Answer to referee comments from Referee#1 regarding:
Particulate emissions from residential wood combustion in Europe – Revised estimates and an evaluation

Hugo Denier van der Gon (1)*, Robert Bergström (2,3), Christos Fountoukis (4), Christer Johansson (5), Spyros N. Pandis (4,6), David Simpson (7,8), and Antoon Visschedijk (1)

We are glad that the referee found the paper interesting, well-written and recommends it for publication. We thank the referee for useful comments for improving the manuscript.

Specific Comments:

1) I would like to see some discussion of how well the CTM models simulate the boundary layer during cold calm night time conditions when residential wood combustion emissions tend to be maximum. Do the CTM's overestimate or underestimate the PM concentrations in these cases?

Answer:

This is a good point. It is difficult to model the boundary layer height accurately in large scale CTMs and there is a risk that the models will underestimate PM from residential wood combustion (and other local, low stack-height sources) during calm, cold nights with strong temperature inversions. If the model overestimates the mixing height during these conditions (which is very likely) the PM concentrations due to local residential wood combustion will be underestimated close to the source.

However, the turbulence parameterization and calculation of the atmospheric boundary layer (ABL) height in the EMEP MSC-W model were updated a few years ago (Jeričević et al., 2010) and evaluation against radiosound data, and data from the Cabauw tower, showed that the EMEP model is able to reproduce spatial and temporal mixing height variability fairly well.

Since measurements of the ABL are usually not available for the same sites (and time periods) as the PM concentrations we cannot directly compare the model bias for PM in the present study to observed ABL. Instead we have compared the model bias for OC to the modelled mixing height (H_{mix}) and air temperature at 2m height (T_{2m}). This may give some indication if there are general problems with under or overestimation in the model during cold and stable conditions.

The correlation between the model bias (for OC) and H_{mix} or T_{2m} varies between the six different sites included in the 2007-2009 evaluation of the EMEP MSC-W model. When looking at all (winter-half year) data from all six sites (Fig. A1), there is a tendency that the model underestimates OC for periods with low model H_{mix}, when using the old emission inventory for RWC (but the correlation between OC-bias and H_{mix} is fairly low, R^2=0.084). When using the updated RWC emission inventory (TNO-newRWC) the underestimation at low modelled H_{mix} decrease markedly and the correlation between the OC-bias and H_{mix} drops to a very low value (R^2=0.017, see Fig. A2).

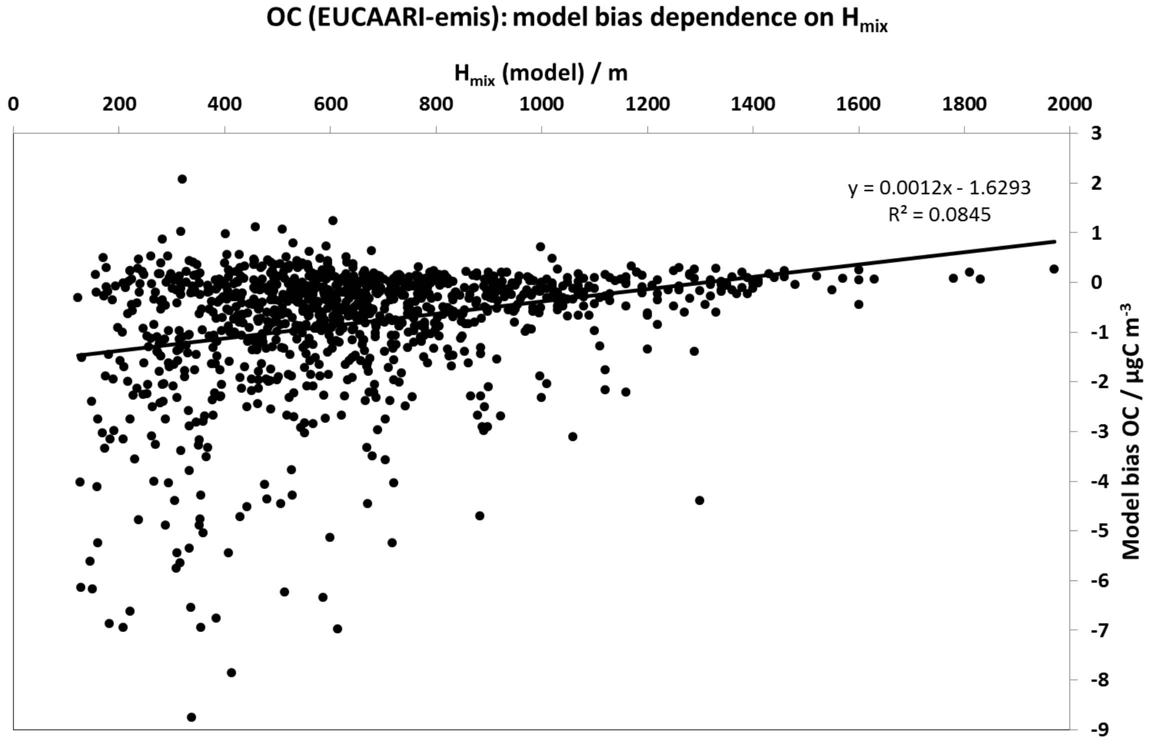

Figure A1. Scatter plot of model bias for particulate organic carbon (OC) concentrations with the old (EUCAARI) emission inventory for residential combustion [model OC - measured OC] and modelled boundary layer height (H_{mix}) using winter half-year (Nov-Apr) data from 2007-2009 from the six stations Hyytiälä, Aspvreten, Vavihill, Melpitz, Overtoom and Birkenes (see manuscript for further details).

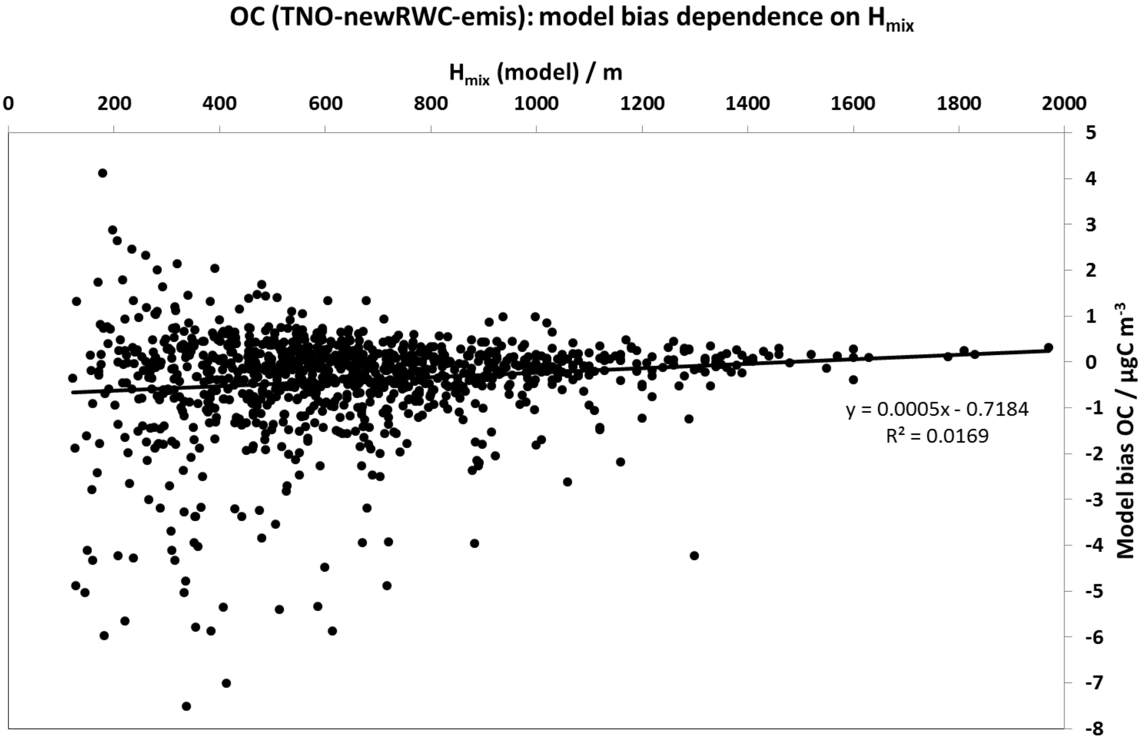

Figure A2. As Fig. A1 but model OC bias based on simulation with the new (TNO-new RWC) emission inventory.

The correlation between the model bias for OC and near surface temperatures is close to zero, both with the old and with the revised RWC emission inventory (see Fig. A3 and A4). So our final answer to referee 1 on this point is that at least for total OC, the EMEP MSC-W model does not seem to produce significantly worse results at low temperature periods than during milder conditions. Since OC is by far the dominant component in PM from RWC, this conclusion holds for PM2.5 or PM10 as well.

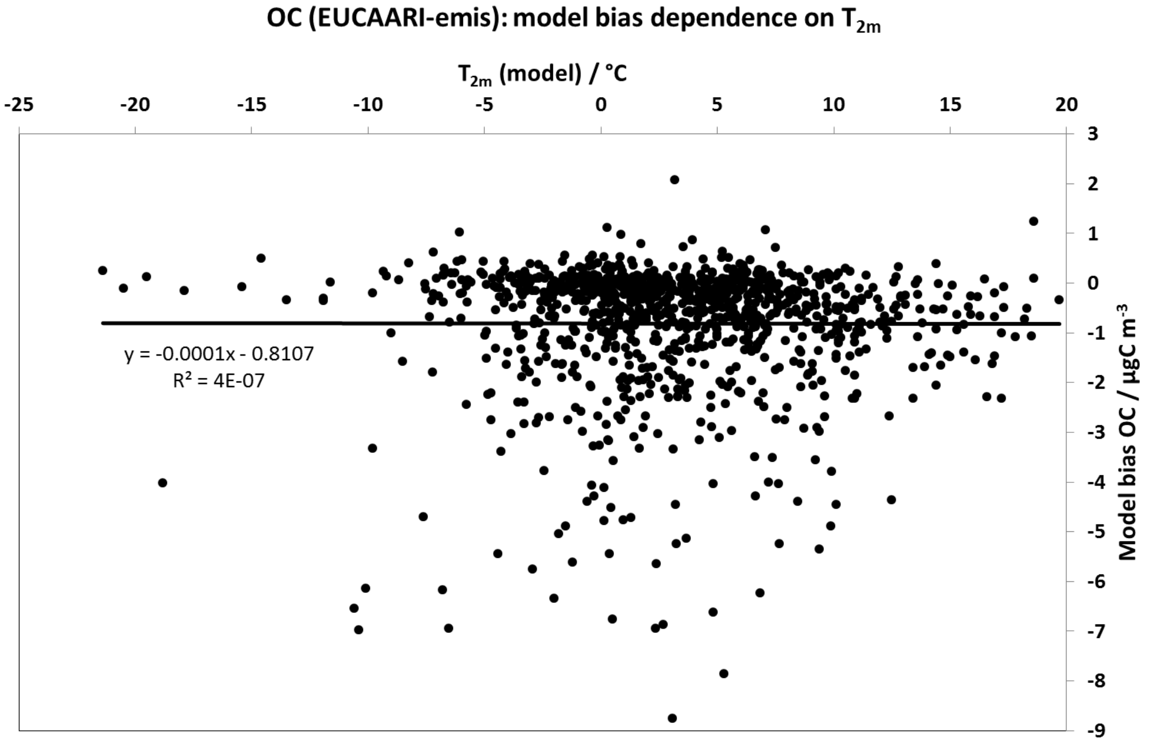

Figure A3. As Fig. A1 but plotting model OC bias against modelled 2m-temperature (T_{2m}).

