Authors' response

Regarding the updated manuscript based on the GMDD paper: GMD-2014-63, by Andersson et al. entitled "MATCH-SALSA – Multi-scale Atmospheric Transport and Chemistry model coupled to the SALSA aerosol microphysics model. Part 1 – model description and evaluation".

In this document follows comments and concerns by Referees #1 (pages 2-6), #2 (pages 7-10) and #3 (pages 11-17). After each concern our original replies follow, as previously submitted. Our replies in these pages are exactly the same as previously submitted to GMDD; they are included here for the convenience of the readers.

While working on updating the manuscript, we have decided to incorporate section 5 within the other sections (mainly section 4), thus section 5 is removed as suggested by two of the reviewers. A few of the formulations as we suggested them in the replies have been slightly modified for language. Otherwise our replies are still valid. For all changes between the new manuscript and the GMDD paper, see tracked changes at the end of this document (after page 17).

Reply to review 1

We are happy for the positive response and we thank the reviewer for insightful comments and good suggestions that will help us improve our manuscript.

Replies to major comments

R1) Section 4 of the article focusses on comparing model results with measurements. Unfortunately, possible reasons for deviation of model results from measurements and corresponding suggestions for model improvements are mostly provided in section 5. This considerably affects the readability of the article since the reader already expects such information when reading section 4. Some explanations are provided in section 4 but the corresponding discussions are comparably sparse. For instance, the overestimation of PNC in Melpitz due to nucleation is briefly explained in section 4.2.1, but reasons for underestimation at other sites are not discussed. As another example, in section 4.2.3 it is discussed that the reason for the maximum occurring at too small sizes may be too little condensation onto nucleating particles in the model. However, the reader misses a subsequent discussion why condensation is too inefficient. The reader misses such information when reading section 4 but is surprised to find such details in section 5 later on. To enhance readability of the article I would suggest skipping section 5 and discussing the reasons for discrepancies and possible model improvements directly in the context of the model comparisons with the observations (section 4). A summary of the major improvements needed could be included in the Conclusions section. If the authors decide to refrain from merging section 4 and 5 in this manner, I would urgently change the title of section 5 since ‘Identified issues’ sounds somewhat meaningless. A possible title could be ‘Major reasons for discrepancies and suggestions for model improvement’. Choosing such a title would show readers of the previous sections that this important information is given later in the paper.

Answer, Remark 1: We chose to separate these into two sections in the paper for a clear 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 were not clear enough in pointing this out in the text. We will change the title of section 5 to Major discrepancies and suggestions for model improvements, and explain this in a leading text in section 4.

There is an underestimation in all size ranges in Melpitz, Hyytiälä and Aspvreten. This may be due to problems with wet scavenging or a combination of problems. For the accumulation and Aitken modes the problem can be due to underestimated primary emissions. The underestimation in the nucleation mode implies either a low-biased nucleation mechanism or a too efficient removal (deposition). Further, EC is not included in the Aitken mode in the model (the mass and resulting particle number emissions are distributed on larger particle sizes). This is a model deficiency leading to underestimated total particle number concentration (in the Aitken mode and subsequently in larger sizes as well). Further organic nucleation is not included as a nucleation process in the evaluated base case simulation resulting in possible underestimation of nucleation in areas of high BSOA. Sensitivity tests including organic nucleation will be discussed in part 2 of the paper (Andersson et al., 2014) but a lot of the material is available in the Supplement to the present manuscript (which is available for the reviewer). The sensitivity tests indicate increases of the PNC when including organic nucleation, but there is still underestimation at most sites.

We will add a similar discussion on possible reasons for the underestimation of PNC at Melpitz, Hyytiälä and Aspvreten to the manuscript, including mentioning it in the abstract and conclusions.

R2) It should also be discussed in the manuscript how the results of MATCH-SALSA compare to other European-scale aerosol model results described in the literature. This would show whether the discrepancies found by the authors are model specific or common features of regional aerosol models. If some of the discrepancies occur also in other models they could be due to external forcings, such as an underestimation of emissions or general lacks of knowledge e.g. about SOA formation. Such analysis would help to evaluate the overall quality of MATCH-SALSA.

Answer, Remark 2: Thank you for the suggestion; we will add a section, with a comparison of MATCH-SALSA performance to other aerosol models.

Replies to minor comments

R1. Abstract: The acronym SALSA should be explained.

A1: The acronym SALSA (Sectional Aerosol module for Large Scale Applications) will be explained.

R2. Page 3269, line 20: The statement 'PNC was not described' should be discussed in more detail. Since fixed particle sizes were assumed PNC could have been derived from total mass of the respective particles. The authors probably mean that prognostic equations for PNC were not included.

A2. Yes, that is what we mean. The text will be modified to clarify this.

R3. Page 3269, line 25: Is this really an iteration (i.e. are the different operations passed multiple times within each time step)? If not, the term 'integration' might be more appropriate.

A3. Yes, integration is more appropriate. The sentence will be changed to: After initializations are completed the model integrates over time.

R4. Figure 1: It should be specified which parts of the flow chart show MATCH and which parts characterize SALSA operations. It is also not clear why output from the aerosol microphysics module is needed as input for the meteorological part of the model. This should be specified in more detail and Figure 1 should be modified accordingly.

A4. We will specify the SALSA components (the Aerosol Microphysics box) in the figure. We will clarify that the arrows show the model integration order rather than data flow; the figure caption will be changed to: Model integration and time stepping in MATCH-SALSA.

Since the MATCH-SALSA model is an offline model, the aerosol microphysics output does not affect the meteorological processing. The calculated CDNC can however be coupled to the wet scavenging of particles, but that feature is not included in this base case simulation that is evaluated in this paper. Such a simulation is evaluated and described further in the supplement report.

R5. Page 3271, lines 15-16, '. . .and a few heterogeneous reactions for nitrogen compounds are included in the model.': Since no reference is provided here, some more details should be added (which nitrogen compound react on what kind of surfaces? Which uptake coefficients are used?).

A5. A very simplified scheme is used for modelling heterogeneous loss of gaseous HNO_3 and N_2O_5:

$HNO_3(g) \rightarrow NO_3^-$(pm, coarse mode)

$N_2O_5(g) \rightarrow 2\ HNO_3(aq)$ [the nitric acid formed in the reaction is assumed to immediately evaporate to the gas phase]

The treatment is based on the original EMEP MSC-W model chemistry (see, e.g., Simpson et al., 1992) with two adjustments:

We apply the pressure-scaling factor $[M]/2.55\times10^{-19}$ for both reactions (Strand and Hov, 1994) and for the HNO_3-reaction we use the reaction rate for low relative humidity conditions (RH<0.9), $k=5\times10^{-6}\times[M]/2.55\times10^{-19}\ s^{-1}$, regardless of the actual RH (as Strand and Hov, 1994).

Note that the nitrate formed in this HNO_3-reaction is considered as *coarse* mode nitrate. This coarse nitrate is treated as bulk particles in the MATCH-SALSA model.

Ammonium chemistry is also handled by means of a simplified treatment:

$NH_3(g)$ reacts instantaneously (and irreversibly) with available sulfate and form ammonium sulfate $(NH_4)_2SO_4$, which is distributed over different particle sizes according to the sulfate distribution in MATCH-SALSA.

If there is excess NH_3(g) available, ammonium nitrate can be formed via the reaction:

$NH_3(g) + HNO_3(g) \leftrightarrow NH_4NO_3(p)$.

The ammonium nitrate is assumed to be in equilibrium and the dissociation constant of NH_4NO_3 is dependent on relative humidity and temperature, using the equations and parameters from Mozurkewich (1993).

We will add this information, together with the full description of the gas-phase photochemistry scheme, in a Supplement to the article. We will also update the text in Section 2.2 as follows:

Particulate nitrogen species are described by a simplified chemistry scheme (see Supplement X), currently handled outside SALSA. Ammonium bound to sulfate was distributed according to the size-distribution of particulate sulfate. Ammonium nitrate was distributed according to the available aerosol surface area. Coarse nitrate was treated separately as a simple tracer compound (not included in the MATCH-SALSA particle modes).

Further we will discuss the consequences of these simplifications in the revised manuscript: This leads to underestimation of both condensational growth of the particle size distribution, and hygroscopicity and thus an underestimation of the cloud droplet number concentration (CDNC) as well as impacts on the PNC and PM.

R6. Page 3272, lines 13-14: '. . . size bins with a constant internal volume ratio.' Should be explained in more detail. What is meant exactly?

A6: The description of the size bins will be clarified. The sentence will be changed to: … size bins with equidistant distribution of the bins on the log-normal scale.

R7. Page 3272, line 15, 'are that are': Skip first 'are'.

A7: This will be corrected.

R8. Page 3272, line 25, 'shrinkage of particles': It should be explained how particles can shrink in the model. Since semivolatile species as nitrate or ammonium seem to be neglected, this could only happen due to water evaporation. Or are other mechanisms relevant here? Are the simplified treatments of nitrate and ammonium (see next point) capable to simulate shrinkage?

A8. The particles can not shrink in the present version of the model. We will update the text.

R9. Aerosol nitrate and ammonium are included by means of a simplified treatment. Since these compounds can be quite important, possible consequences of this simplification need to be discussed. The simplified treatment should be explained in more detail.

A9. See answer to remark 5.

R10. Page 3274, line 6, '. . . sub-cloud scavenging is neglected for these species': It should be discussed why this simplification is justified.

A10. For ozone sub-cloud scavenging is likely to be negligible; O_3 has a very low solubility in water and wet deposition is not an important sink process for this species – ozone concentrations typically also increase with altitude meaning that the falling raindrops are perhaps more likely to evaporate O_3 at lower altitudes than to scavenge it from the air. For SO_2 the neglect of sub-cloud scavenging is likely leading to a slight underestimation of the wet-deposition losses; but SO_2 also has a relatively low solubility and a modelling study of wet scavenging of sulfur Berge (1993) found that sub-cloud scavenging by precipitation was small (only about 1% of the total S-deposition was due to sub-cloud scavenging). The neglect of sub-cloud scavenging for hydrogen peroxide probably leads to a substantial underestimation of wet deposition for this species. In recent MATCH-model runs (without SALSA), that included sub-cloud scavenging of H_2O_2, it was found that sub-cloud scavenging contributed about 20-40% to the total wet deposition of H_2O_2. We will add a discussion on this in the revised manuscript.

R11. Page 3276, lines 3-8: In the description of the size distribution settings it is mentioned that different bins are used for soluble and insoluble particles but mixtures of these particle types seem to be not considered. In the beginning of section 2, however, the authors mention that also mixed particles can be represented in the model. This is also suggested by Figure 2 where ‘aged’ particles are mentioned. Hence it is not clear how aged or mixed particles are considered in the model runs. It seems that the bins termed soluble here include also the mixed particles. This however would imply that the model is not capable to represent purely soluble particles. This should be explained in more detail in the manuscript.

A11. We will take out term "aged" in Figure 2 to avoid confusion. We will also include a more detailed explanation on how mixing of compounds is handled in the model: Size distribution is divided into three subregions and for these different subregions the level of external mixing is different. In the smallest size bins (diameter < 50 nm), all particles are internally mixed. In the second subrange (50 nm < diameter < 700 nm), there are two parallel externally mixed size bins for each size. In the largest subrange (diameter > 700nm), there are three externally mixed size bins: 1) soluble, where above-mentioned soluble compounds are emitted, 2) cloud active insoluble particles, which are mainly composed of insoluble compounds, but which have enough soluble material to activate as cloud droplets, and 3) freshly emitted insoluble, where insoluble compounds are emitted to.

R12. Page 3277, lines 1-2, ‘The emitted sulfate mass was distributed over particle sizes in the same manner as OM.’: It should be explained how these compounds are distributed over the different particle sizes and appropriate references should be given.

A12. OM (and sulfate and EC) emissions are distributed over particle sizes according to emission source sector resolved mass size distributions from Visschedijk et al. (2009). This is explained and referenced on page 3276, lines 22-24. Details about the size distributions are also given in the Supplement (Table 4, page 16). Emissions from most SNAP sectors are described by uni-modal distributions; emission from two sectors (international shipping and SNAP sector 4: production processes) are described by bimodal distributions. We will add this information to the revised manuscript.

Replies to editorial comments

R1. Figures 1 and 8: Some fonts used are hardly visible even when the figure is enlarged. Larger fonts need to be used.

A1. Figure 1 will be updated with larger font size in the revised manuscript. Figure 8 will be divided into 2 figures to achieve larger font.

R2. Page 3279, line 2: Replace ‘is general’ by ‘in general’.

A2. Ok, will be fixed!

R3. Figure 5: The legend (description of colour bars) is hardly visible and should be enlarged. The colours are hard to distinguish and should be replaced.

A3. We will make the legend text larger. We will change the color of the bars for the observed PNC to make them more visible and easier to distinguish from the model bars.

R4. Figure 6: The legend (description of colour) is hardly visible and should be enlarged.

A4. The figure will be updated to make the legend larger.

References:

Berge, E.: Coupling of wet scavenging of sulphur to clouds in a numerical weather prediction model, Tellus B, 45, 1–22, 1990.

Mozurkewich, M.: The dissociation constant of ammonium nitrate and its dependence on temperature, relative humidity and particle size, Atmos. Environ., 27A, 261–270, 1993.

Simpson, D.: Long period modelling of photochemical oxidants in Europe. Calculations for July 1985, Atmos. Environ., 26A, 1609–1634, 1992.

Strand, A and Hov, Ø.: A two-dimensional global study of tropospheric ozone production, J. Geophys. Res. 99, 22877–22895, 1994.

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.

Replies to technical comments

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

Reply to review 3

We thank the referee for a very careful and thorough review of the manuscript and for many comments and questions that will help us improve the paper.

Answer to the general referee comments

R1: The manuscript gives an overview of the new model, but many details on the respective processes are not provided in the manuscript. Instead, it is referred to the Supplement, which turns out be a rather long SMHI report by the same authors. This impairs the readability of the manuscript since one has to search for the corresponding parts in the Supplement. Even so, many details on the aerosol dynamics modelling are still missing. For instance, the description of condensation in the Supplement does not extend over what is already stated in the manuscript text.

A1: Methods for solving aerosol microphysics are listed in Section 2.2. These methods are commonly used and we do not see it necessary to decribe them in detail in this manuscript. The full description of solving aerosol microphysical processes in SALSA are given by Kokkola et al. (2008), which is referred to in the end of Section 2.2. However, the method for solving condensation and nucleation when both sulphuric acid and organics are involved in the nucleation process has not been described previously. We have added the description for it as a supplement for the manuscript as well as to this response to the reviewer.

R2: Unfortunately, all sensitivity tests that could help to evaluate the assumptions made in MATCH-SALSA are presented in part 2 of the manuscript, not accessible to the reviewer.

A2: The sensitivity tests will be discussed in part 2 of the paper (Andersson et al., 2014), as referred to in the manuscript, but a lot of the material is available in the Supplement to the present manuscript (which is available for the reviewer).

R3: The SALSA model has some focus on the prediction of activating cloud droplets. However, the prediction of activating cloud droplets is not included in the current evaluation of MATCH-SALSA. It is mentioned that a more advanced cloud activation scheme can be coupled online, but I got the impression that this is computationally too expensive to be applied operationally. On the other hand, if the prediction of PNC and size distributions is the focus of the new model, then maybe an aerosol dynamics model better suited for prediction of new particle formation should have been preferred for the implementation in MATCH.

A3: The reviewer questions our choice of SALSA for describing aerosol microphysics. SALSA has been developed with the focus of describing particle number concentration and e.g. includes several nucleation mechanisms. Especially the fact that SALSA uses sectional approach for describing aerosol size distribution gives it an advantage over modal aerosol models in simulating new particle formation (see e.g. Korhola et al., 2014). We will add a discussion of this in the introduction of the manuscript.

SALSA was included for description of aerosol dynamics including PNC, size distirbution and for prediction of cloud droplet number concentration (CDNC). There is an option in MATCH-SALSA to couple the CDNC to wet scavenging of particles as described in the manuscript (section 2.3 deposition). Presently there is no online coupling of MATCH-SALSA to a meteorological or climate model. We will clarify this in the manuscript (see also reply A6).

Replies to concerns

R1. The authors state that they do not expect to model BSOA formation in a realistic way and justify this by the need to make progress in the model development. Instead of consequently using the best yield estimate available, the authors chose 30%. By doing so, they ignore yield values currently applied in other models or recommended in literature. The value of 30% would not be so critical if they had decided to use a reasonable saturation vapor pressure for the semi-volatile compounds, instead of setting it to zero. The decision for treating SOA formation in this way might be motivated by the wish to match observed OC, but that is not a justification for making unrealistic assumptions. The SOA parameterization in the model should be revised for example by using the simple parameterization as applied in GLOMAP (Scott et al., 2014), with a fixed molar yield of 13% (equivalent to a 14.3% mass-based yield) of SOA from the oxidation of monoterpenes.

A1. Unfortunately the description of the BSOA treatment was confusing and partly in error. An extremely simplified BSOA-scheme was chosen for the model development phase of MATCH-SALSA, to be updated in future work with the model.

The initial idea was to assume a 10% (instantaneous) yield of non-volatile BSOA from monoterpene oxidation by OH, O_3 and NO_3 (loosely based on Tunved et al., 2006). However, the BVOC-emissions were considered uncertain by (at least) a factor of three (see Langner et al., 2012, for an illustration of the range of model estimated isoprene emissions in Europe – four different chemical transport models predict emissions within about a factor of five; we do not expect the uncertainty in the monoterpene emissions to be lower than for isoprene). Furthermore, we included only the contribution of monoterpenes to BSOA, which means that we excluded some potentially important BSOA-precursors, such as sesquiterpenes and other BVOC emitted from stressed plants.

Considering the large uncertainties in biogenic VOC emissions we performed tests with varying terpene emissions and found improved model performance when using three times larger emissions than those taken from the EMEP MSC-W model. This sensitivity test turned out to become the base-case simulation for the present study. Future development of the MATCH-SALSA model will include a more realistic treatment of SOA-yields from BVOC.

We will clarify these assumptions in the text.

R2. Why is a rather outdated isoprene chemistry scheme used instead of the detailed scheme which is included in the EMEP MSC-W model's EmChem09 scheme? Further, isoprene is not included as SOA precursor in MATCH-SALSA. The authors should justify the use of an outdated isoprene scheme and state whether it is planned to include SOA formation from isoprene oxidation.

A2. The isoprene chemistry scheme used in the MATCH-model is somewhat more condensed, i.e., uses fewer model species and reactions (7 species, 26 reactions) than the EMEP MSC-W scheme (currently, EmChem09: 19 species, 32 reactions). Both are, however, very small compared to more explicit chemical mechanisms, such as, e.g., the MCM scheme (http://mcm.leeds.ac.uk/) with hundreds of species and reactions for the isoprene chemistry.

The EMEP isoprene scheme is based on the isoprene chemistry scheme by Paulson and Seinfeld (1992) (with a few reactions omitted, as described by Simpson et al., 1993); the scheme has been updated with new reaction rate constants and some other changes of the chemical mechanism during 2008-2009 (Simpson et al., 2012).

The MATCH model isoprene scheme was constructed in 1998 (Langner et al., 1998) and is based on the Carter 1-product scheme (Carter, 1996). The MATCH isoprene scheme was updated simultaneously as the EMEP scheme (EmChem09) taking into account the same new reaction rate data. We will add a detailed description of the MATCH chemistry scheme as a Supplement to the paper, including the isoprene chemistry with the reaction rates presently used.

