Response to reviewers on "Modelling the dispersion of particle numbers in five European cities" by J. Kukkonen et al.

- 5 Anonymous Referee #1
 - Received and published: 3 September 2015

9 The authors present an overview of the modelling of particle number concentrations (PNC) in 10 five cities in Europe. The simulations have been performed on a regional scale with the LOTOS-11 EUROS model and on a local scale with different local models for every city. Model simulations 12 focus on the years 2005, 2008 and 2020. The simulation results of the regional and local models 13 were compared with measurements of the year 2008.

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From the current version of the paper it is very hard for the reader to assess the main result of the paper, i.e. the five maps of UFP concentrations for the different considered European cities, because the reader does not have enough information about the difference between the local models and the input of the emissions. We simply can't see and understand what is driving the differences between the results for the different cities, and how important these differences are.

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21 <u>Response</u>: In the original manuscript, we tried to clarify the differences and similarities of the 22 various emission inventories and models in Table 1, and the associated text. This Table 23 summarizes the treatments of emissions, meteorological data, dispersion models, source 24 categories included, etc. According to the reviewer's comment, we have revised and clarified 25 Table 1 and its associated discussion, in section 2.1.

26

The urban scale modelling systems used in various target cities are different. However, all of the modelling systems used for Helsinki, Oslo, London and Rotterdam are urban, multi-source Gaussian dispersion and transformation systems. These systems can also allow for dispersion in street canyons. The modelling system for Athens is based on the combined use of a meteorological model and a chemical transport model. All these modelling systems have previously been extensively evaluated against experimental data. This has been more clearly stated in the revised section 2.1.

34

We are therefore confident that the major differences of the numerical results in various cities are caused by (i) the differences of the structure and distribution of emissions, (ii) differences of meteorological conditions and (iii) differences of other specific characteristics of the cities, instead of the differences of the dispersion modelling systems.

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We also examined in detail all the descriptions of urban modelling in section 2.3.2 (Urban scaledispersion modelling), and have made major revisions to several of those texts, especially in the

42 case of modelling of Rotterdam, London and Athens. In our view, the revised descriptions

- 43 illustrate much more clearly the treatments of the various urban modelling systems.
- 44

- We have also completely re-written the interpretation of the results, especially that of Fig. 8 (concentration maps). We have presented more clearly the differences and similarities between different cities, and the main causes of these differences, in terms of the source contributions and spatial distributions. Fig. 8 was also presented in a harmonized form, using the same concentration legend for all the cities, for an easier city inter-comparison.
- 6

7 In our opinion the paper therefore needs major revisions in which the material should be 8 structurally re-organised such that the material is presented in a more uniform way. We 9 recommend that the authors consider the following points:

10

- in the description of the urban-scale emission inventories (2.2.2) there should be more emphasis
on the major differences and similarities between the inventories. Are the differences such that
the output of the maps for the five cities can be objectively compared or are there serious
omissions in some of the inventories. For example the inclusion/exclusion of harbours and
airports, 2 important sources next to road traffic.

16

17 <u>Response</u>:18

19 We have done a concrete major improvement to the original manuscript: also the shipping 20 emissions in Rotterdam have been modelled in the revised manuscript. The corresponding 21 changes were of course made to the description of methods (the section on the emission 22 inventory for Rotterdam), to the section on model evaluation, and other relevant sections.

23

PN emission and dispersion modelling had not previously (before this study) been done in the
target cities. This is also true for almost all other European cities. Only a few simple preliminary
attempts have been published, before this study. We have therefore attempted a pioneering study
in this field, to improve this situation. However, the state of knowledge and information on
emissions of PN for various source categories is currently far from complete.

29

30 The state of the emission information for PN is also variable in terms of the target cities, and in terms of the source categories. This is the reason why the collections of the source categories that 31 32 were included could not be better harmonized. Instead of requiring a complete harmonization in 33 this respect (which would require conducting this study only for regional background and 34 vehicular traffic), we felt that it would be better to allow the inclusion of those source categories 35 in each city, for which this information was available. We have therefore examined all available 36 sources of information for all of the target cities, and included all those source categories, for 37 which sufficiently reliable emission information was available. This has been more clearly stated 38 in the revised manuscript (in section 2.1., the sections on urban emission inventories, and section 39 3.1.2).

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41 At the moment, we have included:

- 42 1. vehicular traffic for all cities,
- 43434344442. shipping (explicitly or implicitly; the latter referring to an evaluation on the importance of shipping) for all cities
- 45 3. small-scale combustion or evaluation of its importance for Oslo and Hki (which is sufficient, as this source category is not substantial for the other three cities)

- 4. major stationary sources as separate sources for Hki, Oslo, Athens, and as part of the regional background for Rotterdam and Athens
- 5. aviation explicitly only for Athens, but its importance has been evaluated for Helsinki
- 3 4

2

5 Wood burning is known to be relevant in Oslo and potentially relevant also in Helsinki. In this 6 study, its influence was explicitly allowed for in case of Oslo. For Helsinki, a sufficiently 7 accurate emission inventory of wood burning was not available. We have therefore used the best 8 available information for Helsinki; that was an estimate of the total emissions from wood 9 burning in that area (without the information of its spatial distribution). This was used for an 10 indirect estimate of the contribution of wood burning emissions in that area, although it is not 11 possible to conduct detailed dispersion computations.

12

Actually, there are also substantial differences between Oslo and Helsinki in terms of wood burning: that is substantially more important for Oslo. Although the climatic region is similar, there are differences caused by socio-economic reasons, such as the abundance of wood-burning

- 16 facilities in housing, traditions and customs, local and governmental policies, etc.
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19

Absolute Contribution (t/yr) of Emissions Sources to Total PM10 Emissions 4,000 3 500 3.000 Agriculture-Nature Airport Boilers 2,500 Coal Gas 2.000 Oil Part A Processes ■Part B Processes Rail 1.500 Road Transport Ships ■Solvents-Building 1.000 500 0 2004 2006 2008 2011 2015



Figure R1. Contributions (t/year) of emissions to total PM10 emissions in London during 2004 2015. 'Part A Processes' are large industrial processes regulated by the Environment Agency,
'Part B Processes' are smaller industrial processes regulated by the local authorities. 'Boilers'
refers to large industrial boiler plants. Ref.: GLA London Atmospheric Emissions Inventory
(LAEI).

1 It is evident based on Fig. R1 that the most important source categories of PM in London are 2 road transport, agriculture-nature and industrial processes. According to this emission inventory, 3 the PM_{10} emission from shipping is 2 ton/year, which is 0.08 % of total emissions. We therefore 4 considered it appropriate to neglect the influence of shipping in case of London (this has been 5 revised in section "Emission inventory for London" in the revised manuscript).

6

For Helsinki, the Helsinki Metropolitan Area (HMA) aviation PM2.5 emissions were about 17 %
of the total road traffic PM2.5 emissions in the HMA in 2008 (ref. official statistics of Finland).
This has been added to the revised manuscript to section "Emission inventory for Helsinki".

10

We have also clarified the importance of other sources, such as airports and refineries forRotterdam in the section "Emission inventory for Rotterdam".

13

- in the description of the urban scale dispersion modelling (2.3.2) the major differences and
similarities should be explained. Again, as above, the questions is whether the differences
between the models are such that the output of the maps for the five cities can be compared in a
meaningful way.

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Response: We have substantially revised these descriptions, especially for London, Rotterdam and Athens. We also included an overview of these differences and similarities to section 2.1.
 The concentration maps were harmonized; these are now presented in a uniform manner. We also evaluated better the effects of different kinds of modelling on the numerical results, in section 3.2.2.

24

- can the authors indicate whether the uncertainties in the presented maps from city to city aredue to the used emission inventory or due to the local model which was used.

27

Response: Our expert judgment is that the largest contributor to the uncertainties is by the urban
 scale emission inventories for Helsinki, Oslo and Athens. However, all the local scale modelling
 systems used in this study have been previously evaluated against experimental data; their
 uncertainties regarding meteorology, chemistry and dispersion processes are therefore fairly well
 known.

33

However, in street canyon locations, the dispersion modelling is expected to under-estimate the concentrations (though, in case of Rotterdam, also a street canyon model was applied). We have clarified also the description of the uncertainties caused by street canyon conditions in the revised manuscript (section 3.2.3). The uncertainties caused by the coarser resolution in Athens were also discussed in the revised section 3.2.3.

39

40 The regional background concentrations are clearly lower than the urban concentrations in 41 Helsinki, Oslo and Athens, although not in Rotterdam, and partly not in London. This can be 42 more clearly seen from revised Fig. 9 and its associated discussion. The uncertainties caused by 43 regional emission inventories and regional scale dispersion modelling are therefore also 44 relatively smaller in Helsinki, Oslo and Athens, compared with the uncertainties caused by the

45 urban scale emission inventories.

We have added discussion on the modelling uncertainties to section 3.3.

- the authors show that the correspondence between measurements and calculations for the
LOTOS-EUROS model still needs some significant improvement. In our opinion it is therefore
not very meaningful at this stage to present a future scenario for 2020, Instead, we suggest that
the 2008 map of LOTOS-EUROS is presented in figure 7, such that the same year is used as for
the local calculations (figure 8).

9 <u>Response</u>: We agree with the reviewer on the former point, and have removed the scenario 10 results for 2020 from the revised manuscript. Regarding the latter point, we have added a 11 concentration map for 2008, and also two additional maps that show the differences of 12 concentrations between these two years. We also replaced the former Fig. 6 with a more 13 extensive model evaluation figure.

14

- make the figures of the city maps uniform (figure 8), ideally use the same visualization tool, it
is the 'heart of the paper'. Choose a scaling which is 'smart' such that the five panels can easily
and meaningfully be compared. Indicate locations of airports and harbour areas (or other
significant local sources) in the maps where appropriate.

Response: We have completely re-drawn Fig. 8. using only one visualization tool, according to
 the reviewer's suggestions. Scaling is smart i.e. identical for all cities, and still showing well the
 concentration contrasts. We drew the locations of harbours and airports to a separate figure that
 is in Annex 1.

In figure 9 the results should be presented with for example stacked bars, such that the reader
can see which part of the modelled concentration is from the LOTOS-EUROS model, and which
part is from the local model.