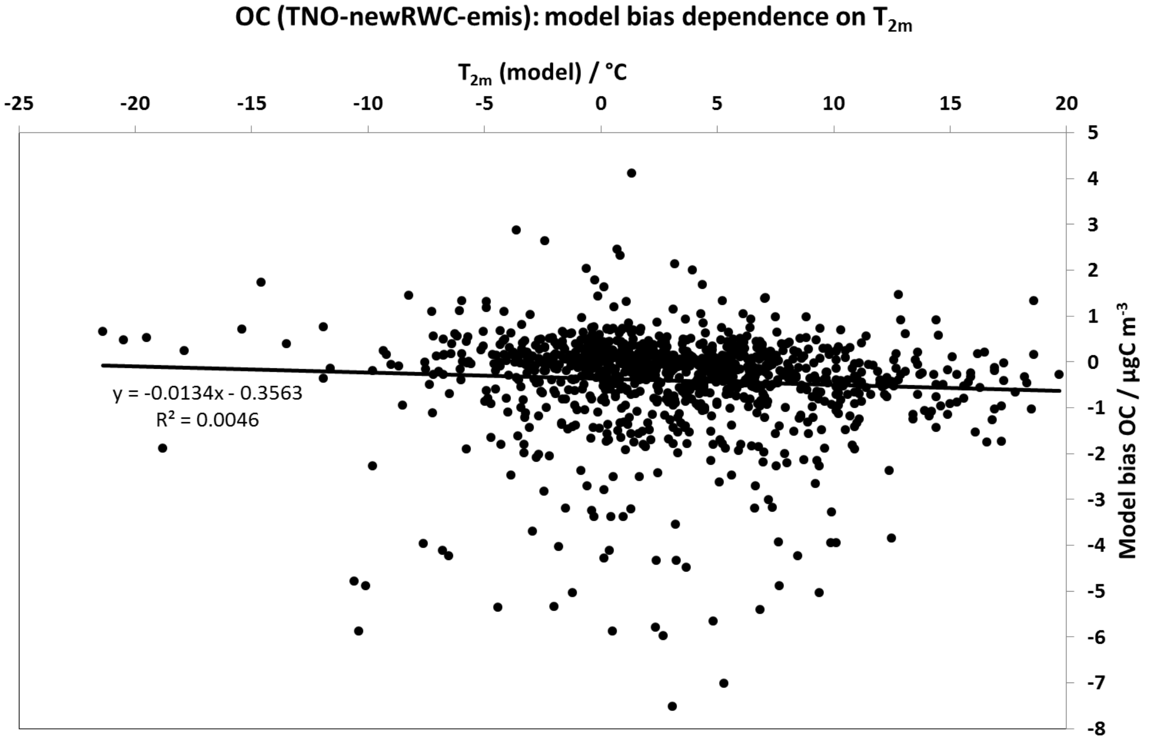

Figure A4. As Fig. A3 but model OC bias based on simulation with the new (TNO-new RWC) emission inventory.

Reference: Jeričević, A., Kraljević, L., Grisogono, B., Fagerli, H., and Večenaj, Ž.: Parameterization of vertical diffusion and the atmospheric boundary layer height determination in the EMEP model, Atmos. Chem. Phys., 10, 341–364, doi:10.5194/acp-10-341-2010, 2010.

We will add the above discussion to the Supplement of the paper and add the following text to the manuscript (in Section 4.2):

It is difficult to model the boundary layer height accurately in large scale CTMs and there is a risk that the models will underestimate PM from residential wood combustion (and other local, low stack-height sources) during calm, cold nights with strong temperature inversions. However, using the EMEP MSC-W model we find essentially no correlation between either mixing height or temperature and model bias for OC (see Supplementary information).

2) The abstract is a bit vague when referencing the different emission inventories (i.e. use of the word “new” somewhat overused in a “new inventory”. Perhaps a better label might be employed and referenced in the abstract.

Answer:

Probably a better phrasing would be “revised” since we do not start from scratch and all other sources were kept constant. In the abstract and the discussion text we will replace “new” with “revised” and will introduce “TNO_new_RWC” as the abbreviation of the TNO emission inventory with revised emission estimates for residential wood combustion. We prefer to keep this abbreviation as it is more compact, remaking all figures is additional work but mostly to avoid confusion since some other researchers / papers already refer to the new-RWC using the ACPD reference. Completely removing this abbreviation might suggest this is yet again another inventory (e.g. a “revision” of the “new” inventory). We hope this is a satisfactory solution to the referee.

Technical Comments:

1) Wood use factor units are sometimes referred to as GJ inhabitant-1 or GJ person -1 In the article and supplement. Use consistent units throughout.

Answer:

We will change to consistent units (GJ person^{-1}) in both the MS and the supplementary material.

2) The last sentence in the conclusions section "For a global assessment we would have to more carefully study the origin of emissions factors used, but global OA emissions from biofuel use could also increase significantly if condensable PM is fully taken into account." Does not add much to the article, is a bit confusing and seems like it doesn't belong to the rest of the paper. Perhaps it might be removed.

Answer:

We will remove this sentence from the conclusions. We also slightly modified the final sentence in the abstract:

Old:

"Since usage of biofuels, such as wood, in small combustion units is a globally significant source, this insight may also dramatically change global estimates of organic aerosol emissions."

New:

"Since usage of biofuels in small combustion units is a globally significant source, the findings presented here are also relevant for regions outside of Europe."

Answer to referee comments from Referee#2 regarding:
Particulate emissions from residential wood combustion in Europe – Revised estimates and an evaluation

Hugo Denier van der Gon (1)*, Robert Bergström (2,3), Christos Fountoukis (4), Christer Johansson (5), Spyros N. Pandis (4,6), David Simpson (7,8), and Antoon Visschedijk (1)

We are glad that the referee found the paper well-written and of interest for ACP readers. We thank the referee for useful comments for improving the manuscript.

General Comments:

1) The authors mention several times the formation of organic aerosol due to condensation of semi-volatile organics upon cooling of the flue gas. When hot flue gas is emitted from the chimney there are to simultaneous processes: cooling and dilution. Where cooling will lead to condensation the dilution will lead to evaporation. Although dilution is mentioned briefly separately (P. 31723 L. 25), a brief discussion of these counteracting processes should be included.

Answer:

The referee is correct that cooling and dilution have an opposite effect on the amount of particulate OC in the atmosphere. At the same time there is confusion because the “dilution” of flue gases coming out of the chimney results effectively in cooling. As the referee notes, flue gases coming out of the chimney are never only cooled, the cooling and dilution goes together. So, what we try to address in our paper is the net effect of the cooling and dilution immediately after exiting the chimney or stack. It is <u>not</u> the effect of aging (which causes a change in volatility of organic vapours, and if these subsequently condense form secondary organic aerosol, SOA) or evaporation under the influence of further dilution or temperature change in the ambient atmosphere (e.g. from day to night). This we assume to be part of the model set-up, for example the VBS approach. So, the condensable OA that we calculate using or DT emission factors does initially not belong in the category of SOA because it does not involve aging or chemical transformations for them to become OA. We refer to these emissions as condensable (organic) PM and will argue that they should be seen as part of the primary PM emissions. Since they are condensable they may however later evaporate, undergo transformations etc. and thus become SOA. This process is covered in the present work since both AQ models used in this study simulate the cooling (by calculating the equilibrium between the organic vapors and particulate matter) at the temperature of the grid cell at the time of emission and the dilution (by mixing the emissions with the rest of the pollutants in the corresponding grid cell initially and then following their evolution as they are transported in the modeling domain).

We will integrate the above explanation in the introduction in connection with the section where dilution and VBS is now discussed (bottom of page 31723)

2) As mentioned in the manuscript, emission factors depend on burner type, operation and sampling method. Secondary organic aerosol (SOA) has been produced from the emissions of different types of burners under different conditions (Hennigan et al., 2011; Grieshop et al., 2009; Heringa et al., 2011). However, the volatility distribution of the organic emissions can vary as well as no SOA formation was observed for a pellet burner. It would be nice if the authors would touch on this as well.

Answer: The volatility distribution of the organic emissions will likely vary very much, both between different fuel and burner types and between different operation conditions/practices as is supported by the references suggested by the referee. To use a single volatility distribution for all types of residential biomass combustion is a simplification. We will point this out in the revised version of the manuscript. In the introduction, we will now also refer to 2 of the 3 additional studies suggested, [we will not include Hennigan et al. (2011), although a good paper, as it discusses open burning of biomass which is different from the burning of logs and pellets for heating which is the focus of our paper].

Specific Comments:

P. 31721: Condensation due to cooling is mentioned in the abstract whereas SOA formation is not. Both processes have an influence on OA concentrations (as described in 3.3).

A: We will change the sentence: “This suggests that primary organic aerosol emission inventories need to be revised to include the semivolatile OA that is formed almost instantaneously due to cooling of the flue gas or exhaust.”

to

“This suggests that primary organic aerosol emission inventories need to be revised to include the semivolatile OA that is formed almost instantaneously due to cooling of the flue gas or exhaust and can form secondary organic aerosol (SOA) after chemical processing.”

P. 31722, ln. 28: Please define the abbreviation EEA.
A: The abbreviation will be defined (European Environment Agency).

P. 31724, ln. 15: Please define RWC.
A: The abbreviation will be defined (residential wood combustion).

P. 31730, ln. 19: SP and DT were already defined above.
A: This will be corrected.

P. 31730, ln. 26: It would be nice to include the emission factors used by EUCAARI for comparison purposes (e.g. as additional column in table 2).
A: Unfortunately this is not possible. The revised RWC emission inventory is a “bottom-up” inventory using the same emission factors for each type of application for each country (as presented in Table 2). The EUCAARI inventory, as explained in more detail in section 2.1.1 was derived from the GAINS model (Klimont et al., 2002; Kupiainen and Klimont, 2004, 2007). As part of the development of GAINS, the responsible team at IIASA conducts “country consultations”. Such consultations may, and often do, result in country-specific adjustments of the emissions factor for a particular process or activity. This means that in the end quite a complex mixture of country-specific emission factors is used which cannot be simplified into an additional column in Table 2. The country consultation process is mentioned in the final paragraph of section 2.1.1. For more detail we refer to Kupiainen and Klimont (2007). The effect of country specific data is also seen when comparing for example the ratio between the EUCAARI and the revised RWC inventory by country as shown in Fig. 3 and Fig. 5. In the revision of the MS we will, next to the current discussion of Table 2, make a clear reference to Kupiainen and Klimont (2007) and the Table 2 in their paper which gives ranges of the emission factors they used but (to make the point again) these ranges originate partly from different methods that countries use and cannot be directly compared with the solid particles (SP) and dilution tunnel (DT) emission factors as presented in our Table 2.

P. 31735, ln. 15/17: VBS has been defined before, SOA was not.

A: This will be corrected.

P. 31741: Some abbreviations are defined again some are not. Please make it consistent.

A: We will make this consistent by removing all repetitions.

Fig. 4: The lower end of the scale disappears in the color legend.

A: Thank you for spotting this. We corrected the legend in the graph to solve the problem (see below).

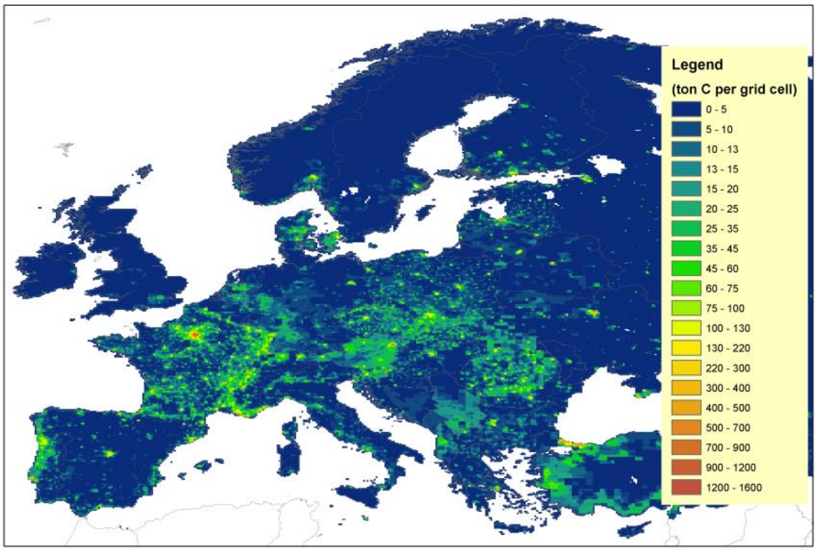

Fig. 6: The figure is really hard to read because of its size.

A: We will try to get the Figure set in larger format in the ACP version of the article. For this we will discuss with the editorial office of ACP. Fig 6 could possibly be printed in landscape format.

