

## Response to reviewers' comments

MS No.: gmd-2015-130

Title: Modelling the dispersion of particle numbers in five European cities

Author(s): Kukkonen et al.

Our responses to reviewers have been written in blue font. The page and line numbers below refer to the new, revised manuscript that has been sent together with this response.

### Reviewer 3

**For final publication, the manuscript should be accepted subject to MINOR REVISIONS**

#### **SUGGESTIONS FOR REVISION**

This is a description of the application of five separate models to the prediction of particle number concentrations in five different cities. The only common features are the objective to predict particle number concentrations, and the use of a single model to estimate spatially resolved regional background across Europe. The models differ in their design and input data and show varying skill in the estimation of particle number concentrations for the few sites with available data for comparison.

The paper has been very substantially revised from the original version and now contains a great deal more information and relevant discussion. In this context, it is much improved although its expansion inevitably leads to further questions of clarification being raised.

Further points which need to be addressed are the following:

- (a) Page 4, line 25-26 – it is stated that “coagulation was found to contribute to losses of PNCs ..... as compared to inert particles”. The term “inert” is normally taken to mean unreactive whereas I assume what is meant here is “compared to particles which are assumed not to coagulate”.

Corrected.

- (b) Page 4, lines 41-42 – the statement that there are only concentration-response functions based upon expert judgement is not correct. The Hoek et al. (2010) study was an expert elicitation but the Stolzel et al. and Atkinson et al. studies were original epidemiological studies.

Corrected.

- (c) Page 8, line 27 – this refers to aerosol transformation processes being taken into account but does not specify which processes. If those processes are coagulation and deposition, then the former is a transformation process but the latter is a sink process.

The text has been corrected to be clearer. The included aerosol transformation processes are nucleation and condensation of H<sub>2</sub>SO<sub>4</sub>, and coagulation of particles (these of course can be called transformation processes). The chemical transport model (LOTOS-EUROS) also includes treatments for the dry and wet deposition (sink processes).

- (d) Page 10, first paragraph – it seems very strange to use a single emission factor for light duty vehicles when this is typically a mix of gasoline and diesel vehicles for which the emission factors are very different. Some discussion of this point is needed. The same point applies to page 11, lines 18-22.

In case of Rotterdam, we had to use so-called composition emission factors, due to the available traffic flow data. This has been explained on p. 11, lines 27-34. Quote:

“As mentioned above, two composite emission factors were used for passenger cars, one for motorway traffic, and the other one for traffic in urban roads. This was necessary, as the available traffic flow data was also in composite form, including a value for each street for each of the following vehicle categories: passenger cars, lorries and busses. The assumption of composite emission factors implies that the fractions of passenger cars equipped with diesel, petrol and vehicle technologies are not spatially variable within the city. However, these composite emission factors take into account, e.g., the differences between the emission factors of cars using gasoline and diesel fuels.”

So, we emphasize that the composite emission factors take into account the differences between the emission factors of cars using gasoline and diesel fuels. We hope that this replies to the reviewer’s question.

Similarly, in case of Helsinki, composite emission factors were used. Unfortunately, we had to do this, as our best applicable emission data (from Gidhagen et al., 2005) did not include separate emission factors for diesel and gasoline vehicles. However, these composite emission factors, if correctly measured, will in our view represent fairly accurately the combined emissions of diesel and gasoline vehicles.

Giechaskiel et al. (2012) (reference information below) have tested real-time emissions of non-volatile particles from light-duty (LD) vehicles. Total particle number emissions from diesel LD vehicles depend on the applied after-treatment technology and those from gasoline LD vehicles depend on injection technology. With the introduction of diesel particle filter, PN emissions were

found to decrease by 2-3 orders of magnitude. However, emission factors for total particle number based on laboratory measurements are strongly dependent on dilution rates, which make them difficult to use for dispersion modelling purposes. Gidhagen et al. (2005) reported fleet aggregate emission factors of particle number for LD vehicles based on measurements of a typical 'urban' fleet at roadside in different urban micro environments. The latter kinds of measurements are actually better applicable for use in urban dispersion modelling.

Giechaskiel, B., Mamakos, A. , Andersson, J. , Dilara, P., Martini, G., Schindler, W., and Bergmann, A., 2012. Measurement of Automotive Nonvolatile Particle Number Emissions within the European Legislative Framework: A Review, *Aerosol Science and Technology*, 46:7, 719-749, DOI: 10.1080/02786826.2012.661103.

