Response to Referee comments

We thank both referees for their comments. The common concern was that this manuscript was a pure documentation paper. We argue below that such a paper is needed (indeed, well overdue) to accompany the wealth of other EMEP-model related papers, and that the EMEP special issue is the proper place for this paper.

Point by point responses to both referees are given below.

1 Response to Referee #1 comments

1.1 Referee comment:

This paper provides a description of a new version of the EMEP model. The model is described in detail. I can find no serious errors in the paper. However, the paper is purely a description of the model and does not include any science. That is, the paper does not address relevant scientific questions or reach any substantial conclusions. Model evaluation is left for a second paper. Therefore whilst the paper will be of great interest to the atmospheric modelling community and would act as a very useful reference to the EMEP model the paper does not seem to be strictly within scope of ACP. As written, it is my opinion that the paper would be more suited to a journal such as GMD.

Reply

We have submitted this paper part of the ACP special issue ”EMEP - an integrated system of models and observations in support of European air quality and policy”. Although we agree that GMD would be a suitable journal for this paper as a stand-alone item, we feel that it makes sense to document the EMEP model as a part of this special issue. This paper was also welcomed to the EUCAARI ACP special issue by the editors of that issue. As the referee notes, this paper should be a very useful reference for the EMEP model (something our colleagues have been strongly demanding for years), and the ACP special issues was always intended to accommodate such model documentation.

Further, now that this paper has been accepted for ACPD, and had an discussion period (linked from 2 special ACPD issues) of over 2 months, we are keen to continue the publication process within ACP and not switch journal at this late stage.

1.2 Referee #1 minor comments:

1. P3782, L8 and P3783, L15: Should this be ”50 × 50 km”, not ”50 × 50 km2”. Same comment applies at several other places e.g. P3784, L5; P3785, L7; P2785, L10

   Answer: We found differing advice on this in different sources. According to the AGU style guide we should have used 50 km × 50 km.

2. P3783, L14-L17: These 4 lines are a direct repeat of the abstract.

   Answer: or rather, the abstract repeats these lines. We believe that this is an important statement, and as both the abstract and the introduction are supposed to be read independently of each other, we feel that this repetition is worthwhile.

3. P3787, L2-4. Can you provide a reference to support this statement?

   Answer: The statements that convection is very difficult to parameterise is taken from the cited Stevenson paper, but there are many papers which support this. We have moved this statement to the end of the convection section, and add the following extra support:

   Willett et al. (2008), Zhao et al. (2009) and Monks et al. (2009) (and references cited therein) also give examples where significant differences in precipitation and mass transport were found between different parameterisations of convection in NWP models.

4. P3788, L8. eta?

   Answer: The ECMWF model uses a so-called eta coordinate system in the vertical. Rather than go into the details of how this differs from the sigma coordinates used in EMEP, we will simply omit the term eta, and just inform that ECMWF uses 91 vertical layers.

5. P3808, L10. Please clarify what you mean that DMS emissions are treated as SO2 on input to the calculations

   Answer: The EMEP model DMS treatment is based entirely on the work of Tarrasón et al. (1995). We have re-phrased the paragraph to make this clearer:

Biogenic emissions of dimethly sulphide (DMS) can sometimes make a significant input to European sulphate levels. As discussed in detail by Tarrasón et al. (1995), the EMEP model uses a very simplified treatment, in which DMS is not modelled explicitly, but rather we assume that most DMS enters long-range transport already as sulphur dioxide. Monthly emission fields of DMS-derived SO_2 are taken from the work of Tarrasón et al. (1995).

6. P3808, L12. What emissions data for volcanoes do you use?

   Answer: This information has been added to the text:

   For the standard European-scale runs, volcano emissions are based upon officially reported data. These have been provided by Italy for many years, and recently by Iceland. (For global and regional scale calculations, a new module for volcanic eruptions with default values based upon Mastin et al. (2009a,b) has recently been implemented and is currently in testing.)

