9:00LT for daily-averaged filters."
The superscript "3" should be "d", that has been corrected.

R2.10) Figure 4: This legend I could hardly understand because it is so condensed. Please describe every panel separately, even if there is some repetition. It is not immediately clear that panel A is for T0 and panel C is for T1. x-Axis label missing in panel b and d.

A2.10) The x-Axis labels have been added in panels b and d. The figure caption was simplified to read: "Figure 4: Comparison of EC diurnal profiles (left panels: a,c) and averaged EC concentrations (right panels: b,d) for the PM_1 US samples as predicted by the CHIMERE model (red line) and as measured (black dots, Aethalometer-Marley, OCEC Doran, see Fig-S1) at the URBAN and SUBURBAN sites. Sampling time periods are given in Table 3. In plots b and d, model values are given only for available US dataset points to allow a more focused comparison with radiocarbon data."

R2.11) Figure 5: x-Axis label missing.

A2.11) The x-Axis label has been added.