Reply to questions/comments from Anonymous Referee #3

We thank the referee for good comments and questions that will help improve the paper.

Referee Q1:
Section 3.1: The authors provide two references for the range of measured MAC values. The first one is a review paper and the second one, giving a much broader range, is a study representing a single site. This is confusing: how does a review paper end up a much narrower range than a single paper? Some explanation of the variability is needed.

Reply Q1:
This was indeed a bit confusing. The narrow range (9-12 $m^2 g^{-1}$) from Bond and Bergstrom does not cover the full span of reported MAC values; rather it is the range of the most prevalent observed atmospheric MAC in polluted regions. Bond and Bergstrom also cite a number of papers that have shown large variability in atmospheric MAC (about 5-25 $m^2 g^{-1}$)

The wide range given in the second study (Cheng et al., 2011) is a summary of previously published studies, referred to in the paper; the actual single-site measurements (in Beijing) by Cheng et al. gave small variability in MAE (8.45±1.71 $m^2 g^{-1}$, during winter, and 9.41±1.92 $m^2 g^{-1}$, during summer).

We will revise the text in Section 3.1 to make this clear. We will replace the sentences on page 9065, lines 13-16 by the following:

MAC values for ambient aerosols in polluted regions are often in the range 9 - 12 $m^2 g^{-1}$, however, the variability of reported MAC is large, from about 2 to 25 $m^2 g^{-1}$ (Bond and Bergstrom, 2006 and references therein; Cheng et al., 2011).

Referee Q2:
Page 9066, lines 20-26: Five factors for model-measurement differences are provided. The authors should mention that the factors i and v are not really factors that have anything to do with the model itself, but uncertainties in the model input data.

Reply Q2:
Since the emissions are taken from available emission inventories we agree that they can be considered as not truly being model factors (in the sense that they are not internal factors within the EMEP MSC-W model) but in some ways all model inputs (emission amounts and temporal variation, meteorological driving data etc.) could be considered as part of the *model system*; in order to not lengthen the paper too much we suggest just changing ”(model) factors” to ”(model related)” factors, as follows:

There are many other factors that can also contribute to model-measurement disagreement. The main (model related) factors are (i) emission amounts and distribution, (ii) model transport (wind directions and strength), (iii) vertical dispersion, too much or too little, (iv) wet and dry deposition, and (v) time-variation of emissions.

Referee Q3:
Section 3.3: Beside analyzing each location separately, the authors should also state something general about model-measurement comparison. For example, when looking at Figure 4, it seems that measured values have much larger variability than modeled ones. Any particular reason for this?

Reply Q3:
It is correct that the measured EC-values (and even more so BC_e) have larger variability than the model predicted concentrations. There are several reasons for this; the most important is the model resolution (spatial and temporal). The smoothing of regional scale model results, compared to point observations, is unavoidable, given the large grid-sizes (in this case each model grid box is 50 km × 50 km × 90 m) and use of, e.g., 3-hourly meteorology and simplified descriptions of the temporal (and vertical) variation of the emissions. This tends to average out variations that occur on smaller geographical scales and short time scales (especially important for primary emissions, such as BC/EC).

This also explains why the problem with too small model variability is not as big when comparing to the EC measurements as when comparing to BC_e. The measured BC_e used in this study are hourly averages with large variability while the EC samples are usually collected for one day or more. This longer-time sampling leads to a similar averaging of the observed concentrations as in the model. However, some local phenomena can still not be reproduced by the regional scale model. Much higher model resolution is needed to model primary emitted species, such as EC, on short time scales.

We will add some more text to Section 3.3 about general model-performance and model variability compared to the observations as follows.

Updated beginning of Section 3.3:

When comparing model EC concentrations (or any air pollutant) to measurements it is important to remember that the model results represent grid point average concentrations (50 km × 50 km × 90 m, in this study), which limits the ability to reproduce local concentration gradients or short-term fluctuations. This limitation is especially important for primary emitted species, such as EC, and means that model predicted concentrations will have a lower variability than observations.

Page 9067, lines 19-22, replaced by:

The model performs well when compared to measurements of (long term average) EC concentrations at most sites included in this study; for EC the model bias is relatively low, within ±20 %, except for Melpitz (-69 %) and Vavihill (+66 %) (see below). As expected, the model variability is lower than the observed one (Fig. 4) and the mean absolute error (MAE) of the model concentrations compared to the EC-measurements is 36-45%, at five of the eight sites, but higher for Melpitz, Vavihill and Harwell (as discussed below). The correlation coefficients, r, between modelled and measured EC, range from 0.45 at Harwell to 0.91 at Mace Head (Table S2a; note that Mace Head has only nine EC measurements).

Referee Q4:
Conclusions: The authors summarize the main results. Do the authors have any specific recommendations for the future work?

Reply Q4:
Some recommendations for needed future work were included in the conclusions (e.g., the need for more observations of tracers of wood burning and the need for harmonization of measurement techniques for EC and BC) but we will add the following to the Conclusions:

Future plans involve work on investigating the RWC emissions contribution to organic aerosol in Europe and further detailed comparisons of the EMEP MSC-W model to measurements that include tracers of wood burning, in an effort to improve the modelling of both RWC and the impact of open biomass burning.

Minor/technical issues
Referee Q5:
Page 9066, line 9: fossil sources? Maybe fossil fuel sources

Reply Q5:
Yes, this will be fixed in the revised version.

Referee Q6:
The authors could check out the use of tense when reporting the results. In many cases, imperfect would be more suitable than present tense.

Reply Q6:
We will go through all of the text and change from present tense to imperfect where appropriate.