2 Response to referee #2

2.1 Referee comment:

The paper contains a relatively detailed description of the most recent version of the EMEP chemical transport model. It documents modelling choices made by the EMEP developers for the various components of the model; in some places there is some interesting discussion of the reasons for these choices, but in most cases there is just a list of the corresponding equations and formulations. A lot of the material presented can be found in previous papers, reports with descriptions and/or applications of the model but also in textbooks. There is little that is new here (no substantial new concepts, ideas, methods, or data) something that is obvious in the conclusions section.

I do understand the need to document the contents of a complex CTM for future reference. However, I believe that this could be accomplished in a few pages of supplementary material in the forthcoming EMEP evaluation paper using the references to previous work. This shorter description could be quite helpful for the reader too, because I had a hard time identifying the most recent changes in the model.

Reply:

We are happy that the referee sees the need to document the contents of a complex CTM for future reference. We strongly disagree that this material could be presented in a few pages of supplementary material! We hope it is obvious from the comments listed below that we are not dealing with a few cosmetic changes to the 2003 model, but a significant number of changes.

The last full documentation of the EMEP MSC-W model is almost ten years old (Simpson et al. 2003). The model has changed in numerous ways since this document was written, and many changes (both large and small) have not been documented until now. It is a very confusing and in fact impossible task to understand the current model formulation by reference to the multiple snippets of information presented in articles and report chapters over the last 10 years. Indeed, EMEP status reports 1/2010 and 1/2011 used about 5 pages just giving lists of bullet-points of all the changes in those years. Both reports referred to the forthcoming documentation article for proper details – the one which we are now trying to publish.

Since this manuscript has appeared on ACPD, we have received numerous comments from colleagues, welcoming this article as a thorough description of the code as it now is. Many people were also surprised at the changes which have taken place in the EMEP model, and the flexibility we now have with regard to model resolution, meteorological inputs, and chemical mechanisms.

The EMEP MSC-W chemical transport model is one of the key models used in policy support in Europe. It is central to UN-ECE work, is the sole provider of source-receptor matrices to the IIASA GAINS model (which is central to EU policy work), and is used in many EU projects alongside other chemical transport models. The model is public domain, and it behoves the model developers to document their code, so that both scientific and policy-interested users can review the formulations. We also feel that this documentation is an important part of the EMEP special issue on ACP.

We summarise here a list of the main changes since the 2003 report:

- New calculation method for the boundary layer height, based upon the work of Jeričevič et al. (2010).
- New K_z values for stable and neutral PBL, again based upon Jeričevič et al. (2010).

- Changed parameter values for the land-cover types, coupled with new land-cover databases.
- For anthropogenic emissions, many changes were made in 2010, 2011 which have not previously been documented:
    - New vertical distribution of emissions (sec. 6.1.1, Table A2)
    - New temporal variation factors applied for SNAP-1 and SNAP-2 (sec. 6.1.2), and (in the revised manuscript) new hourly factors were introduced. An alternative form of the degree-day method (suggested by B. Bessagnet) was also added as an option in early 2012; this will be mentioned in the revised manuscript.
    - New VOC speciation (sec. 6.2)
    - EC, OC is now part of the default PM speciation used (sec. 6.3, Table A5), instead of the previous 'PM'. We have also introduced EC ageing (from Tsyro et al. 2007) along with this change.
    - Aircraft emissions now from QUANTIFY (sec. 6.4)
    - New shipping emissions (sec. 6.5)
- The biogenic VOC emissions treatment is now based upon gridded maps of 115 species in Europe, combined with emission factors which have been updated based upon recent literature. This system was introduced in 2010, further updated in 2011, and this manuscript is the first documentation of the methods used (sec. 6.6).
- The soil NO emissions treatment here is completely new, introduced in late 2011 and this is the first documentation (sec. 6.6).
- Sea-salt (sec. 6.8) is new to the standard MSC-W model, although this implementation has been discussed in a 2011 article, so we shorten the text in the revised submission.
- Forest-fires (sec. 6.9) were introduced to the model. Further, the system used for forest fires was updated in 2011 to use globally stored FINNv1 data, with the previously available GFED-3 data as an option.
- Organic aerosols were added to the EMEP model. Although organic aerosol (SOA) schemes have been in research versions of the EMEP model for

