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 measurement 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.

General comments:

1) **Use of f_M vs. f_{NF}** (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 f_{NF} detailed in section 2.1 I would argue that the use of f_M would be scientifically 'cleaner'. This parameter is directly measured. The conversion of the measured f_M to f_{NF} with a constant factor of 1.1 for the whole measurement period is less accurate than a conversion of the modeled f_{NF} values to f_M. 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 f_{NF}, 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).

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/IVOCs 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 station, 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?

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.

Specific comments:

1) 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 fC_{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 'f_M' 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.

2) 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 just confusing here.

3) 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).

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

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

6) 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

7) 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.

8) Figure 5: x-Axis label missing