29 **<u>Response</u>**: Done as suggested.

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34 Anonymous Referee #2

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36 Received and published: 15 September 2015

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There have long been suggestions that ultrafine particles, often assessed by particle number count (PNC) are more toxic per unit mass than coarser particles. Measurements of PNC are relatively scarce, emission factors few, and hence modelling is at a relatively early stage of development. In this study, the authors report a model study (actually five separate studies) of PNC in five cities of Europe, using an array of local urban models, supported by a single model to evaluate the regional background concentrations.

44

45 Modelling particle concentrations is very challenging, and this paper makes a useful attempt at 46 doing so. The results compare surprisingly well with measurements, but a number of key issues

1 have been given insufficient consideration, which reduces the overall value of the study. The 2 most important issue which is largely ignored is the sulphur content of motor fuel. This has a 3 major impact upon the emission factor for particle number, and has been reducing for many 4 years in Europe. The year chosen for modelling in three of the five cities is 2008, which was 5 around the time that the sulphur content of motor fuels was decreasing rapidly in many European 6 countries, from < 50 ppm S to < 10 ppm S. In late 2007, this was associated with a reduction in 7 particle number concentration of around 65% at London, Marylebone Road, and a substantial but 8 lesser decline _39% at London, North Kensington (Jones et al., 2012). Hence, defining the 9 sulphur content of fuel in each city is essential, but is not currently considered. The corollary to 10 this, is that emission factors determined with the fuel content at the time of the measurements should be used. For London, the emission factors from Jones and Harrison (2006) are used, 11 12 which refer to the higher (< 50 ppm S) fuel sulphur, while the measurement year (2008) is after 13 the transition to low S fuel. The suggestion (p5902, line 23-26) that these emission factors may 14 underestimate those on this congested road is incorrect, as the field measurements were made on 15 Marylebone Road! No doubt also of importance is the canyon nature of the site, which the 16 authors recognise. For Helsinki, emission factors from Gidhagen et al. (2005) are used, which 17 may also overestimate 2008 emissions. 18

19 **Response**:

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We have addressed in more detail the issue of the sulfur content of the motor fuels. We have added text to the manuscript that reports the values, which were used in this study, and discusses their usage, in the section 3.1.2.

24

25 For the Helsinki case, calculations were based on EFs given by Gidhagen et al. (2005) 26 (references are listed at the end of this response text) for Stockholm. The measurements that are 27 the basis for these EFs were made in Stockholm in 1999 for heavy duty vehicles (HDV) and in 28 2003 for light duty vehicles (LDV). Sweden introduced its Environmental Class 1 (EC1) diesel 29 fuel in 1991, with maximum sulfur content of 10 ppm (weight). At the time of its introduction, 30 the EC1 diesel was the first ultra-low sulfur diesel fuel in the world. The EC1 grade reached nearly full market penetration in Sweden already in the nineties, due to a strongly supportive tax 31 32 policy.

33

The EF's used for Helsinki therefore refer to fuel with less than 10 ppm sulphur content. As also
Finland (similarly to Sweden) used the lower S content vehicular fuel in 2008, the EF's used in
the manuscript are valid in this respect, despite the rapid decrease of sulphur content of motor
fuels at somewhat later years in many other European countries.

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We have also examined the situation regarding the fuel S content in all the other target cities, andincluded some discussion to section 3.1.2.

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42 We have also revised the discussion of the results for the Marylebone street.

The second most important issue which gets no mention is the vehicle fleet mix. For Rotterdam, a single emission factor is used for passenger cars, apparently ignoring the huge difference between gasoline and diesel fuels. This needs to be explicitly considered, and if a composite
emission factor is used, this needs to be justified.

4 **Response:**

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For Rotterdam, the COPERT IV emission factors were used. These have been specified
separately for motorway and for urban road traffic; for both cases there are categories both for
heavy and light duty vehicles, and passenger cars. It is correct that for Rotterdam, a composite
emission factor was used for passenger cars. This was the only possible choice, as the available
traffic flow data was also in composite form: a value per street for each of the following vehicle
categories: passenger cars, lorries and busses.

12

However, this does not mean that we would ignore the difference between the emission factors of diesel and petrol cars in Rotterdam. We have only assumed that the fractions of passenger cars equipped with diesel, petrol and vehicle technologies (compared with the total number of passenger cars) are not spatially variable within Rotterdam. We have therefore NOT ignored the difference between the emission factors of cars using gasoline and diesel fuels. The manuscript text was revised to explain these assumptions more clearly in section "Emission inventory for Rotterdam".

20

21 The authors recognise the distinction between the solid particle mode and the nucleation mode 22 particles formed in the exhaust plume by condensation, but give it insufficient attention. The 23 nucleation mode particles comprise semi-volatile organic compounds with a very small solid 24 core. Such particles can evaporate if entering an environment with low concentrations of the 25 associated vapour phase component (Dall'Osto et al., ACP, 6623-6637, 2011). Current 26 knowledge of such processes is insufficient to include a deterministic description or even a 27 meaningful parameterisation in numerical models. However, measurements of particle number 28 concentrations in cities include these particles, and one implication is that the measurement 29 method for particle number counts need to be specified. PNC measured by a CPC normally 30 exceeds that measured by an SMPS, even if the greater losses in the latter instrument are 31 accounted for. The usual reason is that the lower size cut of a CPC (depending upon model) will 32 be 2.5-7 nm, whereas most SMPS used in network monitoring have a lower size cut of 15 nm 33 (except Helsinki, which is specified), which cuts off the lower tail of the size distribution, 34 particularly important if size reduction due to evaporation has occurred. 35

- 36 **Response:**
- 37

38 The reviewer is correct, and we have therefore added a new section: "2.4 The measurements of 39 PN concentrations in target cities". The measurement methods and their size ranges have been 40 described in detail in this section.

41

42 Consequently, a modelling paper of this type should specify clearly what PN size range it is 43 seeking to model. The evaporative shrinkage and loss of nucleation mode particles may explain 44 why regional models tend to overestimate concentrations in this size range (p5888, lines 4-8).

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46 **Response:**

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- We totally agree that the paper should specify clearly what PN size range it is seeking to model.
 We have therefore written in the beginning of the section 2.2.1. the following: "The PN emission inventory includes particles in the 10 300 nm size range." The earlier part of this section was also clarified in the revised manuscript (to indicate that the inventories of this study address both anthropogenic and natural emissions).
- 7
- 8 We have also clarified the treatment of the particle size range used in the dispersion modelling.9 We added the following clarification to section 2.3.1. (2nd paragraph in the revised manuscript):
- 10

Although the size range of the anthropogenic emissions was assumed to be from 10 to 300 nm, the dispersion computations were performed for the size range from 10 to 1000 nm. There are several reasons for the relatively wider size range of the computations. First, due to condensation and coagulation, particles may grow to larger sizes than 300 nm. Second, small particles interact with larger particles (even larger than 300 nm); the latter can be originated from natural sources, such as, for instance, sea salt. The structure of the M7 model also includes the Aitken and accumulation size modes, with no strict separation at 300 nm.

18

19 While in the case of Oslo, a correction is made for double counting the model results for LOTOS

- and the urban model, it needs to be more explicit for other cities as to whether this was an issue,or whether LOTOS was used solely to provide a boundary condition for the urban model.
- 21 22

Response: Double counting due to the evaluation of regional background is not an issue for any of the target cities, as the LOTOS-EUROS predictions have in all the cases been taken from grid squares that surround the city (instead of the squares inside the city). Within the EPISODE modelling system, there is double counting between the two modelling components used within that system (but not with the LOTOS-EUROS model). However, that inaccuracy exists in only small part of the domain and is fairly small (as explained in the manuscript in section 'Dispersion and transformation modelling in Oslo').

30

Two lesser points: (1) the Hoek et al. (2010) study is not the only source of exposureresponse functions for PNC. These can also be taken from Atkinson et al. (2010), cited in this paper and from Stolzel et al., J. Expos. Sci. Environ. Epidemiol., 2007, 17, 458-467. (2) It is not acceptable for the maps in Figure 8 to use different scales, as this makes comparison between cities very

- 35 difficult.
- 36

37 <u>Response:</u> We added a citation to the recommended references. Stolzel et al 2007 was added to
 38 the list of references. We have also revised Fig. 8 so that the same scale is used in all the panels.
 39

40

41 Anonymous Referee #3

42

43 Received and published: 18 September 2015

44

45 The authors present an overview of the particle number concentrations (PNC) modelling

46 activities performed within the FP7 project TRANSPHORM.

- The PNC modelling is definitely a challenging activity and the proposed work is interesting because it verifies the possibility to model PNC at continental and city scales with state-of-theart air quality models without introducing relevant aerosol model developments. Therefore, the authors investigate the possibility to realize PNC evaluations to support air quality management.
- 6

The simulations have been performed at regional scale with the chemical transport model
LOTOS-EUROS and at city scale with different types of air quality models in different cities.

9

The heterogeneity of the modelling approaches used to reproduce PNC concentrations in the different cities strongly limits the comprehension of study results and the significance of the proposed conclusions. The reasons why a more harmonized analysis was not possible should be illustrated. If the use of different models in different cities can be understood on the basis of previous local tools development and use, the reason of different approaches in emission estimate and background concentration evaluation is hardly understandable.

16

The authors should revise the manuscript making efforts to explain the reasons why different sources like house heating, ports and airports activities are taken into account in some cities and not in the others. The paper revision should enable the reader to understand the reason of similarities and differences among the results obtained for the different target cities.

21

Response: For a detailed response, we would like to ask the reviewer to read our responses to the
 first and second comments of the reviewer number 1.

24

In short, the main reason why a more harmonized analysis was not possible is that the state of the emission information for PN is variable in terms of the target cities, and in terms of the source categories. However, we have substantially re-structured the manuscript to make this analysis as harmonized as possible in practice. For instance, we have added the contribution of shipping for Rotterdam, and evaluated much better the influences of other source categories for all the cities.

30

We have also completely re-written the interpretation of the results, especially that of Fig. 8 (concentration maps), to present more clearly the differences and similarities between different cities, in terms of source contributions and spatial distributions. Fig. 8 was also presented in a

harmonized form, using e.g. the same concentration legend for all the cities.