For the European wide (TRANSPHORM) inventory we calculate separately emission factors for the gasoline and diesel vehicles. However, the city emissions used in urban scale assessments are most often not specified by fuel type. The reason for this is that the exact fleet composition and fuel type of cars driving in the city is commonly not known, it is therefore better to use measurements in the city (these will be representative for the fleet in that city), in combination with the information on fleet composition. As detailed engine info is commonly not available, the only separation that you can then make is personal cars, LD and heavy duty (HD) vehicles.

Discussion on this issue is already included in the section for Rotterdam (as described above). Some additional comments and discussion about this issue were added also to the section for Helsinki (in section 2.2.2, p 10, lines 5-17).

(e) Page 13, second and third paragraphs – does the aerosol nucleation scheme take account of the local condensation sink?

Yes. The nucleation scheme as such is only dependent on the gaseous  $H_2SO_4$  concentration. However, the  $H_2SO_4$  concentration that is used in the modelling is that after taking into account the condensation on existing particles (as explained in Vignati et al 2004 p4).

We have added a clarification to p. 13, lines 24-27.

(f) The revised paper includes far more information on the emissions inventory for particle number, including the future projections. Since particle number emissions from combustion sources are critically dependent upon the sulphur content of the fuel, it would be advisable to include information on the sulphur content used in the inventory and in the future projections. The criticality of sulphur content of shipping fuels is brought out on page 23, and similar information regarding motor fuels is desirable.

There is fairly extensive discussion on the sulphur content of motor fuels on p. 25, lines 4-28 (and regarding shipping fuels, also on p 24, lines 24-28). This was added to the previous manuscript version on reviewer's request. This discussion addresses both the criteria for assuming the sulphur content values used in the modelling in various cities, and the validity of these assumptions.

The predicted concentration results of future projections were removed from the revised manuscript, due to a request of one of the reviewers.

- (g) Further discussion of the way in which emissions from aircraft are dealt with would be useful. There is a comment on page 32, line 27-28 that in Athens the highest predicted concentrations of PNC occurred in the vicinity of the Athens International Airport. Was this because of road traffic or aircraft emissions? It appears that for London the aircraft emissions were not included. What was the rationale for this?

We agree; this has been explained a little later in the same section (3.2.3), on p. 34, lines 11-23. The text reads as follows: "... In Athens, there were substantially elevated PNC's near the Athens International Airport, located to the east from the centre of the city (it is clearly visible in Fig. 8e). Detailed computations showed that aviation emissions were responsible for the largest share of the concentrations within this airport and in its immediate vicinity.

The above text replies to the reviewer's question on aviation in Athens.

The text continues in revised form (the latest revision shown below as underlined): "The influence of the Heathrow airport in London is also visible in the PNC map (near the outer ring road on the western part of the city). However, these higher predicted concentrations were caused by the emissions from the congested roads leading to the Heathrow airport. The emissions originated from aviation in London were included in the regional background concentrations (the LOTOS-EUROS predictions), but not explicitly in the urban scale computations."

It was unfortunately not possible in practice to include the detailed aviation emissions as a specific urban source in London. The reasons for this were that (i) the detailed PNC emissions were not available (these would have to be converted from the aviation PM emissions, and it is not clear how to do this accurately) and (ii) the OSCAR modelling system does not currently provide treatment of line sources that are not horizontal but tilted in the vertical direction (during the takeoff and landing).

- (h) A statement appears in several places that the shipping in the London area has a negligible impact on overall PNCs (e.g. page 32, lines 38-40). It is stated earlier that this conclusion is based upon the PM<sub>2.5</sub> emissions that are included in the inventory. Since this source of PNCs was not included in the model for London, a clearer statement would be preferable.

We have added some arguments on why this conclusion is very probable, and wrote this conclusion more cautiously and more accurately, on p. 33, lines 41-45. This includes a short discussion of a recent source apportionment study of PNC in London.

It is correct of course that statements on PM<sub>2.5</sub> can not necessarily be converted to the same conclusions on PNC's. E.g., the temporal hourly values of the concentrations of PM<sub>2.5</sub> and PN do not necessarily correlate. However, a negligible contribution of shipping on the total PM<sub>2.5</sub> emissions on an annual average level indicates very likely also a negligible or very small contribution on the total PNC on an annual average level, for the whole city. As the same result was found also using source apportionment (although for one location only), we feel that this conclusion is probable on an annual average level over the whole city. Regionally and temporally the results may of course be different; these aspects have not been studied here.