many years (Andersson-Sköld and Simpson 2001, Simpson et al. 2007), this is the first time that SOA has been included in the ’standard’ EMEP model. The main documentation of SOA schemes is reported elsewhere (Bergström et al. 2012), so here we give just 3 paragraphs. Still, the standard EMEP model uses a simplified POA representation compared to Bergström et al., 2012, see the discussion below.

- Windblown dust was introduced to the MSC-W model in 2011. Although early versions of scheme have been tested for several years with research model versions, we have only recently included near-surface soil water in the model, a key input to dust modelling, and now feel that this component is a useful addition to the ‘official’ PM modelling.
- The EmChem09 mechanism given here is an update of the 2003 mechanism. Many rates and some reactions have been changes, and HONO added to the gas-phase species. These changes have not previously been documented.
- Several other chemical schemes were implemented, ranging in complexity from CBM-IV to CRI v2 (Table 5).
- Changes were made to the particle sizes assumed for different components, so that now we have five classes as given in Table 6. (These parameters were updated somewhat in early 2012. We will give the latest parameters in the revised version.)
- The use of MARS (sec. 7.8) is new in the model, replacing the EQSAM system we had previously.
- The basics of the dry-deposition for ozone (and stomatal conductance) have been documented in several papers, but many small changes have been made over the years, and there is a clear need for a summary of the implementation as it is now. For sulphur and nitrogen compounds the equations have changed substantially in recent years (e.g. for the non-stomatal conductance, treatment of humidity, snow, etc.). The current setup has not been documented.
- The aerosol dry deposition scheme (sec. 8.9) is completely new, it has not been previously presented.

- The wet deposition scheme uses the same equations as in Simpson et al., 2003, but the values of the collection efficiency (Table A19) of fine particles has been reduced substantially.

- The section on model outputs was added because of the rather confusing list of possibilities concerning in particular the meaning of ecosystem-outputs, and of AOT and POD type outputs. It is important that users of EMEP model outputs understand what we are providing, so we feel this explanation will be helpful.

- Water associated with PM is now calculated with the same MARS model as used for gas/aerosol partitioning calculations. More comments on MARS below.

Some aspects of the model have remained constant over the years, in particular the basic advection structure (which stems from Berge and Jakobsen, 1998), photolysis (sec. 7.3, although e.g. HONO was added) and sulphate production (sec. 7.4–7.6). However, these sections have been kept relatively short. We feel that given the sheer amount of new material, it is better to have essentially all documentation in one place. to cover everything, than to force the reader to chase up these items in previous documents.

However, from the referee's comments we appreciate that we should have made more effort to highlight the changes which have been over the last ten years. We agree with the referee that the conclusions section does not reflect all these changes, and that the historical development could be clearer. We have therefore added text along the lines presented above to the introductory section, with reference to this in the conclusions. We have also expanded the description of some of the newer entries (adding some extra Figures) and reduced the description of some items (especially sea-salt) that are available in other peer-review literature.

2.2 Referee:

Unfortunately there is little discussion of the effects that these changes had on the CTM predictions. A discussion of the effects (e.g., the sensitivity of the model to process descriptions) could be a lot more interesting for the readers than the current list of model contents.

Reply:

To a large extent, such descriptions can be found in papers which are already published, currently in discussion, in preparation, or planned. As is obvious from the number of equations and data-sets though, a systematic discussion of the effects of all processes would be a mammoth undertaking. Any comparison of processes usually merits a paper of its own, requiring extensive explanations. We (or EMEP model users) have in fact being writing up such studies, as seen for organic aerosol schemes (Bergström et al. 2012), sea-salt modelling (Tsyro et al. 2011), water-content of aerosols (Tsyro 2005), ozone deposition (Tuovinen et al. 2004, 2009, Tuovinen and Simpson 2008), aerosol deposition schemes (Flechard et al. 2011), boundary layer physics (Jeričevič et al. 2010) or soil water modelling (Büker et al. 2012). The chemical schemes mention in section have been compared and a write-up is in progress (Hayman et al. 2012).