- 35 Section 2.2.2
- 36

The reasons of the different emission estimate for the different cities should be explained. If wood burning for house heating is considered relevant for PN emission in Oslo, why the general approach should be different in Helsinki, that is located in similar climatic area, and in the other cities. Why the harbor activities are not taken into account in Rotterdam? Why airport emissions have relevant effects in Athens and are not considered in London?

42

43 <u>Response</u>: Please see our response to the second comment of reviewer number 1 on the almost
 44 same topic.

1 Different emission factors for traffic source sector have been used in different cities. It is not 2 clear why it has been not possible or advisable to use the same emission factors for all the target 3 cities.

4

Response: Unfortunately, the PN emission factors (EF) are currently not sufficiently well
 known. There are no universal EF's that would be reliably valid for the various traffic fleets and
 climatic conditions throughout Europe. The best option was therefore to select the EF's that were
 considered to be the best applicable ones for each target city.

- 9 10 Section 2.3.1, pag 5886
- 11

12 The sentence "The PN emissions were converted to values that are compatible with the M7 13 module, using assumptions on the chemical composition of particulate matter." is rather obscure. 14 The used assumptions should be mentioned explicitly.

15

16 <u>Response:</u> We have revised and elaborated this description (in revised manuscript, the second to
 17 last paragraph in section 2.2.1).

- 18
- 19 Section 2.3.2, pag. 5891 20

From the description of the model simulation performed for Rotterdam it is not clear if the model computed hourly concentration time series like e.g. in Helsinki or if an annual average concentration was directly estimated has suggested by the sentence "The contribution of traffic emissions to annual average concentrations has been assumed to depend on the emission rate, the annual average wind speed and the road type."

26

27 <u>Response:</u> The modelling system in Rotterdam was used to compute only annually averaged 28 concentrations; this has been more clearly stated in the revised manuscript. We have also 29 checked and substantially revised the whole section "Dispersion modelling for Rotterdam", to be 30 more accurate and clear. We have also specified which models were used, by using model 31 acronyms; this is now in that respect consistent with the model descriptions for the other target 32 cities.

- 33 C
- 34 Pag 5893
- 35

The authors say that "The magnitude of these evaluated values for the urban background were checked, by comparing these with the measured PNC values at the station of North Kensington" but no information is provided on the results of the mentioned verification. It is not specified if any correction has been applied to the background concentration values.

40

41 **<u>Response:</u>** The LOTOS-E hourly values were scaled by multiplying them with the ratio of

42 annual average measured / predicted concentrations. These measured values were taken from the

43 regional background station of Harwell. This has been stated more clearly in the revised

- 44 manuscript.
- 45

- 1 We removed the comment that the urban background values were checked, as the final predicted
- 2 values were later on not only checked, but evaluated (as described in a later section in the3 article).
- 3 8 4

5 The sentence "For evaluating the annual concentration means, a weighting scheme was applied 6 on the daily concentration fields, based on a classification of local meteorological patterns" 7 makes the reader think that a limited number of days have been simulated for Athens, but no 8 detail on the number of days and their selection method is provided.

9

10 Response: Yes, a limited number of days were simulated for Athens, and the results were then 11 extended for the whole year. We have substantially revised and expanded the description of how 12 exactly this has been done. A few references were also added that include a more detailed 13 description of these methods.

14

15 It is not clear why LOTOS-EURO simulation results have not been used to estimate PNCbackground values in Athens.

17

18 **Response:**

19

The information in Table 1 regarding the use of LOTOS-EUROS values was not sufficiently clear in the original manuscript. We have presented the correct information more clearly in the revised manuscript. For clarify, we also specified for each city, which values were measured or modelled, and which represented urban, which regional background.

24

However, the methods for estimating regional or urban background were not identical in all the target cities. Our main aim was not a total harmonization of the methods, but instead the achieving of as realistic final results as possible, using physically well-founded methods. As the LOTOS-EUROS values were not as accurate as required in some cases, we either used measured values instead (for Helsinki and Athens), or scaled the predicted LOTOS-EUROS values using measured values (for Oslo). This has been presented more clearly in the revised manuscript, in section 2.1.

32

33 Section 3.1.1, pag 5894, lines 8-1034

After the evaluation of the 60% PN emissions attributed to the transport sector it would be interesting to add the estimate of the contributions attributed to the other major sectors.

37

38 **Response:** This interesting result, including the contributions of the other sectors, can be seen

directly from Fig. 2a. (this figure is included both in the original and the revised manuscript).

40 The transport sectors are represented by the columns 'road transport' and 'non-road transport'.

- 41 The other sectors include industry, residential combustion, etc.
- 42

43 For clarity, we added a comment to the first paragraph of section 3.1.1: "The other most 44 important sectors include industry (defined here excluding energy industries), residential

- 45 combustion, fugitive emissions and energy industries."
- 46

1	Line 24
2 3 4	The reference to Fig.2a should be probably to Fig. 3a.
5 6 7	<u>Response</u> : Yes, this has been corrected.
7 8 9	Pag 5895, line 4
10 11	The reference to Fig. 2b should be probably to Fig. 3b.
12 13	<u>Response:</u> Yes, this has been corrected.
14 15	Line 27
16 17 18 19 20	The meaning of the sentences "Although PN emission factors were not included in the uncertainty evaluation of the above mentioned study, it is possible to indirectly estimate also the uncertainties of the PN emissions. The latter were derived by combining the available experimental data on mass and PN emissions with COPERT PM emission factors" is not clear.
21 22 23 24 25	Response: The meaning is that although particle NUMBER was not included in the above mentioned study, particle MASS-based results can be used for indirectly evaluating the uncertainties of particulate number emissions. We have revised this paragraph in the manuscript to be clearer.
26 27	Section 4, pag 5904
28 29 30 31 32 33	The authors say that "the present knowledge is not sufficiently accurate regarding the variation of PN emission factors in terms of the various source categories, especially for shipping and small-scale combustion, and for various environmental conditions.". They should try to quantify the impact of these sources on the PNC in the cities where they have been included in the emission inventory.
34 35 36 37 38	Response: We agree that such a result would be interesting. Unfortunately, it was not possible (and outside the scope of this study), to perform a detailed source apportionment of PN concentrations for these cities. Performing such an analysis would involve a large amount of additional work.
39 40 41 42 43 44 45	Clearly, the fractions of the source contributions vary in terms of the part of the city, the traffic and street environment (e.g., street canyons vs. more open surroundings) and the season of the year. For instance, the influence of small-scale combustion is larger in winter and commonly larger in residential, suburban areas. There is also presently not a sufficient amount of information for analyzing the source contributions of PN for all sectors, for all cities. This could be a continuation study of the present work.

However, we have indicated the fractions of PN in emissions explicitly for the whole of Europe in Fig. 2, and for Oslo in Fig. 5. These are totally new results. We have also completely rewritten the discussion associated with Fig. 8 (spatial concentration distributions), including the best available estimates on the source contributions to concentrations. The abstract and conclusions sections were also revised accordingly.

6

7 The sentence "As expected, the most important local source category in terms of the PNC's was 8 local vehicular traffic in all the target cities." and the following discussion is quite questionable 9 in the proposed form because in some cities traffic emissions where the only one to be 10 considered.

11

12 <u>Response:</u> We have completely re-structured this analysis and its associated discussion to be 13 more specific, and better argumented. The importance of harbours and shipping was also 14 evaluated for all the cities in the revised manuscript (as well as that of several other source 15 categories for several cities).

- 16
- 17
- 18 References

19

Gidhagen, L., Johansson, C., Langner, J., Foltescu, V.L., 2005, Urban scale modeling of particle
number concentration in Stockholm Atmospheric Environment 39, 1711-1725.

Kumar, P, Garmory, A, Ketzel, M, Berkowicz, R and Britter, R (2009) Comparative study of
measured and modelled number concentrations of nanoparticles in an urban street canyon
Atmospheric Environment, 43 (4). 949 - 958. ISSN 1352-2310.

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- 27 28

29 Additional revisions

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We have improved the model evaluation on a regional scale, by considering the comparisons of predictions and measurements at 8 stations, instead of 3 as in the original manuscript. In the revised manuscript, we have considered also the correlations of the predicted and measured hourly timeseries of concentrations (not only monthly averaged concentration values). The text in section 3.2.1. and Fig. 6 were therefore revised.

36

We have redrawn Fig. 5, to be more easily readable. We also clarified the descriptions of thevarious emission sectors in the figure and in the text.

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1

List of all relevant changes made to the manuscript

- 2
- 3 Authors: One of the affiliations changed
- 4 Abstract: Revised according to the reviewers' comments: more source analysis added
- 5 Introduction: 2 references to concentration-exposure functions added
- 6 Table 1: texts clarified, and written more specifically
- 7 Section 2.1: Common features and differences of modelling systems discussed in more detail
- 8 Section 2.2.1: The use of the M7 module described more clearly
- 9 Emission inventories for Rotterdam and London: Discussed more thoroughly, according to
- 10 reviewers' comments
- 11 Section 2.3.1: The particle size distributions discussed better
- 12 Dispersion modelling for Rotterdam, London and Athens: written in more detail, and allowing
- 13 for the reviewers' comments
- 14 Section 2.4: A new section, added due to a reviewer's request
- 15 Section 3.1.2: The Sulphur content of fuels discussed
- 16 Fig. 5: A better version of fig. replaced
- 17 Section 3.2.1: better versions of figs. 6 and 7 added, and discussion revised accordingly.
- 18 Section 3.2.2: Better version of Fig. 8 added. Discussion of results re-written, especially
- 19 including a better description of source contributions.
- 20 Section 3.3: Better version of Fig. 9 added. Influence of regional background discussed better.
- 21 Modelling uncertainties discussed better.
- 22 Conclusions: The source contributions discussed better.
- 23 References: A few updates and miscellaneous corrections made.
- Annex added to show the locations of the most important emission sources.
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Modelling the dispersion of particle numbers in five European cities

