Reply to review 2

We are happy for the positive feedback and we thank the reviewer for helpful suggestions that will help us improve our manuscript. In particular we are grateful for the references provided; they will help in comparing our results to other models and drawing conclusions on what our future development work should focus on.

Replies to general comments

R1. Articles in GMD are required to represent a sufficiently substantial advance in modelling science; therefore the authors need to a better job of communicating the importance of this model and how it will extend/advance previous modelling work. For instance, what are existing regional/global sectional models lacking compared to MATCH-SALSA and what are the major benefits of using this model over the others available? At the very least, it would be good to get an idea of how the model set-up and performance (against observations) of MATCH-SALSA compare to other similar models (particularly the PMCAMx-UF model, which is also a regional sectional model focussed on the European domain). The authors have communicated the technical aspects of the model well, but discussion of how MATCH-SALSA fits in with and compares to existing aerosol models is lacking.

A1. The MATCH model contains a number of advanced features including variational data assimilation (Kahnert 2008) and inverse modelling of aerosol optics (Kahnert 2009) of both surface observations and satellite data. These assimilation techniques are uncommon in models that include advanced aerosol dynamics. The coupling of the aerosol dynamics model SALSA to MATCH leads to a unique modeling system at the scientific frontline.

We will include a discussion of other models and how MATCH-SALSA compares to these (as also suggested by Referee#1).

R2. The Introduction (Section 1) needs some further attention in terms of the number of citations and the quality of the written language. In comparison with the rest of the article, this section is not particularly well written and steps should be taken to make improvements. I have given some specific comments and technical corrections below for more guidance.

A2. We will improve the language of the introduction and update the citations while seeing to remark 1. We thank you for the particular comments and corrections which will help us in doing so.

R3. I strongly agree with Referee 1's comment regarding the layout and order of Sections 4 and 5. When reading through the article I made several comments regarding the lack of reasons given for the model discrepancies (particularly in Section 4.3.1), but realised when reading on to Section 5 that some of these discrepancies were discussed later in the article. To improve the readability of the article I would also suggest moving the discussion of model discrepancies into the relevant sub sections in Section 4 (or at the very least, add comments at appropriate points in the text to state that the model discrepancies are discussed further in Section 5).

A3. We chose to separate these into two sections in the paper for a clearer overview of the discrepancies, as compared to the text in the supplement report which is integrated. We prefer to keep the two separated. However, we realize that we were not clear enough in pointing this out in the text. We have decided to change the title of section 5 (as suggested by reviewer 1) and include more references to this section in section 4 (as suggested by you).

R4. Throughout the article there are numerous references to the supplementary material (report). The supplementary report is extensive and is an important accompaniment to the article. However, to aid the reader and prevent the need to go back and forth between the documents I suggest including some of the sections/tables/figures in the main paper.

A4. We restricted the number of figures and tables in order to keep the manuscript from becoming too long. We also tried to keep down the number of references to the supplement. We may have been too restrictive and we agree with the Referee that some material from the Supplement should be moved to the main article to aid the reader.

Replies to specific comments

R1. Abstract: The sentence on L12-13 "Elemental and organic carbon concentrations are underestimated at many of the sites." contradicts sentence before. I suggest that you alter or combine the sentences on L11-13 e.g. "On the other hand the model performs well for inorganic particle mass (including secondary inorganic mass), but elemental and organic carbon concentrations are underestimated at many of the sites."

A1. We will revise the sentences as suggested by the referee.

R2. Section 1, P3268, L16 L19: Please provide some references of previous studies that have used/described/developed bulk and modal models. See for example the models compared (and corresponding references) in Mann et al. (2014).

A2. We will add the following text to the introduction:

In bulk schemes, typically the total mass concentration of particles, or the mass in a certain size interval is modeled – which has been a method of choice in MATCH (before the present work). LOTUS-EUROS (Schaap et al., 2008) and DEHM (Christensen, 1997; Frohn et al., 2002) are two other examples of bulk scheme models.

In modal schemes, the aerosol size distribution is represented with a small number of modes, typically assuming lognormal size distribution shapes for the modes. The description of new particle formation is limited in modal schemes. Modal schemes are computationally more expensive than the bulk approach, but less than the sectional, which is why they are common in regional and global CTMs and climate models, e.g. the Regional Particulate Model (Binkowski and Shankar, 1995), CMAQ (Byun and Schere, 2006), CAM5-MAM3 (Liu et al., 2012), TM5 (Aan de Brugh et al., 2011), GLOMAP-mode (Mann et al., 2012), EMAC (Pringle et al., 2010), ECHAM5-HAM2 (Zhang et al., 2012), GISS-MATRIX (Bauer et al 2008).

The sectional scheme, in which the size distribution is represented by a large number of discrete bins, is the most flexible and accurate choice – but computationally the most expensive. Many modern CTMs and global climate models (GCMs) include the sectional approach, e.g. PM-CAMx (Fountokis et al., 2011), GLOMAP-bin (Spracklen et al., 2005a, 2011; Reddington et al, 2011), ECHAM5-SALSA (Bergman et al., 2012), and GISS-TOMAS (Lee and Adams 2010). Mann et al. (2014) compare the performance of 12 global aerosol microphysics models using modal and sectional approaches. We will discuss our performance in relation to theirs.

