

Reply to questions/comments from Anonymous Referee #1

We thank the referee for a careful review and good comments and questions that will help improve the paper.

Referee Q1:

- Page 9061, the last paragraph, the authors pointed out the two different meteorological drivers lead to difference for PM_{2.5} and worse model results can be expected for the latter one. Can the authors explain more why they didn't use the same meteorological driver over all the years?

Reply Q1:

The earlier PARLAM-PS driver was taken out of operation at the Norwegian Meteorological Institute (met.no) when the year 2006 simulations were finished, and it was replaced by the ECMWF-IFS model. The latter model was used for the years 2006 onwards, and it is only recently that met.no has begun to re-run for years prior to 2006. These earlier data were not available at the time of our study.

Referee Q2:

- Page 9066, Line 26, references are needed to demonstrate that emission and deposition are better known for NO_x.

Reply Q2:

On the emissions side, NO_x emissions comprise mainly the well-defined and relatively easy to measure compounds NO and NO₂, arising from well understood emission sectors such as road traffic and power stations. Emissions of NO_x have been the subject of significant harmonisation efforts within the European region (UNECE and EU) for decades, and are subject to a regular review process (e.g. Reis et al., 2009). As an example, the UK estimates an uncertainty for NO_x emissions of just 7% (as reported by Reis et al, 2009). EC emissions on the other hand are more difficult to measure, and even define, with major sources from hard-to-quantify sectors, such as residential wood burning (Kupiainen and Klimont, 2007, Granier et al. 2011, Bond et al. 2012, and indeed this work). EC emissions have not been the subject of UNECE Protocols, or the EU air pollution process, and such emissions are not reported officially to EMEP. Such emissions are sometimes estimated directly (e.g. Kupiainen and Klimont, 2007), but sometimes also as fractions of PM emissions, as we discuss in Sect. 3.4. Various studies suggest major uncertainties with EC emissions (Simpson et al., 2007, Granier et al, 2011, Bond et al., 2012), as does the re-evaluation of RWC emissions presented in our text.

For deposition the story is more complex. Deposition of gaseous components such as HNO₃ is relatively well known (Fowler et al., 2009). Deposition of NO₂ is somewhat more complicated since emissions of NO_x (mainly as NO) from soil microbes confounds simple deposition estimates. Deposition rates of all particles, whether nitrate or EC, are very uncertain (ibid.).

To avoid lengthening the current manuscript, we simply added references to some of the papers mentioned above:

Emissions and deposition are expected to be better known for NO_x than for EC (e.g. Granier et al., 2011, Reis et al., 2009, Simpson et al., 2007, Fowler et al., 2009) so if the model works well for NO₂ at a given site it is likely that points (ii) and (iii) are relatively well modelled.

References:

Fowler, D., Pilegaard, K., Sutton, M., Ambus, P., Raivonen, M., Duyzer, J., Simpson, D., Fagerli, H., Fuzzi, S., Schjoerring, J., Granier, C., Neftel, A., Isaksen, I., Laj, P., Maione, M., Monks, P., Burkhardt, J., Daemmgen, U., Neiryneck, J., Personne, E., Wichink-Kruit, R., Butterbach-Bahl, K., Flechard, C., Tuovinen, J., Coyle, M., Gerosa, G., Loubet, B., Altimir, N., Gruenhage, L., Ammann, C., Cieslik, S., Paoletti, E., Mikkelsen, T., Ro-Poulsen, H., Cellier, P., Cape, J., Horváth, L., Loreto, F., Niinemets, Ü., Palmer, P., Rinne, J., Misztal, P., Nemitz, E., Nilsson, D., Pryor, S., Gallagher, M., Vesala, T., Skiba, U., Brüeggemann, N., Zechmeister-Boltenstern, S., Williams, J., O'Dowd, C., Facchini, M., de Leeuw, G., Flossman, A., Chaumerliac, N., and Erisman, J.: Atmospheric composition change: Ecosystems-Atmosphere interactions *Atmos. Environ.*, 43, 5193-5267, 2009.

Granier, C., Bessagnet, B., Bond, T., D'Angiola, A., van der Gon, H. D., Frost, G. J., Heil, A., Kaiser, J. W., Kinne, S., Klimont, Z., Kloster, S., Lamarque, J.-F., Liousse, C., Masui, T., Meleux, F., Mieville, A., Ohara, T., Raut, J.-C., Riahi, K., Schultz, M. G., Smith, S. J., Thompson, A., van Aardenne, J., van der Werf, G. R., and van Vuuren, D. P.: Evolution of anthropogenic and biomass burning emissions of air pollutants at global and regional scales during the 1980-2010 period *Climatic Change*, 109, 163-190, 2011.

Kupiainen, K. and Klimont, Z.: Primary emissions of fine carbonaceous particles in Europe, *Atmos. Environ.*, 41, 2156-2170, 2007.

Reis, S., Pinder, R. W., Zhang, M., Lijie, G., and Sutton, M. A.: Reactive nitrogen in atmospheric emission inventories, *Atmos. Chem. Physics*, 9, 7657-7677, 2009.

Referee Q3:

- Page 9069, Line 28, I suggest the authors provide one sentence to briefly summarize the results of EC due to vegetation fires in Birkenes instead of only referring to Section 3.5.