Thus, in our opinion, the MATCH isoprene scheme is not more *outdated* than the EmChem09 scheme. Both the EMEP and MATCH model isoprene schemes were chosen to be computationally efficient and still model ozone formation well (compared to larger chemical mechanisms). The compact MATCH isoprene scheme has been shown to yield comparable ozone concentrations as the somewhat larger EMEP scheme (Langner et al., 1998) and has been successfully used in many studies focused on ozone (e.g., van Loon et al., 2007).

The isoprene emission scheme in the MATCH-SALSA model is different from the completely revised biogenic emission module in the EMEP MSC-W model. The emissions of isoprene in the MATCH and EMEP MSC-W models are compared in Langner et al. (2012); the total European isoprene emissions are about twice as large in the EMEP MSC-W model as in the MATCH model.

The MATCH SOA chemistry is presently under development. The new version of MATCH includes a new isoprene emission model and SOA formation from isoprene will be included in future versions of MATCH-SALSA. We will add information about this in the article.

R3. A so-called "nitrogen gas-particle partitioning" is introduced in this manuscript. However no explanation on the nitrogen gas-particle partitioning and how it is solved in MATCH-SALSA can be found in the manuscript or in the Supplement. Furthermore, it needs to be stated which species are involved in the nitrogen gas-particle partitioning.

A3. The description of the treatment of nitrogen chemistry in MATCH and its (missing) coupling to the aerosol microphysics module in MATCH-SALSA was not clear in the manuscript. We will clarify it in the revised manuscript.

So far, particulate nitrogen species (ammonium, fine and coarse mode nitrate) are not directly included in the aerosol microphysics routines; the MATCH-chemistry routines calculate mass concentrations of these species as a bulk (not particle size-resolved). *After* the aerosol dynamics steps in the MATCH-SALSA model they are partitioned to different particle-sizes. The model particle radii are not affected by this “post-dynamics” addition of nitrogen species and the partitioning on different sizes are only introduced as a way of describing the particulate nitrogen mass size distribution.

The following particulate nitrogen-species are included in MATCH-SALSA: ammonium sulfate [$(NH_4)_2SO_4$], ammonium nitrate [NH_4NO_3] and coarse mode nitrate.

These species are all treated outside the SALSA-module and the ammonium chemistry scheme and gas-particle partitioning are very simplified:

NH_3(g) reacts instantaneously (and irreversibly) with available sulfate and form ammonium sulfate $(NH_4)_2SO_4$, which is distributed over different particle sizes according to the sulfate distribution in MATCH-SALSA.

If there is excess NH_3(g) available, ammonium nitrate can be formed via the reaction:

$NH_3(g) + HNO_3(g) \leftrightarrow NH_4NO_3(p)$.

The ammonium nitrate is assumed to be in equilibrium and the dissociation constant of NH_4NO_3 is dependent on relative humidity and temperature, using the equations and parameters from Mozurkewich (1993). Ammonium nitrate is distributed over different particle sizes according to the available aerosol surface area.

We will add this information, together with the full description of the gas-phase photochemistry scheme, in a Supplement to the article. We will also update the text in Section 2.2 as follows:

Particulate nitrogen species are described by a simplified chemistry scheme (see Supplement), currently handled outside SALSA. Ammonium bound to sulfate was distributed according to the size-distribution of particulate sulfate. Ammonium nitrate was distributed according to the available aerosol surface area. Coarse nitrate was treated separately as a simple tracer compound (not included in the MATCH-SALSA particle modes).

R4. More details on the treatment of the emission of primary particle components, in terms of mass and number need to provided, and how consistency between mass and number of emitted particles is ascertained. Elemental Carbon (EC) is not defined in the first size range (nucleation and Aitken modes) despite EC is emitted from various combustion sources (mainly from residential biomass burning and traffic) in the Aitken size range. The attribution of EC should be revised for the PNC modelling on the European scale where many regions are under the influence of combustion sources.

A4. Particle number emissions are calculated based on the sectoral mass-based emission size distributions from Visschedijk et al. (2009), as referred to in the manuscript. We will add the following sentences to Section 3 of the manuscript to further clarify how the emissions were treated: Details about the size distributions are also given in the Supplement (Table 4, page 16). Emissions from most SNAP sectors are described by uni-modal distributions; emission from two sectors (international shipping and SNAP sector 4: production processes) are described by bimodal distributions.

EC is not included in the nucleation and Aitken modes in SALSA. The fact that EC is not included in the Aitken mode is a shortcoming in the model, which will be updated in future model versions of MATCH-SALSA. We will add a discussion of the missing EC-emissions in the Aitken mode in the revised manuscript.

R5. Nucleation is solved concurrently with condensation using the methodology by Jacobson (2002). Coupling nucleation with growth avoids that one of these processes is favored over the other in the operator splitting. However, the manuscript states that several nucleation options exist in MATCH-SALSA, including for example the activation of sulfuric acid and organic vapors, while the original methodology by Jacobson (2002) was derived for homogenous binary nucleation. Despite the authors mention that the alternative nucleation options are only used in part 2 of the manuscript, it is recommended to present the detailed algorithm for the coupling of sulfuric acid – organic nucleation with growth (give equations!) in part 1.

A5. Since the organic nucleation scheme is not used in the model simulations presented in Paper 1 we will add the detailed algorithm in a Supplement to the article and as an appendix to this reply.

R6. It is suggested to split section 2.3 into two sections, one that deals with deposition and one that deals with cloud droplet activation. If there is any interaction between the two processes in MATCH-SALSA, it has to be stated more clearly as it is the case now. Based on the current description it is not obvious why there is a need for online calculation of cloud condensation nuclei, since the fraction of activated cloud droplets in the standard version of MATCH-SALSA is only inferred from the fraction of particles that are located in grid boxes covered with cloud. That can be done equally well in a simple post-processing of the model output.

A6. One of the wet scavenging schemes (though not used in the evaluated version) is coupled to the CDNC formation, and for this reason it is needed online. This is also the reason for including this in the deposition section. We will make an effort to clarify this to avoid future confusion.

R7. Provide reasons for the underestimation of total PNC at Northern and Central European sites and of accumulation mode particle numbers at all sites, in the Abstract and in the Conclusions. The list of planned developments given at the end of the manuscript is useful, but it does not replace a discussion on how missing processes or inadequate parameterizations have affected the presented model results. Specifically, it has to be assessed how the obvious shortcomings in the treatment of BSOA and nucleation mechanism affected the prediction of total PNC.

A7. There is an underestimation in all size ranges in three of the northern and central European sites. This may be due to problems with wet scavenging or a combination of problems. For the accumulation and Aitken modes the problem can be due to underestimated primary emissions. The underestimation in the nucleation mode implies either a low-biased nucleation mechanism or a too efficient removal (deposition). Further, EC is not included in the Aitken mode in the model (the mass and resulting particle number emissions are distributed on larger particle sizes). This is a model deficiency leading to underestimated total particle number concentration (in the Aitken mode and subsequently in larger sizes as well). Further organic nucleation is not included as a nucleation process in the evaluated base case simulation resulting in possible underestimation of nucleation in areas of high BSOA. Sensitivity tests including organic nucleation will be discussed in part 2 of the paper (Andersson et al., 2014) but a lot of the material is available in the Supplement to the present manuscript (which is available for the reviewer). The sensitivity tests indicate increases of the PNC when including organic nucleation, but there is still underestimation at most sites.

We will add a similar discussion on possible reasons for the underestimation of PNC at Melpitz, Hyytiälä and Aspvreten to the manuscript, including mentioning it in the abstract and conclusions.

Replies to technical comments

P 3268 L17-20 when discussing modal vs. sectional schemes, examples for the application of both in aerosol dynamics models and the corresponding literature references should be given. How does the sectional approach compare with the modal approach? Mention some advantages and disadvantages of both.

A. We will update the text as follows (partly also based on comments by Referee#2):

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.

P3268 L21-23 PM-CAMx and GLOMAP are mentioned as two examples of CTMs that include aerosol dynamics and are applied on the European scale. Give a short summary of the shortcomings of these models with respect to their capability to predict observed PNC in Europe.

A. We will add a description on the performance of other models and compare these to our own.

P 3269 L2 Exemplify briefly what the new model features of MATCH-SALSA are.

A. We will specify that the new features are the description of aerosol microphysics and particle number size distribution.

P 3269 L22-23 "makes it possible to describe PNC and the mixing state of the particles." Revise language. It has to be explained more precisely what this entails: physical treatment, computation, model output, etc. It should also be stated briefly which size distributions are defined, with reference to section 3 where this is explained in more detail.

B. We will update the manuscript: The coupling of SALSA to MATCH introduces a model description of particle microphysics and aging in the model. New features include particle nucleation, condensation, coagulation and activation; leading to a description of the temporal evolution of the particle number size distribution in a number of bins, through the sectional approach. Further the model describes the mixing state of the particles. For further details on the new physical treatment of aerosol microphysics and the particle size distribution see Section 2.2 as well as further details on the specific set up in this study in Section 3. SALSA was chosen for this task since it was developed with the focus of describing the particle number concentration and e.g. includes several nucleation mechanisms. Especially the fact that SALSA uses the sectional approach for describing the aerosol size distribution gives it an advantage over modal aerosol models in simulating new particle formation (e.g. Korhola et al. 2014).

P3269 L26 "New emissions are emitted"; revise language of this sentence.

A. We will change the sentence to: The integrations are based on the meteorological time step (dtmet) starting with reading or interpolation of weather data, reading emissions, and setting of lateral and top boundary concentrations of chemical species.

P3270 L2 Replace "model chemistry" by "model gas-phase chemistry".

A. Not only gas-phase chemistry is included, also a few aqueous phase and heterogenous reactions are included in the chemistry scheme. These are described in section 2.1 and we will add a complete list of the chemical reactions included in the model in a Supplement to the article.

P3270 Footnote 1 The footnote should be included in the main text because otherwise it is difficult for the reader to comprehend the statement on P. 3286, line 1; which explains the underestimation of OC and PM peaks at Melpitz by a vegetation fire episode.

A. Ok. We will move the text from the footnote to the main text.

P3271 L5. Describe the coupling between gas-phase chemistry and aerosol dynamics. How frequently are gas-phase concentrations of the relevant species (e.g. sulfuric acid) updated by the changes due to condensation and nucleation?

A. The chemistry (e.g. oxidation of SO_2 to H_2SO_4) is solved prior to SALSA using the kinetic pre-processor (KPP). There is no sub-time-step in SALSA. Some further details are given in the Supplement on the model time steps, in addition to what is given in the manuscript. We will add a reference to the Supplement in the manuscript, and clarify that there is no internal sub-time stepping between the chemistry and SALSA or within SALSA itself.

P3271 L17-19. A complete list of the reactions of the MATCH-SALSA model is missing in the manuscript and in the Supplement.

A. We chose to exclude the reaction list since the chemistry is basically the same as in previous MATCH versions but we will add the list of reactions as supplementary material to the revised manuscript.

P3273 L5 The reference to the paper by Lehtinen et al. (2007) is missing in the list of References.

A. We will add the reference: Lehtinen, K.E.J., Dal Maso, M., Kulmala, M. and Kerminen, V.-M. Estimating nucleation rates from apparent particle formation rates and vice versa: revised formulation of the Kerminen-Kulmala equation. Journal of Aerosol Science, 28, 988-994, 2007.

P3273 L17 "accurate over time step length of 7200 s" - presumably this accuracy is only achieved with condensation is the only operative aerosol dynamical process.

A. The reviewer is correct in that Jacobson (2002) has demonstrated the scheme to be accurate over time step length of 7200 s, when condensation is the only operative aerosol dynamical process. We will clarify that it is meant for condensation as the only operative process in the manuscript.

P3277 L7 Replace "($PM_{2.5}$ and PM_1)" by "(PM_1 and $PM_{2.5}$)".

A. Ok, the order will be changed.

P3277 L18 Why was Mace Head chosen as a station for evaluation of PNC? It is known that new particle formation at Mace Head occurs via nucleation of iodine oxides (e.g. Saiz-Lopez et al., 2006). Therefore it cannot be reproduced by a model that uses nucleation parameterizations for sulfuric acid clusters.

A. We will remove the Mace Head PNC evaluation.

P3278 L9 High PNC in nucleation mode along shipping lanes are probably artificial since kinetic and activation nucleation parameterizations tend to overestimate the nucleation rate in the ship plume (e.g. Metzger et al., 2010).

A. We will add the following sentence to the manuscript: Metzger et al. (2010) have shown that the high PNC found in oceanic regions with large ship emissions could be caused by overestimated nucleation when traditional activation type nucleation schemes are used; they found that a nucleation scheme involving both organic molecules and sulfuric acid led to much lower PNC over oceans in better agreement with observations.

P3280 L4-5. This explanation is in contradiction with the fact that the formation of <3 nm particles is parameterized as J3 according to Lehtinen et al., 2007 (see P. 3273, line 4-5).

A. Here we refer to the fact that 3nm particles do not grow to large enough sizes by condensation. This is why we do not see the contradiction between us using the 3nm particle formation according to Lehtinen et al. (2007) and a modelled maximum occurring at too small sizes compared to observations being explained by underestimated condensation in the model.

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MATCH-SALSA – Multi-scale Atmospheric Transport and CHemistry model coupled to the SALSA aerosol microphysics model. Part 1 – model description and evaluation

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Abstract

We have implemented the sectional aerosol dynamics model SALSA (Sectional Aerosol module for Large Scale Applications) in the European scale chemistry-transport model MATCH (Multi-scale Atmospheric Transport and Chemistry). The new model is called MATCH-SALSA. It includes aerosol microphysics, with several formulations for nucleation, wet scavenging and condensation.

The model reproduces observed higher particle number concentration (PNC) in central Europe and lower concentrations in remote regions. The modeled PNC size distribution peak occurs at the same or smaller particle size as the observed peak at four measurement sites spread across Europe. Total PNC is underestimated at Northern and Central European sites

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and accumulation mode PNC is underestimated at all investigated sites. The low nucleation rate coefficient used in this study is an important reason for the underestimation. On the other hand the model performs well for particle mass (including secondary inorganic aerosol components), while elemental and organic carbon concentrations are underestimated at many of the sites.

Further development is needed, primarily for treatment of secondary organic aerosol, in terms of biogenic emissions and chemical transformation. Updating the biogenic SOA scheme will likely have a large impact on modeled $PM_{2.5}$ and also affect the model performance for PNC through impacts on nucleation and condensation.

1 Introduction

Most aerosol properties relevant to climate are both size and chemical composition dependent. Thus, there is a need to resolve the size distributions of particle mass, number and chemical composition in climate models (e.g. Chen and Penner, 2005; Roesler and Penner, 2010). Aerosol particles also have adverse effects on human health (e.g. Pope and Dockery, 2006), which depend on particle size and chemical composition (WHO, 2013). In particular, ultrafine particles (with diameter less than 100nm) may be important for impacts on human health (e.g. Oberdörster et al., 1995; Peters et al., 1997; Knol et al., 2009), but there is still limited epidemiological evidence on their effects on health (WHO, 2013). The ultrafine particles do not contribute significantly to the particle mass concentration (PM) but they constitute a large proportion of the particle number concentration (PNC). Aerosol microphysical processes need to be considered in greater detail in order to describe PNC and size distributions accurately (e.g. Adams and Seinfeld, 2002). This has led to an increased need for realistic treatment of aerosols in atmospheric models.

A number of CTMs, which are used operationally for simulating atmospheric chemistry in Europe, were recently reviewed by Kukkonen et al. (2012). The aerosol descriptions in such models can be classified into three main categories: bulk schemes, modal schemes (Whitby and McMurry, 1997) and sectional schemes (Gelbard et al., 1980). In bulk schemes, typically the total mass concentration of particles, or the mass in a certain size interval, is modeled. LOTUS-EUROS (Schaap et al., 2008), DEHM (e.g. Frohn et al., 2002) and the EMEP MSC-W model (Simpson et al., 2012) are examples of bulk type models.

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Borttaget: for predicting health impacts of regional or global particle concentrations, such detail may be even more crucial than for climate models. ¶
CTMs that are commonly used

Borttaget:), with a previous version of our CTM MATCH as one of the models included in the study.

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In modal schemes, the aerosol size distribution is represented by a small number of modes, typically assuming lognormal size distribution ~~for the modes.~~ The description of new particle formation is limited in modal schemes. Modal schemes are computationally more expensive than simple bulk schemes, but less than the sectional approach, which is why they are commonly used 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 (Fountoukis et al., 2011), GLOMAP-bin (e.g. Reddington et al., 2011), ECHAM5-SALSA (Bergman et al., 2012), and GISS-TOMAS (Lee and Adams 2010). PM-CAMx and GLOMAP-bin make the assumption of internally mixed particles, in GLOMAP described by 20 size bins, whereas GISS-TOMAS includes externally mixed particles described by 30 size bins. Such a high size bin resolution is computationally demanding. GLOMAP uses prescribed monthly-mean oxidant fields. Mann et al. (2014) compared the performance of 12 global aerosol microphysics models using modal and sectional approaches.

The standard version of the MATCH (Multi-scale Atmospheric Transport and Chemistry) model (Robertson et al., 1999; Andersson et al., 2007) uses a simple bulk scheme for treating aerosols, with four size bins for primary particles, without any aerosol dynamics treatment (except hygroscopic growth in some model versions), but with dry and wet deposition of primary particles being dependent on particle size. The particle species considered in previous applications (e.g. Andersson et al., 2007; Andersson et al., 2009) were primary anthropogenic elemental carbon (EC), organic carbon (OC) and non-carbonaceous particles, as well as secondary inorganic aerosol (sulfate, nitrate, ammonium) and sea salt particles. Secondary organic aerosol was not included in the model. PNC formation and growth was not described. MATCH was adapted to assess anthropogenic ultrafine particles in an urban environment in a previous study (Gidhagen et al., 2005); seven monodisperse sizes were used and the aersosol

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Flyttad nedåt [1]: we highlight the main new features and present the results from evaluation tests. In a second paper (Andersson et al.,

Borttaget: 2014) results from various sensitivity tests, with the MATCH-SALSA model, will be presented.

Flyttad nedåt [2]: The aim of MATCH-SALSA is to describe particle mass and number concentrations, and particle size distribution on the European scale.

Borttaget: This is the first of two papers presenting a new aerosol dynamics

Borttaget: a European scale Eulerian CTM; the new model is called MATCH-SALSA and was detailed in a SMHI RMK report (Andersson et al., 2013), which is included as Supplement to this paper. In this paper

Borttaget: The new model features are developed with intention of coupling the model to climate models and radiative transfer calculations, and can also be utilized for the estimation of human exposure to particles. ¶

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Borttaget:). An earlier urban application of MATCH was applied to assess anthropogenic ultrafine particles in an urban environment (Gidhagen et al., 2005); seven monodisperse sizes were used and the aersosol dynamics considered water uptake, coagulation and dry deposition, but no nucleation or condensation processes. In earlier European scale MATCH versions (e.g. Robertson et al. 1999, Andersson et al. 2007; 2009), particles were handled with

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dynamics considered water uptake, coagulation and dry deposition, but without inclusion of nucleation or condensation processes.

The MATCH model includes photo-chemistry for calculating oxidant fields that can be used for online coupling to oxidation of organics and sulphur compounds, resulting in a coupled photo-chemistry and aerosol dynamics description. Further, MATCH contains a number of advanced features, including data assimilation (Kahnert 2008) and inverse modeling of aerosol optics of both surface observations and satellite data (Kahnert 2009). These assimilation techniques are uncommon in models that include advanced aerosol dynamics.