R3. Section 2.3, P3274, L24 – P3275, L6: The text describes that MATCH-SALSA can be coupled to an online cloud activation model. I assume this coupled model is only used for quantifying cloud drop number concentration and is not used in this study? Please clarify this.

A3. The cloud activation model is used for quantifying the cloud droplet number concentration. The activated fraction of particles is coupled to one version of the wet scavenging scheme. We will explain this more clearly in the revised manuscript.

R4. Section 3, P3276, L1: Are the vertical levels in the model terrain following? Please state this in the text.

A4. We will include the following information in the revised manuscript: The vertical distribution is inherited from the meteorological model, which in this case is hybrid (η) coordinates, with shallow terrain following layers close to the ground and thicker pressure levels higher up.

R5. Section 3, P3277, L1: Please include reference(s) after "95–100% in European scale models".

A5. This is by Spracklen et al. (2005), which will be clarified.

R6. Section 3 (general): How are oxidants treated in the model? Are they online or specified from offline fields?

A6. The oxidants are calculated online in the model using the photochemistry scheme described in section 2.1. Some further details about the chemistry scheme will be added as Supplementary material as requested by Referee #3.

R7. Section 4 (general): What model level is used to compare with observations? Is the model output interpolated to the location of the ground station? Please give details.

A7. We will clarify that we use first model level results everywhere (with no interpolation to the height of the measurement stations).

R8. Section 4.2.2, P3279, L4-6: Firstly, is the correlation coefficient quoted here r or r^2? If these values are not squared, they indicate particularly low correlations between the model and observations. How do these values compare to other models (including ECHAM5-HAM-SALSA) that have been evaluated against observations from the same ground stations (e.g. Spracklen et al., 2006, 2010; Fountoukis et al, 2011; Reddington et al., 2011; Bergman et al., 2012)? In particular with regards to the comments on model resolution, do the global models (with grid sizes on the order of 200 km x 200 km over Europe) show weaker correlation with these observations relative to MACTH-SALSA? Please add some discussion on this.

A8. Our correlation coefficient is the Pearson r-value, and we agree that it is low. We will add a discussion on this and compare the MATCH-SALSA model performance to other models as suggested.

R9. Section 4.2.4, P3280, L12-14: Again, can these results be compared to any of the modelling studies listed in the comments above? How does the performance of MATCH-SALSA at simulating nucleation events compare to e.g. the performance of the GLOMAP model (presumably on a coarser grid) at Hyytiala in Spracklen et al. (2006), which captures nucleation events relatively well?

A9. We will revise the text regarding the problems with capturing nucleation events. Further, we will compare and discuss the MATCH-SALSA model performance of nucleation to that of other models.

R10. Section 4.2.4, P3280, L14: The size of the grid cell is quoted here to be $2x10^3$ km^2, but in the description of the model set-up the spatial resolution of the model over Europe is quoted to be 44 km. Please clarify/explain this.

A10. 44x44 km^2 is ca 2000 km^2, but to avoid misunderstandings we will keep to 44kmx44km instead.

R11. Section 4.3.1, P3281, L23: The bias is defined in the supplementary report, but should be defined in the main text (or at the very least the reader should be directed to the supplementary material for the definition).

A11. We will add a sentence in the beginning of Section 4 explaining that the definitions of all the statistical measures used in the article are given in the Supplement.

R12. Section 6 (Conclusions), P3286, L17-18: “The model peak PNC occurs at the same or smaller particle size as the observed peak.” To be clearer that this sentence refers to the particle size distribution I suggest changing the sentence to the following: “The model peak in the particle number size distribution occurs at the same or smaller particle size as the observed peak.”

A12. We will modify the manuscript as suggested.

<u>Replies to technical comments</u>

R1. Section 1, P3268, L1: “Especially” should be changed to “In particular,”.

R2. Section 1, P3268, L2: Change “. . .importance for the health impacts..” to “. . .importance for impacts on human health. . .”.

R3. Section 1, P3268, L5-7: Sentence does not read well. I suggest changing it to the following: “As the dynamics of these ultrafine particles are particularly sensitive to the various aerosol microphysical processes, they need to be considered in as high detail as possible in order to describe PNC accurately (e.g. Adams and Seinfeld, 2002).”

R4. Section 4.2.2, P3279, L2: “is general” should be “in general”.

R5. Section 4.2.4, P3280, L9: “Especially” should be changed to “In particular,”.

A1-5. We thank the referee for these corrections. We will change the text as suggested.

R6. Figure 6 Figure 9: Please increase the text size of the legends to make them

more visible.

A6. We will improve the figures and make the legends more visible (as also discussed in the answer to Referee#1).

References

Kahnert, M. Variational data analysis of aerosol species in a regional CTM: background error covariance constraint and aerosol optical observation operators. Tellus 60B: 753-770, 2008

Kahnert, M. On the observability of chemical and physical aerosol properties by optical observations: Inverse modelling with variational data assimilation. Tellus 61B: 747-755, 2009