J. Kukkonen (1), M. Karl (2), M.P. Keuken (3), H.A.C. Denier van der Gon (3), B.R. Denby (4,
9), V. Singh (5,10), J. Douros (6), A. Manders (3), Z. Samaras (6), N. Moussiopoulos (6), S.
Jonkers (3), M. Aarnio (1), A. Karppinen (1), L. Kangas (1), S. Lützenkirchen (7), Tuukka Petäjä
(8), Vouitsis, I. (6), R. S. Sokhi (5)

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4 5 6

- 12 (1) Finnish Meteorological Institute, Helsinki, Finland
- 13 (2) Helmholtz-Zentrum Geesthacht, Institute of Coastal Research, Geesthacht, Germany
- 14 (3) TNO, Netherlands Organization for Applied Research, Utrecht, The Netherlands
- 15 (4) Norwegian Institute for Air Research, Kjeller, Norway
- 16 (5) Centre for Atmospheric and Instrumentation Research (CAIR), University of Hertfordshire,
- 17 Hatfield, UK
- 18 (6) Aristotle University of Thessaloniki, Thessaloniki, Greece
- 19 (7) City of Oslo Agency for Urban Environment, Oslo, Norway
- 20 (8) University of Helsinki, Helsinki, Finland
- 21 (9) Norwegian Meteorological Institute, Oslo, Norway
- 22 (10) National Atmospheric Research Laboratory, Gadanki, Andhra Pradesh, India
- 23
- 24 Correspondence to: J. Kukkonen (jaakko.kukkonen@fmi.fi)
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26

27 Abstract

28

29 We present an overview of the modelling of particle number concentrations (PNC's) in five 30 major European cities, namely Helsinki, Oslo, London, Rotterdam and Athens, in 2008. Novel 31 emission inventories of particle numbers have been compiled both on urban and European scales. 32 We used atmospheric dispersion modelling for PNC's in the five target cities and on a European 33 scale, and evaluated the predicted results against available measured concentrations. In all the 34 target cities, the concentrations of PN were mostly influenced by the emissions originated from 35 local vehicular traffic. The influence of shipping and harbours was also significant for Helsinki, 36 Oslo, Rotterdam and Athens, but not for London. The aviation emissions in Athens were also 37 notable. The regional background concentrations were clearly lower than the contributions 38 originated from urban sources in Helsinki, Oslo and Athens. The regional background was also 39 lower than urban contributions in traffic environments in London, but higher or approximately 40 equal to urban contributions in Rotterdam. It was numerically evaluated that the influence of 41 coagulation and dry deposition on the predicted PNC's was substantial for urban background in 42 Oslo. The predicted and measured annual average PNC's in four cities agreed within 43 approximately ≤ 26 % (measured as fractional biases), except for one traffic station in London. 44 This study indicates that it is feasible to model PNC's in major cities within a reasonable 45 accuracy, although major challenges remain in the evaluation of both the emissions and 46 atmospheric transformation of PNC's.

4 1. Introduction

5

1 2 3

6 Airborne particulate matter (PM) affects human health and climate (e.g. Smith et al., 2009). 7 While a large base of scientific information exists on particle mass, especially for PM₁₀ and 8 PM_{2.5}, there are substantially less studies on particle number (PN) and in particular on modelling 9 dispersion of PN's in urban areas (e.g., Kumar et at., 2013). This may be attributed to (i) scarcity 10 of reliable information on emissions, (ii) the greater complexity of physical and chemical 11 atmospheric processes and (ii) lack of monitoring data of PN. The majority of urban particles - in 12 terms of number concentration - are ultrafine particles (UFP), i.e., particles with diameter (D_p) < 13 100 nm, originating mainly from traffic-related emission (e.g., Morawska et al., 1998). The rapid 14 transformation processes of PN after emissions in ambient air, such as condensation and 15 evaporation, coagulation, dry deposition and dilution pose challenges for dispersion modelling, especially on an urban scale (e.g., Pohjola et al., 2003, Ketzel et al., 2004; Kittelson et al., 2004; 16 17 Kumar et al., 2011, von Bismarck-Osten et al. 2013). In addition, PN measurement techniques 18 are also more complex and resource-consuming, compared with the measurements of particulate 19 mass fractions.

20

21 Although attention on the health effects of particulate matter has been focused on particle mass 22 fractions, a number of studies are indicating that UFP's may have specific health effects. UFP's 23 are poorly filtered in the human respiratory tract after inhalation, and such particles can penetrate the epithelial cells of the lungs and accumulate in lymph nodes (Nel et al., 2006). 24 25 Epidemiological and toxicological studies show a strong correlation between exposure to 26 ultrafine particles and various health endpoints, such as cardiovascular hospital admission (short-27 term exposure), mortality (long-term exposure) and neurological effects (Oberdörster et al., 28 2004; Delfino et al., 2005; Atkinson et al., 2010; Franck et al., 2011; Daher et al., 2013; Loane et 29 al., 2013).

30

There is a severe lack of representative sets of urban measurements of particle number concentrations (PNC's) that could be used in epidemiological studies, when compared to particle mass. Similarly, the scientific literature is scarce on predicting the dispersion of PN in urban environments. It is therefore necessary to develop and evaluate dispersion modelling systems capable of reliably predicting PNC's.

36

37 Combustion is a direct source of UFP's, and secondary particle formation may occur via 38 atmospheric reactions and condensation of semi-volatile components produced in photochemical 39 reactions (Kulmala et al. 2013, Kulmala et al. 2014, Kumar et al., 2014). Combustion of carbon-40 based fuels for power generation, heating and transport are important sources for PN emissions 41 (Shi et al., 2001; Obaidullah et al., 2012; Kittelson et al., 2006; Maricq, 2007; Buzea et al., 42 2007; Kumar at al., 2013; Pant and Harrison, 2013, Keuken et al., 2015a and b). In most 43 European cities, road traffic emissions of PN are expected to be the most important source for 44 exposure of the population, due to the near-ground emissions and the vicinity of road traffic to 45 populated areas.

1 The importance of aerosol processes has been analyzed via aerosol process time scales by Zhang 2 and Wexler (2004) and Ketzel and Berkowicz (2004), and with aerosol dynamics model 3 simulations by Jacobson and Seinfeld (2004). Pohjola et al. (2003) simulated the transformation 4 and dilution of particulate matter on distance scale of less than 100 m from a road in an urban 5 area. As expected, dilution was found to be the most important process affecting the PNC's; 6 however, condensation of an insoluble organic vapour was also found to be important, if its 7 concentration exceeds a certain threshold value. Ketzel and Berkowicz (2004) evaluated that the 8 influence of dry deposition would be irrelevant on an urban time scale. Kerminen et al. (2007) 9 evaluated that coagulation, condensation and evaporation could be important in conditions, 10 where dilution with cleaner background air is restricted.

11

12 Small-scale combustion may also be a prominent source of PNC's in winter (Glasius et al., 13 2008). Elevated levels of PN have also been found in specific areas, such as, near harbors, 14 refineries and in particular near airports (González and Rodríguez, 2013; Westerdahl et al., 2008; 15 Zhu et al., 2011; Keuken et al., 2012; Hsu et al., 2014). Whereas most of the state-of-the-art 16 chemical transport models include treatments for aerosol size distributions and microphysics 17 (Kukkonen et al., 2012), such treatments are substantially less commonly included in urban scale models. There are currently very few models, which are especially designed to predict particle 18 19 number concentrations by taking into account particle dynamics. Kumar et al. (2012) presented a 20 review on the importance of aerosol transformation processes at various urban scales and 21 environments.

22

A first European size-resolved anthropogenic PN emission inventory was compiled in the framework of the EU-funded EUCAARI project (Denier van der Gon et al., 2010). Consolidated emission factor data bases (e.g., COPERT, PARTICULATES and TRANSPHORM) have recently become available to establish PN emission inventories in Europe; these have been reviewed by Kumar et al. (2014). According to the inventory by Paasonen et al. (2013), for the 28 EU countries in 2010, road transport contributed over 60 % of the total PN emissions, nonroad transport (including partly also shipping) 19 % and domestic combustion 13 %.

30

31 The first stage between the point of emission (vehicle tailpipe) and the kerbside is characterized 32 by strong turbulence generated by the moving vehicles. According to Zhang and Wexler (2004), 33 the initial stages of dilution within a few first seconds would be accompanied with nucleation. 34 On-road measurements by Rönkkö et al. (2007) demonstrated that the nucleation mode was 35 already present after 0.7 s residence time in the atmosphere. However, the modelling of 36 nucleation will require detailed information about the environmental conditions very near the 37 tailpipe (e.g., temperature gradient, and chemical composition and concentrations of volatile 38 nucleating vapours). Nucleation mode particles grow rapidly by condensation of high-molecular 39 weight low-volatile hydrocarbons from the unburned lubrication oil and sulphur compounds

- 40 (Kittelson et al., 2006).
- 41

42 In the second stage between the street and a few hundred meters away from the street, 43 atmospheric turbulence, induced by wind and atmospheric instability, is the main cause for 44 dilution of particle concentrations. In this stage, condensation/evaporation and dilution become 45 the major mechanisms in altering the particle size distribution, while coagulation and deposition

46 play minor roles (Zhang et al., 2004). In the third stage, between street canyon/street

neighborhood and the urban background, the number size distribution is altered by multiple
processes, such as dilution with cleaner air, entrainment of polluted air, condensation of vapors,
oxidative ageing, and coagulation of particles (e.g., Wehner et al., 2002).

4

5 Asmi et al. (2011) examined aerosol number size distribution data from 24 European field 6 monitoring sites in 2008 and 2009. The data was collected from the stations at the EUSAAR 7 (European Supersites for Atmospheric Aerosol Research) and GUAN networks (German 8 Ultrafine Aerosol Network), and represented mainly regional background or remote locations. 9 They categorized the aerosol to several types: central European aerosol, Nordic aerosol, 10 mountain sites and southern and western European regions, and analyzed the seasonal 11 characteristics and patterns of the various size modes.

12

Hussein et al. (2007) and Pohjola et al. (2007) conducted a field measurement campaign near a major road in an urban area in Helsinki in February, 2003. Measured PNC data at various distances from the road was compared with dispersion and aerosol process model predictions. A similar measurement campaign was conducted downwind of a motorway in Rotterdam (Keuken et al., 2012). Size-resolved PNC measurements were compared with dispersion modelling and an aerosol process model (Karl et al., 2011). Both these studies concluded that dilution was shown to be the most important process.