Fig. 9: Give a description of the bars (measured range / 95% / . . .).

A: The description of the red bars in the Figure caption will be updated as follows:

Red bars show the measurement-estimated bbOC range (lower limit: 5.5 × the measured levoglucosan concentration, upper limit: 14 × levoglucosan; the estimated range for the bbOC/levoglucosan ratio, 5.5-14, is taken from Szidat et al., 2009).

To be included in the introduction and reference list:

Grieshop, A. P., Logue, J. M., Donahue, N. M., and Robinson, A. L.: Laboratory investigation of photochemical oxidation of organic aerosol from wood fires 1: measurement and simulation of organic aerosol evolution, Atmos. Chem. Phys., 9, 1263-1277, 2009.

Heringa, M. F., DeCarlo, P. F., Chirico, R., Tritscher, T., Dommen, J., Weingartner, E., Richter, R., Wehrle, G., Prévôt, A. S. H., and Baltensperger, U.: Investigations of primary and secondary particulate matter of different wood combustion appliances with a high-resolution time-of-flight aerosol mass spectrometer, Atmos. Chem. Phys., 11, 5945-5957, doi:10.5194/acp-11-5945-2011, 2011.

Particulate emissions from residential wood combustion in Europe – Revised estimates and an evaluation

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Abstract

Comment [HD1]: Abstract is revised

Currently residential wood combustion (RWC) is increasing in Europe because of rising fossil fuel prices but also due to climate change mitigation policies. However, especially in small-scale applications, RWC may cause high emissions of particulate matter (PM). Recently we have developed a new high-resolution (7 × 7 km) anthropogenic carbonaceous aerosol emission inventory for Europe. The inventory indicated that about half of the total $PM_{2.5}$ emission in Europe is carbonaceous aerosol and identified RWC as the largest organic aerosol source in Europe. The inventory was partly based on national reported PM emissions. Use of this organic aerosol inventory as input for two Chemical Transport Models (CTMs), PMCAMx and EMEP MSC-W, revealed major underestimations of organic aerosol in winter time, especially for regions dominated by RWC. Interestingly, this was not universal but appeared to differ by country.

In the present study we constructed a revised bottom-up emission inventory for RWC accounting for the semi-volatile components of the emissions. The revised RWC emissions are higher than those in the previous inventory by a factor of 2 – 3 but with substantial inter-country variation. The new emission inventory served as input for the CTMs and a substantially improved agreement between measured and predicted organic aerosol was found. The revised RWC inventory improves the model calculated organic aerosol significantly. Comparisons to Scandinavian source apportionment studies also indicate substantial improvements in the modeled wood-burning component of organic aerosol. This suggests that primary organic aerosol emission inventories need to be revised to include the semivolatile organic aerosol that is formed almost instantaneously due to cooling of the flue gas or exhaust and can form secondary organic aerosol after chemical processing. Since RWC is a key source of fine PM in Europe, a major revision of the emission estimates as proposed here is likely to influence source-receptor matrices and modelled source apportionment. Since usage of biofuels in small combustion units is a globally significant source, the findings presented here are also relevant for regions outside of Europe.

1. Introduction

There is growing evidence of associations of adverse health effects with particles originating from combustion sources (e.g., Hoek et al., 2002; WHO, 2005). Particulate matter (PM) emissions from combustion sources, such as traffic and residential combustion, contain a large fraction of carbonaceous material, consisting of elemental carbon (EC) and organic carbon (OC). Carbonaceous aerosol is predominantly present in the sub-micron size fraction (Echalar et al., 1998; Hitzenberger and Tohno, 2001). In the last two decades a growing number of studies highlight the importance of this carbonaceous fine fraction of PM in relation to adverse health effects (Hoek et al., 2002; Miller et al., 2007; Biswas et al., 2009; Janssen et al., 2011). Moreover, atmospheric fine particulate matter ($PM_{2.5}$) also has climate-forcing impacts, either contributing to or offsetting the warming effects of greenhouse gases (Kiehl and Briegleb, 1993; Hansen and Sato, 2001). In particular, black carbon (BC) has been identified as an important contributor to radiative heating of the atmosphere (Myhre et al., 1998; Jacobson, 2001; Bond et al., 2013). Organic aerosol (OA), which is always emitted along with BC, may act to offset some of the global warming impact of BC emissions (Hansen and Sato, 2001; Bond et al., 2013). So, both from a climate and an air quality and health impact perspective there is a need for size-resolved emission inventories of carbonaceous aerosols.

There have been a number of efforts to develop emission inventories for EC and OC (e.g. Bond et al., 2004, Schaap et al., 2004; Kupiainen and Klimont, 2007; Juncker and Liousse, 2008). However, these inventories are for the year 2000 or earlier, and not gridded on a resolution that facilitates detailed comparison of model-predicted and measured concentrations with specific source sectors, like separating the coal and wood fired residential combustion. An advantage of a more recent base year is that it is closer to years with detailed measurements, including source apportionment studies with organic molecules that can act as a tracer for certain processes, such as levoglucosan for wood combustion (Simoneit et al., 1999). Emissions of particulate matter or carbonaceous aerosols are notoriously uncertain. The European Environment Agency (EEA, 2013a) concluded in its European Union emission inventory report 1990–2011 that as only a third of the Member States report on their uncertainty in emissions, it was not possible to evaluate uncertainty overall at the EU level. The countries that do report use quite different methodologies. The most advanced, like the UK, evaluate uncertainty by carrying out a Monte Carlo uncertainty assessment (EEA, 2013a). Quantitative estimates of the uncertainties in the UK emission inventory were based on calculations using a direct simulation technique. For PM_{10} this resulted in an uncertainty of -20% to + 50% in the UK. Other countries, however, report different values sometimes well exceeding 100% (EEA, 2013a). Moreover, this recent European emission inventory report also highlights that residential combustion is now the most important category for $PM_{2.5}$ emissions, making up 44% of the total $PM_{2.5}$ emissions in the EU (EEA, 2013a and Figure 2.7 therein). The origin of the uncertainty is only partly an instrument measurement uncertainty. More important are the conditions under which the emission factor measurements take place. Whereas the instrument to do the measurement may be defined or prescribed, the exact conditions of sampling and sample treatment are often not well defined but may have a great impact on the total measured PM or aerosol. Key environmental conditions include humidity, temperature, and dilution ratio during sampling (e.g. Lipsky and Robinson, 2006; Nussbaumer et al., 2008a).

Due to the importance of PM for both air quality and climate impacts there has been an increased interest in developing models that can describe PM concentrations in the atmosphere under present conditions and predict the impact of emission changes. A major challenge for chemical transport models (CTMs) is to simulate OA. The ability to model OA is crucial for predicting the total concentration of $PM_{2.5}$ in the lower atmosphere since a large fraction of fine PM is organic material (typically 20-90%, Kanakidou et al., 2005; Jimenez et al., 2009). Current understanding of organic aerosol emissions suggests that more than half of the organic matter emitted from transportation sources and wood combustion actually evaporates as it is diluted in the atmosphere (Robinson et al., 2007). The resulting organic vapors can be oxidized in the gas phase and recondense forming oxygenated organic aerosol. Further oxidation ("chemical aging") of semi and intermediate volatility organic compounds (SVOCs and IVOCs) can be important (Robinson et al., 2007) and has been previously neglected in most modelling efforts. The volatility basis set (VBS) framework has been developed to describe the OA formation and atmospheric processing and is now used by a number of CTMs (Fountoukis et al., 2011; Bergström et al., 2012; Zhang et al., 2013).

In this paper we briefly describe the construction of the EUCAARI inventory – a high resolution emission inventory of EC and OA for UNECE-Europe for the year 2005. UNECE-Europe includes the EU27 countries and Albania, Armenia, Azerbaijan, Belarus, Bosnia Herzegovina, Croatia, Georgia, Moldova, Macedonia, Norway, Russian Federation, Serbia and Montenegro, Switzerland, Turkey and Ukraine. An important characteristic of this inventory was the update of activity data for residential wood combustion and an improved spatial distribution. The EUCAARI inventory was used as input for two CTMs, PMCAMx and the EMEP MSC-W model (Fountoukis et al., 2011; Bergström et al., 2012). The evaluation of the model results revealed a significant underestimation of OA in winter time, especially for regions dominated by residential wood combustion (RWC). These results are consistent with an earlier study with the EMEP model (Simpson et al., 2007) comparing model predictions to measurements of the wood-burning tracer levoglucosan and other source apportionment data from the EU CARBOSOL project (Gelencsér et al., 2007). The study clearly demonstrated that almost all of the OA measured during winter-time at low elevation sites (K-Puszta in Hungary and Aveiro in Portugal) in the CARBOSOL project could be attributed to wood-burning emissions. The authors concluded that wood-burning contributions were much higher than could be accounted for with the emission inventory available at the time.

These findings were the motivation to revisit the EUCAARI EC/OC inventory, especially critically looking at the emission factors used. While the VBS framework deals with transformation and fate of organic aerosol due to evaporation, aging and transport, this framework does not describe the changes in condensable PM emissions immediately at the point of emission (chimney or exhaust). Here two processes are important; cooling and dilution, which have an opposite effect on the amount of particulate OC in the atmosphere. However, the “dilution”, of flue gases coming out of the chimney, itself leads to cooling. Flue gases coming out of the chimney are never only cooled, the cooling and dilution goes together. In this paper we address the net effect on emission factors for RWC, of the cooling and dilution *immediately after exiting the chimney or stack*, leading to a revised emission inventory. The improved inventory (TNO-newRWC) using another type of emission factors for residential wood combustion was tested in two CTMs and evaluated using available measurement data.

2. Carbonaceous particulate matter emissions in Europe

Air emissions inventories are fundamental components of air quality management systems used to develop and evaluate emission reduction scenarios. A transparent and consistent emission inventory is a prerequisite for (predictive) modelling of air quality. The combination of air emissions inventories, source sector contributions and predictive modelling of air quality are all needed to provide regulators, industry and the public with access to the best possible data to make informed decisions on how to improve air quality.

2.1 The EUCAARI EC and OC inventory

Recently, improvements were made in the spatial distribution of European emission data, as well as in completeness of country emissions in Europe (Pouliot et al., 2012; Kuenen et al 2011). The spatial distribution used in the present study is a 1/8 º × 1/16º longitude – latitude grid. The area domain is Europe from -10° to +60° Lon and +35° to +70° Lat (excluding Kazakhstan and the African continent, but including Turkey). The set of gridding tools used in this study is described in Denier van der Gon et al. (2010). The exception is residential wood combustion for which a new distribution map has been compiled (see Sect. 2.3.1). For gridding a distinction is made between point and area sources. Point source emissions are distributed according to location, capacity, and fuel type (when applicable). Area sources are distributed using distribution maps of proxy data such as population density. For a detailed description of the gridding we refer to Denier van der Gon et al. (2010). The point sources and area sources used to distribute the emissions for individual source categories are presented in the Supplementary material Table S1. The emission inventory database provides the emissions at a detailed level of about 200 sub source categories. Each subcategory was spatially distributed using the most appropriate proxy map and then aggregated to Standardized Nomenclature for Air Pollutants (SNAP) level 1 source categories (Table 1).

2.1.1 Primary PM_{10}, $PM_{2.5}$ and PM_1 Emission Inventory and EC and OC fractions

Size-fractionated EC and OC emission factors (carbonaceous mass per unit of activity) are available only for a limited number of sources and technologies and can vary widely due to different measurement protocols and analytical techniques (Watson et al., 2005). Although a direct calculation of emissions as activity times the EC/OC emission factor would be preferable, this would give widely varying, inconsistent and incomplete results. This problem is tackled by starting from a size-fractionated particulate matter ($PM_{10}/PM_{2.5}/PM_1$) emission inventory, followed by deriving and applying representative size-differentiated EC and OC fractions to obtain the EC and OC emissions in the size classes, <1 μm, 1-2.5 μm, and 2.5-10 μm.