We have implemented the sectional aerosol dynamics model SALSA (Sectional Aerosol module for Large Scale Applications; Kokkola et al., 2008) in the European scale CTM MATCH (Robertson et al., 1999; Andersson et al., 2007). SALSA was chosen since it was developed to describe the PNC well; it includes several nucleation mechanisms and the sectional approach used in SALSA, to describe the aerosol size distribution, is an advantage for simulating new particle formation (e.g. Korhola et al. 2014). The coupling of SALSA to MATCH introduces a description of particle microphysics and aging in the model. New features include particle nucleation, condensation, coagulation and activation; leading to a description of the temporal evolution of the particle number size distribution in a number of bins, through the sectional approach. The model also describes the mixing state of the particles. The physical treatment of aerosol microphysics and the particle size distribution is described in Section 2.2; further details about the specific set-up used in this study are given in Section 3. We discuss the performance of MATCH-SALSA in relation to other models in Section 4.

This paper presents the resulting new aerosol dynamics version of the MATCH model; the new model is called MATCH-SALSA. The model was detailed in a report from SMHI (Andersson et al., 2013), which is included as Supplement to this paper (Supplement A). In this paper, we highlight the main new features and present the results from evaluation tests. In a second paper (Andersson et al., 2014) results from various sensitivity tests will be presented. The aim of MATCH-SALSA is to describe particle mass and number concentrations, and particle size distribution on the European scale. The new model features – inclusion of sectional descriptions of aerosol microphysics and particle number size distributions – are developed with the aim to couple the MATCH-SALSA model to climate models and radiative

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Flyttad (infogning) [2]

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Borttaget: SALSA to MATCH introduces a description

Borttaget: aging in the model and makes it possible

Borttaget: describe PNC and the mixing state of transfer calculations; the new model can also be utilized for the estimation of human exposure to particles of different sizes.

2 Description of MATCH-SALSA

The layout of MATCH-SALSA is illustrated in Fig. 1. After initializations are completed the model integrates over time. The integrations are based on the meteorological time step (dtmet), starting with reading or interpolation of weather data, reading emissions, and setting lateral and top boundary concentrations of the chemical species. After this, the emissions are injected and model transport fluxes are calculated with the internal sub-stepping time steps. Subsequently, the model gas- and wet-phase chemistry, aerosol microphysics and cloud droplet number concentrations are calculated. Meteorological data are read at regular intervals, typically every three or six hours. Boundary conditions may be updated at compound dependent time intervals.

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Borttaget: iterations

Borttaget:) where

Borttaget: are read or interpolated, new

Borttaget: are emitted

Borttaget: are set

Natural and anthropogenic emissions are included in the model. Sea salt and isoprene emissions are calculated online, whereas anthropogenic and other emissions (volcanic sulfur, marine DMS and biogenic monoterpenes) are given as input data to the model in the set-up used in the present study. All primary particle components are emitted both as mass and number. Sea salt emissions are modeled as described by Foltescu et al. (2005) but modified to allow arbitrary size bins. For the smallest bins (diameter ≤ 1 µm) the description by Mårtensson et al. (2003) was used; for larger sizes the sea salt generation function was taken from Monahan et al. (1986). Biogenic emissions of isoprene are calculated using the E-94 isoprene emission methodology proposed by Simpson et al. (1995). Emissions from wildfires and agricultural burning are not included in the present version of the model.

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The transport model includes advective and turbulent transport. Particle number and mass are transported independently in MATCH-SALSA. The transport scheme is described in detail in Robertson et al. (1999).

2.1 Chemistry

The original MATCH photochemistry scheme (Langner et al., 1998) was, to a large extent, based on the EMEP MSC-W (European Monitoring and Evaluation Programme

Meteorological Synthesizing Centre - West) scheme (Simpson, 1992; Simpson et al., 1993), but with an alternative treatment of isoprene chemistry, using an adapted version of the Carter 1-product mechanism (Carter, 1996; Langner et al., 1998). A simplified mixture of a dozen representative compounds (“lumped molecules”) is used to model all organic molecules emitted to the atmosphere (e.g., o-xylene represents all emitted aromatic species).

The gas-phase chemistry scheme in MATCH has remained mostly the same since 1998, but a number of reaction rates have been updated, taking into account new recommendations from IUPAC (Atkinson et al., 2006) and the Master Chemical Mechanism, MCM v3 (Jenkin et al., 1997; Saunders et al., 2003, via website: http://mcm.leeds.ac.uk/MCM); a few new gas phase components have also been added to the scheme. The revision of the MATCH chemistry scheme was based closely on the updates done in the EMEP MSC-W model, during 2008-2009, as documented by Simpson et al. (2012); the updated gas-phase reaction scheme in MATCH is mostly identical to the EMEP MSC-W EmChem09 scheme of Simpson et al. (2012), but, for isoprene the scheme from Langner et al. (1998) is retained (with some reaction rates updated to new recommended values from IUPAC (Atkinson et al., 2006), see Supplement B).

In addition to gas-phase chemistry, aqueous-phase oxidation of SO_2 in cloud water (based on Berge, 1992) and a few heterogeneous reactions for nitrogen compounds are included in the model. For MATCH-SALSA some further modifications related to particle formation have been made and the scheme used in the present work consists of ca 140 thermal, wet and photolysis reactions, including ca. 60 different chemical species.

The chemistry code includes a simple scheme for secondary organic aerosol (SOA) formation from biogenic monoterpene emissions; α-pinene is used as a surrogate for all monoterpenes. In the present study, we assume rapid formation of condensable SOA after gas-phase oxidation of α-pinene (by O_3, OH or NO_3; oxidation rates are based on MCM v3.2, http://mcm.leeds.ac.uk/MCM); we assumed that all oxidation paths for α-pinene produce low-volatility SOA-forming compounds, with 10% (mass-based) yield. These compounds are included in the condensation scheme for organic compounds in SALSA. The SOA-yield used here for α-pinene is relatively high compared to some reported SOA-yields for this monoterpene in smog-chamber experiments (e.g., Mentel et al., 2009, find about 5% yield). However, recent findings by Ehn et al. (2014), regarding formation of extremely low-volatility organic compounds from ozonolysis of α-pinene, indicate that SOA-yields from this

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Borttaget: unrealistic for real α-pinene emissions; typical

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process may be higher than 10% above forest canopies. We also note that there are recent studies that indicate that SOA-yields based on smog-chamber studies may be underestimated by up to a factor of four, due to wall losses of gas-phase semi-volatile organic (Kokkola et al., 2014; Zhang et al., 2014). Note that the simplified BSOA "scheme" used in the present study is included to test the organic-aerosol parts of MATCH-SALSA, with minimal changes to the standard photochemistry scheme; it is not expected to model BSOA formation in a very realistic way compared to real-world conditions but, given the high uncertainties in monoterpene emissions and the neglect of other BSOA-forming emissions, it was considered a reasonable approach for the development phase of MATCH-SALSA.

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The chemical equations are solved prior to SALSA. There is no internal sub-stepping between the chemistry and SALSA (cf. Figure 1). For a detailed description of the MATCH chemistry scheme, including a full list of the reactions and reaction rates, see Supplement B.

2.2 Aerosol microphysics

The SALSA model was designed to obtain a balance between computational efficiency and numerical accuracy. This was reached by keeping the number of tracer variables low, by using a relatively coarse particle size resolution, and including only the relevant chemical compounds in different particle size ranges (see Kokkola et al., 2008). The size resolution is varying across the size spectrum, with higher resolution for particles that are crucial in cloud activation and for aerosol radiative properties.

Aerosol number and mass concentrations are described by three size ranges, divided into size bins with equidistant distribution of the bins on the log-normal scale. The number of bins in each subrange and the size limits of the subranges are flexible. The level of mixing differs between the subranges:

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Borttaget: size ranges

i. In the smallest subrange, all particles are internally mixed.

ii. In the second subrange, there are two parallel externally mixed size bins for each size. In this subrange, we assume that soluble compounds (sulfate, sea salt, and soluble organics) are emitted to so called soluble bins whereas insoluble compounds (black carbon, mineral dust, and insoluble organics) are emitted to the insoluble bins.

iii. In the largest subrange, there are three externally mixed size bins: 1) soluble, into which the above-mentioned soluble compounds are emitted, 2) cloud active insoluble particles, which are mainly composed of insoluble compounds, but which have enough soluble material to activate as cloud droplets, and 3) freshly emitted insoluble range, into which insoluble compounds are emitted.

In addition, the chemical compounds that are treated in each size range are chosen depending on the compounds that are relevant to that size of particles in the atmosphere (for details, see Kokkola et al., 2008):

i. The first size range (nucleation and Aitken modes) includes sulfate (SO_4^{2-}) and OC.

ii. The second (accumulation mode) size range includes SO_4^{2-}, EC, OC, sea salt (NaCl) and mineral dust in two externally-mixed parallel size bins for each size section.

iii. The third (coarse mode) size range also includes SO_4^{2-}, EC, OC, sea salt (NaCl) and mineral dust in three externally-mixed particle types: sea salt, “insoluble dust” and “soluble dust”; all water soluble compounds, including SO_4^{2-} and OC, are combined in the “soluble dust” type.

Note that EC is not included in the Aitken mode, which is a shortcoming of MATCH-SALSA. The reason for this choice in SALSA was to reduce the CPU burden.

The hygroscopicity of the aerosol is calculated using the Zdanowskii-Stokes-Robinson method (Jacobson, 2002). At the end of each microphysical time step the size distribution is updated to take into account growth of particles due to dynamic and chemical transformation processes.

Nitrate in coarse mode particles is treated separately as a simple tracer compound. Other particulate nitrogen species are described by a simplified chemistry scheme (see Supplement B), currently handled outside SALSA, i.e. ammonium salts (e.g. ammonium nitrate) are not taken into account in the modeling of the aerosol microphysical processes. After the aerosol microphysical processes have taken place, ammonium bound to sulfate is distributed according to the size-distribution of particulate sulfate and ammonium nitrate is distributed according to the available aerosol surface area. However, this condensation of ammonium and nitrate do not affect the particle radius in the model, thus they do not influence shape of the size distribution. A possible consequence of the simplified treatment can be underestimation of condensational growth, which may cause overestimation of nucleation, due to a too small

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Borttaget: on particle sizes. Ammonium

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Borttaget: distribution and coarse nitrate was treated separately.

condensational sink for the nucleation mode particles. The lack of ammonium nitrate condensation in the aerosol microphysics could cause underestimation of cloud droplet number concentration (CDNC).

In this study nucleation is simulated through an activation type nucleation formulation (Kulmala et al., 2006; Riipinen et al., 2007) and the formation rate of 3 nm particles (J3) is calculated according to Lehtinen et al. (2007). Nucleation is solved concurrently with condensation, using the methodology of Jacobson (2002); this takes into account the competition of nucleation and condensation in the mass transfer of volatile species between gas and particle phase. The MATCH-SALSA model also includes other nucleation schemes, for example binary nucleation (Vehkamaki et al., 2002), ternary nucleation (Napari et al., 2002a, 2002b) and activation of both H_2SO_4 and organic vapors (Paasonen et al., 2010; Supplement C). Tests of these alternative nucleation schemes will be presented in the companion paper (Andersson et al., 2014).

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Borttaget: scheme options

The scheme used for gas-to-particle transformation is the Analytical Predictor of Condensation scheme, with saturation vapor pressure set to zero (Jacobson 1997). The scheme solves condensation and evaporation of semi-volatile compounds over a discrete time step. It is very well suited for large scale atmospheric models, such as MATCH, since it requires no iteration, it is mass conserving, and it has been shown to be accurate over time step length of 7200s when condensation is the only active process (Jacobson, 2005).

Borttaget: The method solved non-equilibrium transfer

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Borttaget:) it is very well suited for large scale atmospheric models such as MATCH.

Coagulation is described using a semi-implicit scheme (Jacobson 1994). Similarly to the condensation scheme, a semi-implicit coagulation scheme does not require iteration and it is mass conserving. Since coagulation is the (computationally) most time consuming microphysical process, it is neglected between aerosol pairs for which the coagulation efficiency is low. The detailed list of selected collision pairs accounted for in the coagulation routine is given in Kokkola et al. (2008).

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Further details of the SALSA model is given by Kokkola et al. (2008) and Bergman et al. (2012).

2.3 Deposition

Dry deposition of trace gases is calculated with a simple resistance approach (Chamberlain and Chadwick, 1965), which depends on land use and season. Wet scavenging of most gaseous species is proportional to the precipitation intensity. For ozone, hydrogen peroxide and sulfur dioxide, in-cloud scavenging is calculated assuming Henry's law equilibrium; sub-cloud scavenging is neglected for these species. For ozone sub-cloud scavenging is likely to be negligible; O_3 has a very low solubility in water and wet deposition is not an important sink process for this specie. For SO_2 the omission of sub-cloud scavenging is likely leading to a slight underestimation of the wet-deposition losses; but SO_2 also has a relatively low solubility and a modeling study of wet scavenging of sulfur (Berge, 1993) found that sub-cloud scavenging by precipitation was small (only about 1% of the total S-deposition was due to sub-cloud scavenging). The absence of sub-cloud scavenging for H_2O_2 probably leads to a substantial underestimation of wet deposition for this compound. In recent MATCH-model simulations, that included sub-cloud scavenging of H_2O_2, it was found that sub-cloud scavenging contributed about 20-40% to the total wet deposition of H_2O_2. Wet and dry deposition of gases in the MATCH-model is described in detail by Andersson et al. (2007).

Particle dry deposition (including the effects of hygroscopic growth) is calculated using a scheme based on Zhang et al. (2001), adapted to a smaller set of land use classes (Water, Forest, Low vegetation and Vegetation-free land areas). More details regarding the dry deposition of particle species are given in Supplement A.

Particles are wet deposited through incloud and subcloud scavenging. The incloud scavenging depends on the fraction of cloud water (or ice) that is precipitated in each grid box, the fraction of the box that is cloudy, the concentration of particles and the fraction of particles in each particle size bin that are inside the cloud droplets. MATCH-SALSA includes a simplified scheme, based on Seinfeld and Pandis (1997), to estimate the fraction of particles that are activated as cloud droplets (and thus are located inside the droplets) – in-cloud particles larger than 80nm in diameter are considered activated as cloud droplets. This simplified description is used in the present study.

A more advanced (and CPU-time consuming) formulation for cloud activation is also implemented in MATCH-SALSA. The model can be run coupled to an online cloud activation model that computes CDNC based on the prognostic parameterization scheme of Abdul-Razzak and Ghan (2002). The number of activated particles in each size bin is

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In

Borttaget: the fraction of particles that are inside the cloud droplets is assumed to be

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determined by the particle size distribution, their number concentration and chemical composition, as well as the updraft velocity and the maximum supersaturation of the air parcel. Running the model with particle activation is optional. Optionally, the resulting activated particle fraction in each size bin can be used for calculation of incloud scavenging of particles. In this formulation the activated fraction of each particle class is calculated in each time step for each grid point. The online cloud-activation scheme was not used in the present study, but in Supplement A it is compared to the simplified scheme used here.

Borttaget: There is an option to use

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Borttaget: F_s is the activated fraction of each particle class.

The subcloud scavenging in the model is treated in a similar way as by Dana and Hales (1976). In MATCH-SALSA, a simplified approach is used, where a monodisperse washout coefficient is calculated for each particle bin, and a standard rain drop spectrum[b] is assumed for all precipitation. The washout coefficient (i.e., the fraction of a species that is removed by precipitation below clouds) depends on precipitation amount and takes into account particle collection by Brownian diffusion, inertial impaction and interception. The total wet deposition is the sum of the incloud and subcloud scavenging.

Further details on the wet scavenging of particles are given in Supplement A and in the companion paper Andersson et al. (2014).

Borttaget: Alternatively, more parameterized formulations for the particle wet scavenging can also be used.

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3 Model set up

In this section we describe the setup of the simulation used to evaluate MATCH-SALSA in Section 4.

Borttaget: this paper

Meteorological data is input at regular time intervals; here we used three-hourly fields from the HIRLAM (Hi-Resolution Limited-Area Model; Undén et al., 2002) weather forecast model. The meteorological data are interpolated to hourly resolution. The model domain covers Europe with a spatial resolution of ca 44km. The lowest model level is ca. 60m thick, and, in total, 22 vertical levels are used; the top level is at about 5km height. The vertical structure of MATCH-SALSA is the same as in the meteorological model; in this case hybrid (η) coordinates, with shallow terrain following layers close to the ground and thicker pressure levels higher up.

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[b] A representative frontal rain spectrum is used, R_g=0.02 cm, Σ_g=1.86 (Dana and Hales, 1976).

For the aerosol size distribution, the following settings were used (see Fig. 2): The first subrange covered the diameter interval 3-50nm, with three log-normally distributed size bins; the second subrange covered the diameter interval 50-700nm, with four bins each for soluble and insoluble particle types; the third subrange covered the diameter size range 700nm-10µm, with three size bins for each of the following three particle types: seasalt, soluble particles and insoluble particles.

The top and lateral boundary concentrations of gaseous and particle species, including seasonal variation for some species, were set as described in Andersson et al. (2007). However, boundary concentrations of particulate organic matter (OM) on the southern, western and northern boundary were set based on marine OM measurements (O'Dowd et al. 2004).

Borttaget: OM

In the present study, biogenic emissions of monoterpenes (MT) were based on monthly emissions of MT taken from the EMEP MSC-W model (Bergström et al., 2012; Simpson et al., 2012). The BVOC-emissions are highly uncertain. With four different chemical transport models Langner et al. (2012) predicted European isoprene emissions within about a factor of five; we do not expect the uncertainty in the monoterpene emissions to be lower than for isoprene. Considering the large uncertainties, emissions tests with varying terpene emissions were performed; decreased underestimation in March and July 2007 for PNC and accumulation mode PNC, and improved temporal variation in March 2007 was found at the four measurement sites (see Supplement A) when using three times larger emissions than those taken from the EMEP MSC-W model. For this reason, the MT emissions in the base-case simulations in the present study were chosen to be three times higher than the corresponding emissions in the EMEP MSC-W model. We stress once more that the biogenic SOA description in the present MATCH-SALSA model set-up is incomplete and simplified – the aim is to test the first versions of MATCH-SALSA without introducing a complex and uncertain SOA scheme at the same time as introducing the aerosol dynamics module. The fact that model performance improved when the MT-emissions were tripled should not be interpreted as an indication that the MT-emissions are underestimated in the EMEP MSC-W model. A number of BVOC-emissions are missing in the MATCH-SALSA model (e.g., sesquiterpenes and other VOCs emitted by plants subject to stress; e.g. Bergström et al., 2014). We also miss some other potentially important OA sources, such as wild fires (and other open burning), anthropogenic secondary OA and multigenerational aging of organic

Borttaget: Monthly biogenic emissions of monoterpenes were taken from the EMEP MSC-W model (Bergström et al., 2012; Simpson et al., 2012). α-pinene is used here as a surrogate species for all biogenic monoterpenes. The anthropogenic emissions of gass compounds in the atmosphere. The increased BVOC-emissions in the model may lead to improved model results by compensating for other missing sources of OA or for too low SOA yields from BVOC-oxidation.