Reply Q3:

We agree that the description of the model results for Birkenes was a bit too short in Section 3.3. We have modified the text slightly and added one sentence, as follows:

Model results for EC_{2.5} at Birkenes (Figs. 4b, 5b, S4) are in fairly good agreement with observations, clearly better than the results for NO₂. The model bias for EC_{2.5} is low (-12 %), the MAE is only 0.05 µg m⁻³ (45 %), and the correlation between model and measurements is reasonably good (r=0.71). Although the results for this site are in general in good agreement with EC measurements, there are some episodes when the model overestimates EC severely; this seems to be mostly due to overestimation of the contribution from some fossil source(s). The Birkenes results are discussed in more detail in Sec. 3.5.

Referee Q4:

- Page 9070, Line 10, references are need to explain the irregular behavior of the Aethalometer due to high amounts of sea salt and high humidity.

Reply Q4:

The original comment in the text was in error. We thank the referee for pointing this out. Neither sea-salt or relative humidity are the most important factors to explain the discrepancy between the two instruments (sea-salt explains only 5% of the variance and RH only 3%, so neither are likely to be the main reason for the discrepancy). The problems with the Aethalometer during clean marine conditions at Mace Head are due to its detection limit (71 ng m^{-3} , Arnott et al., 2003).

We will add a reference and replace the sentence with the following:

The irregular behaviour of the Aethalometer during clean conditions is related to its minimum detection limit (71 ng m^{-3} for an hourly average, according to Arnott et al., 2003).

Reference:

Arnott, W. P., H. Moosmüller, P. J. Sheridan, J. A. Ogren, R. Raspert, W. V. Slaton, J. L. Hand, S. M. Kreidenweis, and J. L. Collett Jr., Photoacoustic and filter-based ambient aerosol light absorption measurements: Instrument comparisons and the role of relative humidity, *J. Geophys. Res.*, 108(D1), 4034, doi:10.1029/2002JD002165, 2003.

Referee Q5:

- Page 9072, Line 10 – 14, could the authors provide for details about the Norwegian and Swedish measurement protocols, such as some references if possible? I understand that cooling would results in more condensable organics to be captured. However, dilution can cause the evaporation of semi-volatile organic carbon, which leads to less condensable organics to be captured.

Reply Q5:

Essentially the difference between the Norwegian and Swedish measurement protocols is that the latter only considers solid particles. This is a methodology consisting of sampling of particles on a heated filter, through a probe, from undiluted flue gas in the chimney at gas temperatures of e.g. 160°C (German norms, VDI) or 120°C (US EPA; EPA Method 5; <http://www.epa.gov/ttnemc01/methods/method5.html>). The Norwegian method includes dilution of the sample in a dilution tunnel (DT) and thereby measures solid particles as well as condensable particles. (Haakonson and Kvingedal, 2001, and references therein; Sternhufvud et al., 2004). It will always give higher values than sampling only solid particles, however, considerable variation in methods is possible as dilution ratios as well as cooling temperatures may vary between methods. The Norwegian standard NS 3058-2 (Norsk Standard, 1994) describes sampling of filterable particles in a dilution tunnel with a filter holder gas temperature at $< 35^{\circ}\text{C}$. An overview of the different emission factor values and origin is given chapter 4 of a report by the Nordic Council of Ministers (Sternhufvud et al., 2004). Elemental carbon will be present in the solid particles only. Therefore, the *fraction* of EC present in PM from wood combustion depends on the sampling methodology; the absolute amount of EC is independent as solid particles are included in both methodologies.

We will modify the text in Chapter 3.4 to include the above description and references to the revised version of the paper.

References:

Haakonson G. and Kvingedal E., 2001. Utslipp til luft fra vedfyring i Norge. Utslippsfaktorer, ildstedsbestand og fyringsvaner. Statistisk sentralbyrå - Statistics Norway, 2001/36. Available at http://www.ssb.no/a/publikasjoner/pdf/rapp_200136/rapp_200136.pdf

Sternhufvud C., Karvosenoja N., Illerup J., Kindbom K., Lükewille A., Johansson M., and Jensen D.: Particulate matter emissions and abatement options in residential wood burning in the Nordic Countries, ANP 2004:735, Nordic Council of Ministers, Copenhagen 2004. Available at, e.g.: http://www.norden.org/en/publications/publikationer/2004-735/at_download/publicationfile

Norsk Standard (1994) Norwegian Standard for Enclosed wood heaters; NS 3058-2; available at <http://www.standard.no/en/>.

Referee Q6:

- Supplemental Information, Figure S2- S8, line plots may better show the time variation of modeled and observed data than scatter plot, just as what the authors did for Figure S1.

Reply Q6:

We agree that it is somewhat inconsistent to use a line plot for Figure S1 and showing individual data points in the other time series plots (without connecting) lines. The reason for choosing a line plot for Melpitz is that the number of measurements is so large that individual points would not be visible. Each point in the different plots (Figure S2-S8) represents one measurement (that can be variable in duration even for an individual site, such as alternating daily and 6-daily-measurements) and the corresponding model calculated concentrations (for the same time period, using hourly model resolution). At some measurement sites there are numerous gaps in the measurement data (missing or non-valid measurements). For these reasons we prefer plots that show the individual data points rather than line plots.