A consistent set of PM_{10}, $PM_{2.5}$ and PM_1 emission data for Europe is obtained from the GAINS (Greenhouse Gas - Air Pollution Interactions and Synergies) model (Klimont et al., 2002; Kupiainen and Klimont, 2004; 2007). GAINS accounts for the effects of technology (such as emission control measures) on PM emissions, which would otherwise be difficult to assess from the EC/OC literature. The detailed source categorization in GAINS enables the use of highly specific EC and OC fractions which increases the accuracy of the final emission inventory. For a description of the relevant GAINS PM emission data used here, we refer to Klimont et al. (2002) and Kupiainen and Klimont (2004; 2007). Further documentation can be found at the IIASA web page (http://www.iiasa.ac.at/). PM_1, $PM_{2.5}$ and PM_{10} emissions by source sector often vary by country in GAINS, due to different degrees of emission control. The size-differentiated PM emission estimates (PM_{10}, $PM_{2.5}$, PM_1) from GAINS have been combined with EC and OC fractions, resulting in EC and OC emission estimates for 230 source categories and the three particle size classes.

Although EC and OC fractions may also vary with control technology, the reviewed EC and OC literature does not allow further technology-dependent fractions of EC and OC. Therefore, EC and OC fractions were assumed to be independent of control technology. Since the absolute PM_1, $PM_{2.5}$ and PM_{10} emission level is control technology dependent, the most important impact of control technology on EC and OC is taken into account in this approach by a corresponding reduction of PM. The used EC and OC fractions were partly based on previous compilations (Streets et al., 2001; Bond et al., 2004; Schaap et al., 2004; Kupiainen and Klimont, 2004; 2007). For the EUCAARI EC and OC inventory, Visschedijk et al. (2009) concentrated on adding new information if available, and estimating the EC and OC fractions when no information was available.

The term EC is often used for measurements based on thermal analysis to indicate the carbon that does not oxidize below a certain temperature. OC refers to the non-carbonate carbonaceous material other than EC. OC content is usually expressed on a carbon mass basis. Full molecular mass (OM=Organic Matter) can be estimated by multiplication with a factor to account for the other, non-C elements present in organic matter like O and N; however, the OM/OC ratio varies (Simon et al., 2011); freshly emitted primary organic aerosol typically have OM/OC varying between about 1.2 – 1.8 (Aiken et al., 2008) and the ratio increase as the aerosol ages (OM/OC ratios of 2.5 have been observed for aged ambient oxygenated organic aerosol in Mexico, (Aiken et al., 2008)). Total carbon (TC) is the sum of EC and OC (C mass-basis).

The IIASA GAINS PM emission data have been subject to a country consultation and review process and therefore for many countries these PM emissions are in line with national reported emission data as available at the EMEP Centre on Emission Inventories and Projections (CEIP) (http://www.ceip.at/). The EUCAARI OA inventory (Fig. 1~~Fig. 1~~) was

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derived from the IIASA GAINS PM emission data base in combination with the EC and OC fractions derived by Visschedijk et al. (2009).

2.2 Residential Wood Combustion in Europe

Wood, woody biomass, and wood pellets are extensively used as fuel in European households. However, reliable fuel wood statistics are difficult to obtain because fuel wood is often non-commercial and falls outside the economic administration. Therefore, fuel wood consumption has been notoriously underestimated in the past. Since combustion of wood is a key source of EC and OC we improved the available wood usage data through a stepwise approach. Specific wood use by country (GJ person^{-1}) was primarily taken from GAINS. Estimates from the International Energy Agency (IEA; 2008) were used when GAINS data were lacking. By comparing industrial and residential use of fuel wood in GAINS we conclude that only the residential use is important on a European scale; industry and power generation both consume less than 1% of the total amount of wood used annually in Europe (IEA, 2008; GAINS, 2009). Moreover, combustion appliances in the residential sector have much higher PM emission factors per unit of fuel. Therefore, our focus is on residential combustion of wood and we neglect its minor use as a fuel in industrial combustion or power generation here.

Grouping the available statistical data resulted in five country cluster averages, based on geographical location and tradition, with wood use varying between 1.6 – 8.6 GJ person^{-1} (Fig. 2). The observed differences between countries and country clusters can be related to the availability of local sources of fuel wood. We define "wood availability" by the geographical intersection (arithmetic product) of population and local fuel wood sources, modelled by overlaying a map of gridded population on $1/16^{o} \times 1/8^{o}$ resolution (from the Center for International Earth Science Information Network, CIESIN; for details see Denier van der Gon et al., 2010) with gridded land cover of woodlands (combination of landcover maps described by Denier van der Gon et al., 2010) on a coarser $0.25^{o} \times 0.5°$ resolution and subsequent summing by country. Based on analysis of reported specific wood use versus expected specific wood use (given a country's estimated average wood availability and cluster average specific consumption) we singled out and corrected wood use for countries where reported usage data substantially differed from expected wood use. In addition, we made default usage estimates based on country cluster averages and wood availability for countries with no reported wood use. Ultimately for the Slovak Republic, Cyprus, Malta, Russia, Ukraine, Moldova and the Caucasus states, specific wood consumption was based on our own estimates of specific consumption instead of using GAINS/IEA data. For documentation of the underlying assumptions we refer to Visschedijk et al. (2009). The estimated residential fuel wood use by country is presented in Fig. 2~~Fig. 2~~. Total wood use in UNECE Europe after reviewing the activity data and gap filling was about 20% higher than the old data set.

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Various types of appliances are used in Europe for residential wood combustion and this has a significant impact on the EC/OC and PM emissions. In this study we adopt the split in appliance types given by Klimont et al. (2002) and Kupiainen and Klimont (2007) who distinguished seven appliance types and provided relative shares of their use in different countries. In terms of emission of particulate matter these technologies are ranked:

Fireplace > Conventional stove > Newer domestic stoves and manual single house boilers > Automatic single house boilers and 50 kW - 100 kW medium boilers > 1 MW - 50 MW Medium boilers.

Especially the fraction of fireplaces and conventional stoves has important implications for the PM/EC/OC emission because of the corresponding relatively high emission factors (Kupiainen and Klimont, 2007). For countries within our domain where no ratios between different appliances were given by Klimont et al. (2002) we used values for neighbouring or comparable countries (see Table S2). For several Eastern European countries the wood usage of fireplaces was reported as 0%, we adjusted this by assuming 5% application in fireplaces (the country cluster average). From the activity data for fuel wood consumption by appliance type by country, it is evident that Western European countries with a relatively high use of fuel wood also have the highest market penetration of more modern combustion equipment.

2.3 The TNO-newRWC emission inventory

The activity data described earlier, in combination with the adjusted allocation of wood by appliance type were used to develop a revised RWC emission inventory by selecting emission factors for each appliance type, independent of the country (Table 2~~Table 2~~). This is a first order approach because it neglects the importance of combustion conditions and "cultural" differences in how to burn wood. Nevertheless it leads to a more transparent and comparable emission inventory.

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Emission factors for wood combustion vary widely even for the same appliance type. This is partly due to the influence of combustion type, fuel parameters and different operation conditions. However, another important factor is the different sampling and measurement protocols or techniques. Nussbaumer et al. (2008 a, b) made a detailed survey and review of the various emission factors in use in Europe, also in relation to the type of measurement technique. A total of 17 institutions from seven countries (Austria, Denmark, Germany, Norway, The Netherlands, Sweden, and Switzerland) participated in the survey and contributed data to the questionnaire. In addition, data for national emission factors were reported or gathered from the literature. Nussbaumer et al. (2008 a, b) describe various sampling methods and the respective emission factors. The most important are filter measurements, measuring only solid particles (SP), and dilution tunnel (DT) measurements, measuring solid particles and condensable organics (or semi-volatile organics). An example of the latter is the Norwegian standard NS 3058-2 which samples filterable particles in a dilution tunnel with a filter holder gas temperature at less than 35°C and at small dilution ratios (DR) of the order 10. Due to the cooling, condensable organic material in the hot flue gas condenses on the filter or the solid particles. The impact of the choice of SP or DT emission factors is large as illustrated in detail in Table 2~~Table 2~~. For example, for conventional woodstoves, one of the most important categories in Europe, the average solid particle emission factor is 150 g GJ^{-1} (range 49–650) whilst the average of the dilution tunnel measurements, that include both solid and condensable particles, is 800 g GJ^{-1} (range 290–1932).. This implies a factor of five difference between the absolute PM emissions depending on the choice to use an SP or DT-based emission factor. National emission factors, used in official reporting, show a considerable range, even if they are of the same type (DT or SP) as is reflected in the range presented in Table 2~~Table 2~~ and documented in detail in Nussbaumer et al. (2008 a, b). In the TNO newRWC emission inventory, the average DT emission factors were used for the respective appliance types (Table 2~~Table 2~~), for all other EC and OC emissions sources the EUCAARI emission values (Visschedijk et al., 2009; Kulmala et al., 2011) remained unchanged; In Fig. 1~~Fig. 1~~ only the sector SNAP 2-wood is different. It should be noted that we revised the primary PM$_{10}$ emissions to be used in the CTMs but as emission factors depend on burner type, operation and sampling method, also the secondary organic aerosol (SOA) produced from the emissions of different types of burners under different conditions will differ (Grieshop et al., 2009; Heringa et al., 2011). These studies showed that the volatility distribution of the organic emissions can vary substantially, both between different fuel and burner types and between different operation conditions/practices. To use a single volatility distribution for organic aerosol emissions for all types of residential biomass combustion as is done here, is a simplification. The result was

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a revised inventory with a consistent approach for residential wood combustion, independent of individual country emission factor choices used for official reporting. A detailed example is presented in Sect. 4.3 A case study: Norway and Sweden~~4.3 A case study: Norway and Sweden~~.

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2.3.1 Spatial distribution

To spatially distribute the emission from residential wood combustion we assumed that within a country the specific fuel wood use per inhabitant is higher in rural regions than in urban areas. The latter have more apartment and high-rise buildings, which often have no wood stoves and/or chimneys. This assumption is confirmed by overlaying gridded urban and rural population with the regional spatial distribution of wood combustion units for Sweden (Segersson, 2008) and the Netherlands (ER, 2008). In both cases, the wood combustion unit distribution was based on chimney sweeps statistics. For the Netherlands, a survey among clients of the wood stove sellers organization was also used. Overall, an urban house is about half as likely to be fitted with a wood combustion unit as a house in a rural environment. A factor of two difference may seem rather low, but this is an average value and it is consistent with data for Germany (Mantau and Sörgel, 2006).

Spatial distribution of wood use will also be influenced by the earlier discussed local wood availability that we derived by spatial analysis of population and woodland distribution. A relationship was derived between the country specific fuel wood use (GAINS/IEA, see Sect. 2.2) and the summed wood availabilities of that country, as discussed in detail in Visschedijk et al. (2009). Thus the population contained in each cell of the population distribution grid was given a weight factor based on the surrounding woodland coverage. Taking local wood availability into account, and differentiating between urban and rural environments, leads to a distribution pattern that significantly deviates from the distribution of total population. Further improvements in the distribution may be feasible by accounting for local factors such as legal restrictions, cultural traditions and the connection of remote areas to energy distribution networks but this has not been attempted within the present study.

3. Chemical Transport Modelling

Two chemical transport models are used in this study, the EMEP MSC-W and the PMCAMx models, both described below. As well as lending more robustness to this study (especially for the modelling of such uncertain components as organic aerosol), these two models have different and complementary strengths. The EMEP model has been evaluated extensively in Europe for many pollutants and across many years (Jonson et al., 2006; Fagerli and Aas, 2008; Aas et al., 2012; Bergström et al., 2012; Genberg et al., 2013). The model is known to work well for compounds where the emissions are well characterised. The EMEP model is readily run for periods of many years, and in this study we will present results from annual simulations. PMCAMx has been widely evaluated in North America, but it has recently been shown to perform well also in Europe (Fountoukis et al., 2011). The model is typically run for shorter periods than EMEP (e.g. one month), and was evaluated against high time resolution (1-hr) measurements. PMCAMx has an advanced aerosol scheme, with full aerosol dynamics and a 10-bin sectional approach.

3.1 The EMEP MSC-W model

The EMEP MSC-W chemical transport model used in this work is based on the 3-D CTM of Berge and Jakobsen (1998), extended with photo-oxidant, inorganic and organic aerosol chemistry (Simpson et al., 2012; Bergström et al., 2012). The model domain used in this study covers the whole of Europe, and includes a large part of the North Atlantic and Arctic areas, with a horizontal resolution of 50 km × 50 km (at latitude 60°N). The model includes 20 vertical layers, using terrain-following coordinates; the lowest layer is about 90 m thick. Meteorological fields are derived from the ECMWF-IFS model (European Centre for Medium Range Weather Forecasting Integrated Forecasting System, http://www.ecmwf.int/en/research/modelling-and-prediction).