Comparison of the model for sulphur and nitrogen compounds and illustrations of the ecosystem-specific depositions can be found in Simpson et al. (2006a,b) and Fagerli and Aas (2008). Comparison against trends of inorganic species and EC can be found in Fagerli et al. (2007) and for ozone in Jonson et al. (2006). Comparison against AMS and other data-sets can be found in Aas et al. (2012). Processes affecting large scale CO (and to a lesser extent C_2H_6) have been presented in Angelbratt et al. (2011). A discussion of the fine-scale applications and performance of the model can be found in Vieno et al. (2009) and Vieno et al. (2010). Further, the EMEP model has been taking part in a large number of intercomparisons in recent years (e.g. Cuvelier et al. 2007, Fiore et al. 2009, Huijnen et al. 2010, Jonson et al. 2010, Colette et al. 2011).

There is a clear need for a documentation of the model used in all of these studies, and we believe the manuscript we have produced could not be made much shorter without compromising this important aim. It would be beyond the scope of the current paper to include the effects of changes in each process, since any such additions could only be very superficial and inevitably extend the size of the paper. This paper is intended indeed to serve as a reference document, so that we have a new ’base’ EMEP model to act as a reference against which test versions can be compared.

We have added some text to make the above points (and others) to an extended Discussion and Conclusions section in the new manuscript.

2.3 Referee:

Sections that are quite long and could be shortened significantly using mainly references to previous include: meteorology, biogenic emissions (including dust and sea salt), gas and aqueous-phase chemistry, dry deposition, outputs,

Reply:

As noted above in detail there have been changes in almost all of these sections.

Of the examples given by the referee, it is only the aqueous-phase chemistry which is unchanged in methodology since the 2003 report, and even there we have changed the values of the collection efficiencies. In the interests of presenting one model documentation we feel it is worth the few extra pages that inclusion of this material involves.

However, we have significantly reduced one section, that concerning sea-salt emissions. These have been presented in this ACP special issue (Tsyro et al. 2011) and so we agree there is no need to duplicate the equations here.

2.3.1 Referee:

The treatment of convection is a little confusing. Is it used during simulations over Europe or not? If it is not used why is it described?

Reply:

It is a little confusing, we agree. Convection is implemented in the model, but as an optional module. Used with global-scale IFS meteorological data, use of this module seems to give more realistic results compared to measurements. However, we find that if used with European-scale simulations, we obtain somewhat worse model results compared to observations. This is of course an unsatisfactory situation, but given that all cumulus schemes in NWP models have major uncertainties, we adopt a pragmatic approach and by default switch convection off for the European scale, and on for global scale. The option to switch this module on and off in any case affords some valuable information on the importance of convection, and the uncertainties associated with its implementation.

As noted in the reply to Ref #1, we have added extra references to back up the statements about uncertainty, and added some text to discuss the above points.

2.4 Referee:

There is little discussion of issues related to the aerosol size/composition distribution and processes that depend on particle size. Does the current EMEP model still use a two-mode approach describing just the fine and coarse PM? What is the effect of this simplification on its PM predictions?

Reply:

Yes, basically we use two modes, although our definitions of particle-size depend a little on the compound. The present version of model is mainly designed to calculate PM_{10} and $\mathrm{PM}_{2.5}$ mass closure (i.e. concentrations and chemical composition), which over the last decade has been the highest priority within the EMEP/LRTAP Convention framework. A research version of the EMEP model exists (Tsyro 2008), which distributes aerosols between four size bins and accounts for aerosol dynamics processes. In our earlier works, PM calculations with those two models were compared. Then it was shown that the two mode (i.e. fine and coarse) representation of aerosol size distribution was a sound approximation for the given purpose.