- (i) Page 37 – there is some discussion of the influence of coagulation and deposition upon PNCs but it needs to be acknowledged that particles are also subject to evaporation and condensation processes which have not been considered but which may significantly influence their ambient concentrations.

Corrected.

**Submitted on 29 Dec 2015**  
**Anonymous Referee #3**

The manuscript has been improved for almost all the items that were critically highlighted by reviewers.

The heterogeneity of the modelling approaches used to reproduce PNC concentrations in the different cities remains a limit, but is now properly commented and discussed by the authors. The paper can be published after minor revision and technical corrections.

Page 4, line 34:

The reference to TRANSPHORM web site “(ww.transphorm.eu)” needs a correction.

Corrected.

Page 7, Table 1:

Reference stile in Athens column “Petzold et al. (2010) and (Lee et al, 2010).” should be made uniform.

Corrected.

Page 8, lines 8-9:

It would be better to explicitly specify which models include emissions from stationary sources and residential heating, as it is specified in the previous text lines for other sources.

Corrected.

Page 8, lines 22-24:

The sentence “For obtaining an improvement...” causes the following question: the use of local data for Oslo, London and Rotterdam was not necessary because LOTOS-EUROS provided better results for those cities, or this choice only depended on the local modelers approach and choice?

This formulation of the text was somewhat vague, and we have therefore removed this explanation. This choice depended on each modeller’s judgement and choice.

Page 10, lines 2-4:

This sentences contain a repetition and can be simplified/shortened.

Corrected.

Page 10, lines 4-6:

Why emission factors for passenger cars are not specified and commented? Only heavy and light-duty vehicles emission factors are specified.

Please see our response to the comment (d) above.

Page 10, lines 13-14:

The reference to the web site needs to be corrected, probably “indexe” should be “index”.

Corrected.

Page 11, lines 33-38 and Page 12, lines 15-18:

What about possible house heating contribution for Rotterdam and London? No comment is provided about this source at variance with Helsinki and Oslo subsections.

The small-scale house heating contribution for London is very small, based on the local urban emission inventory for PM mass, compared with the other main source categories in this city. More details of this data of emissions specified for various emission sectors in London were included in our previous response to reviewers.

We included a couple of comments on this issue to p12, lines 28-31; the new text is more accurate and more cautiously written than the previous one.

For Rotterdam, the detailed information on house heating was not available.

Regarding Helsinki, the available information has been presented on p. 10, lines 36-41. Quote: “Emissions from small-scale combustion were not taken into account, as their spatial distribution was not known with sufficient accuracy. The contribution of small-scale combustion to the total PM<sub>2.5</sub> emissions in Helsinki Metropolitan Area has been estimated to be 23 % in 2009 (Malkki et al, 2010).”

For Oslo, the influence of house heating was explicitly evaluated (p. 11, lines 16-19).

Page 13, lines 30-31:

The reason why model simulation for year 2005 had to be performed is not clear.

The reason was that the main target year in the TRANSPHORM project was 2005 for the regional scale computations, and 2008 for the urban computations. The earlier year of 2005 was selected in the project for the regional scale, caused by the better availability of emission data at the time, when this was decided. A clarification was added on p. 13, lines 44-45.

Page 21, line 16:

The references stile should be checked.

Corrected.

Page 15, Figure 5 caption:

Does “particles/a” mean particles/year ?

Yes. Corrected.

Page 26, line 7:

The number of mentioned sites should turn from three to eight.

Corrected.

Page 26, line 29:

The generic mention to “different settings of the measurements.” Is quite obscure.

Yes, we agree. During these months (from Jan to Apr) the instrument was not mounted near-surface, but instead at the height of 60 m. The text has been revised to be clearer.

Pag 37, lines 13-18:

I would reinforce the sentence on emission inventories explicitly saying that they should be completed for all the source sectors. Some sectors are still missing, not only characterized by low accuracy.

Yes, we agree. This has been revised.

Figures editing should be checked.

Checked.

## **Other revisions**

The previous Figs. 2a-b have been replaced with technically better versions.