The most recent version of the EMEP MSC-W model includes an organic aerosol scheme that uses the volatility basis set (VBS) approach (Donahue et al., 2009; Robinson et al., 2007) described in Sect. 3.3. An extensive sensitivity analysis of this model has been presented by Bergström et al. (2012). In the present study we used an OA scheme with a nine-bin VBS for the primary OA (POA), including semi-volatile and intermediate volatility (IVOC) gases (see Sect. 3.3 and Table 3). The IVOCs are missing in traditional OA and VOC emission inventories and for the standard emission scenario (referred to as EUCAARI) the total emissions of semivolatile POA and IVOCs were assumed to amount to 2.5 times the POA inventory (based on Shrivastava et al., 2008), i.e., an IVOC mass of 1.5 times the POA emissions was added to the total emission input in the model. For the EMEP model simulations that used the revised RWC emissions, with emission factors based on dilution tunnel measurements, a slightly different emission split was applied for the RWC POA. We assumed that the DT methodology captures a larger fraction of the total semivolatile POA and IVOC emissions than traditional inventories (48% for the new DT emissions, compared to 40% for the EUCAARI emissions); the same volatility distribution of the OA emission was used in both cases but for the revised RWC emission inventory total emissions are assumed to be 2.1 times the inventory (compared to the factor 2.5 for EUCAARI emissions).

The EMEP inputs used in the present study is based on Bergström et al. (2012) with a few updates. The most important changes are the following:

- The background concentration of organic aerosol is set to 0.4 µg m^{-3}. Bergström et al. (2012) used a higher OA background concentration (1 µg m^{-3}) but found that this led to overestimations of OA at many sites during some periods.
- Emissions from open biomass fires (including vegetation fires and open agricultural burning) are taken from the “Fire INventory from NCAR version 1.0” (FINNv1, Wiedinmyer et al., 2011).
- Hourly variations of anthropogenic emissions are used (as in Simpson et al., 2012); Bergström et al. (2012) used simple day-night factors.

The organic aerosol emissions from RWC (given as OC-emissions, in carbon-units, in the inventories) are assumed to have an initial OM/OC ratio of 1.7 (based on data from Aiken et al., 2008). Further details about the EMEP OA model setup are given by Bergström et al. (2012).

3.2 The PMCAMx model

PMCAMx (Fountoukis et al., 2011; 2013, Skyllakou et al., 2014) uses the framework of the CAMx air quality model (Environ, 2003) describing the processes of horizontal and vertical advection, horizontal and vertical dispersion, wet and dry deposition, gas-, aqueous- and aerosol-phase chemistry. For the aerosol processes, three detailed aerosol modules are used. The approach of Fahey and Pandis (2001) is used for the simulation of aqueous-phase chemistry. The inorganic aerosol growth is described in Gaydos et al. (2003) and Koo et al. (2003). These aerosol modules use a sectional approach to dynamically track the size evolution of the aerosol mass across 10 size sections ranging from 40 nm to 40 µm. The aerosol species modelled include sulfate, nitrate, ammonium, sodium, chloride, potassium, calcium, magnesium, elemental carbon, primary and secondary organics. The chemical mechanism used in the gas-phase chemistry is based on the SAPRC99 mechanism (Environ, 2003). The version of SAPRC99 used here includes 211 reactions of 56 gases and 18 radicals and has five lumped alkanes, two olefins, two aromatics, isoprene, a lumped monoterpene species, and a lumped sesquiterpene species. In the current version of the model primary organic aerosol in PMCAMx is assumed to be semivolatile using the VBS scheme (see Sect. 3.3). For the inorganics a bulk equilibrium approach is used in which equilibrium is assumed between the bulk inorganic aerosol and gas phase. At a given time step the amount of each species partitioned between the gas and aerosol phase is determined by applying the multicomponent aerosol thermodynamic equilibrium model ISORROPIA-II (Fountoukis and Nenes, 2007) and is then distributed over the aerosol size sections by using weighting factors for each size section based on their surface area (Pandis et al., 1993).

The PMCAMx modelling domain covers a 5400 × 5832 km^2 region in Europe with 36 × 36 km grid resolution and 14 vertical layers covering approximately 7 km with a surface layer thickness of 55 m. PMCAMx was set to perform simulations on a rotated polar stereographic map projection. The necessary inputs to the model include horizontal wind components, vertical diffusivity, temperature, pressure, water vapor, clouds and rainfall all created with the meteorological model WRF (Weather Research and Forecasting). The biogenic emissions were produced by MEGAN (Model of Emissions of Gases and Aerosols from Nature) (Guenther et al., 2006). A marine aerosol emission model (O'Dowd et al., 2008) was also used for the estimation of mass fluxes for both accumulation and coarse mode including an organic fine mode aerosol fraction. The model was successfully evaluated against hourly Aerosol Mass Spectrometry (AMS) data from various stations in Fountoukis et al. (2011) during a photochemically intense period. The model was also used to assess the importance of horizontal grid resolution and the use of high resolution emissions on the predicted fine PM in a European Megacity (Fountoukis et al., 2013).

3.3 The volatility basis set framework

Both the EMEP MSC-W and PMCAMx models employ the volatility basis set (VBS) framework (Donahue et al., 2009; Robinson et al., 2007), using methods similar to those of Lane et al. (2008) and Shrivastava et al. (2008). Secondary organic aerosol (SOA) production from VOCs (Lane et al., 2008) is simulated using four semivolatile surrogate SOA products for each VOC, with 4 volatility bins (effective saturation concentrations C^* of 1, 10, 100 and 1000 $\mu g\ m^{-3}$ at 298 K). The models treat all organic aerosol species in the gas phase (primary and secondary) as chemically reactive. Each reaction with OH radical is assumed to decrease the volatility of the vapor material by a factor of 10 with a small net increase in mass (7.5%) to account for added oxygen (Robinson et al., 2007). The primary OA emissions in the models were distributed by volatility (Table 3~~Table 3~~) using the volatility distributions of

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Shrivastava et al. (2008). This distribution was derived by fitting gas particle partitioning data for diesel exhaust and wood smoke (Lipsky and Robinson, 2006; Shrivastava et al., 2006).

Primary organic gases, formed due to evaporation of semivolatile POA in the atmosphere, are assumed to react with OH radicals reducing their volatility. Chemical aging through gas-phase oxidation of primary OA vapors is modelled using a rate constant of $k = 4 \times 10^{-11}$ cm^3 molec^{-1} s^{-1} (Atkinson and Arey, 2003).

4. Results, evaluation and discussion

The result of the work presented here was a revised RWC emission inventory as well as the use of that emission inventory in two CTMs and subsequent comparison with observational data. These results are described and discussed in the following sections.

4.1 Revised RWC emission data

We constructed a revised bottom-up RWC emission inventory for Europe that also accounted for condensable organic material. Since EC is present as solid particles the change between the EUCAARI and TNO-newRWC for EC is rather small in most countries (Fig. 1~~Fig. 1~~; for a detailed discussion of the EC emissions see Genberg et al., 2013). However for OC the change is large as most OC emissions are condensable PM (Fig. 1~~Fig. 1~~). The revised RWC inventory changed the total primary OC emissions from wood combustion significantly but not in the same way for all countries. This is illustrated in a more detailed comparison for selected countries in Fig. 3~~Fig. 3~~. Note that in this figure the sum of all anthropogenic OC sources are shown but the difference between the two bars for each country is only due to the new assessment of the wood burning OC emissions. The spatial distribution, using the new distribution proxy described in Sect. 2.3.1, of the European anthropogenic OC emissions is presented in Fig. 4~~Fig. 4~~. Important population centres in countries with high wood consumption can be easily recognized such as Oslo, Paris and Moscow, despite the fact that rural homes were allocated a relatively higher wood use. However, a distinct rural pattern can also be seen with for example high emissions in the eastern part of France. To give a spatial impression of the changes in emissions upon using the TNO-newRWC inventory we calculated the ratio of the revised emissions over the EUCAARI emissions (Fig. 5~~Fig. 5~~). The TNO-newRWC emissions are higher than the EUCAARI emissions in all countries except Norway. This is discussed in detail in Sect. 4.3. The methodology of the EUCAARI inventory for RWC was not a consistent bottom-up inventory but partly involved country

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specific data and expert judgements as explained in section 2.1. A direct emission factor comparison with the emission factors used in the revised inventory (Table 2) is not possible. For more details on the different emission factors used in the EUCAARI inventory we refer to the underlying PM emission factors from Kupiainen and Klimont (2007) and more specifically to Table 2 in their paper. Here ranges of the PM emission factors that were used are presented. These ranges originate partly from different methods that countries use and cannot be directly compared with the SP and DT emission factors used in this study as no detailed description of measurement methodology is given. Another feature of Fig. 5~~Fig. 5~~ is that the relative increase is a fixed number for each country. This is due to the fact that we have an estimate for the appliance types (Table 2~~Table 2~~) and their associated wood use for each country but no information how these appliance types are distributed within the country. Hence, within a country the fraction of each appliance type per grid cell is constant. This may not be realistic as, for example, more affluent regions may use less wood or have more modern wood stoves. However, we are not aware of reliable and documented data on this subject.

4.2 Chemical transport modelling results

The two CTMs were run with the two different RWC emission inventories. Fig. 6~~Fig. 6~~ shows the modelled OA concentrations over Europe with the EUCAARI emission inventory, and the relative change in OA when using the revised RWC inventory instead. For the EUCAARI late winter/early spring AMS campaign (25 Feb.-23 Mar., 2009) PMCAMx predicts mean OA concentrations of 1-2 μg m^{-3} in most of Europe and above 2 μg m^{-3} in western France, Portugal, a fairly large region in western Russia and parts of the Baltic countries, and around some cities (e.g., Oslo, Lyon and Barcelona). The peak concentration, above 6 μg m^{-3}, is predicted in the Paris area. The EMEP MSC-W model predicts higher OA concentrations than PMCAMx in most of the modelling domain, above 2 μg m^{-3} in most of southern and eastern Europe, and a remarkably high concentration in the Oslo region in southern Norway (above 4 μg m^{-3}; a considerably higher level than anywhere in the neighbouring Nordic countries). However, in Paris and western France, the British Isles, Moscow, and the northernmost part of the domain, EMEP gives lower concentrations than PMCAMx.

Both models show a significant increase in OA in large parts of Europe with the revised RWC inventory (and a substantial decrease in southern Norway). The response to changing the RWC emissions is larger in the EMEP model than in PMCAMx. For the Feb.-

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Mar. period, the EMEP model predicts more than 50% higher total OA concentrations with the revised RWC inventory than the old one in large parts of central and southern Europe, and Latvia, Lithuania and southern Finland. The impacts of changing the RWC emissions are much more “local” (and, especially for southern Europe, smaller) for PMCAMx than for EMEP (at least for the areas with increasing emissions); this indicates that the wood burning OA is removed more rapidly in PMCAMx than in the EMEP MSC-W model.

One of the models (EMEP MSC-W) was run for an extended period to evaluate the revised emission inventory against long-term measurements of OC at selected sites in Europe during the three-year period 2007-2009. The revised RWC inventory has much larger emissions of OC in Finland, Sweden, Germany and the Netherlands compared to the EUCAARI inventory (Fig. 3~~Fig. 3~~). For this reason we focus on available observations in these countries, where relatively large changes in modelled total OC are expected. RWC has a strong seasonal variation with the largest emissions during the cold seasons. In the EMEP MSC-W model about 80% of the RWC emissions occur during the winter half-year, November-April. During the warmer months other sources of OC are more important (especially secondary organic aerosol from oxidised biogenic VOC). It is difficult to model the boundary layer height accurately in large scale CTMs and there is a risk that the models will underestimate PM from residential wood combustion (and other local, low stack-height sources) during calm, cold nights with strong temperature inversions. However, using the EMEP MSC-W model we find essentially no correlation between either mixing height or temperature and model bias for OC (see Supplementary information).

