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.
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> 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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Appendix

[see next page]

Solving condensation and nucleation concurrently, we use a method developed by Jacobson (2007), which solves these processes over a discrete time step h. Here we modified the method to solve these processes for two compounds, sulphuric acid and nucleating organic compound.

Jacobson (2007) assumed that the mass transfer rate of compound q in the smallest size bin is the sum of mass transfer rate of condensation and mass transfer rate of homogeneous nucleation (Equation (16.73) in Jacobson (2007)). Since in our model, the smallest diameter in the size distribution is 3 nm, we modify this equation so that the mass transfer rate in the smallest size bin is the sum of mass transfer rates of condensation $k_{q,cond}$ and mass transfer rate of 3 nm particle formation $k_{q,form}$.

$$k_{q,1,t-h} = k_{q,cond,1,t-h} + k_{q,form,1,t-h}. \tag{0.1}$$

The mass transfer rate of compound q during formation of 3 nm particles is analogous to homogeneous nucleation mass transfer rate (Equation (16.74) in Jacobson (2007)):

$$k_{q,form,1,t-h} = \frac{\rho_q v_1}{m_q} \left(\frac{J_{3,q}}{C_{q,t-h} - S'_{q,1,t-h} C_{q,s,1,t-h}} \right), \tag{0.2}$$

where rho_q is the mass density, m_q is the molecular weight of compound q, v_1 is is the volume of a 3 nm particle, $C_{q,t-h}$ is the gas phase concentration, $S'_{q,1,t-h}$ is the Kelvin effect, and $C_{q,s,1,t-h}$ is the equilibrium concentration at the particle surface. We assume the equilibrium concentration to be zero for both condensing compounds. $J_{3,q}$ is the contribution of compound q (sulphuric acid SU and organic OC) to the formation rate of 3 nm particles, J_3

$$J_3 = J_{3,\mathrm{SU}} + J_{3,\mathrm{SU}} = \frac{(n^*_{\mathrm{SU}} + n_{3,\mathrm{SU}}) v_{\mathrm{SU}}}{v_1} J_3 + \frac{(n^*_{\mathrm{OC}} + n_{3,\mathrm{OC}}) v_{\mathrm{OC}}}{v_1} J_3 \tag{0.3}$$

where n^*_q is the number of molecules in the critical cluster, $n_{3,q}$ is the number of molecules in a 3 nm particle, v_q is the molecular volume. The number of molecules of individual compounds in a 3 nm particle is assumed follow the ratio of gas phase concentrations

$$n_{3,\mathrm{SU}} v_1 \approx \frac{C_{\mathrm{SU},t-h} v_{\mathrm{SU}}}{C_{\mathrm{SU},t-h} v_{\mathrm{SU}} C_{\mathrm{OC},t-h} v_{\mathrm{OC}}} (v_1 - v^*), \tag{0.4}$$

where v^* is the volume of the critical cluster.

Now, gas and size-resolved aerosol mole concentrations can be solved using equations (16.67)-(16.72) by Jacobson (2007). The new number concentration of particles in the smallest size bin is

$$n_{1,t} = n_{1,t-h} + \sum_{q=1}^{2} \mathrm{MAX} \left[(c_{q,1,t} - c_{q,1,t-h}) \frac{m_q}{\rho_q v_1} \frac{k_{q,form,1,t-h}}{k_{q,1,t-h}}, 0 \right], \tag{0.5}$$

where c_q is the mole concentration of compound q in the particle phase.