The anthropogenic emissions of gases and primary aerosols are taken from the TNO-MACC emission inventory (Kuenen et al., 2011; Pouliot et al., 2012; see also the MACC - Monitoring the Atmospheric Composition and Climate - project web page http://www.gmes-atmosphere.eu/). The TNO-MACC emissions are given as annual totals. Seasonal, weekday and diurnal variations of the emissions are based on results from the GENEMIS project (http://genemis.ier.uni-stuttgart.de/; Friedrich and Reis, 2004).

The particle emissions of EC and OM[c] are distributed over different particle sizes according to sector resolved mass size distributions described by Visschedijk et al. (2009). Details about the size distributions are given in Supplement A (Table 4, page 16). Emissions from most SNAP sectors are described by uni-modal distributions, while emission from two sectors (international shipping and SNAP sector 4: production processes) are described by bimodal distributions.

The emissions of oxidized sulfur (SO_x) were split into 99% SO_2 and 1% H_2SO_4. The split is intended to account for subgrid scale processes of gas phase transformation and gas-to-particle partitioning. The distribution of SO_x emissions between SO_2 and more oxidized compounds is discussed in Spracklen et al. (2005b) – the fraction of SO_2 increases with grid resolution and it is typically set to between 95-100% in European scale models. The assumed fractions have large uncertainties and it is not clear from the literature how to better partition SOx emissions between SO_2(g), H_2SO_4(g) and particulate sulfate in modeling studies. The best distribution depends on model resolution (Spracklen et al., 2005b). Lee et al. (2013) have shown that the uncertainties in the sub-grid production of sulfate particles in plumes are more important for CCN uncertainty than the uncertainties in the total anthropogenic SO_2 emissions. Since we expect that the choice of distribution of SOx emissions has a large impact on the model results, we investigate this further in a companion paper (Andersson et al., 2014). The size distribution of the emitted sulfate is the same as for OM. NOx and NMVOC emissions were handled in the same way as in Andersson et al. (2007).

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Flyttad nedåt [3]: The emissions of oxidized sulfur (SO_x) were split into 99% SO_2 and 1% H_2SO_4.

Borttaget: The distribution of SO_x emissions between SO_2 and more oxidized compounds was discussed by Spracklen et al. (2005); the fraction of SO_2 increases with grid resolution and is typically set to between 95-100% in European scale models. The emitted sulfate mass was distributed over particle sizes in the same manner as OM. NOx and NMVOC were emitted as in Andersson et al. (2007).

Flyttad (infogning) [3]

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[c] OM emissions are assumed to be distributed over different particle sizes in the same way as OC.

4 Evaluation of MATCH-SALSA

In this section we compare our model results to observations at a number of measurement sites throughout Europe. The evaluated model results are extracted from the lowest model level. The statistical measures used are defined in Supplement A. We evaluate the PNC, both in terms of total number concentration, accumulation mode number concentration, and temporal and spatial distribution. We also evaluate the particle mass, including speciation of secondary inorganic aerosol, EC and OC.

4.1 Measurement data

Most measurement data were extracted from EBAS (http://ebas.nilu.no). Details of the stations used in the evaluation of particle number size distribution, PM_1, $PM_{2.5}$, EC and OC are given in Supplement A (Table 5). The secondary inorganic aerosol (SIA) components (nitrate, sulfate and ammonium) were evaluated against available measurements in the EMEP network for 2007 (http://www.emep.int).

For evaluation of PNC, four stations from EBAS were chosen to represent different parts of Europe; all classified as rural background sites. Two of the measurement sites: Melpitz (in eastern Germany) and K-Puszta (in central Hungary), are relatively close to regions with large emissions. Hyytiälä (in the inland of southern Finland) and Aspvreten (ca 70 km south west of Stockholm, in south eastern Sweden) were chosen as regional background stations occasionally impacted by aged particles due to transport from large emission sources in Europe.

4.2 Model evaluation of PNC

Fig. 3 shows the modeled annual mean PNC in Europe; both total PNC (Fig. 3a) and the PNC in the different model size bins up to 700nm are shown (Fig 3b-g). Corresponding measured annual mean PNC at the four measurement sites are also displayed in circles, for particle sizes where measurements are available.

The largest modeled total PNC (Fig. 3a) are found in areas with high SOx emissions (e.g., areas around large point sources in Spain, Poland, south-eastern Europe, the Ukraine, Russia

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and the area around Etna; as well as along shipping routes around the Iberian Peninsula and the Gibraltar strait). These results are in line with other model studies (e.g. Yu and Luo, 2009; Spracklen et al., 2010; Ahlm et al., 2013).

Most of the total PNC in the model resides in the Aitken mode bins (particle diameters 7-20nm and 20-50nm; Figs. 3c and 3d). The highest PNCs in the smallest bin (Fig. 3b), indicating recent nucleation, are found in in Russia and Ukraine Increased values in this bin are also seen along the shipping lanes; the modeled high nucleation in marine areas is not in agreement with observations (Heintzenberg et al., 2004). Metzger et al. (2010) found similar nucleation over oceanic regions with large sulfur emissions when traditional activation type nucleation mechanisms were used; their results with a new organic activation mechanism captured the observed lack of nucleation in marine areas, indicating that organic molecules may have a critical role in the nucleation.

The Aitken mode PNC pattern (Figs. 3c and 3d) is similar to the total PNC distribution (Fig. 3a). The highest concentrations are found in areas in Spain, Turkey, Former Yugoslavia, Bulgaria, and north-eastern Russia, and around the volcano Etna. The highest accumulation mode (50-700nm) PNCs (Figs. 3e-h) are found in southern Europe. This is partly due to relatively large emissions of primary fine particles and gaseous SOx, and partly due to less precipitation in southern Europe, compared to the north and west, allowing accumulation mode particles to reside longer in the atmosphere.

We evaluate the model performance (see Figs. 4 - 6) in terms of total and accumulation mode particle number concentration (PNC and PNC$_a$, respectively) against observations at the four European surface sites. Due to seasonal differences in emissions and atmospheric processes, we separate performance during summer half-years (April-September) from winter (October-March). For example, residential biomass burning emissions are much higher during winter than during summer, while biogenic VOC emissions are largest during summer. Both these sources are associated with large uncertainties regarding the emissions and modeling. It should be noted that the size ranges for PNC and PNC$_a$ vary between the stations depending on the measurement interval.

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4.2.1 Spatial distribution

Modeled total PNC shows moderate to poor agreement with the observations (Fig. 4a). At most sites the deviation between observed and modeled mean is large both in summer and winter, and the correlation coefficients for daily mean PNC are low (r range from 0.05 to 0.66).

The model captures the general observed features of lower total and accumulation mode PNC in the northern and north-western parts of Europe (Fig. 3). Aspvreten and Hyytiälä have the lowest modeled and observed PNCs (Fig. 4a). However, looking in more detail at the stations (Fig. 4) there are some discrepancies, Melpitz clearly has the highest *observed total PNC* (during both winter and summer; Fig 4a); the model severely underestimates the PNC at Melpitz and predicts much higher total PNC at K-Puszta than at Melpitz. The highest *observed accumulation mode PNCs* are found at K-Puszta and Melpitz (the PNC are at similar levels for both seasons and both sites; Fig. 4b); just as for total PNC, the model predicts much higher accumulation mode PNC at K-Puszta than at Melpitz.

Thus, the spatial distribution of PNC in the model is not in agreement with the observations. There may be many reasons for this. One important reason for the high modeled total PNC at K-Puszta is a high rate of nucleation (Fig. 5c), which is caused by the large emissions of SOx in the area. For the other three northern and central European sites, there is an underestimation in all size ranges. This may be due to too weak nucleation rate, too efficient wet scavenging or a combination of various problems. For the Aitken and accumulation modes, the problem can also be due to underestimated primary emissions. The underestimation in the nucleation mode implies either a low-biased nucleation mechanism, a too efficient removal (deposition) or underestimated precursor emissions. Further, EC is not included in the Aitken mode in the model. This leads to underestimated total particle number concentration (in the Aitken mode and subsequently in larger sizes as well).

Spracklen et al. (2010) investigated the impact of different nucleation mechanisms, including the impact of using different nucleation rate coefficients in the activation mechanism. They chose to investigate three rate coefficients, A=2×10^{-7} s^{-1}, 2×10^{-6} s^{-1} and 2×10^{-5} s^{-1} for which they evaluated the bias to global observations in the free troposphere, and marine and continental boundary layers. In the continental boundary layer the two lowest nucleation rate coefficients resulted in mean underestimations by -48% and -29% respectively, whereas the highest rate resulted in a slight overestimation on the average (12%). The nucleation rate

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coefficient used in MATCH-SALSA in the present study is near the lower end of the interval (A=7.3*10^{-7} s^{-1}), which may explain our underestimation of nucleation at the central and northern sites. In fact, the nucleation rate coefficient in the activation scheme should be site and time dependent in the European boundary layer (e.g. Sihto et al., 2006; Riipinen et al., 2007): observations of this coefficient vary by ~4-5 orders of magnitude for different European measurement sites, ranging from 3.3*10^{-8} to 3.5*10^{-4} s^{-1} (Riipinen et al., 2007). Thus, a more advanced description of the nucleation, e.g. time and space-varying rate coefficients, should be included in MATCH-SALSA.

Organic nucleation is not included as a nucleation process in the evaluated base case simulation, resulting in possible underestimation of nucleation in areas with high BVOC-concentrations and possibly overestimated nucleation in regions with low concentrations of organic aerosol precursors (similar to the overestimated nucleation in the model in oceanic high-SOx regions, discussed above). This may also be an explanation for the overestimated nucleation at K-Puszta. Sensitivity tests including organic nucleation will be discussed in the companion paper (Andersson et al., 2014); a lot of the material is also available in Supplement A.

4.2.2 Size distribution

The modeled and observed size distributions at all four stations are shown in Fig. 5. A common feature for the PNC size distribution is that PNC are underestimated, or on the same level as the measurements, except at K-Puszta, where the PNC of the smallest particles is overestimated both during winter and summer (Fig. 5c). At K-Puszta the mean total PNC is overestimated but the PNC in the accumulation mode is underestimated (Fig. 4). At all stations, the shape of the size distribution is captured relatively well, but during winter at K-Puszta (Fig. 5c) and during summer at Aspvreten (Fig. 5a) and Hyytiälä (Fig. 5b) the modeled size distribution peaks at smaller sizes than in the observations. The reason for the maximum occurring at too small sizes, in combination with underestimated accumulation mode PNC, may be too weak condensation onto nucleating particles in the model. Bergman et al. (2012) also evaluated the modeled particle number size distribution at measurement sites, including Aspvreten, Melpitz and Hyytiälä, and found that the model ECHAM5-HAM underestimated the number concentrations at all three measurement sites for sizes larger than about 20nm,

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both when using the aerosol dynamics modules of M7 and SALSA. SALSA performed better than M7 for PNC above 100nm at the dirtier measurement sites (e.g. Aspvreten and Melpitz) while M7 performed better at cleaner sites (e.g. Hyytiälä), but the differences between the two models were not large. Bergman et al. (2012) concluded that the growth in SALSA probably was too slow.

4.2.3 Temporal evolution

Fig. 6 shows the modeled and observed temporal variation of the daily mean PNC at the four sites. New particle formation in the model is seen in the form of peak concentrations of the smallest particles sizes. These peaks coincide with the observed maximum total PNC on some occasions; sometimes there is a time shift of a few days between the modeled and observed peaks. Many of the observed nucleation peaks at Hyytiälä (Fig. 6a), Aspvreten (Fig. 6b) and Melpitz (Fig. 6d) are not seen in the model results. Reddington et al (2011) simulated hourly PNC with diameters larger than 15nm using the GLOMAP model and evaluated these against measurements from one month (May 2008). Depending on the nucleation parameterization, the correlations (R^2) between model and measured PNC were less than 0.03 at Aspvreten, Hyytiälä and Melpitz, and less than 0.10 at K-Puszta. For PNC with larger sizes (>100nm), the correlations were less than 0.01 at K-Puszta and higher at the other sites (<0.13 at Aspvreten, <0.20 at Melpitz and <0.45 at Hyytiälä). Spracklen et al. (2006) on the other hand captured the nucleation at Hyytiälä very well with GLOMAP, however, they only studied a short period (22 days) in May with clear sky conditions. With MATCH-SALSA the hourly correlations (R^2), for single months of 2007, for PNC with a diameter larger than 50nm range from 0 to 0.17 for Hyytiälä (for May: 0), <0 - 0.20 for Aspvreten (May: <0), <0 - 0.20 for K-Puszta (May: 0.01) and <0 - 0.41 for Melpitz (May: 0.41). These low correlations illustrate that nucleation events are difficult to capture by models when running over long time periods for a large region. One reason for this is the coarse scale of the model – each grid cell is representative of a large area (for MATCH-SALSA, ca 44 × 44 km^2, and for GLOMAP 2.8° x 2.8°). Another reason is that the simple activation type nucleation scheme needs a site and time varying nucleation parameter to work well (Riipinen et al., 2007). Furthermore, the wintertime nucleation peaks in the observations that are absent in the model may also be explained by a temperature dependence in the nucleation, that is not accounted for in the

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model (Dal Maso et al., 2005), or the observed peaks could be of local origin that can not be captured by a regional scale CTM.

The best correlation between modeled and observed daily mean PNC is found at Melpitz (r=0.70; Fig. 6d) but the model underestimates PNC most of the time; the observed PNC is almost always high at this site. The model grossly overestimates the total PNC at K-Puszta (Fig. 6c) during summer, but the temporal variation for particles sizes >20nm follows the measurements fairly well (r=0.32); during winter the model PNC is in better agreement with the observations. At Hyytiälä (Fig. 6a) a lot of nucleation is observed; this is not captured by the model, possibly due to the lack of organic nucleation in this simulation; this will be discussed in detail in the companion paper (Andersson et al., 2014).

Borttaget: At Mace Head some of the observed peaks are fairly well modeled but the overall correlation coefficient is modest (r=0.46); the timing of some peaks is shifted in the model compared to the observations and some model peaks are not seen in the observations and vice versa.

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Spracklen et al. (2010) calculated the correlations (R^2) between monthly mean modeled and observed PNC for sites where the monthly means varied by more than a factor of two during the year 2000 (Aspvreten was excluded due to too small variation). K-Puszta was not included in the assessment. Their results were R^2=0.39 and 0.28 for the sites Hyytiälä and Melpitz, respectively. With MATCH-SALSA we get R^2=0.67 and 0.08, respectively, for the same sites (for PNC with diameter >50nm). Using kinetic nucleation description Spracklen et al. (2010) achieved higher monthly correlations than with activation type nucleation at most evaluated sites, including Hyytiälä and Melpitz.

4.3 Model evaluation of particle mass and composition

Simulated annual average total PM_{10}, and the chemical components that constitute PM_{10}, are displayed in Fig. 7. The largest concentrations of total PM_{10} (Fig. 7a) are found at anthropogenic emission hotspots (e.g., northern Italy, Moscow and the eastern Ukraine) and over the Atlantic Ocean and parts of the Mediterranean Sea. The highest modeled concentrations over land are due to large anthropogenic emissions of primary anthropogenic inorganic aerosol (Fig. 7d), except in northern Italy, where there is a large contribution from ammonium nitrate (Figs. 7f-g), and in southeastern Europe, and some sulfur emission hotspots, where sulfate (Fig. 7e) dominates PM_{10}. Over the oceans (and in large parts of western and northern Europe), the largest contribution to PM_{10} is from sea salt particles (Fig. 7h); important sulfate contributions are also seen, especially around Etna and the eastern Mediterranean Sea. OM (Fig. 7c) gives the largest modeled non-sea salt contributions to PM_{10}

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in northern Europe and also in some parts of southern/western Europe. In the following subsections we present evaluation statistics for the different particle components.

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4.3.1 Secondary inorganic aerosol (SIA)

Statistics from the evaluation for SIA components (particulate sulfate, SO_4^{2-}; nitrate, NO_3^-; and ammonium, NH_4^+) are shown in Table 1 and in Supplement A (Tables A15-A19 and Figs. A32-A36). In order to avoid biases due to possible incorrect separation of gas and particle phase nitrogen in the measurements, we also include evaluation results for total nitrate (TNO3: HNO_3(g) + NO_3^-(p)) and total reduced nitrogen (TNHx: NH_3(g) + NH_4^+(p)).

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Sulfate has a low mean bias (4%) whereas the root mean square error normalized to the observed mean (CV(RMSE)) is around 50%. The average (Pearson) correlation coefficient (average r at the different sites, based on daily means) is 0.52 and the spatial correlation coefficient ("spatial" r for the annual mean concentration at all the stations) is 0.57. The model performance for the nitrogen compounds (NO_3^-, HNO_3+NO_3^-, NH_4^+ and NHx) at individual stations is of similar quality as that of sulfate. The model underestimates the concentration of the nitrogen components by about 10-20%, while the CV(RMSE)s are a bit lower than for sulfate (range from 36 to 49% for the four N-components). The average r at the measurement sites vary between 0.44 and 0.59 for the N-components, whereas the spatial correlation coefficients are higher (between 0.79 and 0.87).

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4.3.2 Elemental and organic carbon

The organic aerosol measurements used for model evaluation in this study are organic carbon (OC) measurements. The model describes organic matter (OM). In the evaluation we assume an OM:OC ratio of 1.4. The actual ratio varies with location and season (e.g., Simon et al., 2011) and is usually between 1.25 and 2.5, with a greater ratio for more aged OM (Turpin et al., 2000; Kupiainen and Klimont, 2007; Aiken et al., 2008). The choice of a fixed OM:OC ratio for the evaluation will lead to model under- or overestimation, depending on the measurement site and time of year. Fig. 8 and Fig. 9 show the annual observed and modeled mean concentrations of EC (Figs. 8a-b) and OC (Figs. 9a-b) at individual measurement sites,

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as well as the associated correlation coefficients, based on daily data; detailed results are given in Table 2.

Both EC and OC are underestimated at many of the sites. The underestimation is especially large at the Italian sites and Payerne (Switzerland) during winter, for both EC (Fig. 8b) and OC (Fig. 9b), and for EC at Melpitz (Figs. 8a-b). Correlation coefficients are higher for EC than OC; OC is more complicated to model than EC, since it is a combination of primary and secondary components, many of them semi-volatile. The reasons for the model – measurement differences are likely to vary between seasons and locations; e.g., wintertime emissions from residential combustion are often underestimated (e.g. Simpson et al., 2007; Gilardoni et al., 2011; Bergström et al., 2012), during the summer half-year biogenic VOC emissions and wildfires may be more important sources of carbonaceous particles.

At Ispra (IT04) in northern Italy, the model performs fairly well for carbonaceous aerosol during summer but greatly underestimates both EC and OC during wintertime (Fig. 8, Fig. 9 and Fig. A15 in Supplement A). One reason may be the underestimation of residential wood combustion emissions (e.g. Bergström et al., 2012). The model also underestimates NO_2 (by 43% in summer and 51% in winter). Both the observations and the model results show a clear seasonal cycle with higher concentrations during winter for NO_2 as well as for EC and OC. However, for EC and OC the model underestimation during winter is much larger (-74 and -87%, respectively) than during summer (-20 and -37%, respectively) (Supplement A, Fig. A15). The poor model performance for EC and OC during winter is likely due to lacking emissions from one or more emission sectors, with greater emissions of EC and OC during winter, but relatively small contribution to NO_2. This work therefore supports the results of previous studies (e.g. Gilardoni et al., 2011) that have concluded that residential wood combustion emissions are likely underestimated in current emission inventories, at least in the area around Ispra.