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The modelled OC is in better agreement with observations at most sites when using the revised RWC emissions compared to the EUCAARI emissions (Fig. 7 and Table 4, and supplement Fig. S1). The model-measurement correlation is higher with the revised inventory, at all of the five investigated sites in the countries with large increases in RWC emissions (the average correlation coefficient, *r*, for the winter half-year data, is 0.69 with the EUCAARI inventory and 0.75 with the revised RWC inventory); the mean absolute error (MAE) of the model OC is lower with the revised inventory, except in Hyytiälä, where it is slightly higher (the average relative MAE for the five sites, for Nov-Apr data, decrease from 52% to 42% when updating the RWC emission inventory).

The Norwegian site Birkenes was also included in the comparison (Fig. S1); Birkenes is situated in the southernmost part of Norway and it is to a large extent influenced by long-range transport from other parts of Europe. The long-term average (cold-season) model results and statistics for this site are very similar with the old and revised RWC emission inventories; the reduced Norwegian emissions are balanced by increased emissions in the neighbouring countries.

The revised emission inventory leads to higher correlation between modelled and measured total OC concentrations and lower model MAE also when comparing to full-year data (Supplement Table S3). Since the measurements of OC are not source-specific, improved model predictions can be caused by the increased RWC-emissions compensating for other missing OC emissions in the model. However, the fact that both winter and full-year results are improved, in combination with the strong seasonal variation of the RWC emissions, is an indication that the revised emissions are indeed more realistic than the old ones. Fig. 8~~Fig. 8~~ shows a comparison of predicted (PMCAMx) vs. Observed (AMS) PM$_1$ OA concentrations (Crippa et al., 2014) from 7 measurement sites in Europe during the EUCAARI winter 2009 campaign (25 February – 23 March). Even for this short term period the revised emission inventory improves the model performance against the AMS data. The slope of the linear fit increases from 0.51 to 0.82 with the use of the revised inventory for RWC emissions. The average fractional bias is substantially reduced (from -0.3 to 0.1) with the use of the revised RWC inventory. The model predicts 35% to 85% higher OA concentrations as a result of the update in the RWC emission inventory.

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4.3 A case study: Norway and Sweden

In order to test the revised RWC emission inventory in more detail we performed a case study using data from published source apportionment studies for the two neighbouring countries Norway and Sweden. Genberg et al. (2011) measured levoglucosan (LG) levels at Vavihill, in southern Sweden, during the period April 2008 – April 2009. We compare the model calculated bbOC (OC from wood burning, including both residential combustion and wildfires) for both inventories to the observed LG concentrations in Fig. 9~~Fig. 9~~. The amount of LG that is emitted during wood combustion varies (see, e.g., Genberg et al., 2011, and references therein); here we assume that bbOC lies in the range 5.5 to 14 times the LG concentration (as in Szidat et al., 2009). Observations at Vavihill show a clear seasonal variation with high concentrations of LG during the winter and, mostly, low concentrations during summer. Modelled bbOC with the EUCAARI emissions underestimate the winter observations severely. Using the TNO-newRWC emission inventory improves the model results a lot; there is still a tendency to underestimate bbOC but not as badly as with the older inventory. Modelled total OC is also improved compared to measurements (Table 4 and Fig. S1).

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We also compared predicted OC from wood burning to source apportionment data from winter campaigns in and near Gothenburg in Sweden (Szidat et al., 2009) and in the Oslo-region in Norway (Yttri et al., 2011). A more extensive comparison to these campaigns (including other sources and both summer and winter periods) was done by Bergström et al. (2012); in the present study we focussed on the wood burning part and the impact of the choice of RWC emission inventory. The results are shown in Fig. 10~~Fig. 10~~. For the Norwegian sites (Oslo and Hurdal, 70km NE of Oslo) modelling with the EUCAARI emission inventory led to large overestimations of wood-burning OC; results were clearly improved for both sites when the TNO-newRWC inventory model was used. Model results for the Swedish campaign were quite different, with underestimated wood-burning OC with the EUCAARI inventory while the results with the revised RWC inventory were within the 10-90 percentile of the source apportionment estimate, based on the measurements, for the rural background site Råö, but still somewhat underestimated for the urban background site (Gothenburg). The revised RWC inventory also improves model results for wood-burning EC at these sites as shown by Genberg et al. (2013).

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Although the two source apportionment campaigns were relatively short, and limited to two regions, the model improvement using the revised RWC emission inventory is consistent with the findings for modelling total OC over longer time periods. These results further support the need to update and harmonize the official estimates of wood burning emissions in Europe.

5. Conclusions and Discussion

Combustion of biofuels, like wood, for cooking or heating is one of the major global sources of organic aerosol (OA). In Europe, residential wood combustion (RWC) is the largest source of OA. Robinson et al. (2007) proposed an alternative framework for the treatment of OA in regional chemistry transport models (CTMs), commonly known as the Volatility Basis Set (VBS) approach. This acknowledged the semi-volatile nature of OA and significantly improved our ability to reproduce observed OA concentrations. However, these new insights have so far had no, or little, impact on the primary particulate matter emission inventories used in Europe. In our opinion this issue needs to be addressed. Currently RWC is increasing in Europe because of rising fossil fuel prices and stimulation of renewable fuels in the framework of climate change mitigation policies. On the basis of the work presented here we conclude that European emissions from RWC are significantly underestimated.

In the present study we constructed a revised bottom-up emission inventory for RWC accounting for the semi-volatile components of the emissions. The revised RWC emissions are higher than those in the previous inventory by a factor of 2–3 but with substantial inter-country variation. The revised emission inventory served as input for the CTMs and a substantially improved agreement between measured and predicted organic aerosol was found. Our comparison of model results with observations suggest that primary aerosol (PM) inventories need to be revised to include the semi-volatile OA that is formed almost instantaneously due to cooling of the flue gas or exhaust. We note that Murphy et al. (2014) suggest that in a simplified framework all emitted semi-volatile organics (effective saturation concentration, C^*, in the range 0.32 - 320 μg m^{-3}) should be considered as primary OA. In this study we adjusted only the RWC emissions while keeping other sources constant. The total European OA emission estimates increased by almost a factor of two. This will have important implications for PM$_{2.5}$ emissions as OA is an important contributor to PM. Interestingly, the EEA/EMEP emission inventory guidebook (EEA, 2013b) was recently updated for wood combustion PM emission factors. We analysed the new emission factors and conclude that they are in line with what was used in this study. For example, for the most important appliance type, the conventional wood stove (about 50% of all wood consumption), the emission factor (DT) used in the TNO-newRWC is 800 g PM GJ^{-1} wood. EEA/EMEP (EEA, 2013b) presents exactly the same value but as total suspended particles (TSP) with the remark that PM$_{2.5}$ ≈ 90 - 95% of TSP. This is very similar compared to the previous gap of a factor 2-4. There are also a few larger differences, e.g. our emission factor for conventional log boilers is a factor 2 higher than EEA/EMEP. However, given the ranges in emission factors, shown in Table 2~~Table 2~~ this is not surprising. A quick calculation suggested that the TNO-newRWC emissions for RWC are likely to be only slightly higher (~10-15%) than when all countries would apply the new EEA/EMEP guidebook factors. As we have shown this leads to a factor 2-3 higher emissions from RWC than currently reported. It will increase total European PM$_{2.5}$ emissions by about 20%. Those are dramatic changes, and this will certainly help reducing the gap between modelled and observed PM, which has often been reported during cold seasons (Aas et al., 2012).

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Acknowledgments

Most measured campaign data originate from EUCAARI partners (Kulmala et al., 2011). Most of the long-term OC filter measurement data were downloaded from the EBAS database (http://ebas.nilu.no); thanks to Hans Areskoug for OC data from Aspvreten, Dave de

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Table 1: Description of source categories in the inventory

SNAP	Description
1	Public electricity and other energy transformation
2_other	Residential and small combustion plants; non-wood fuels
2_wood	Residential and small combustion plants; wood / biomass
3	Industrial combustion
4	Industrial process emission
5	Fossil fuel production
6	Solvent and product use
7	Road transport
8	Non-road transport and mobile machinery
9	Waste disposal
10	Agriculture
11*	Nature

* Emissions for SNAP 11 (nature) are not included in the EUCAARI inventories. Modules for handling these biogenic are typically included in the chemical transport models.

Table 2: Wood use by appliance type in Europe in 2005 and related solid particle (SP) and dilution tunnel (DT) particle emission factors

Appliance type(a)	Wood use in Europe in 2005 (PJ)	Fraction of wood consumption	Emission factor (g GJ^{-1}) (b)			
			SP		DT	
			avg	range	avg	range
Fire place	140	6%	260	23 – 450	900	(d)
Traditional heating stove	1167	52%	150	49 – 650	800	290 – 1932
Single house boiler automatic	198	9%	30	11 – 60	60	(d)
Single house boiler manual	348	15%	180	6 – 650	1000	100 – 2000
Medium boiler automatic	267	12%	40	(c)	45	(c)
Medium boiler manual	141	6%	70	30 – 350	80	30 – 350
Total Europe	2262	100%				

(a) Following IIASA GAINS stove type definition (Klimont, 2002)

(b) Derived from Nussbaumer (2008a, b)

(c) Range in emission factor is determined by end-of-pipe emission control

(d) Not enough data available to indicate range

Table 3: Parameters used to simulate partitioning of primary organic aerosol in PMCAMx and EMEP MSC-W

Parameter [a]	Surrogate Species								
C^* (μg m^{-3})	10^{-2}	10^{-1}	1	10	10^{2}	10^{3}	10^{4}	10^{5}	10^{6}
MW (g mol^{-1})	250	250	250	250	250	250	250	250	250
ΔH_v (kJ mol^{-1})	112	106	100	94	88	82	76	70	64
Base case emission fraction	0.03	0.06	0.09	0.14	0.18	0.30	0.40	0.50	0.80

[a] C*: Saturation concentration at 298 K; MW: Molecular weight; ΔH_v: Enthalpy of vaporization

Table 4: Evaluation of EMEP MSC-W model prediction results (with two different inventories for residential wood combustion emissions) to data from observations during the winter half-years (Nov-Apr) in 2007–2009. Obsvd = Average Measured OC concentration, Model = Average modelled OC concentration (for the periods with measurements, see footnotes), R^2=coefficient of determination, MAE=Mean of Absolute Error. Unit for Obsvd, Model and MAE: µg(C) m^{-3}. The relative MAE = MAE/Obsvd is given within brackets (in %).

		EUCAARI emissions			TNO new RWC emissions		
Site	Obsvd	Model	R^2	MAE	Model	R^2	MAE
Hyytiälä (FI)[a]	1.12	0.80	0.55	0.43 (38%)	1.21	0.61	0.47 (42%)
Aspvreten (SE)[b]	1.77	0.92	0.47	0.91 (51%)	1.35	0.48	0.75 (43%)
Vavihill (SE)[c]	1.68	0.92	0.28	0.84 (50%)	1.30	0.43	0.56 (33%)
Melpitz (DE)[d]	2.12	0.97	0.48	1.20 (57%)	1.51	0.52	0.88 (41%)
Overtoom (NL)[e]	2.37	0.91	0.62	1.52 (64%)	1.34	0.76	1.16 (49%)
Birkenes (NO)[f]	0.58	0.65	0.66	0.24 (41%)	0.66	0.69	0.25 (42%)

a) 14 Feb. 2007 – 18 Feb. 2008, 129 measurements: Measured OC_1, Model $OC_{2.5}$, Aurela et al. (2011);

b) 18 Apr. 2008 – 30 Dec. 2009, 114 measurements: OC_{10};

c) 24 Apr. 2008 – 31 Dec. 2009, 29 measurements: OC_{10}, Genberg et al. (2011);

d) 1 Jan. 2007 – 31 Dec. 2009, 544 measurements: $OC_{2.5}$;

e) Note: Urban background station in Amsterdam (the station is not heavily influenced by RWC, and OC concentrations are similar to surrounding rural background sites, Schaap and Denier van der Gon, 2007), 18 Feb. 2007 – 31 Dec. 2008, 63 measurements: $OC_{2.5}$;

f) 2 Jan. 2007 – 29 Dec. 2009, 140 measurements: $OC_{2.5}$.