20

21 Gidhagen et al. (2005) implemented a three-dimensional dispersion model in Stockholm and 22 presented the spatial distribution of number concentrations over the whole city. Typical number concentrations in the urban background of Stockholm were 10 000 cm⁻³, and approximately three 23 24 times higher close to a major highway and seven times higher within a densely trafficked street 25 canyon. Coagulation was found to contribute to losses of PNC's of only a few percent as 26 compared to inert particles, while including dry deposition resulted in PNC losses of up to 25% 27 in certain locations. However, removal of PN's due to coagulation and deposition was more 28 significant during peak episodes.

29

30 This study is part of the European Union funded research project TRANSPHORM (Transport related Air Pollution and Health impacts - Integrated Methodologies for Assessing Particulate 31 32 Matter). This project was one of the very few international projects, where dispersion models 33 have been developed and applied to predict spatially and temporally resolved concentrations of 34 PN for exposure and health applications (ww.transphorm.eu). The cities Helsinki, Oslo, 35 Rotterdam, London and Athens were involved to test the methodologies developed within the 36 TRANSPHORM project at an urban scale. These cities were selected in order to include at least 37 one major urban agglomeration from the following regions: (i) the Nordic countries (Helsinki 38 and Oslo), (ii) the central and north-western Europe (Rotterdam and London) and (iii) the 39 Mediterranean region (Athens).

40

41 Health studies for PN are scarce, and currently there are only concentration-response functions

42 based on expert judgment. According to Hoek et al. (2010), there will be a 0.3 % increase in all-

43 cause mortality per 10^3 particles per cm³. Source-exposure functions for PN have also been

44 presented by Stolzel et al. (2007) and Atkinson et al. (2010). Von Klot et al. (2005) underlined

45 similar effects for hospital re-admissions of a susceptible population, in cases, for which the

46 aerosol number increased 10^3 particles per cm³ or aerosol mass by 10 µg m⁻³. However, in view

of the potential health effects for exposure to PNC's, there is a need to combine epidemiological
 data and PNC's with a high spatial resolution.

3

4 The aim of this article is to present an overview of the modelling of PNC's on an urban scale in 5 five major European cities, presented in Fig. 1: Helsinki, Oslo, Rotterdam, London and Athens. 6 The target cities represent megacities, such as London (population of approximately 8.3 million) 7 and Athens (we address here Greater Athens, 3.5 million), and other major cities, such as 8 Helsinki Metropolitan Area, Oslo and Rotterdam (populations of 1.0, 0.6 and 0.6 million, 9 respectively). For simplicity, we refer to Helsinki Metropolitan Area simply as 'Helsinki' in the following. The primary year used in the computations is 2008. The modelling of PNC's for these 10 cities has been presented in the present article for the first time. The previous literature also does 11 12 not contain any compilations of PNC modelling for several cities.

13 14



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We address emission inventories and emission modelling of PN, dispersion modelling of PNC's, numerical results on the annual average spatial distributions in the target cities and evaluation of the predicted results against measured PNC's. The main scientific goals were (i) to evaluate the capability of models to predict PNC's in several European cities, (ii) to examine the predicted spatial characteristics of PN in the selected cities, (iii) to evaluate the contributions of various source categories on the concentrations, and (iv) to highlight areas of improvements in modelling PN for health based studies.

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- 28

29 **2. Modelling methods**

2 In this section the computational methods are presented, which were used for the evaluation of 3 PNC's in the five target cities. We address both the methods for the evaluation of emissions, and 4 the atmospheric dispersion modelling systems. For practical reasons, it was not possible to 5 completely harmonize the computations, by using only one modelling system for all the cities. 6 All of the urban emission and dispersion modelling systems were therefore locally or nationally 7 developed ones; these were different for each city. However, the regional background 8 concentrations for all the urban scale modelling systems were computed with the same model, 9 the LOTOS-EUROS chemical transport model (Schaap et al., 2008). We have therefore also 10 briefly discussed a new European-scale emission inventory used as input for the above 11 mentioned regional scale chemical transport model.

12 13

14 **2.1 Overview of the PNC computations in the target cities**

15

16 For readability, selected summary information has been presented in Table 1 on the urban scale

17 computations. The more detailed information will be presented in the following sections.

- Table 1. Overview information on the computational methods and the evaluation of predictions
- 2 in the five target cities for 2008.

	Helsinki	Oslo	Rotterdam	London	Athens
Traffic flows and urban scale emissions	Traffic planning model, vehicular emission factors based on Gidhagen et al.(2005), shipping emission model STEAM2	Local traffic data, TRANSPHORM emission database (Vouitsis et al., 2014) with temperature correction, STEAM2	Local traffic data, COPERT IV (Gkatzoflias et al., 2012) and TRANSPHORM emission database	Local traffic data, emission factors from Jones and Harrison (2006)	Local traffic data, TRANSPHORM emission database, Petzold et al. (2010) and (Lee et al, 2010).
Meteorological data and its pre- processing	Meteorological pre-processor model MPP-FMI, based on measured sounding data and other data from two stations	Diagnostic wind field model, based on measured data at two sites	Measured data from local airport	Meteorological pre-processor model GAMMA-met, based on measured data at one station	Prognostic model MEMO, based on measured data at one location
Urban source categories included	Vehicular traffic, importance of shipping and major stationary sources separately evaluated	Vehicular traffic, shipping, small- scale combustion, industry, other sources	Vehicular traffic, shipping, airports and refineries included in the regional background	Vehicular traffic, all the sources influencing urban background	Vehicular traffic, shipping, aviation, stationary sources
Regional or urban background concentrations and their evaluation	Urban background values measured at an urban background station	LOTOS-EUROS, regional background values at the grid squares that surround the city, scaled using measured regional background values	LOTOS-EUROS, regional background values at a grid square that surrounds the city	LOTOS- EUROS, regional background values at grid squares that surround the city	The measured regional background PNC values by Kalivitis et al. (2008). The values of other relevant compounds were extracted from LOTOS-EUROS at grid squares surrounding the city
Urban modelling system	CAR-FMI, PN treated as tracer	EPISODE, Aerosol process parameterisation included	URBIS:street- canyon and line- source models; PN treated as tracer	OSCAR, PN treated as tracer	MARS-aero, PN treated as tracer
Evaluation of predictions against measured concentrations	At one measurement station for one year	At two measurement stations, for three months	At two measurement stations for one year	At two measurement stations, for one year	Measurements were not available for 2008

1 The TRANSPHORM project emission database was used on an urban scale in three of the target 2 cities. Two urban modelling systems applied a meteorological pre-processing model, two others 3 other meteorological models, and one modelling system applied directly measured data. All the 4 models included the emissions from vehicular traffic. The shipping emissions were explicitly 5 included in the computations of Oslo, Rotterdam and Athens, and the importance of primary 6 shipping emissions was separately evaluated for Helsinki (Soares et al., 2014). For London, the 7 local scale shipping emissions were not taken into account, as its importance was found to be 8 negligible. Several of the models included also the emissions from major and/or small-scale 9 stationary sources and other source categories.

10

11 The urban scale emission and dispersion modelling systems were specific for each target city. 12 All of the urban dispersion modelling systems used for Helsinki, Oslo, London and Rotterdam 13 are multi-source Gaussian dispersion and transformation systems. These can also allow for 14 dispersion in street canyons; however, these street canyon dispersion models were not used in 15 this study (except for using the semi-empirical street canyon model for Rotterdam). The 16 modelling system for Athens is based on the combined use of a meteorological model and a 17 chemical transport model. All these modelling systems have previously been extensively 18 evaluated against experimental data. 19

Regional background concentrations of PN were derived from the LOTOS-EUROS model computations for three target cities (Oslo, London and Rotterdam), based on the predicted values at grid squares that surrounded these cities. For obtaining an improved accuracy of the computations, we used measured values for the urban or regional background for Helsinki and Athens, respectively. The predicted LOTOS-EUROS regional background values were scaled, using the ratios of measured and predicted annual average concentrations, for Oslo and London.

The aerosol transformation processes were taken into account in the LOTOS-EUROS computations. Measured PNC data was available in four of the cities, in three of these for a complete year; however, only at one or two measurement stations for each city.

30 31

32 **2.2 Emission inventories**

34 We describe in this section both a new European scale emission inventory and the urban 35 emission inventories in the five target cities.

36

33

37

38 **2.2.1 European-scale emission inventory**

39 40

40 A new emission inventory was compiled for the EU-wide anthropogenic transport activities, 41 supplemented by the anthropogenic non-transport activities. In addition to this anthropogenic 42 emission inventory, we included various natural emission sources in the LOTOS-EUROS 43 computations. These included sea spray aerosol emissions, and the dust emissions from road 44 suspension, agriculture and bare soils. These were modelled as described by Schaap et al. (2009).

- 1 The baseline emission data in the anthropogenic emission inventory contains the following 2 substances: NO_x , SO_2 , non-methane volatile organic compounds (NMVOC), CH_4 , NH_3 , CO, 3 PM_{10} , $PM_{2.5}$, EC (elemental carbon), B[a]P (benzo[a]pyrene) and PN (Denier van der Gon et al.,
- 4 2014). The anthropogenic PN inventory includes particles in the size range of 10-300 nm.
- 5
- 6 The emission data can be calculated for the individual countries; the official UN ISO3 Country 7 Codes were used. We have used three groups of countries. The EU15+ group is defined to 8 include EU15, and Norway and Switzerland. The EU12+ group contains the New Member 9 States, Malta and European Non-EU countries; the latter refers to the other European countries in 10 the United Nations Economic Commission for Europe domain. The EU27+ group consists of 11 EU15+ and EU12+. Emissions from international shipping have been estimated for the various 12 European sea regions.
- 13

The first European particle number emission inventory was made in the EU FP6 project EUCAARI (Denier van der Gon et al., 2010a; Kulmala et al., 2011). This inventory was used as a starting point for the present study. For the different transport modes (road, rail, air and maritime navigation), a new bottom-up PN emission estimate was made, including also technologies and activities in the future years, 2020 and 2030.