For the German site Melpitz, the model grossly underestimates EC throughout the year (Supplement A, Fig. A37). OC is generally captured fairly well at the station, with underestimation of OC in $PM_{2.5}$ and PM_{10} (but not PM_1) during winter and overestimation for OC in $PM_{2.5}$ and underestimation (-25%) in PM_{10} during summer (Supplement A, Fig. A38). Part of the reason for the relatively high EC measurements at Melpitz is that the measurement technique used at this site, to separate OC from EC, has no charring correction and is expected to lead to too high EC values and to underestimate OC (see Genberg et al., 2013,

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Evaluation of PM_1 and $PM_{2.5}$ at 28 measurement sites is presented in Fig 9. and in the Supplement Table A21 and Fig. A39; detailed time series plots are given in the Supplement Figs. 17, A40-A41. For PM_1 the annual means at the sites with the lowest concentration (Scandinavian sites NO01, FI17, DK41) are overestimated by the model. On the other hand, at the central European sites the PM_1 concentrations are much better captured. The model underestimates $PM_{2.5}$ by 14% (spatial average) and the spatial correlation coefficient is 0.64. Out of the 35 evaluated annual means (PM_1 and $PM_{2.5}$) at the 28 stations, six means (at five stations) deviate by more than 50%. The largest underestimations of $PM_{2.5}$ are seen at the measurement sites with the highest observed annual mean. The underestimation of $PM_{2.5}$ can be due to a number of reasons including underestimated emissions, too short aerosol lifetime or too little secondary aerosol production. There is probably too little EC and OC in the model, at least at some of the sites, which can be explained by underestimated emissions. ¶
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During this work we found that further improvement is needed for a better representation of PNC. Here, in this section we would try to address some of the issues related to model development and ... [1]

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and references therein). There are large peaks during spring and late autumn of OC (and EC) in $PM_{2.5}$ and PM_{10}, which are clearly underpredicted. The peak in the beginning of April coincides with a vegetation fire episode (Genberg et al., 2013); the earlier peaks and the late autumn peaks are perhaps more likely due to residential combustion or other missing/underestimated sources, possibly, also due to fires in eastern Europe (Jönsson et al., 2013). Stern et al. (2008) compared five different chemical transport models to observations from northern and eastern Germany during highly polluted conditions. None of the models could reproduce the very high EC concentrations observed at Melpitz. Stern et al. (2008) suggested that the large underestimations of EC may be an indication that emissions in the central European region were underestimated during these episodes.

4.3.3 Total particulate matter (PM_1 and $PM_{2.5}$)

Evaluation of PM_1 and $PM_{2.5}$ at 28 measurement sites is presented in Fig. 10 and in Supplement A (Table A21 and Fig. A39); detailed time series plots are given in Supplement A Figs. A17, A40-A41. For PM_1 the annual means at the sites with the lowest observed concentration (three Nordic sites: NO01, FI17, DK41) are overestimated by the model. On the other hand, at the central European sites the PM_1 concentrations are much better captured. The model underestimates $PM_{2.5}$ by 14% (spatial average) and the spatial correlation coefficient is 0.64. Six of the 35 evaluated annual means (PM_1 and $PM_{2.5}$) deviate by more than 50% from the measured concentrations. The largest underestimations of $PM_{2.5}$ are seen at the sites with the highest observed annual mean. The underestimation of $PM_{2.5}$ can be due to a number of reasons, including underestimated emissions, too short aerosol lifetime or too small secondary aerosol production. There is probably too little EC and OC in the model, at least at some of the sites, which can be explained by underestimated emissions.

The treatment of sea spray needs to be further evaluated and the model scheme for sea salt particles may need to be updated. For PM_1 the annual means at the sites with the lowest concentrations are overestimated by the model. This seems to be partly due to overestimation of sea salt. Evaluation scores for modeled PM_1 and $PM_{2.5}$ excluding sea salt aerosol in the total PM mass (see Supplement A: Table A21, Figs. A18 and A39) gives higher correlation coefficients for daily mean $PM_{2.5}$ or PM_1 at 22 of the 28 sites (and lower at only one site) than when sea salt is included. This is an indication of too much sea salt at the wrong time. It may

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be due to too strong sea salt emissions and/or too weak sink processes for the sea salt, since substantial improvements in correlation are seen also at some far inland sites.

5 Conclusions

We have implemented the sectional aerosol dynamics model SALSA (Kokkola et al., 2008) in the European scale CTM MATCH (Multi-scale Atmospheric Transport and Chemistry; Robertson et al., 1999). The new model is called MATCH-SALSA. It includes aerosol microphysics with several options for nucleation, wet scavenging and condensation.

In general, the model reproduces observed lower particle number concentration (PNC) in northern and north-western Europe and remote regions than in central Europe. The model peak in the particle number size distribution occurs at the same or smaller particle size as the observed peak. Total PNC is underestimated at northern and central European sites. The low nucleation rate coefficient used in this study is probably one important factor for the underestimation, although other reasons may also contribute, e.g. organic nucleation is not included and EC are not emitted in the Aitken mode. The model performs well for particle mass, including secondary inorganic aerosol components. Particulate elemental and organic carbon concentrations are underestimated at many of the sites.

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Before using the model for simulating total $PM_{2.5}$, the SOA formulation needs further improvements. MATCH-SALSA is computationally heavier than MATCH, which also puts restrictions on when the model can be used.

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The development of the MATCH-SALSA model is continuing and in the near future focus will be on the following areas:

- An updated biogenic emission module is needed for realistic treatment of BSOA formation. Updating the biogenic SOA scheme will likely have a large impact on modeled $PM_{2.5}$ and also affect the model performance for total PNC through impacts on nucleation and condensation.
- Updating the nucleation rate coefficients possibly with time- and space-varying rate coefficients.

- Nitrogen gas-particle partitioning should be coupled to the microphysics. This may increase condensational growth, which is underestimated in the present version of the model.
- Emissions from open fires (wildfires and agricultural burning activities) will be added to the model.
- Dust emissions from road traffic, agricultural activities and non-vegetated soils including desert areas should be included in the model.
- Processes affecting sea salt need further work and evaluation. This study has shown large modeled sea salt peaks that are not seen in the measurements. Both emissions and deposition of sea salt particles should be investigated.
- Emission inventories need to be improved, especially for EC and OC emissions.

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6 Acknowledgements

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Flyttad (infogning) [15]

Borttaget: .,:

Borttaget: .

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Borttaget: .

Flyttad (infogning) [17]

Formaterat: Teckenfärg: Auto, Tyska (Tyskland)

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Flyttad uppåt [16]: G.,

Flyttad uppåt [17]: H.,

Formaterat: Teckenfärg: Auto, Tyska (Tyskland)

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Tables

Table 1. Comparison of modeled secondary inorganic aerosol (SIA) components to daily observed concentrations. Average results covering available measurements for the year 2007 (results for individual stations are given in Tables A15-A19 in Supplement A). In addition to the SIA components also the total nitrate (TNO3=HNO$_3$(g)+NO$_3^-$(p)) and total reduced nitrogen (TNHx=NH$_3$(g)+NH$_4^+$(p)) are evaluated. r: the Pearson correlation coefficient, CV(RMSE): the coefficient of variation of the Root Mean Square Error (RMSE normalized to the observed mean concentrations), #obs: the total number of observations included in the evaluation, #stns: the number of measurement stations included in the evaluation.

	Global/temporal						Spatial			
Measure: Unit:	Mean Obsvd µgS/N m^{-3}	Mean Model µgS/N m^{-3}	Rel. Bias (%)	mean[a] r	mean[a] CV(RMSE) (%)	#obs.	Rel. Bias (%)	r	CV(RMSE) (%)	#stn
SO$_4^{2-}$	0.63	0.65	4	0.52	46	16033	-6	0.57	53	5
NO$_3^-$	0.40	0.32	-21	0.44	49	7249	-22	0.83	48	2
TNO$_3$	0.49	0.40	-19	0.59	36	11039	-21	0.85	41	3
NH$_4^+$	0.72	0.64	-12	0.57	39	9728	-11	0.79	37	3
TNH$_x$	1.27	1.01	-21	0.53	40	10137	-20	0.87	38	3

[a] Weighted average of correlation coefficients and CV(RMSE) at individual stations.

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Table 2. Statistics of the comparison of MATCH-SALSA results to daily observed concentrations of elemental carbon (EC) and organic carbon (OC) in PM_1, $PM_{2.5}$ and PM_{10} for the year 2007. Obs = Measured concentration, Mod = Modeled concentration, MAE = mean absolute error, r = Pearson correlation coefficient (only calculated for sites with more than 10 measurements). Relative bias and MAE are given as percentage of the observed average. For further information about the measurement stations, see Table A5 in Supplement A.

		EC							OC						
	Stations	Obs µg m^{-3}	Mod µg m^{-3}	bias (%)	MAE µg m^{-3}	MAE (%)	r	#meas	Obs µg m^{-3}	Mod µg m^{-3}	bias (%)	MAE µg m^{-3}	MAE (%)	r	#meas
In PM_1 winter	Melpitz	0.54	0.21	-60	0.33	60	0.60	32	0.65	0.76	18	0.23	36	0.83	32
In $PM_{2.5}$ winter	Birkenes	0.12	0.18	47	0.11	87	0.58	73	0.60	0.88	46	0.46	76	0.45	73
	Overtoom	0.75	0.54	-27	0.27	36	0.76	27	2.19	1.15	-48	1.25	57	0.59	28
	Melpitz	1.28	0.29	-77	0.99	77	0.60	182	1.81	1.21	-33	0.95	52	0.59	182
	Payerne	1.45	0.39	-73	1.06	73	0.67	23	5.61	1.33	-76	4.28	76	0.52	23
	Ispra	3.67	0.93	-75	2.76	75	0.28	173	14.1	2.04	-86	12.1	86	0.24	173
	Puy de Dome	0.05	0.36	556	0.31	556	0.43	33	0.99	1.35	36	0.46	46	0.60	21
	Montelibretti	1.10	0.40	-64	0.70	64	0.60	32	17.2	1.22	-93	16.0	93	0.53	32
	Montseny	0.17	0.49	181	0.32	181	0.60	17	1.64	1.74	6	0.48	29	0.68	17
	Campisabalos	0.16	0.27	65	0.10	65	-	9	1.73	1.01	-42	0.72	42	-	9
In PM_{10} winter	Birkenes	0.14	0.19	38	0.10	75	0.62	73	0.76	0.92	22	0.48	63	0.43	73
	Harwell	1.06	0.93	-11	0.68	64	0.50	56	3.23	1.67	-48	1.65	51	0.70	56
	Melpitz	1.65	0.32	-80	1.33	80	0.63	182	2.77	1.40	-49	1.48	53	0.56	182
	Kosetice	0.36	0.25	-30	0.13	37	0.42	30	1.96	0.86	-56	1.13	58	0.62	30
	Montelibretti	1.30	0.44	-66	0.86	66	0.47	31	15.5	1.29	-92	14.2	92	0.65	31
	Montseny	0.21	0.51	143	0.30	143	0.73	17	1.61	2.03	26	0.57	35	0.80	17
	Campisabalos	0.17	0.29	71	0.12	71	-	8	1.92	1.25	-35	0.69	36	-	8
In $PM_{2.5}$ summer	Birkenes	0.09	0.11	27	0.03	40	0.81	51	0.74	0.85	14	0.31	42	0.73	51
	Overtoom	0.57	0.37	-36	0.24	42	0.34	37	1.66	1.17	-29	0.62	38	0.76	37
	Melpitz	0.95	0.17	-82	0.78	82	0.54	183	1.26	1.78	41	0.83	66	0.47	183
	Ispra	0.87	0.68	-21	0.35	40	0.48	165	3.80	2.54	-33	1.91	50	0.34	169
	Puy de Dome	0.09	0.26	171	0.18	192	0.09	33	2.18	2.05	-6	1.57	72	-0.08	11
	Montseny	0.17	0.47	172	0.29	172	0.60	21	1.82	2.72	49	0.91	50	0.60	21
	Campisabalos	0.10	0.14	46	0.05	53	-	5	2.24	1.33	-41	1.28	57	-	5
In PM_{10} summer	Birkenes	0.11	0.12	10	0.04	37	0.76	52	1.04	0.90	-13	0.27	26	0.81	52
	Melpitz	1.60	0.19	-88	1.41	88	0.59	183	2.58	1.93	-25	0.87	34	0.51	183
	Montseny	0.19	0.49	162	0.30	162	0.51	21	1.66	2.89	74	1.23	74	0.62	21
	Campisabalos	0.15	0.14	-9	0.08	52	-	10	2.26	1.48	-35	1.13	50	-	9

Figures

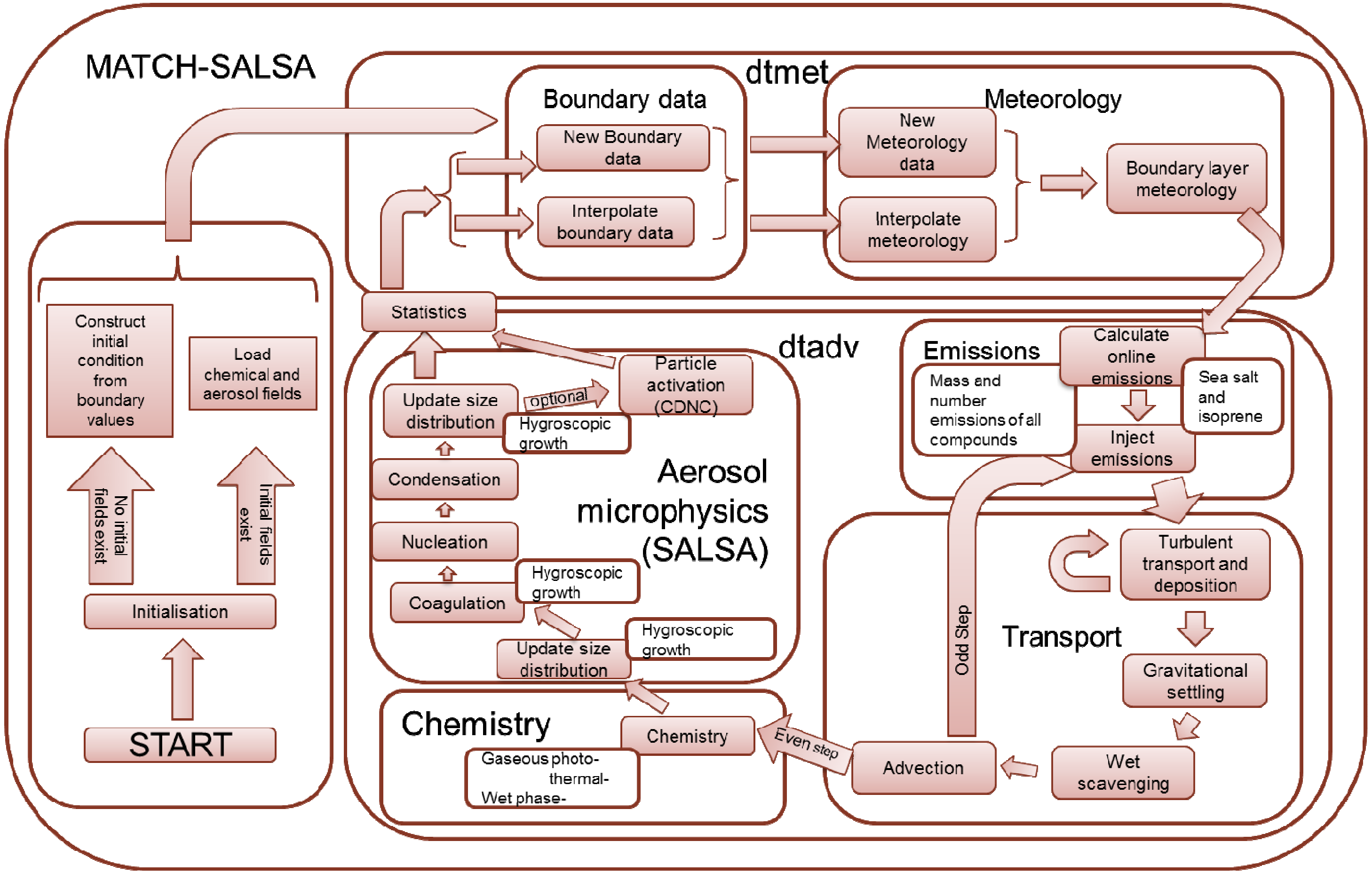

Figure 1. Model integration and time stepping in MATCH-SALSA.

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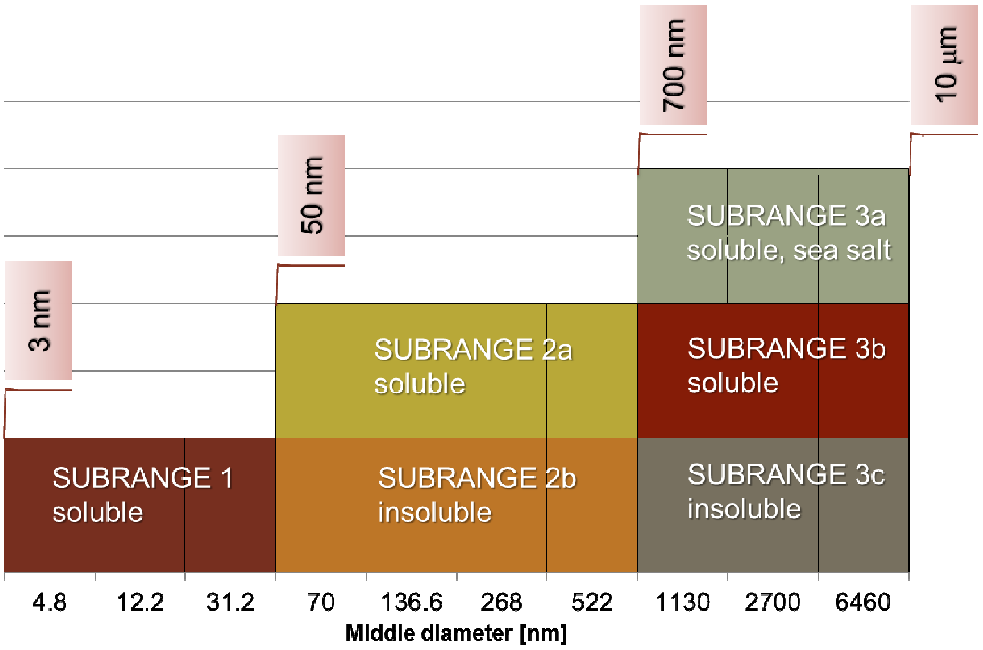

Figure 2. Aerosol division into bins in the three SALSA subranges in the base case set up of MATCH-SALSA.