Finally, an important point is that extensive comparison with measurements of particulate matter has been a feature of EMEP models over the last three decades. Indeed, one of the main reasons for maintaining the extensive EMEP measurement network (Tørseth et al. 2012) is to provide data against which these models are evaluated. In comparison after comparison (e.g. Fagerli and Aas 2008, Simpson et al. 2006b, or EMEP status reports over many years) the EMEP model has been shown to perform quite well against measured PM mass.

We have added some words to the above effect in the revised manuscript.

2.5 Referee:

The new EMEP model uses the MARS thermodynamics model. However, this does not treat sodium chloride and dust components. This should have a significant impact on fine and coarse PM predictions (e.g., partitioning of nitrate) in coastal areas and areas affected by dust. It is not clear how coarse nitrate is treated.

Reply:

Yes, this is partially a weakness of MARS, especially in areas where sea-salt (near coasts) and dust are important, but it should be recognised that there are also significant uncertainties with other approaches. Equilibrium approaches such as MARS (or EQSAM) are probably most appropriate for fine-aerosol, and it is only for such aerosol that we use MARS. In principal we would like to use a kinetic (rather than equilibrium) approach for coarse nitrate formation, with explicit reactions of for example HNO_3 with with NaCl or dust. We have indeed been exploring such reactions, but this is ongoing work. Apart from the difficulty of predicting such components, there are also large gaps in our scientific understanding of nitrate composition - there are hardly any measurements of coarse-mode nitrate to compare against for example.

Further, PM water is now calculated with MARS. Due to MARS's formulation, PM water content is calculated for sulphate, nitrate and ammonium aerosol components, but not for sea salt and mineral dust components. Therefore calculated PM water is expected to be under-predicted over seas and coast areas, where sea salt contributes considerably to PM. The effect of not accounting for mineral components is, however, anticipated to be smaller due to their smaller solubility compared to sea salt.

The above remarks have been added to the revised manuscript.

2.6 Referee:

Despite the recent studies showing that POA is semi-volatile, the current version of the model treats it as non-volatile. This should lead to serious over-predictions of the POA in areas affected by primary anthropogenic emissions based on the available Aerosol Mass Spectrometer measurements.

Reply:

We agree with the reviewer that primary organic aerosol emissions consist of components of varying volatility and for a more realistic modelling of POA this should in principal be taken into account, e.g., by using a VBS-approach. In high-emission areas the non-volatile assumption for POA is likely going to lead to some overestimation compared to a model that allows evaporation of some of the initially emitted POA. Indeed, in the VBS experiments documented in Bergström et al. (2012) we do include such effects.

However, there are two main reasons why we choose to use nonvolatile POA emissions in the ‘standard’ EMEP model code (that used for policy-associated runs):

- The volatility distribution of POA is poorly known; the amount of semi-volatility OC emissions is probably substantial, but so far we have only a very limited number of (American) studies with which to estimate this contribution.
- Official European emission inventories used for policy modelling consist of PM compounds which are assumed to be inert - no consideration of volatility is made in either the PM or VOC inventories. For policy modelling it is necessary to keep these POA and VOC emission totals the same as in the official emission inventories.

One should also remember that there are many uncertainties in all parts of the OM modelling. The volatility question is important, but we believe not the biggest source of uncertainty in this difficult topic.

The EMEP model can in fact treat POA volatility in VBS-schemes, as explored in detail by Bergström et al. (2012). In order to address the referee’s comment, we have used the schemes presented by Bergström et al. 2012 to compare model versions with and without this emissions-volatility assumption. We find that total OM2.5 concentrations over most of the European land-area are 10-20% higher if we use volatility-based emissions. The biggest effects are found over Paris, where we obtain more than 40% higher OM2.5 with the non-volatile assumption. We will include a Figure in the Supplementary information to illustrate these calculations.