19

The above mentioned PN emission inventory includes only anthropogenic sources; the emissions from mainly natural sources such as, e.g., wild-land fires, windblown dust and sea salt are not included. The inventory also does not include vegetation related emissions (e.g., Guenther et al. 1995), or the formation of PNC's from biogenic VOC's (e.g., Paasonen et al. 2012).

24

25 The above mentioned emission inventory describes internally mixed PN emissions originated 26 from several source categories in 12 size bins, covering the particle dry diameter range from 10 27 to 250 nm. The LOTOS-EUROS model in combination with the M7 module uses the PN 28 emission as input; that is converted into the Aitken and accumulation modes used in the M7 29 module. The M7 module additionally requires the associated masses of black and organic 30 carbon, sulfate and mineral dust, and a division to soluble and insoluble material. Using the 31 sulfate content of the internally mixed particles as a proxy, the PN concentrations were attributed 32 to the soluble and insoluble modes.

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35 **2.2.2 Urban-scale emission inventories in the target cities**

36 Emission inventory for Helsinki

The emission inventory included exhaust emissions from vehicular traffic for the network of roads and streets in the Helsinki Metropolitan Area (HMA). The traffic volumes and average travel speeds of each traffic link were computed using the EMME/2 transportation planning system (INRO, 1994). Traffic volume data in 2008 was used as input for the estimation of annual average road traffic emissions in the HMA. The final emission inventory consisted of average hourly emissions for each line source over the year, separately for weekdays, Saturdays and Sundays.

- The emission factors for vehicular traffic determined by Gidhagen et al. (2005) in Stockholm have been used. The emission factors corresponding to Stockholm were used, as these were estimated to optimally correspond to the climatic and traffic conditions in Helsinki. These values are 2.70 10¹⁵ particles/vehicle/km and 1.8 10¹⁴ particles/vehicle/km for heavy and light-duty vehicles, respectively. These values were determined for driving speeds less than 70 km/h; however, we have applied these values for all urban roads included in the computations.
- 8

9 In addition to the computations for 2008, we computed the PNC's at the roadside traffic station 10 at Ring road 1, Malmi (called simply as 'Ring road 1' in the following) in 2012, for model 11 evaluation purposes. For the hourly computations in 2012, the 2008 traffic volume data was 12 scaled using the ratio of the total vehicular mileages (km/a) in the HMA in 2008 and 2012. These 13 mileage values were obtained from the national traffic emissions data archive LIPASTO (http:// 14 http://lipasto.vtt.fi/indexe.htm).

15

16 The importance of the shipping emissions was evaluated based on Soares et al. (2014). They 17 showed using the STEAM2 shipping emission modelling (Jalkanen et al., 2012; Johansson et al., 18 2013) that the contribution of primary shipping emissions of to the concentrations of $PM_{2.5}$ are 19 only 3 % on the average in the Helsinki Metropolitan Area. However, this contribution can be 19 higher than 20 % in the vicinity of the harbours (within a distance of approximately one 20 kilometer).

22

23 Emissions from stationary sources were not included. However, major stationary sources in the 24 area (these are mostly power plants) have previously been shown to have negligible effect on the 25 PM_{2.5} concentrations near the ground level in Helsinki (Kauhaniemi et al., 2008); the same was 26 assumed to be valid also for PNC's. Emissions from small-scale combustion were not taken into 27 account, as their spatial distribution was not known with sufficient accuracy. The contribution of 28 small-scale combustion to the total PM_{2.5} emissions in Helsinki Metropolitan Area has been 29 estimated to be 23 % in 2009 (Malkki et al, 2010). The emissions of PM_{2.5} originated from 30 aviation in the Helsinki Metropolitan Area were about 17 % of the total road traffic PM₂₅ 31 emissions in the area in 2008.

32

33 Emission inventory for Oslo

Emission factors for traffic exhaust (measured at an ambient temperature of +33 °C) were extracted from the emission database of the TRANSPHORM project (Vouitsis et al., 2014) Emission factors for PN in Oslo and in other studies (Klose et al., 2009; Olivares et al., 2007) have been found to have a significant dependence on ambient air temperature. A dependence of -3%/K has been applied to the Oslo traffic emissions, leading to significantly higher emission factors in the cold winter period (approximately double) than those provided in the emissions database.

41

Shipping emissions were based on the STEAM2 emission model (Jalkanen et al., 2012;
Johansson et al., 2013). Emissions for PN were based on the CO₂ emissions, converted firstly

44 back to fuel consumption, and then PN emissions were calculated using an emission factor of

1 1×10^{16} particles/(kg fuel), recommended by Petzold et al. (2010). Shipping emissions were 2 evaluated in a domain of 29 x 18 km² and thus only included shipping in the Oslo fjord area.

3

Domestic heating emissions of PN, due mostly to wood burning, were calculated based on a previously compiled $PM_{2.5}$ inventory. A conversion factor of $4x10^{14}$ particles/(g $PM_{2.5}$) emitted was used to convert $PM_{2.5}$ emissions to PN emissions, based on the data presented in Hedberg et al. (2002). Other emissions concerning combustion sources, i.e. agricultural, industrial and mobile sources use the existing $PM_{2.5}$ emissions inventory and convert to PN using a ratio similar to diesel truck emissions; a conversion factor of $3x10^{15}$ particles/(g $PM_{2.5}$) was applied.

10

11 Emission inventory for Rotterdam

Road traffic data and road characteristics were obtained from a national database (www.nslmonitoring.nl). Road traffic data contains information about the number of vehicles, speed, congestion and fleet composition in-between traffic links for every major road and motorway in Rotterdam. The road characteristics refer to, e.g., the width and height of buildings along the road.

17

18 The following emission factors from COPERT IV (Gkatzoflias et al., 2012) and the 19 TRANSPHORM database have been applied: (i) for motorway traffic, 10^{15} particles km⁻¹ veh⁻¹ 20 for heavy and light duty vehicles, and $0.3*10^{15}$ particles km⁻¹ veh⁻¹ for passenger cars, and (ii) for 21 urban road traffic, $0.5*10^{15}$ particles km⁻¹ veh⁻¹ for heavy and light duty vehicles and buses, and 22 $0.3*10^{15}$ particles km⁻¹ veh⁻¹ for passenger cars.

23

24 As mentioned above, two composite emission factors were used for passenger cars, one for 25 motorway traffic, and the other one for traffic in urban roads. This was necessary, as the 26 available traffic flow data was also in composite form, including a value for each street for each 27 of the following vehicle categories: passenger cars, lorries and busses. The assumption of 28 composite emission factors implies that the fractions of passenger cars equipped with diesel, 29 petrol and vehicle technologies are not spatially variable within the city. However, these 30 composite emission factors take into account, e.g., the differences between the emission factors 31 of cars using gasoline and diesel fuels.

32

Airports and refineries can be potentially important sources for PN emissions (Keuken et al.,
2015a, b). However, the Airport Rotterdam is a relatively small airport. E.g., the annual average
number of passengers is smaller than 10 % of that of the main airport in the Netherlands, the
Schiphol Airport in the vicinity of Amsterdam. Major refineries are located at a distance of 10
km west of the modelling domain. Both the emissions from the Airport Rotterdam and refineries
have therefore been included in the regional background.

1 <u>Emission inventory for London</u>

The road traffic data for London have been obtained from London Atmospheric Emission Inventory (LAEI; GLA, 2010). Each road link was characterised by the amount of vehicles per day per vehicle category and mean speed. The traffic activity data were disaggregated by vehicle categories such as motorcycles, cars including taxis, buses, light goods vehicles (LGV) and heavy goods vehicles (HGV). The HGV's are further subdivided into articulated HGV's and rigid HGV's categories. The fleet compositions have been further subdivided as per fuel type, weight, engine size and emission standards.

9

10 The emission model in current version of the OSCAR system commonly uses the emission 11 functions and factors based on COPERT IV (Gkatzoflias et al., 2012) and Department for 12 Transport (DfT) emission data base. However, due to the unavailability of emissions in that 13 database for PN's, emission factors from Jones and Harrison (2006) have been used in this study.

14

15 According to the LAEI (GLA, 2010), the most important source categories of PM₁₀ in London in

16 2015 were road transport, agriculture-nature and industrial processes. The PM_{10} emission from

17 shipping was only 2 ton/year, which is a negligible fraction (0.08 %) of total emissions. We

18 therefore neglected the influence of shipping in the case of London.

19

20 Emission inventory for Athens

For Athens PN emissions included vehicular traffic, shipping and aviation. Emission factors for traffic exhausts were taken from the TRANSPHORM emission database (Vouitsis et al., 2014). Emissions from shipping and the major ports, and airport emissions were calculated on the basis of the operational action plan for air pollution management in Athens. This plan was developed for 2004, using activity and fuel consumption data (Samaras et al., 2012). The emission factor used for shipping was 10¹⁶ particles /(kg fuel) according to Petzold et al. (2010), and for aviation 6 10¹⁴ particles /(kg fuel), assuming a fuel sulphur content of 1000 ppm (Lee et al, 2010).

28 29

30 2.3 Dispersion and transformation modelling 31

First, we address the dispersion modelling on a continental scale, which provided the regional background concentrations for urban dispersion modelling. Second, we discuss the urban scale dispersion modelling systems used in the five target cities.

35 36

2.3.1 Chemical transport modelling on a European scale

38

The chemistry-transport model LOTOS-EUROS (Schaap et al., 2008) was used in this study to evaluate the regional background PNC's. Compared with other widely used chemical transport

- 41 models in Europe, reviewed by Kukkonen et al. (2012), the model is of intermediate complexity.
- 42 The relevant processes have been parameterized in such a way that the computational demands

are modest. The LOTOS-EUROS model has been included in several international model intercomparison studies that have addressed the dispersion and transformation of ozone and particulate matter (e.g., Stern et al., 2008 and Solazzo et al., 2012). The model performance has in these model inter-comparisons been comparable with other European chemical transport models.