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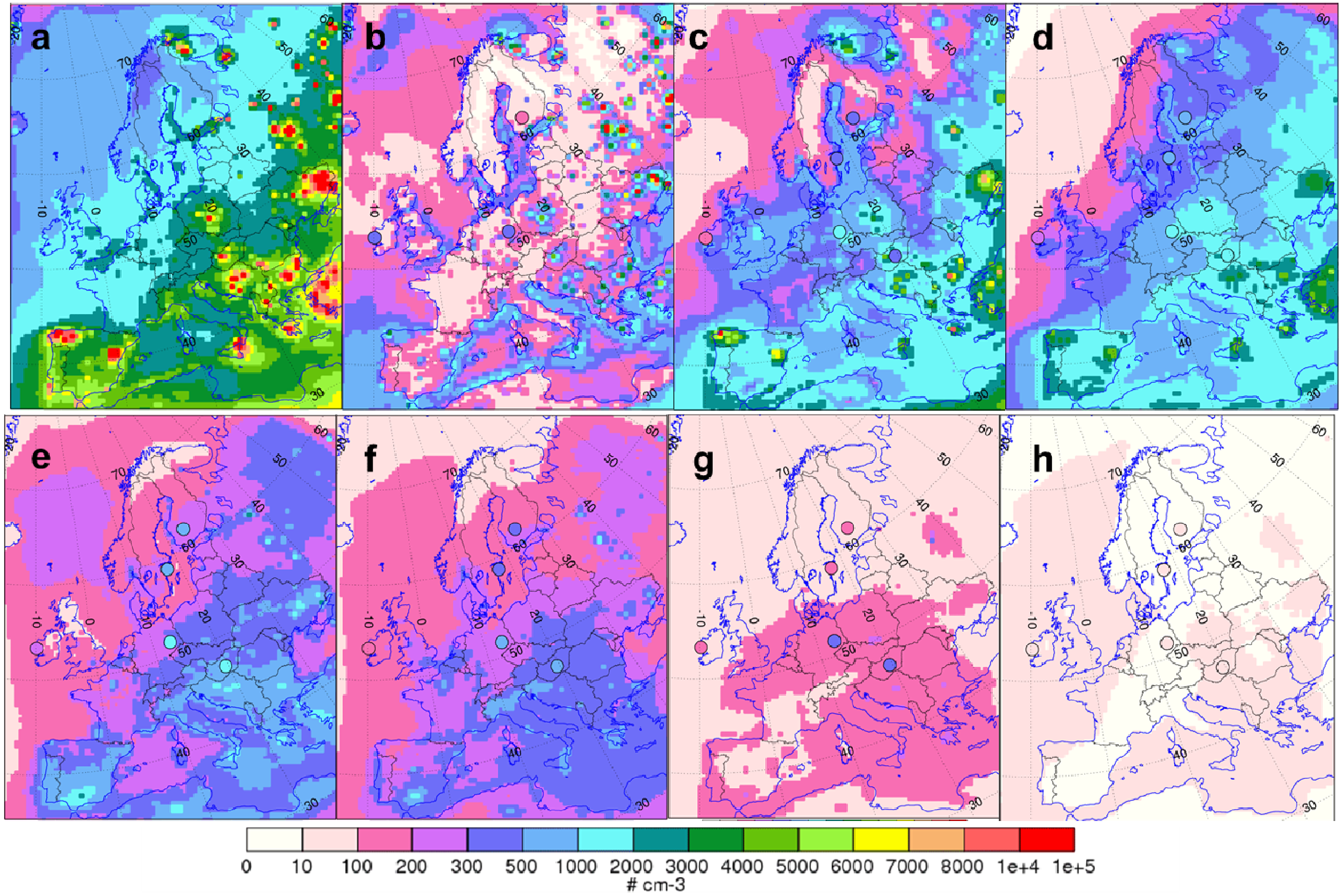
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Figure 3. Calculated annual mean (2007) particle number concentration (PNC) in Europe. Total PNC (sum of all sizes; panel a), and PNC in size bins PNC$_{3<d<7nm}$ (panel b), PNC$_{7<d<20nm}$ (panel c), PNC$_{20<d<50nm}$ (panel d), PNC$_{50<d<98nm}$ (panel e), PNC$_{98<d<192nm}$ (panel f), PNC$_{192<d<360nm}$ (panel g), PNC$_{360<d<700nm}$ (panel h). Observed annual mean PNC (filled circles) at the observation sites: Hyytiälä (Finland), Aspvreten (Sweden), Melpitz (Germany) and K-Puszta (Hungary) when observed numbers exist in the indicated interval. Unit: # cm^{-3}.

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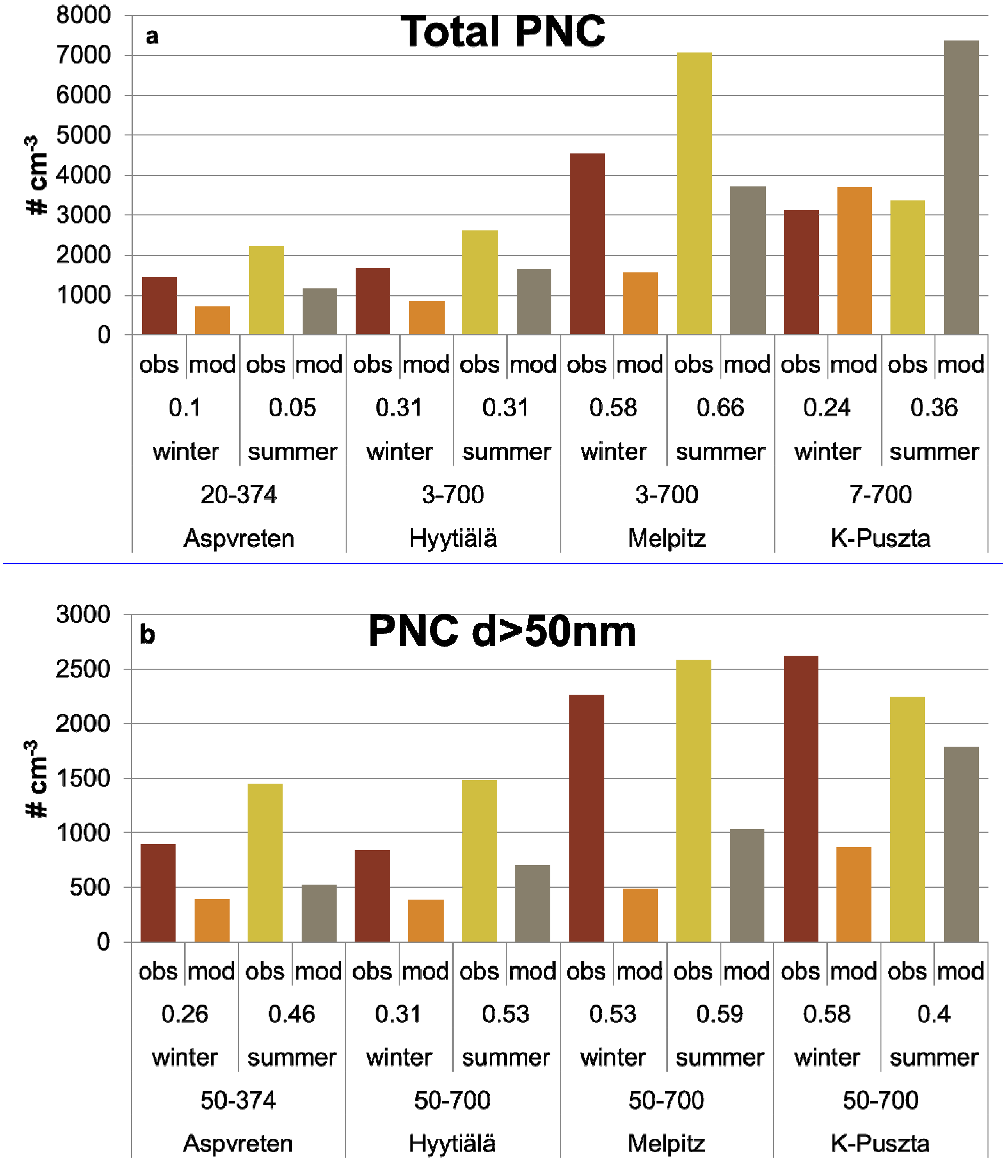

Figure 4. Mean particle number concentration (PNC) during winter (Jan-March; Oct-Dec) and summer (April-September) half years at four sites in Europe. Top panel (a): mean observed and modeled total PNC. Bottom panel (b): mean observed and modeled PNC in the accumulation mode. The interval above the site name indicates the particle size interval (unit: nm). The number above the season shows the (Pearson) correlation coefficient (r) of daily

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mean PNC. Note that the size intervals differ between the stations: the same size interval is used for both modeled and observed values at each site. Unit: # cm^{-3}.

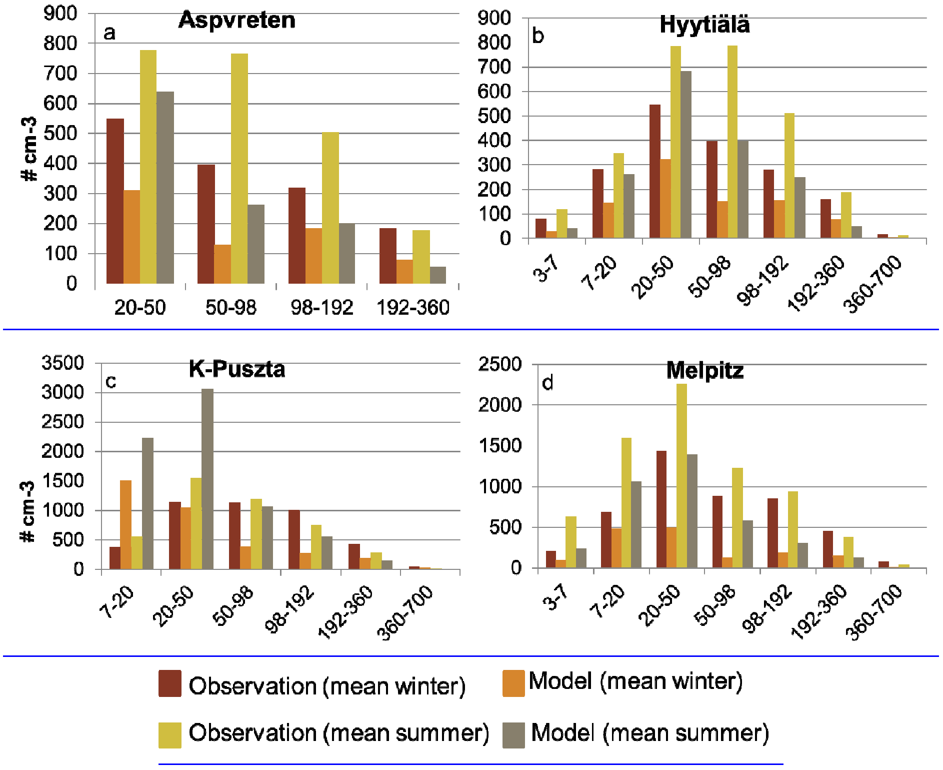

Figure 5. Modeled and measured winter (Jan-March, Oct-Dec) and summer (April-September) half year mean particle number concentration size distribution at four measurement sites in Europe during 2007. Unit: # cm^{-3}.

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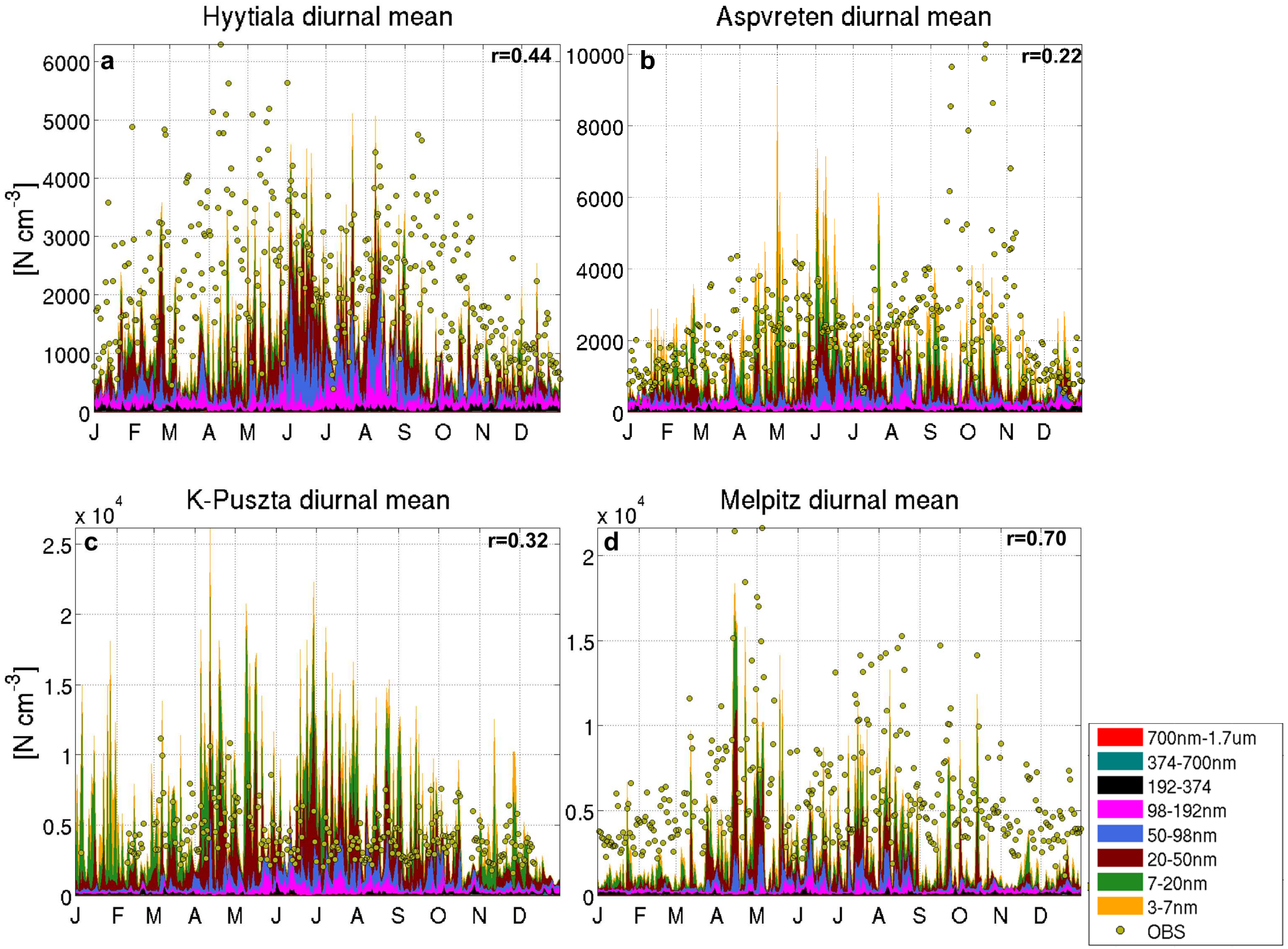
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Aspvreten diurnal mean
b
r=0.22
0
2000
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6000
8000
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J F M A M J J A S O N D
K-Puszta diurnal mean
x 10^4
c
r=0.32
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0.5
1
1.5
2
2.5
J F M A M J J A S O N D
Melpitz diurnal mean
x 10^4
d
r=0.70
0
0.5
1
1.5
2
J F M A M J J A S O N D
700nm-1.7um
374-700nm
192-374
98-192nm
50-98nm
20-50nm
7-20nm
3-7nm
OBS

Figure 6. Observed and modeled daily mean particle number concentrations (PNC) at four sites in Europe during 2007 (panels a-d). Modeled (surfaces) size resolved and observed total (filled circles) daily mean PNC are displayed as a time series. See legend for colors representing the different size bins. Observed PNC limit diameters are: 3.2nm-1µm for Hyytiälä, 11-418nm for Aspvreten, 5.6-1 µm for K-Puszta and 3-859nm for Melpitz. Unit: # cm^{-3}.

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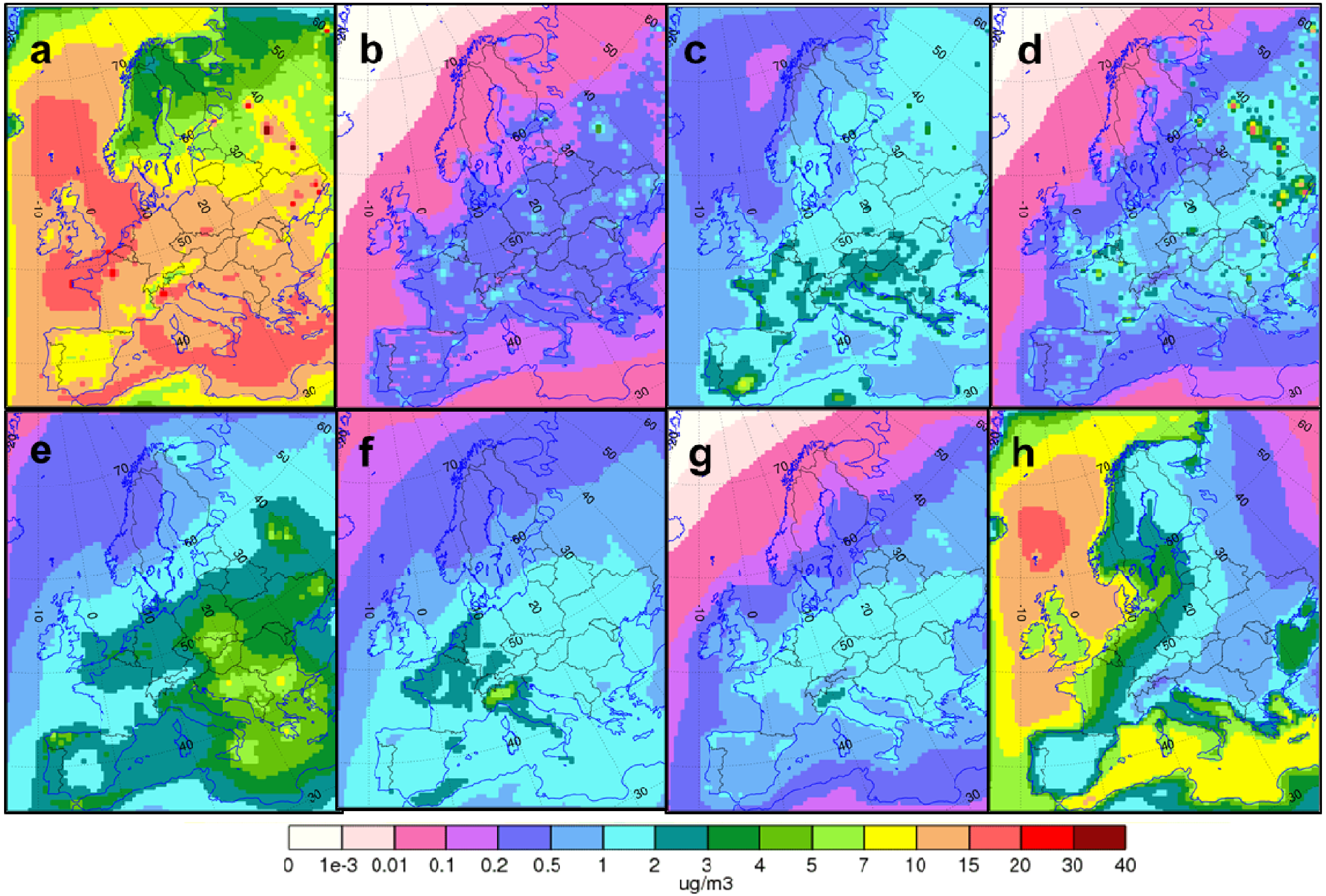
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ug/m3

Figure 7. Modeled annual mean concentrations (for 2007) of PM_{10} (panel a; peak at 37 µg/m^3 in Moscow) and its particle components: elemental carbon (panel b), organic matter (panel c), anthropogenic primary inorganic aerosol (panel d), sulfate (panel e), nitrate (panel f), ammonium (panel g) and sea salt (panel h). Unit: µg m^{-3}.