2.7 Referee:

There is little description of the aerosol-cloud interactions especially given the limited size resolution in the aerosol module. How is the initial cloud composition determined? What happens when a cloud evaporates? How is the effect of sea-salt and dust on the cloud pH taken into account?

Reply:

The aerosol-cloud interactions in the EMEP model are very simplified. We make no assumptions about initial cloud composition. Ions (e.g. sulphate) formed in cloud are simply assigned to the airborne phase once a cloud evaporates. Cloud pH is estimated using the concentrations of the acids and bases including the buffering by bi-carbonate (through CO_2);

$$[\mathrm{H}^+] = 2[\mathrm{SO}_4^2 -] + 2[\mathrm{SO}_3^2 -] + [\mathrm{HSO}_3^-] + [\mathrm{NO}_3^-] + [\mathrm{HCO}_3^-] - [\mathrm{NH}_4^+]$$

The effect of sea-salt and dust on the cloud pH is not taken into account. Although this could easily be implemented in the model code, large uncertainties are associated with especially the calculations of dust. In any case, studies over continents (and especially industrial/agricultural areas) show that over land cloud water was dominated by sulphate and nitrate ions and ammonium and hydrogen cations (Aleksic et al. 2009, Aneja and Kim 1993, Li and Aneja 1992). The results suggested that the cloud water acidity may be coming predominantly from sulphate aerosol and less from nitric acid. Therefore we have chosen to omit sea salt and dust from the pH calculations.

Text to clarify this has been added to the manuscript.

2.8 Referee:

The emission fluxes used are derived from annual emission estimates. The monthly and daily distributions of these emissions are quite important and should be included if possible in the supplementary material. Some summary information about the emissions (e.g., annual emissions) could be useful.

Reply:

The monthly and daily factors vary with emission (SNAP) sector and country, so it is not easy to make a simple tabulation. The data files are however available as part of the public-domain release from the www.emep.int.

We have however added a Table of annual emissions, and also added Figures to illustrate the monthly variation found for various emissions sources. These new Figures allow us to compare the anthropogenic emissions against the new BVOC and soil-NO emissions. In order to keep the main manuscript size down, these Figures replace the current Fig. 5 (maps of annual emissions). We have moved these maps to the Supplementary, information.

2.9 Referee:

Summarizing, my recommendation is to shorten the paper considerably (to the point where it may become supplementary material to a future paper) relying on the references where appropriate. The focus should be on what is new in the model (something that is not clear now) and how these new additions affect the results. Some evaluation of these new pieces would be useful. The paper could use a discussion of the strengths and weaknesses of the model compared to the other available CTMs.

Reply:

As noted above, we strongly disagree that this paper can be reduced to a supplementary to another publication. We also know that there is a large interest in the atmospheric science community for a proper documentation of the EMEP model. However, we have added text to make it clear which parts of the model are new, added extra information in some sections (dealing with the newest updates), and shortened some sections. We have also added an expanded Conclusions (now as Discussion and conclusions), in order to highlight the changes, and added comments on some remaining issues with regard to model formulation.

Documenting the impact of each change is beyond the scope of this article as discussed in more detail above. We have, however, added some text concerning other CTMs in the new section Discussion and conclusions.

3 Other changes

We have made a number of small changes to the manuscript to reflect changes in the EMEP model code made during the first few months of 2012, and also to reflect comments made by colleagues. There are:

- The notation style of the model has reverted to the number-based system, not date-based as originally planned. Thus we are documenting rv4.0, not rv2012-06.
- Unfortunately the planned Fagerli et al evaluation paper has had to be postponed, but we refer instead to the papers mentioned above (Sect. 2.2), and extensive evaluations which are readily accessible in EMEP reports.

- The ability to nest the EMEP model for ‘zooming’ model simulations has now been explictly mentioned.
- We modified the meteorological inputs in early 2012 to make use of soil moisture indices instead of volumetric soil water. We have modified the document to reflect this.
- A few references were improved

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