6

7 The M7 aerosol microphysics module (Vignati et al., 2004) was coupled to the LOTOS-EUROS 8 model. This module accounts for nucleation and condensation of H_2SO_4 , and coagulation of

9 particles. The default nucleation scheme originally based on Vehkamäki (2002) was replaced by
 10 the activation type parameterization of Kulmala et al (2006), which is better suitable for the

- 11 boundary layer.
- 12

Formation of H_2SO_4 was based on the default gas-phase chemistry of LOTOS-EUROS, using emission inventories provided by the MACC project (TNO-MACC emission inventory; Pouliot et al., 2012) and the TRANSPHORM emission inventories. The PN emissions were converted to values that are compatible with the M7 module, using assumptions on the chemical composition of particulate matter (cf. section 2.2.1).

18

Although the size range of the anthropogenic emissions was assumed to be from 10 to 300 nm, the dispersion computations were performed for the size range from 10 to 1000 nm. There are several reasons for the relatively wider size range of the dispersion computations. First, due to condensation and coagulation, particles may grow to larger sizes than 300 nm. Second, small particles interact with larger particles (even larger than 300 nm); the latter can be originated from natural sources, such as, for instance, sea salt. The structure of the M7 model also includes the Aitken and accumulation size modes, with no strict separation at 300 nm.

26

27 Two sets of simulations for Europe were made. (i) The first set was based on the meteorology of 28 2008, and was used for model evaluation. This set had a $0.5 \ge 0.25$ longitude-latitude grid, for a 29 European domain from 15 W to 35 E and from 35 to 70 N. The concentrations for particle 30 number were assumed to be negligible at the boundaries of the domain. (ii) The second set of simulations was performed for the meteorology and the emissions of 2005. Additional 31 32 simulations were performed for each target city, on a finer 0.125 x 0.0625 longitude-latitude 33 grid, for each city in a domain that covered an area of 3° x 1.5°, using the European-scale 34 simulation for boundary conditions.

35

There are several processes that contribute to uncertainties in the model results. Nucleation mode particles contribute substantially to the total particle numbers. However, several parameterizations for nucleation processes are available, and it is not in all cases clear, which are the optimal ones. The uncertainties associated with the modelling of particle nucleation have mainly an impact on the number concentration of particles smaller than 100 nm (e.g., Fountoukis et al., 2012).

42

43 Some atmospheric species are not represented in the M7 module. For example, secondary

44 aerosol formation from biogenic emissions (such as, isoprene and terpene) is not taken into

45 account. Riipinen et al. (2011) investigated the role of condensable vapours on the growth of 46 freshly nucleated particles until the cloud condensation nuclei size, and proposed a semiempirical modelling approach. Secondary organic vapours can condense on existing particles, and thus contribute to their growth. This process increases the probability of such particles to reach the sizes that are cloud condensation nuclei (CCN) active, before getting scavenged by the background particle population. Secondary organic aerosol from biogenic origin therefore may substantially contribute to the PCN's.

6

7 The emissions of condensable gases from combustion processes are also not taken into account 8 in the modelling; these could potentially contribute, e.g., in areas with substantial residential 9 wood burning. In regions with intensive NH_3 emissions (e.g., from agriculture and animal 10 husbandry), the impact of secondary inorganic aerosol may be significant on number and size 11 distribution of particulate matter; this is not accounted for in the M7 module (Vignati et al., 12 2004).

13

The omission of biogenic secondary aerosol causes inaccuracies to the PM size distribution. The inaccuracies are the largest in the case of the smallest particles. The modelled sum of the Aitken and accumulation mode particle number concentrations are therefore considered the most appropriate quantity to represent regional background PNC's in this study (compared with using the number concentration of the nucleation mode particles).

19 20

21 **2.3.2 Urban scale dispersion modelling**

22

For each modelling system, we address (i) the urban dispersion modelling system and its implementation, (ii) the evaluation of meteorological variables (used as input for the urban modelling), and (iii) the assessment of regional background concentrations.

26

27 Dispersion modelling for Helsinki

The urban scale dispersion of vehicular emissions was evaluated with the CAR-FMI model
(Contaminants in the Air from a Road – Finnish Meteorological Institute; Kukkonen et al., 2001,
Härkönen et al., 1996). The model computes an hourly time-series of the pollutant dispersion

from the line source. The dispersion equation for the line source model is based on a semianalytical solution of the Gaussian diffusion equation for a finite line source. The dispersion parameters are modelled as a function of the Monin-Obukhov length, the friction velocity and

35 parameters are modelled as a function of the Mohim-Obuknov length, the friction velocity a 34 the mixing height. Traffic-originated turbulence is modelled with a semi-empirical treatment.

- 35
- The receptor grid intervals range from 20 m in the vicinity of major roads to 500 m on the outskirts of the area. The concentration values were computed at 18 692 receptor points.
- 38

39 Input data needed by the dispersion model was evaluated using a meteorological pre-processing

40 model (MPP-FMI) that has been adapted for an urban environment (Karppinen et al., 2000c).

41 The MPP-FMI model is based on the energy budget method of van Ulden and Holtslag (1985).

- 42 The model utilises meteorological synoptic and sounding observations, and its output consists of
- 43 estimates of the hourly time series of the relevant atmospheric turbulence parameters and the
- 44 boundary layer height. The computation is based on a combination of the data from the stations

1 at Helsinki–Vantaa airport and Helsinki-Kumpula (3-hour synoptic weather observations), and Jokioinen (soundings).

2 3

4 The urban background concentrations of PN both for 2008 in 2012 were estimated to be equal to 5 the measured hourly values at an urban background measurement site located at Kumpula in 6 Helsinki. This station is part of the network of stations called "Station for Measuring Ecosystem 7 - Atmosphere Relations", SMEAR-III (Järvi et al., 2009). This data contained PNC's in the 8 particle size range from 3 to 950 nm. The measurements and data analysis were conducted 9 according to Wiedensohler et al. (2012). For the computations in Helsinki, we therefore did not 10 use the regional background concentration values predicted by the LOTOS-EUROS model.

- 11
- 12 13

Dispersion and particle transformation modelling for Oslo

14

15 Calculations of concentrations were carried out using the EPISODE dispersion model (Slørdal et 16 al., 2003), which is part of the integrated air quality management tool AirQUIS (Slørdal et al., 17 2007). The EPISODE model consists of a gridded Eulerian model coupled with a Gaussian line 18 source model for modelling the local contribution at receptor points near roads. The Eulerian 19 grid model uses a 1x1 km² grid covering Oslo. There are 13 vertical layers in the model, up to 20 the height of 4000 m, with the lowest layer being 10 m thick. Emissions from traffic sources are 21 placed in the lowest layer, whilst emissions from domestic heating, industry and shipping are 22 placed in the layers between 10 and 35 m.

23

24 Receptor points within 500 m of a road include line source calculations, using the Gaussian line 25 source model in EPISODE, otherwise only the Eulerian model contributes. The model coupling 26 leads to a double counting of the emissions near roads, which has been estimated to contribute a 27 maximum increase of 5 - 20% to the model concentrations at receptor points near roads. The 28 receptor points are placed at monitoring sites, and at aggregated home addresses, at the centre of 29 population mass within a $100 \times 100 \text{ m}^2$ grid.

30

31 The air pollution originated in vehicular traffic tunnels has been modelled assuming that there 32 has been no deposition of particles within the tunnels. The tunnel exits are therefore treated 33 simply as exit points of polluted air.

34

35 Meteorology is generated in the model using the diagnostic wind field model MCWIND. The 36 MCWIND model uses meteorological measurements and interpolates these in space, adjusting 37 for topography and atmospheric stability. Measurements from two sites are used (Valle Hovin 38 and Blindern); both sites are centrally located in Oslo. Data required by the dispersion modelling 39 are atmospheric stability, wind speed and wind direction.

40

41 Hourly regional background concentrations were derived using predictions from the LOTOS-

42 EUROS model at a number of grid squares surrounding Oslo. The hourly median concentration

from these grid squares was extracted for this purpose. These values were further adjusted, based 43

44 on a comparison of the predicted and observed annual mean PNC measurements at Birkenes

45 (located about 300 km south of Oslo). This procedure resulted in a rescaling of all LOTOS-

EUROS predictions by a factor of 0.75. 46

In Oslo, a parametrization was applied to account for deposition and coagulation processes. This was only applied in the gridded model calculations, but not in the sub-grid Gaussian modelling. This parametrization is based on calculations using the MAFOR aerosol process model for road traffic emissions (Keuken et al., 2012). First, MAFOR calculations were carried out using the complete aerosol process model description and then, for simplicity, the emissions and calculations were binned into three particle size classes. Based on these computations, deposition and coagulation rates in these three size classes were derived.

9

10 The change of the PNC in each size bin caused by coagulation was parameterized in the 11 following simplified form:

12

13
$$\left. \frac{d\text{PNC}_{i}}{dt} \right|_{coag} = -\text{PNC}_{i}^{2} \overline{K_{c,i}} ,$$
 (1)

where the subscripts *i* and *coag* refer to the particle size class and coagulation, respectively, and $K_{c,i}$ is the coagulation rate derived using the MAFOR model. Dry deposition is described as

17
$$\frac{d\text{PNC}_{i}}{dt}\Big|_{depo} = -\text{PNC}_{i}\frac{v_{d,i}}{H_{grid}},$$
(2)

18

19 where $v_{d,i}$ is the dry deposition rate for the *i*'th size class and H_{grid} is the depth of the lowest 20 model grid layer.

21 22

23 Dispersion modelling for Rotterdam

24

In Rotterdam, the contribution of traffic to air quality near inner-urban roads was modelled with
the urban dispersion modelling system URBIS (Eerens et al., 1993; Vardoulakis et al., 2003).
This modelling system contains various submodules, such as a model for line sources, called
the Pluim Snelweg model, and a model for evaluating the concentrations in street canyons,
called the CAR model.

30

31 Up to a distance of 500 m, contribution from motorways was modelled with the line source 32 dispersion module, Pluim Snelweg (Wesseling et al., 2003; Beelen et al. 2010; Keuken et al., 33 2012). This line source model is a Gaussian plume model. The modelling also takes into account 34 the vehicle-induced turbulence, the roughness of the terrain, the noise screens near the motorway and the atmospheric stability. The treatments of concentration time-series is based on the concept 35 36 of stratified meteorology. A time series of wind speeds and directions, observed at the airport of 37 Rotterdam, are first clustered as a frequency distribution. The contributions downwind of the 38 motorway, based on averaged emission rates, are then weighted using these frequencies; this 39 procedure results in an estimate for the annual average concentration.

