

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 Aspöreten. 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 Aspöreten 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.

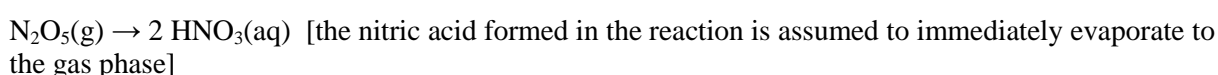
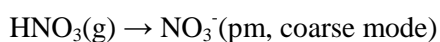
R4. Figure 1: It should be specified which parts of the flow chart show MATCH and which parts characterize SALSA operations. It is also not clear why output from the aerosol microphysics module is needed as input for the meteorological part of the model. This should be specified in more detail and Figure 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 HNO₃ and N₂O₅:



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

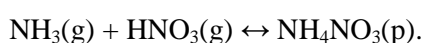
We apply the pressure-scaling factor $[M]/2.55 \times 10^{-19}$ for both reactions (Strand and Hov, 1994) and for the HNO₃-reaction we use the reaction rate for low relative humidity conditions (RH<0.9), $k=5 \times 10^{-6} \times [M]/2.55 \times 10^{-19} \text{ s}^{-1}$, regardless of the actual RH (as Strand and Hov, 1994).

Note that the nitrate formed in this HNO₃-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:

NH₃(g) reacts instantaneously (and irreversibly) with available sulfate and form ammonium sulfate (NH₄)₂SO₄, which is distributed over different particle sizes according to the sulfate distribution in MATCH-SALSA.

If there is excess NH₃(g) available, ammonium nitrate can be formed via the reaction:



The ammonium nitrate is assumed to be in equilibrium and the dissociation constant of NH₄NO₃ 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; O₃ 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 O₃ at lower altitudes than to scavenge it from the air. For SO₂ the neglect of sub-cloud scavenging is likely leading to a slight underestimation of the wet-deposition losses; but SO₂ 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 H₂O₂, it was found that sub-cloud scavenging contributed about 20-40% to the total wet deposition of H₂O₂. 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.