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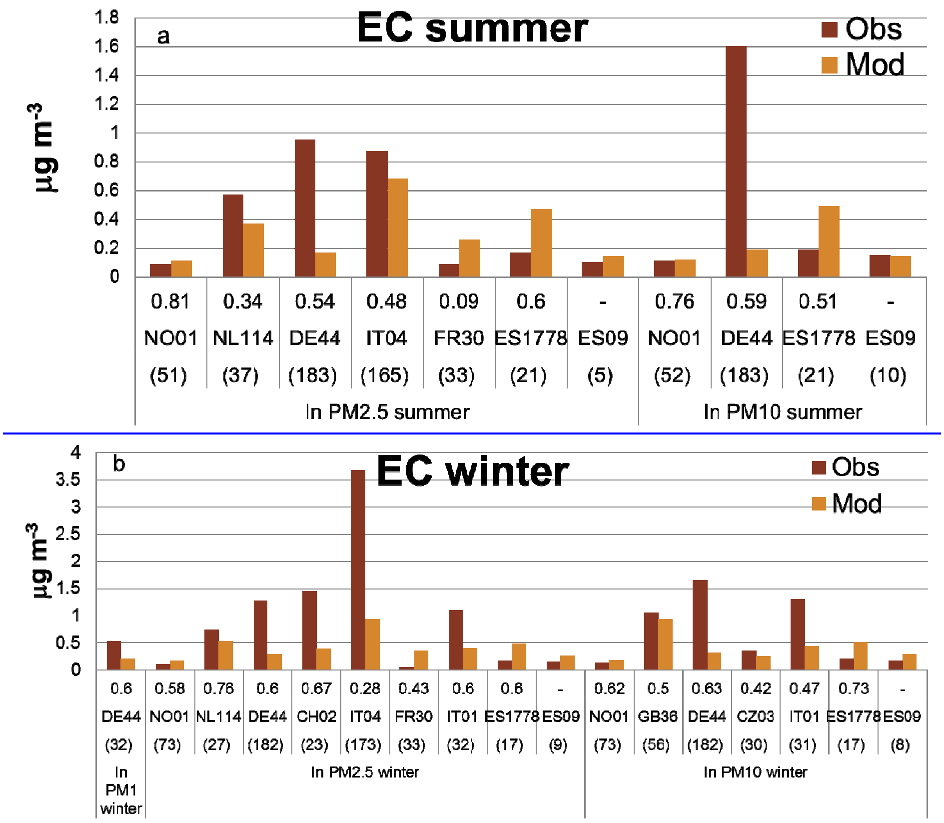

Figure 8. Evaluation of elemental carbon (EC) for 2007 (panel a: April-September mean; panel b: October-March mean). Observed and modeled mean concentrations (unit: µg m^{-3}), correlation coefficients of daily mean concentrations are indicated below the bars. The number of daily mean values is indicated by the numbers in the parentheses. Correlation coefficients were calculated for measurement sites with more than 10 daily observations. Site codes as defined by EMEP, see Supplement A Table 5.

Borttaget: EC (top) and OC (bottom) for 2007.

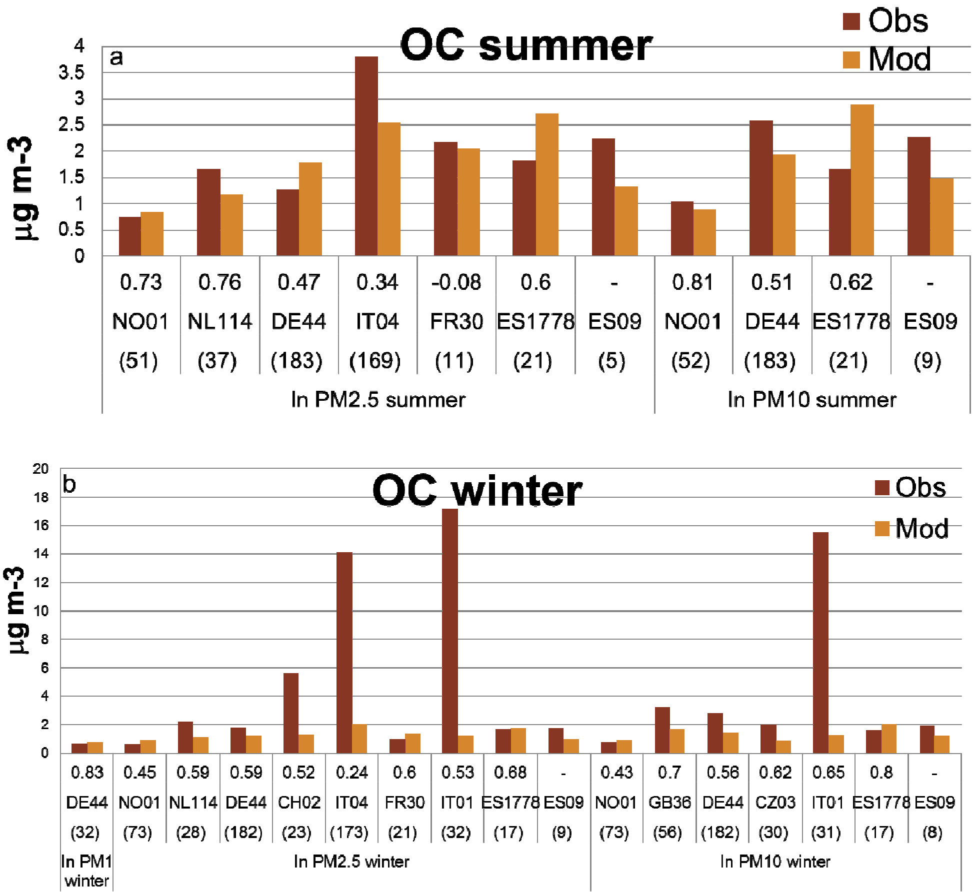

Figure 9. As Figure 8 but for organic carbon (OC).

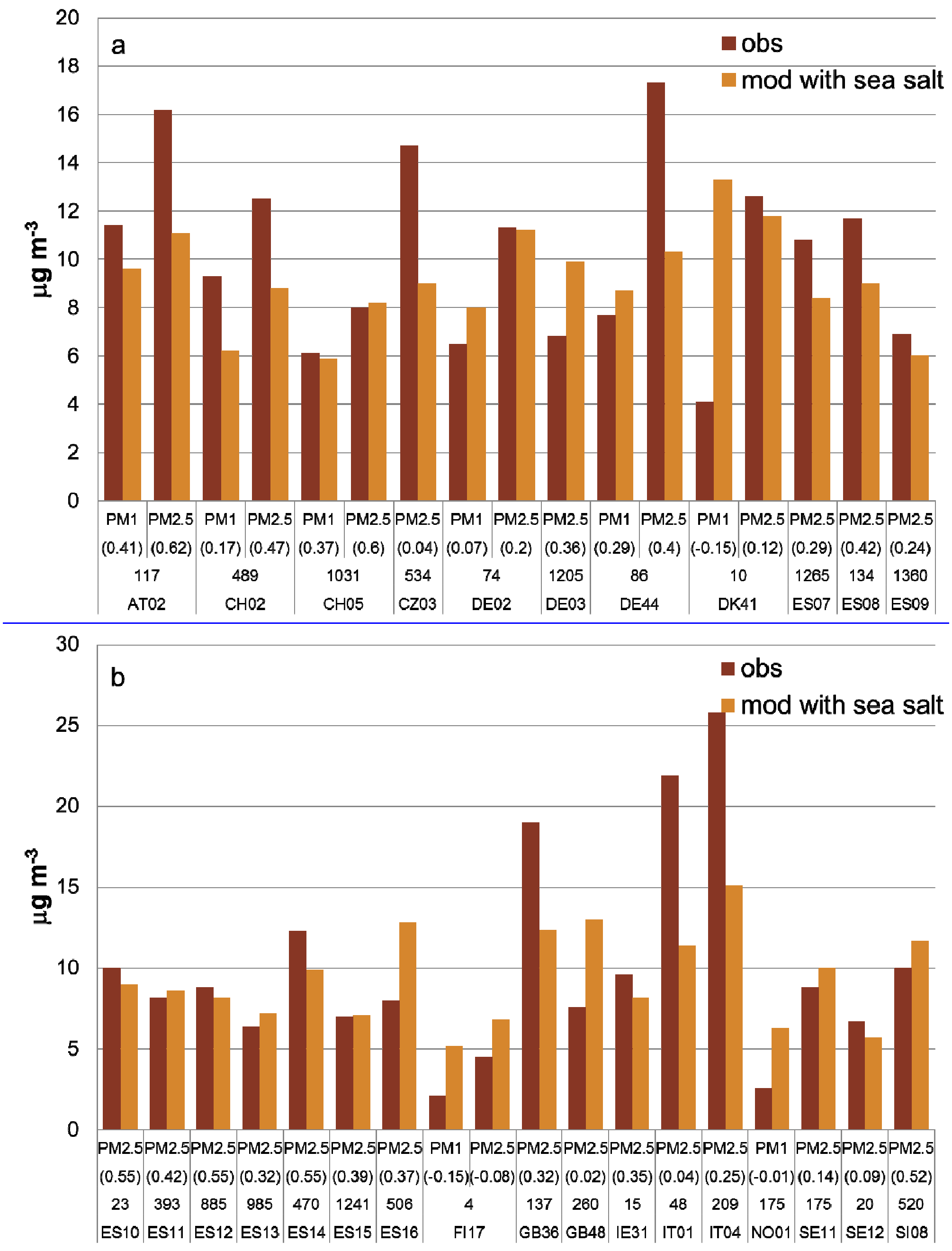

Figure 10. Evaluation of PM_1 and $PM_{2.5}$ for 2007. Observed and modeled mean concentrations (unit: µg m^{-3}); correlation coefficients of daily mean concentrations are indicated below the bars within parentheses. The elevation of each site is included below the

correlation coefficients (unit: m above sea level). Station codes as defined by EMEP, see Supplement A Table 5.

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Total particulate matter (PM_1 and $PM_{2.5}$)

Evaluation of PM_1 and $PM_{2.5}$ at 28 measurement sites is presented in Fig 9. and in the Supplement Table A21 and Fig. A39; detailed time series plots are given in the Supplement Figs. 17, A40-A41. For PM_1 the annual means at the sites with the lowest concentration (Scandinavian sites NO01, FI17, DK41) are overestimated by the model. On the other hand, at the central European sites the PM_1 concentrations are much better captured. The model underestimates $PM_{2.5}$ by 14% (spatial average) and the spatial correlation coefficient is 0.64. Out of the 35 evaluated annual means (PM_1 and $PM_{2.5}$) at the 28 stations, six means (at five stations) deviate by more than 50%. The largest underestimations of $PM_{2.5}$ are seen at the measurement sites with the highest observed annual mean. The underestimation of $PM_{2.5}$ can be due to a number of reasons including underestimated emissions, too short aerosol lifetime or too little secondary aerosol production. There is probably too little EC and OC in the model, at least at some of the sites, which can be explained by underestimated emissions.

Identified issues

During this work we found that further improvement is needed for a better representation of PNC. Here, in this section we would try to address some of the issues related to model development and measurements that could be relevant. The three of these issues will be further investigated in Andersson et al. (2014):

Distribution of SOx. In atmospheric models, given fractions of SOx emissions are assumed as gaseous SO_2, H_2SO_4 and primary sulfate, which is intended to account for subgrid scale processes of gas phase transformation and gas-to-particle partitioning. The assumed fractions have large uncertainty and it is not clear from the literature how to divide SOx emissions between $SO_2(g)$, $H_2SO_4(g)$ and particulate sulfate in modeling studies. Spracklen et al. (2005) discussed that the distribution depends on model resolution. Lee et al. (2013) have shown that the sub-grid production of a few per cent mass of sulfate particles in plumes is much more important for CCN uncertainty than the SO2 emissions themselves. Since we suspect this choice to have impact on the model results, we investigate this further in Andersson et al. (2014).

SOA condensation and nucleation. This version of MATCH-SALSA contains a scheme of formation of SOA, in which SOA precursors are assumed to condense on particles as non-volatile compounds. The SOA formation scheme is simplified and needs further development. For example, atmospheric SOA compounds have a wide variety of volatilities that would affect their partitioning between gas and particles. Also, biogenic emissions are highly uncertain, and the chemistry of SOA formation is complex and modelling of SOA is fraught with great difficulty (e.g. Hallquist et al., 2009; Bergström et al., 2012). For these reasons we test the model sensitivity on the amount of SOA available for condensation in Andersson et al. (2014). Further, MATCH-SALSA contains a scheme including organic nucleation that was not used in this study. In Andersson et al. (2014) the impact of including organic nucleation on modeled PNC is also tested.

Wet scavenging is the most important sink for accumulation mode particles. At many sites particle concentrations are underestimated by the MATCH-SALSA model when the standard wet deposition scheme is used. Several other, more and less advanced, formulations of wet scavenging are implemented in the MATCH model and in the companion paper we also investigate the sensitivity of the of the modeled particle mass and PNC on the wet scavenging formulation.

The treatment of sea spray needs to be further evaluated and the model scheme for sea salt particles may need to be updated. For PM_1 the annual means at the sites with the lowest concentration (Scandinavian sites NO01, FI17, DK41) are overestimated by the model. This seems to be partly due to overestimation of sea salt. Evaluation scores for modeled PM_1 and $PM_{2.5}$ excluding sea salt aerosol in the total PM mass (see Supplement Table A21, Figs. 18 and A39) gives higher correlation coefficients for daily mean $PM_{2.5}$ or PM_1 at 22 of the 28 sites (and lower at only one site) than when sea salt is included. This is an indication of too much sea salt at the wrong time. It may be due to too strong sea salt emissions and/or too weak sink processes for the sea salt, since substantial improvements in correlation are seen also at some far inland sites.

For EC and OC, there is probably a combination of need for model development, uncertainties in measurements and emission inventories.

Sid. 21: [2] Flyttad till sidan 22 (Flytta #6) **Andersson Camilla** **2014-11-27 11:27:00**

Total particulate matter (PM_1 and $PM_{2.5}$)

Evaluation of PM_1 and $PM_{2.5}$ at 28 measurement sites is presented in Fig

Sid. 21: [3] Flyttad till sidan 22 (Flytta #7) **Andersson Camilla** **2014-11-27 11:27:00**

On the other hand, at the central European sites the PM_1 concentrations are much better captured. The model underestimates $PM_{2.5}$ by 14% (spatial average) and the spatial correlation coefficient is 0.64.

Sid. 21: [4] Flyttad till sidan 22 (Flytta #8) **Andersson Camilla** **2014-11-27 11:27:00**

There is probably too little EC and OC in the model, at least at some of the sites, which can be explained by underestimated emissions.

Sid. 21: [5] Flyttad till sidan 22 (Flytta #9) Andersson Camilla 2014-11-27 11:27:00

The treatment of sea spray needs to be further evaluated and the model scheme for sea salt particles may need to be updated.

Sid. 21: [6] Flyttad till sidan 22 (Flytta #10) Andersson Camilla 2014-11-27 11:27:00

This seems to be partly due to overestimation of sea salt. Evaluation scores for modeled PM$_1$ and PM$_{2.5}$ excluding sea salt aerosol in the total PM mass (see Supplement

Sid. 21: [7] Flyttad till sidan 22 (Flytta #11) Andersson Camilla 2014-11-27 11:27:00

and A39) gives higher correlation coefficients for daily mean PM$_{2.5}$ or PM$_1$ at 22 of the 28 sites (and lower at only one site) than when sea salt is included. This is an indication of too much sea salt at the wrong time. It may be due to too strong sea salt emissions and/or too weak sink processes for the sea salt, since substantial improvements in correlation are seen also at some far inland sites.

