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 not 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).

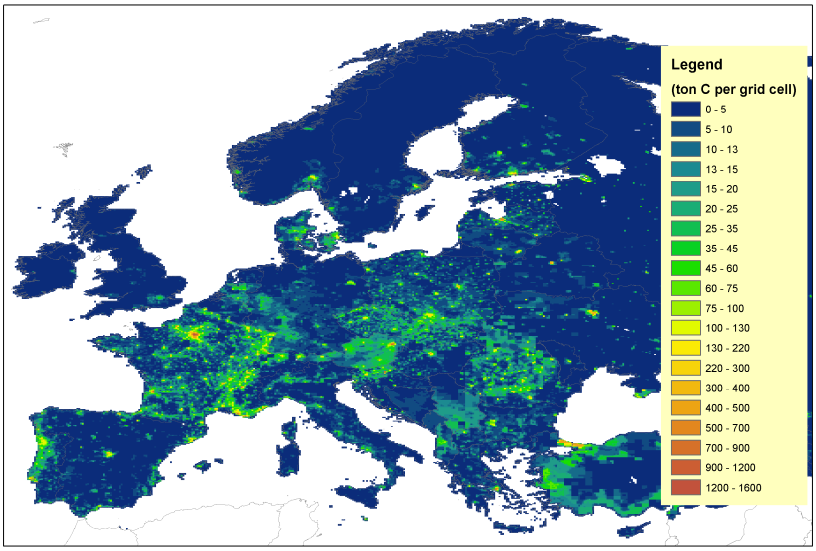

Fig. 6: The figure is really hard to read because of its size.

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.