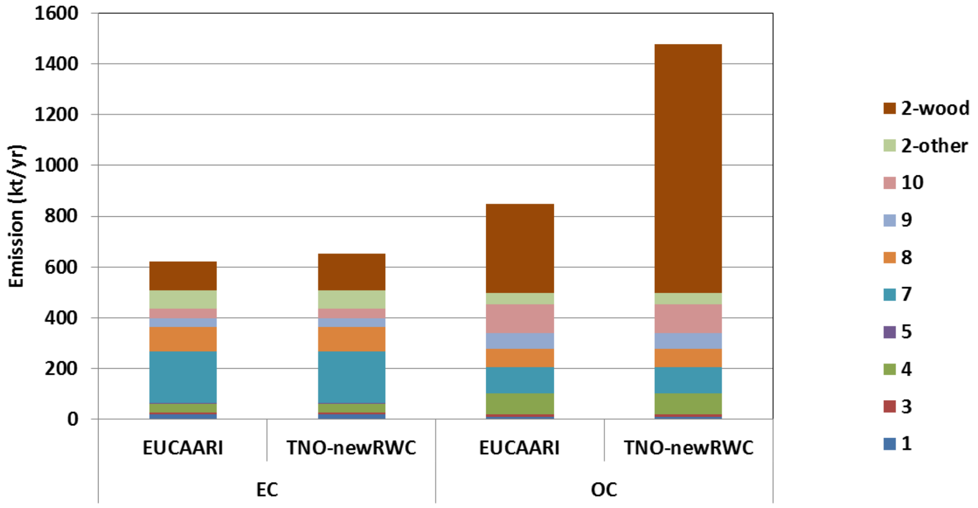


Fig. 1. PM$_{2.5}$ EC and OC emissions (tonnes) for UNECE-Europe in 2005 for each source sector, Table 1) (excluding international shipping) according to the EUCAARI inventory and the TNO-newRWC.

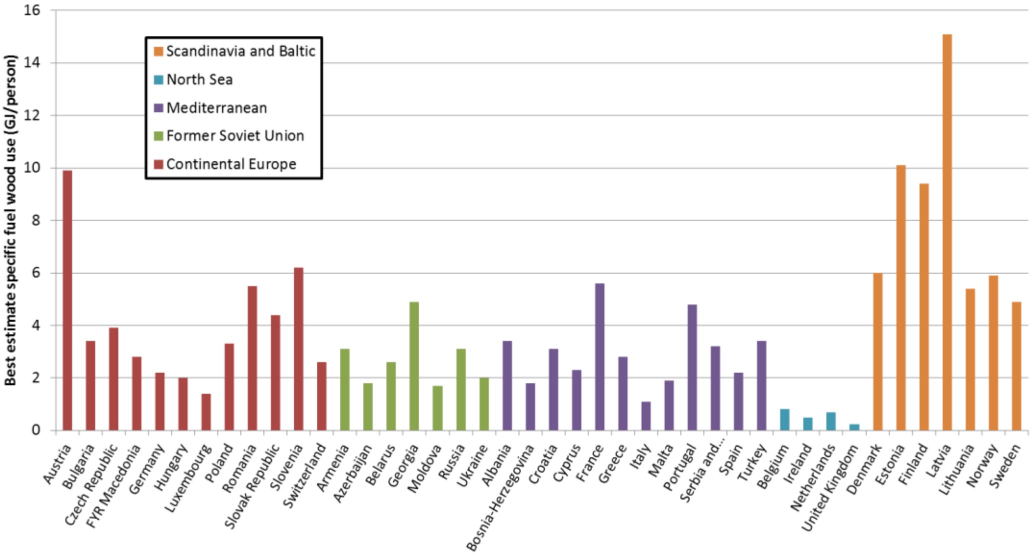


Fig. 2. Estimated specific fuel wood use (in GJ person^{-1}) in UNECE Europe grouped by region.

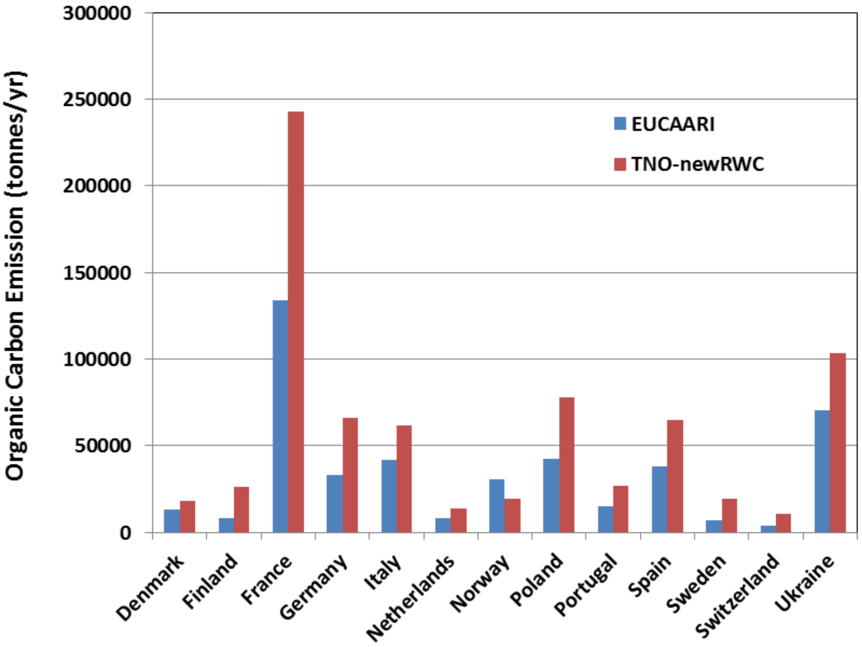


Fig. 3. Total organic carbon emissions (tonnes C yr^{-1}) for selected countries according to the EUCAARI and the TNO-newRWC emission inventories.

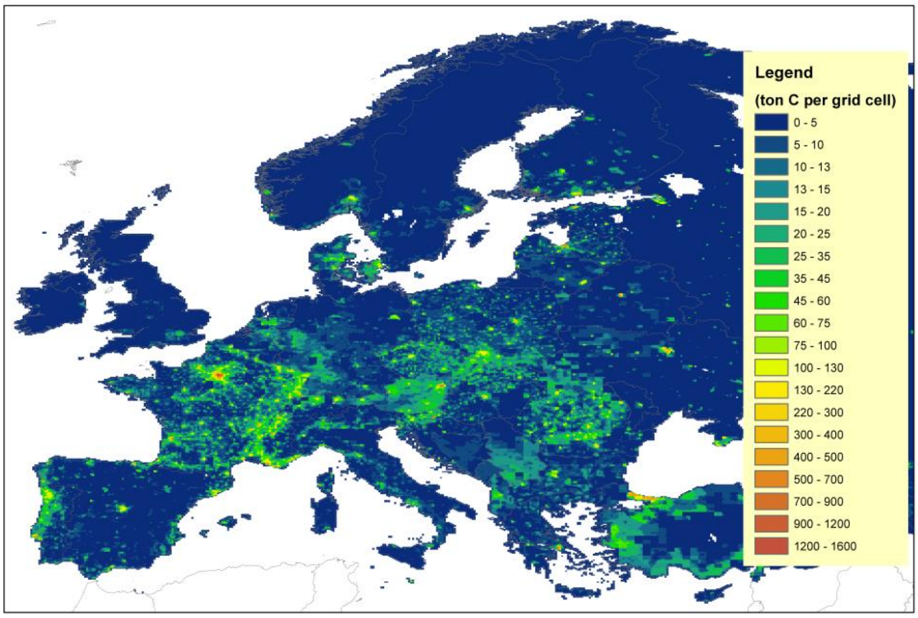


Fig. 4. Spatial distribution of the OC emissions (tonnes C / grid cell; each cell is 1/8° ×1/16° lon-lat, ca 7×7 km) due to residential wood combustion in 2005 based on the TNO-newRWC inventory.

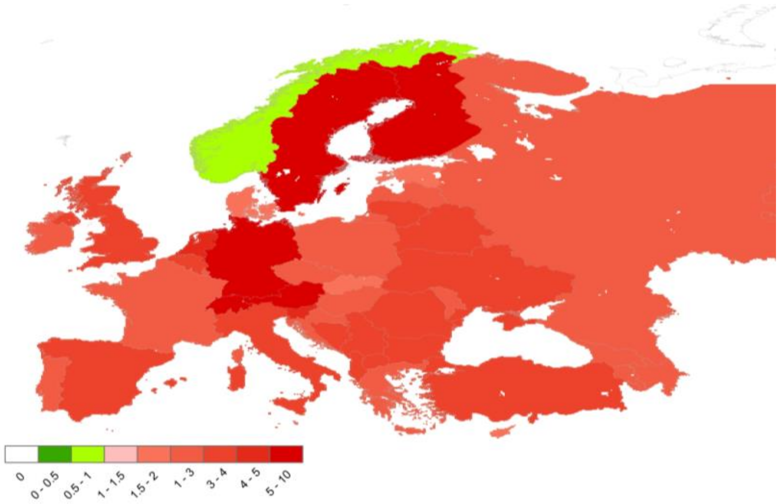
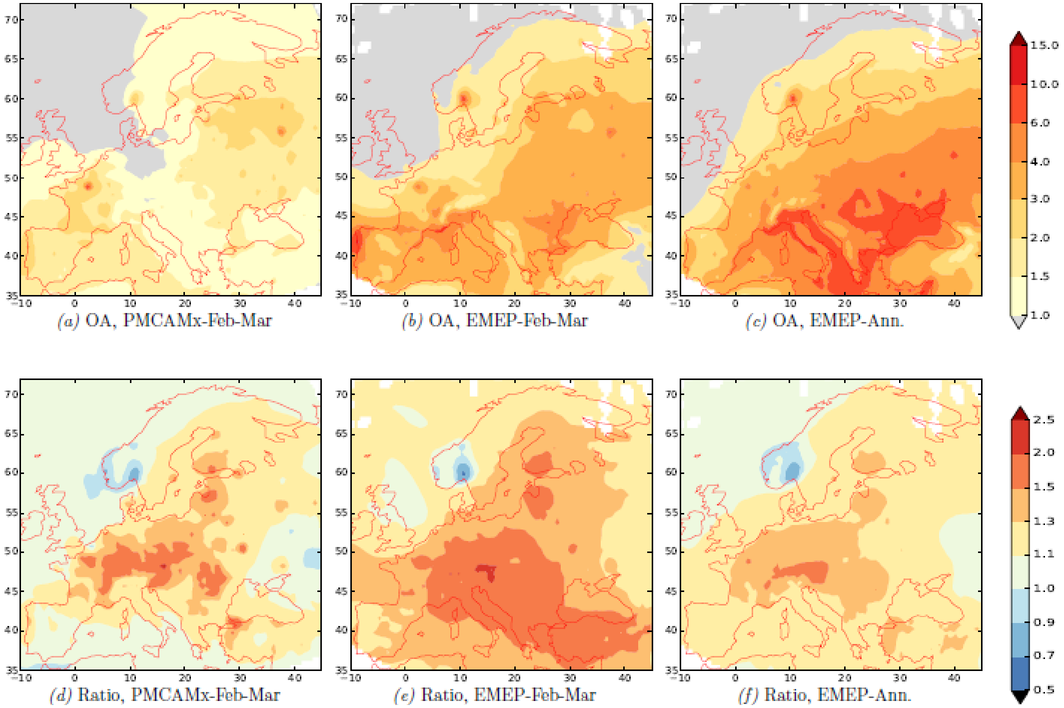


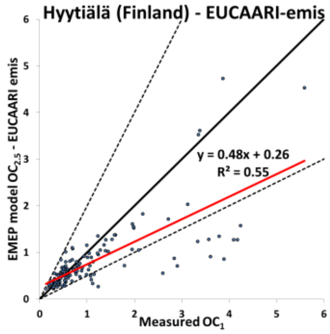
Fig. 5. Ratio of the revised TNO-newRWC inventory relative to the previous EUCAARI OC emission inventory.