Sid. 32: [8] Formaterat Andersson Camilla 2014-11-27 11:27:00

Teckensnitt:Inte Kursiv

Sid. 32: [8] Formaterat Andersson Camilla 2014-11-27 11:27:00

Teckensnitt:Inte Kursiv

Sid. 32: [9] Borttaget Andersson Camilla 2014-11-27 11:27:00

K. S.

Sid. 32: [9] Borttaget Andersson Camilla 2014-11-27 11:27:00

K. S.

Sid. 32: [9] Borttaget Andersson Camilla 2014-11-27 11:27:00

K. S.

Sid. 32: [9] Borttaget Andersson Camilla 2014-11-27 11:27:00

K. S.

Sid. 32: [9] Borttaget **Andersson Camilla** **2014-11-27 11:27:00**

K. S.

Sid. 32: [9] Borttaget **Andersson Camilla** **2014-11-27 11:27:00**

K. S.

Sid. 32: [9] Borttaget **Andersson Camilla** **2014-11-27 11:27:00**

K. S.

Sid. 32: [9] Borttaget **Andersson Camilla** **2014-11-27 11:27:00**

K. S.

Sid. 32: [9] Borttaget **Andersson Camilla** **2014-11-27 11:27:00**

K. S.

Sid. 32: [9] Borttaget **Andersson Camilla** **2014-11-27 11:27:00**

K. S.

Sid. 32: [9] Borttaget **Andersson Camilla** **2014-11-27 11:27:00**

K. S.

Sid. 32: [9] Borttaget **Andersson Camilla** **2014-11-27 11:27:00**

K. S.

Sid. 32: [9] Borttaget **Andersson Camilla** **2014-11-27 11:27:00**

K. S.

Sid. 32: [9] Borttaget **Andersson Camilla** **2014-11-27 11:27:00**

K. S.

Sid. 32: [9] Borttaget **Andersson Camilla** **2014-11-27 11:27:00**

K. S.

Sid. 32: [9] Borttaget **Andersson Camilla** **2014-11-27 11:27:00**

K. S.

Sid. 32: [9] Borttaget **Andersson Camilla** **2014-11-27 11:27:00**

K. S.

Sid. 32: [9] Borttaget **Andersson Camilla** **2014-11-27 11:27:00**

K. S.

Sid. 32: [9] Borttaget **Andersson Camilla** **2014-11-27 11:27:00**

K. S.

Sid. 32: [9] Borttaget **Andersson Camilla** **2014-11-27 11:27:00**

K. S.

Sid. 32: [9] Borttaget **Andersson Camilla** **2014-11-27 11:27:00**

K. S.

Sid. 32: [9] Borttaget **Andersson Camilla** **2014-11-27 11:27:00**

K. S.

Sid. 32: [9] Borttaget **Andersson Camilla** **2014-11-27 11:27:00**

K. S.

Sid. 32: [9] Borttaget **Andersson Camilla** **2014-11-27 11:27:00**

K. S.

Sid. 32: [9] Borttaget **Andersson Camilla** **2014-11-27 11:27:00**

K. S.

Sid. 32: [9] Borttaget Andersson Camilla 2014-11-27 11:27:00

K. S.

Sid. 32: [9] Borttaget Andersson Camilla 2014-11-27 11:27:00

K. S.

Sid. 32: [9] Borttaget Andersson Camilla 2014-11-27 11:27:00

K. S.

Sid. 32: [9] Borttaget Andersson Camilla 2014-11-27 11:27:00

K. S.

Sid. 32: [9] Borttaget Andersson Camilla 2014-11-27 11:27:00

K. S.

Sid. 32: [9] Borttaget Andersson Camilla 2014-11-27 11:27:00

K. S.

Sid. 32: [9] Borttaget Andersson Camilla 2014-11-27 11:27:00

K. S.

Sid. 32: [9] Borttaget Andersson Camilla 2014-11-27 11:27:00

K. S.

Sid. 32: [9] Borttaget Andersson Camilla 2014-11-27 11:27:00

K. S.

Sid. 32: [9] Borttaget Andersson Camilla 2014-11-27 11:27:00

K. S.

Sid. 32: [9] Borttaget Andersson Camilla 2014-11-27 11:27:00

K. S.

Sid. 32: [9] Borttaget **Andersson Camilla** **2014-11-27 11:27:00**

K. S.

Sid. 32: [9] Borttaget **Andersson Camilla** **2014-11-27 11:27:00**

K. S.

Sid. 32: [9] Borttaget **Andersson Camilla** **2014-11-27 11:27:00**

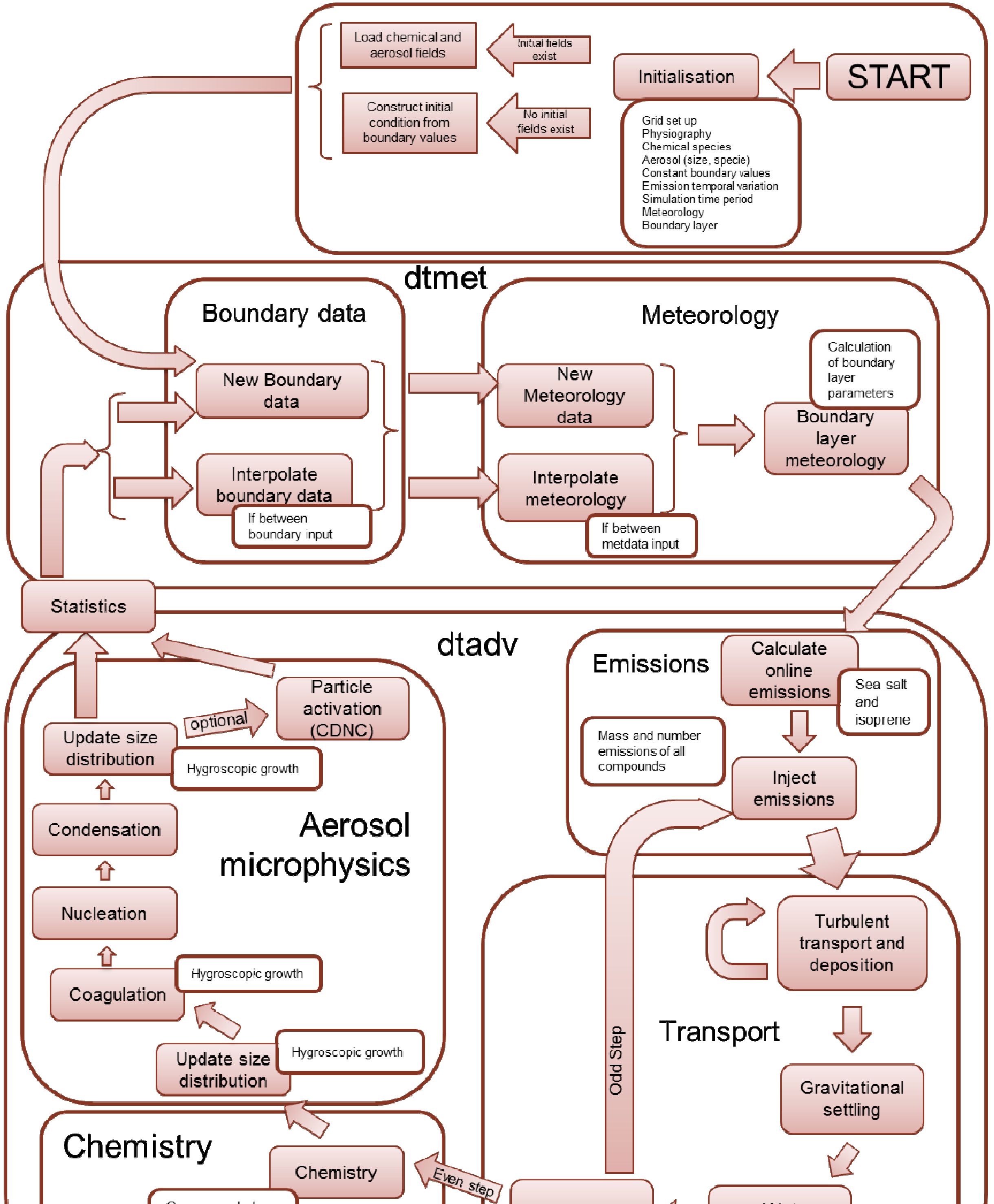
K. S.

Sid. 32: [9] Borttaget **Andersson Camilla** **2014-11-27 11:27:00**

K. S.

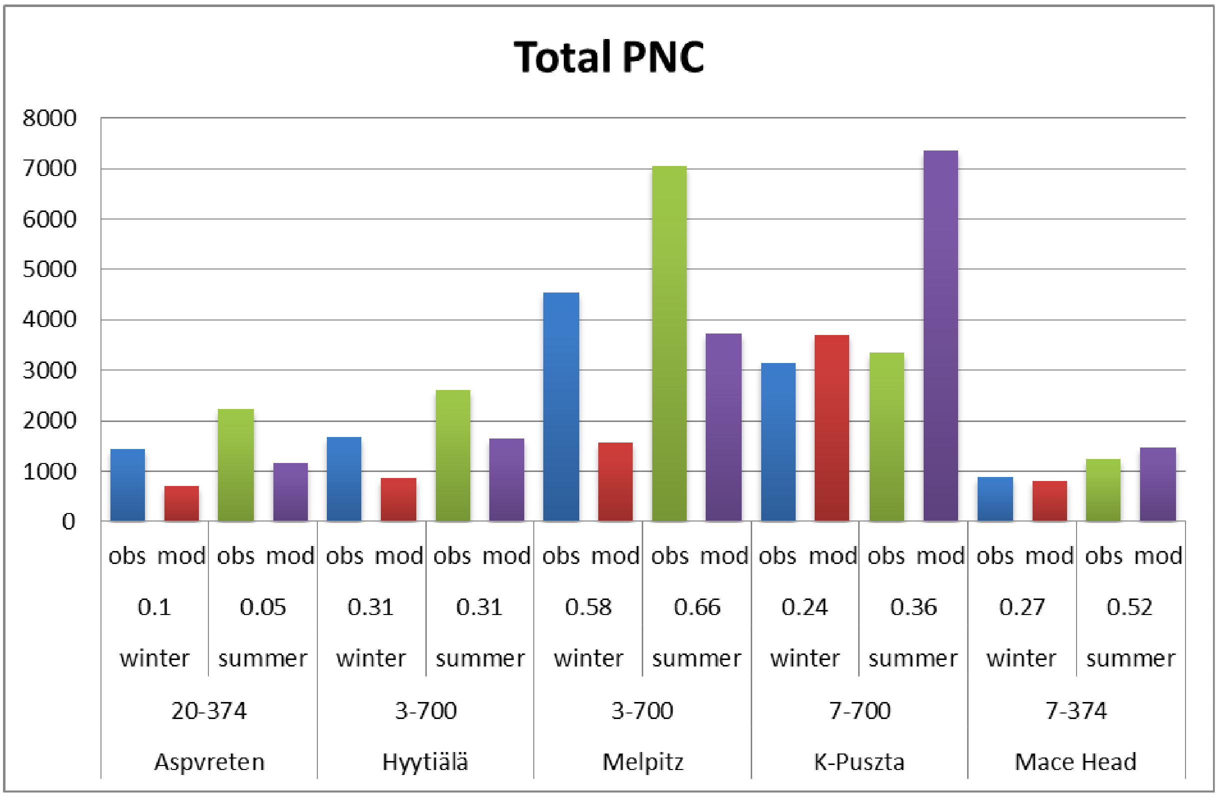
Sid. 37: [10] Borttaget **Andersson Camilla** **2014-11-27 11:27:00**

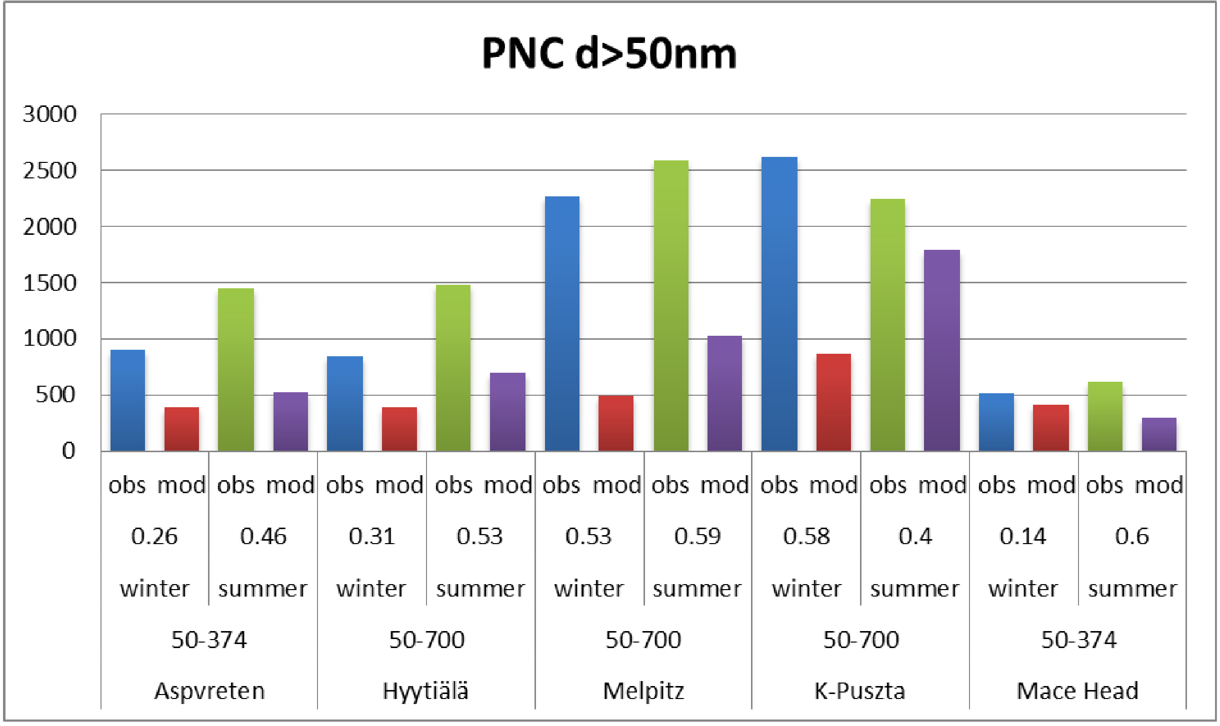
1 Sidbrytning


START
Initialisation
Grid set up
Physiography
Chemical species
Aerosol (size, specie)
Constant boundary values
Emission temporal variation
Simulation time period
Meteorology
Boundary layer
Initial fields exist
Load chemical and aerosol fields
No initial fields exist
Construct initial condition from boundary values
dtmet
Boundary data
New Boundary data
Interpolate boundary data
If between boundary input
Meteorology
New Meteorology data
Interpolate meteorology
If between metdata input
Calculation of boundary layer parameters
Boundary layer meteorology
Statistics
dtadv
Emissions
Calculate online emissions
Sea salt and isoprene
Mass and number emissions of all compounds
Inject emissions
Aerosol microphysics
Particle activation (CDNC)
optional
Update size distribution
Hygroscopic growth
Condensation
Nucleation
Coagulation
Hygroscopic growth
Update size distribution
Hygroscopic growth
Transport
Turbulent transport and deposition
Gravitational settling
Odd Step
Chemistry
Chemistry
Even step

Sid. 43: [11] Borttaget **Andersson Camilla** **2014-11-27 11:27:00**

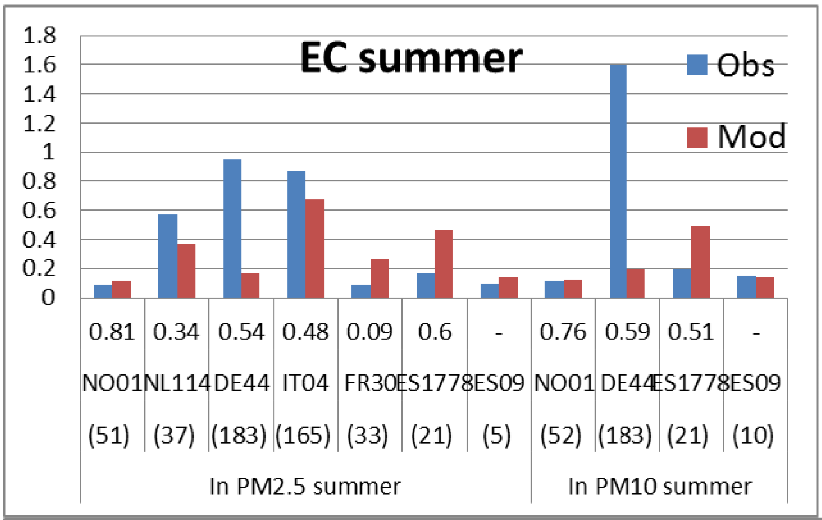
Avsnittsbrytning (nästa sida)

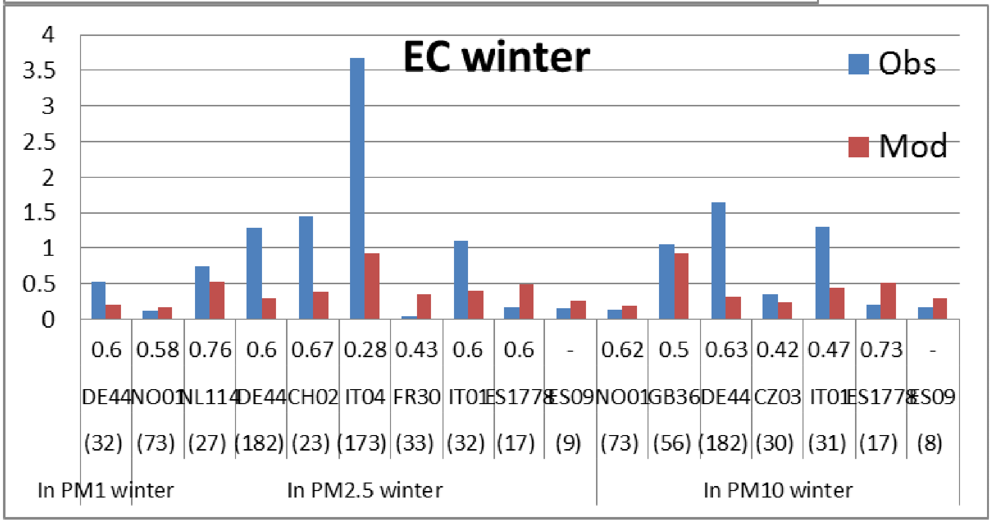


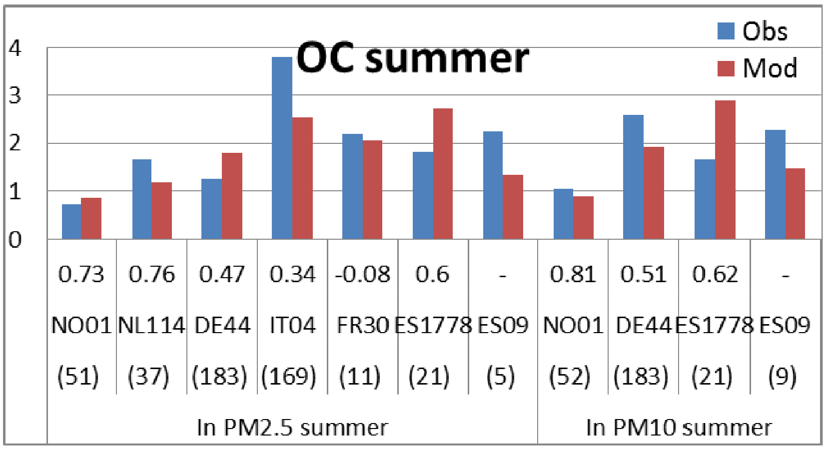


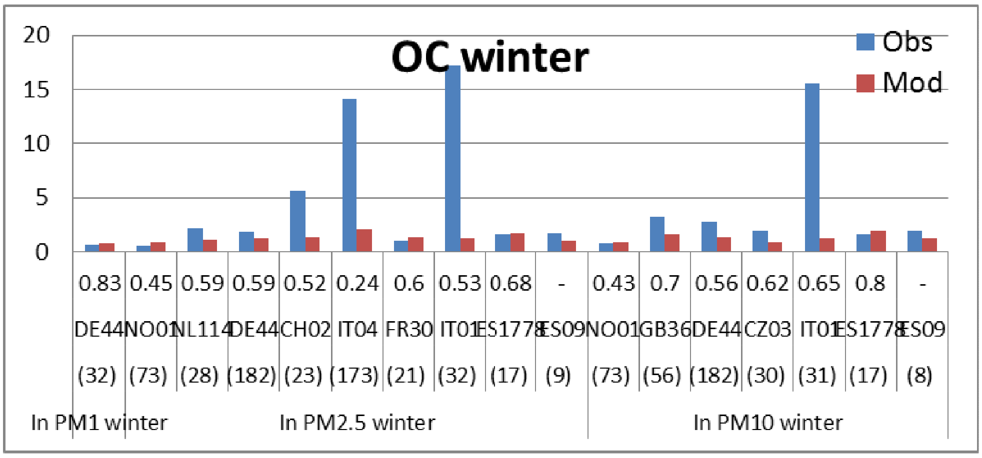
Sid. 50: [12] Borttaget **Andersson Camilla** **2014-11-27 11:27:00**

Sidbrytning


EC summer
Obs
Mod
1.8
1.6
1.4
1.2
1
0.8
0.6
0.4
0.2
0
0.81 NO01 (51)
0.34 NL114 (37)
0.54 DE44 (183)
0.48 IT04 (165)
0.09 FR30 (33)
0.6 ES1778 (21)
- ES09 (5)
0.76 NO01 (52)
0.59 DE44 (183)
0.51 ES1778 (21)
- ES09 (10)
In PM2.5 summer
In PM10 summer


EC winter
Obs
Mod
4
3.5
3
2.5
2
1.5
1
0.5
0
0.6 DE44 (32)
0.58 NO01 (73)
0.76 NL114 (27)
0.6 DE44 (182)
0.67 CH02 (23)
0.28 IT04 (173)
0.43 FR30 (33)
0.6 IT01 (32)
0.6 ES1778 (17)
- ES09 (9)
0.62 NO01 (73)
0.5 GB36 (56)
0.63 DE44 (182)
0.42 CZ03 (30)
0.47 IT01 (31)
0.73 ES1778 (17)
- ES09 (8)
In PM1 winter
In PM2.5 winter
In PM10 winter


OC summer
Obs
Mod
4
3
2
1
0
0.73 0.76 0.47 0.34 -0.08 0.6 - 0.81 0.51 0.62 -
NO01 NL114 DE44 IT04 FR30 ES1778 ES09 NO01 DE44 ES1778 ES09
(51) (37) (183) (169) (11) (21) (5) (52) (183) (21) (9)
In PM2.5 summer
In PM10 summer


OC winter
Obs
Mod
20
15
10
5
0
0.83 0.45 0.59 0.59 0.52 0.24 0.6 0.53 0.68 - 0.43 0.7 0.56 0.62 0.65 0.8 -
DE44 NO01 NL114 DE44 CH02 IT04 FR30 IT01 ES1778 ES09 NO01 GB36 DE44 CZ03 IT01 ES1778 ES09
(32) (73) (28) (182) (23) (173) (21) (32) (17) (9) (73) (56) (182) (30) (31) (17) (8)
In PM1 winter
In PM2.5 winter
In PM10 winter