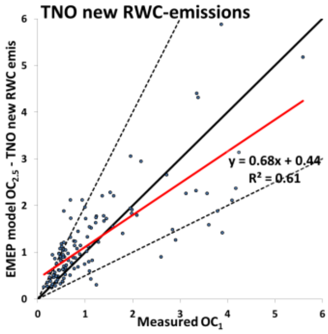


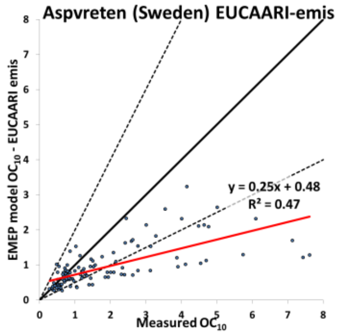
(a) OA, PMCAMx-Feb-Mar *(b)* OA, EMEP-Feb-Mar *(c)* OA, EMEP-Ann.

(d) Ratio, PMCAMx-Feb-Mar *(e)* Ratio, EMEP-Feb-Mar *(f)* Ratio, EMEP-Ann.

Fig. 6. Calculated near-surface organic aerosol (OA, top-row) with the EUCAARI emission inventory (unit: µg m^{-3}), for (a) PMCAMx model, Feb-March, (b) EMEP MSC-W Feb-Mar, and (c) EMEP MSC-W annual, along with the ratio of calculated OA (bottom-row) from TNO-new-RWC/EUCAARI, for (d) PMCAMx, Feb-March (e) EMEP Feb-March, (f) EMEP annual. The Feb-Mar period is 25 Feb - 23 March 2009, the annual period is for 1 Jan - 31 Dec 2009.


Hyytiälä (Finland) - EUCAARI-emis
EMEP model OC2.5 - EUCAARI emis
Measured OC1
y = 0.48x + 0.26
R² = 0.55
0 1 2 3 4 5 6


TNO new RWC-emissions
EMEP model OC2.5 - TNO new RWC emis
Measured OC1
y = 0.68x + 0.44
R² = 0.61
0 1 2 3 4 5 6


Aspvreten (Sweden) EUCAARI-emis
EMEP model OC10 - EUCAARI emis
Measured OC10
y = 0.25x + 0.48
R² = 0.47
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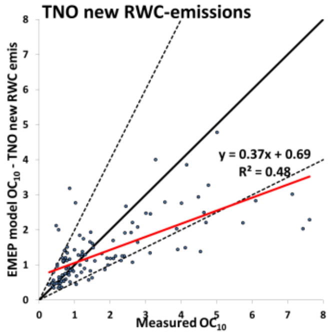
TNO new RWC-emissions
EMEP model OC10 - TNO new RWC emis
Measured OC10
y = 0.37x + 0.69
R² = 0.48
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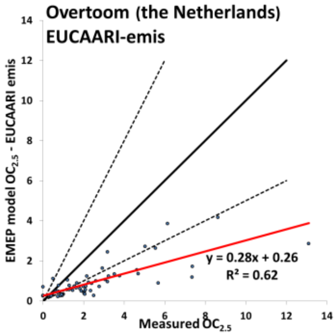
Overtoom (the Netherlands)
EUCAARI-emis
EMEP model OC2.5 - EUCAARI emis
Measured OC2.5
y = 0.28x + 0.26
R² = 0.62
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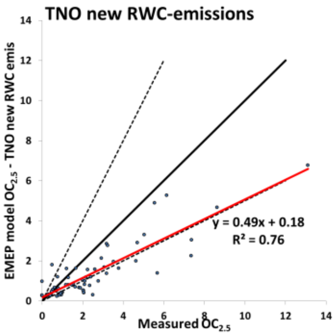
TNO new RWC-emissions
EMEP model OC2.5 - TNO new RWC emis
Measured OC2.5
y = 0.49x + 0.18
R² = 0.76
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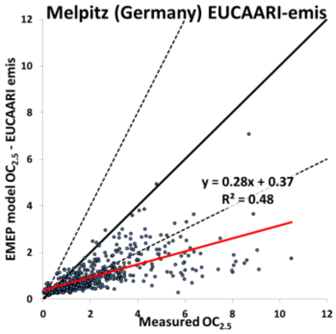
Melpitz (Germany) EUCAARI-emis
EMEP model OC2.5 - EUCAARI emis
Measured OC2.5
y = 0.28x + 0.37
R² = 0.48
0 2 4 6 8 10 12

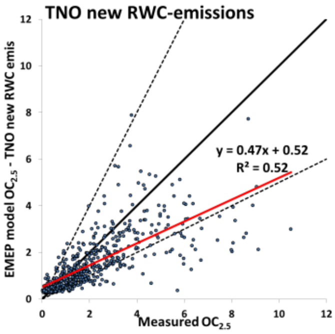
TNO new RWC-emissions
EMEP model OC2.5 - TNO new RWC emis
Measured OC2.5
y = 0.47x + 0.52
R² = 0.52
0 2 4 6 8 10 12

Fig. 7. Measured and modeled organic carbon concentrations (2007-2009, winter half-year data: November-April) at four sites: Hyytiälä (FI), Aspvreten (SE), Overtoom (NL) and Melpitz (DE). The left-side plots show EMEP MSC-W model results using the EUCAARI emissions and the right-side plots results using the revised residential wood combustion emissions. For Hyytiälä measurements are OC in PM_1 and model results are OC in $PM_{2.5}$; for Aspvreten OC in PM_{10} is shown; for Overtoom and Melpitz OC in $PM_{2.5}$. Each point represents one measurement (variable sampling duration, from 17 hours to 2 weeks). Unit: µg C m^{-3}.

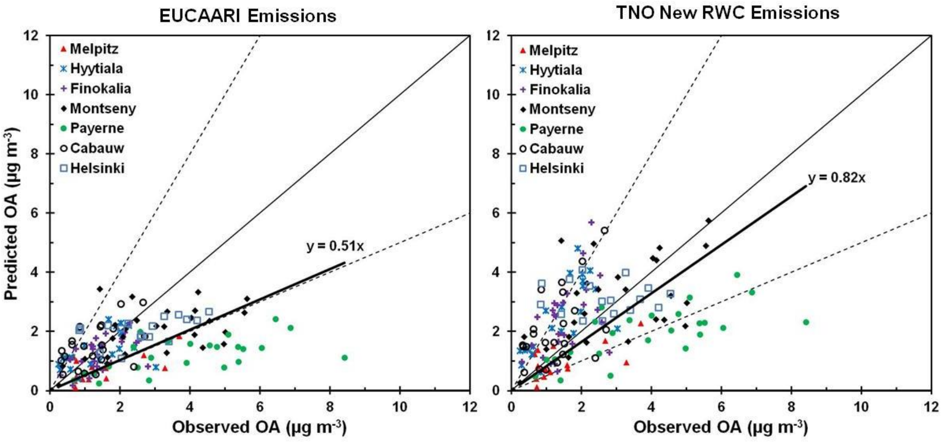

Fig. 8. Comparison of predicted (PMCAMx) vs. observed (AMS) PM_1 OA (μg m^{-3}) from 7 measurement sites during the EUCAARI winter 2009 campaign (25 February – 23 March). Each point is a daily average value. The dashed lines represent 2 : 1 and 1 : 2 lines.

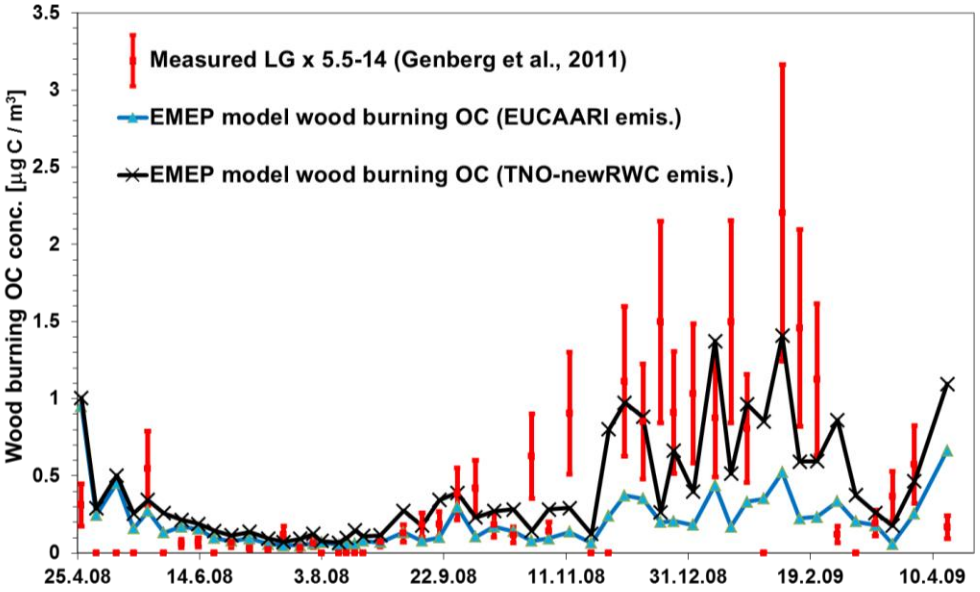

Fig. 9. Comparison of EMEP MSC-W model predicted OC (µg(C) m^{-3}) from biomass burning (bbOC) (including OC from both residential wood combustion and open vegetation fires) to bbOC estimated from levoglucosan concentrations at Vavihill (southern Sweden). Red bars: Estimated bbOC range (measured levoglucosan × 5.5-14); Blue: Model calculated bbOC with the EUCAARI emission inventory; Black: Model bbOC with the TNO-newRWC emission inventory. Red bars show the measurement-estimated bbOC range (lower limit: 5.5 × the measured levoglucosan concentration, upper limit: 14 × levoglucosan; the estimated range for the bbOC/levoglucosan ratio, 5.5-14, is taken from Szidat et al., 2009).

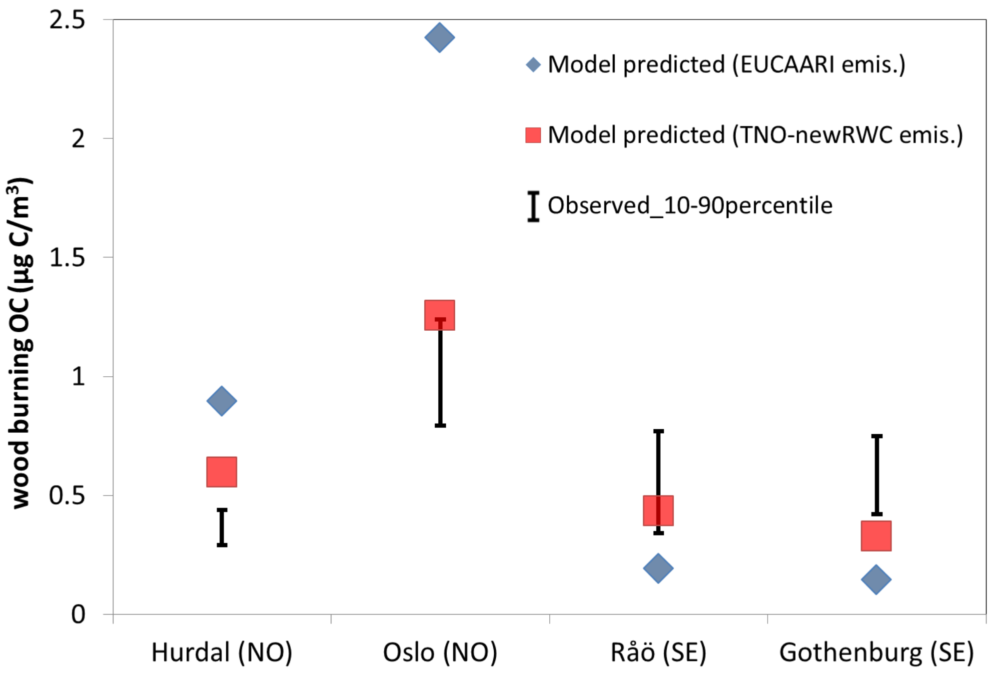

Fig. 10. Comparison of model calculated OC (µg(C) m^{-3}) from wood burning (residential combustion + open vegetation fires) to source-apportionment data from measurement campaigns during winter in Norway (SORGA, 1 - 8 March 2007, Yttri et al., 2011) and Sweden (GÖTE, 11 Feb - 4 Mar 2005, Szidat et al., 2009).