40

41 The street canyon dispersion model CAR is based on the results of wind tunnel experiments at 42 different road types, including street canyons. The ratio of the height of the buildings and the 43 width of the street is used to classify the type of street canyon. A source-receptor relationship has been specified as a function of the distance to the street axis for five different road types. All streets in Rotterdam have been categorized in accordance to the model classification. The model simulates only annually averaged concentrations. The model therefore requires as input values the annually averaged emission rates, and the reciprocal annual average wind speed. The annual average concentration is assumed to be inversely proportional to the wind speed. The wind speed was retrieved from measurements by the National Meteorological Institute at the airport of Rotterdam.

8

9 The contribution of shipping to the PNC's was estimated based on a predicted spatial distribution 10 of the emissions of NO_x from shipping in the Netherlands in 2007 (Snijder et al., 2012). The NO_x emission map was evaluated based on computations using as input the automatic identification 11 12 signals (AIS) of ships. These computations applied for operational shipping parameters, e.g., 13 navigational status and payload, which were based on the AIS signals. The total NO_x emissions 14 were scaled to correspond to the year 2008, using the total amounts of emissions from shipping 15 in the Netherlands in 2007 and 2008 (Denier van der Gon and Hulskotte, 2010). The spatial 16 distribution of the emissions of NO_x was subsequently converted to the emissions of PN's, based on the observations by Petzold et al. (2010). The conversion was done using the average ratio of 17 18 the NOx and PN emissions in the observations of Petzold et al. (2010).

19

The atmospheric dispersion of shipping emissions was evaluated using the Dutch Standard Gaussian dispersion model (van Ham and Pulles, 1998). This model applies the same treatment of atmospheric dispersion as the Pluim Snelweg model. For simplicity, we assumed a constant stack height of 30 m and the heat content of exhausts of 1.0 MW, for all the ships within the region.

- The urban background of PNC's were estimated based on the LOTOS-EUROS model, at a grid square that surrounds Rotterdam. The urban scale modelling has a spatial resolution of 10*10 m², up to a distance of 30 m from the streets, or alternatively at the housing façade along street canyons, and up to a distance of 500 m near motorways.
- 30 31

32 Dispersion modelling for London

33

The OSCAR air quality assessment system (Singh et al., 2013; Sokhi et al. 2008) has been used to estimate traffic related PNC's across London. The models within the OSCAR system consist of an emission model, meteorological pre-processing model and a line source Gaussian dispersion model. The roadside dispersion model within OSCAR system is the CAR-FMI model. The hourly concentrations were predicted at the receptor points placed at varying distances of 10, 40 and 90 m near both sides of the roads, and 100 m apart in the outskirts.

40

41 A range of hourly meteorological parameters are needed, including wind speed, solar radiation,

42 friction velocity and Monin-Obukhov length. These are provided by the dedicated OSCAR 43 meteorological pre-processor GAMMA met, described by Bualert (2002). The meteorological

44 model employs meteorological data, such as solar radiation, roughness length and heat flux, to

44 inoder employs meteorological data, such as solar radiation, roughness length and near flux, to 45 estimate atmospheric stability parameters, including the Monin-Obukhov lengths and mixing

46 heights. Data from the meteorological station at Heathrow was used as input for the model. The

effects of land use characteristics on parameters such as surface roughness, Bowen ratio, Albedo and anthropogenic heat flux are taken into account. The meteorological pre-processor needs six input parameters: time, wind speed, wind direction, ambient temperature, cloud cover and global radiation.

4 5 18

6 The regional background levels were evaluated based on the LOTOS-EUROS simulations. We
7 used the predicted LOTOS-EUROS concentration values surrounding the city. The LOTOS8 EUROS hourly values were scaled by multiplying them with the ratio of annual average
9 measured and predicted concentrations. The measured values used for the scaling were taken
10 from the regional background station of Harwell.

11 12

13 Dispersion modelling for Athens14

15 The modelling system consists of two models: (i) the meteorological model MEMO 16 (Moussiopoulos et al., 1993), and (ii) the chemical transport model MARS-aero (Moussiopoulos 17 et al., 1995; Moussiopoulos et al., 2012). The MEMO model is a three-dimensional Eulerian 18 non-hydrostatic prognostic model. The MARS-aero model can be used to simulate the transport 19 and transformation of gaseous pollutants and atmospheric aerosols in the lower troposphere. The 20 system allows for a finer grid simulation to be nested inside a coarser grid.

21

Meteorological data were generated using the MEMO model. Initialisation and boundary conditions data for the application of the MEMO model were based on upper air soundings for selected meteorological variables (wind speed and direction, temperature); these were performed at the Athens International Airport. Annual mean concentrations were estimated on the basis of computations for eight representative days, combined with a weighting scheme. These days were selected and assigned certain weights based on a classification of synoptic meteorological conditions in the Greater Athens area for 2008 (Helmis et al., 2003; Moussiopoulos et al., 2004).

The classification was done with the application of principal component analysis on a set of six meteorological variables (namely wind speed and direction, surface pressure, mixing layer height, cloud cover and specific humidity), and subsequently using a subtractive clustering algorithm. Using this procedure, the different synoptic weather conditions that prevailed during each day of the year were distributed into distinct groups, which correspond to certain characteristic meteorological features (Sfetsos et al., 2005, Shahgedanova, 1998).

36

37 The day that appeared closer to the mean of each group of synoptic meteorological conditions 38 was considered to be a typical day representing the specific group and was simulated with 39 MARS-aero. The weight assigned to each of the representative days was proportional to the size 40 of the corresponding group. The application of the methodology was based on meteorological fields predicted by the WRF meteorological model (version 3.2.1, Skamarock et al., 2005), 41 which was applied for 2008 with a horizontal grid resolution of 50 km and a temporal resolution 42 43 or 3 hours. The MEMO and MARS-aero models were applied in a computational domain of 50 x 44 50 km^2 , on a spatial resolution of 500 m.

45

46 Both the regional background PNC's and the concentrations of other relevant species are needed

as boundary conditions for the MARS-aero calculations. A spatially uniform annually average
 regional PNC background of 1800 particles cm⁻³ was used for the boundary conditions, based on
 Kalivitis et al. (2008). The regional background values of all other relevant species were
 extracted from the LOTOS-EUROS computations, at the grid squares surrounding the city.

5 6

7

8

2.4 The measurements of PN concentrations in target cities

9 The measurements at the station of Kumpula in Helsinki in 2008 and 2012 were performed using 10 a Differential Mobility Particle Sizer; the particle concentrations were determined at the size 11 range from 3 to 950 nm. Particle number concentrations at the station of Ring Road 1, Malmi 12 were measured using a Grimm butanol condensation particle counter (CPC), with detection limit 13 from 5 nm to larger than 3 μ m.

14

15 In Oslo, the Grimm 565 Environmental Wide Range Aerosol Spectrometer system 16 (www.GRIMMAerosol.com) was used for the measurements. This system combines a Grimm 17 190 aerosol spectrometer OPC (Optical Particle Counter), and a scanning mobility particle sizer 18 with a condensation particle counter (SMPS+C). The entire system in principle covers the range 19 from 5 nm to 30 μ m. For this study only the particle sizes below 350 nm, measured using the 20 SMPS+C instrument, have been used. For the modelling and comparison with measurements we 21 have used a lower cutoff of 8.5 nm.

22

The measured values of hourly PNC concentrations for London were available from Defra's
Particle Numbers and Concentrations Network, which uses CPC. This CPC measures the number
of particles in the size range from 7 nm up to several µm in size.

Total PNC in Rotterdam was measured using a CPC with a lower 50 % cut-off at 3 nm and an
upper limit of 3 µm. Size-resolved PNC was measured with a Scanning Mobility Particle Sizer.
The SMPS consists of a differential mobility analyser (DMA) covering a size range from 10 to
480 nm and a CPC with a lower 50 % cut-off at 4 nm and an upper size limit of 1.5 µm.

31 32

33 **3. Results and discussion**

34

35 **3.1 Emissions**

36

37 **3.1.1 Emissions in Europe and their associated uncertainties**

38

Total anthropogenic PN emissions in UNECE Europe were estimated using a bottom-up methodology (Denier van der Gon et al., 2014). These are presented in Figs. 2a-b, classified according to both source sector and country group. The transport sectors (i.e., road and non-road transport) contributed approximately 60 % to the total land-based PN emissions in UNECE-Europe in 2005 (Fig. 2a). The other most important sectors include industry (defined here excluding energy industries), residential combustion, fugitive emissions and energy industries.

The PN emissions are projected to decrease in 2020 and 2030 to less than a half of their value in 1 2 2005 (Fig. 2b). International shipping was a dominating source in 2005, but its contribution is 3 expected to substantially decline from 2005 to 2020 and 2030, mainly due to the introduction of 4 low sulphur fuels. The contribution of shipping is more dominant in the current inventory, 5 compared with the first European PN emission inventory made in the EU-funded project 6 EUCAARI (Denier van der Gon et al., 2010a; Kulmala et al., 2011). Another remarkable change 7 compared with the previous inventory is that in the new inventory, aviation is a substantially 8 stronger source of UFP's than previously assumed. Most of these shipping and aviation 9 particulate emissions are not solid, but semi-volatile particles, and may therefore have escaped 10 attention in previous emission factor measurements.



Figs. 2a-b. Total anthropogenic particle number emissions in the United Nations Economic Commission for Europe, (a) classified by the source sector for 2005, and (b) classified by the country group for 2005, 2020 and 2030. 'Sea' refers to international shipping.

4 5

6 The PN emission inventory includes in principle all particulate sizes. The PN emissions in two 7 size fractions have been presented in Figs. 3a. The ultrafine particle fraction (UFP) is defined as 8 particles smaller in diameter than 100 nm. As expected, the difference between the total PN 9 emissions and the UFP emissions is relatively small, as the PN emissions are dominated by the 10 smaller size fractions.

11

12 The corresponding emissions solely for the road transport sector have been presented in Fig. 3b. 13 The PN emissions of road transport are projected to significantly decrease in time (Figs. 3b). The 14 PN emissions due to fuel combustion in road transport and shipping are expected to significantly

- 15 decrease as a consequence of motor and fuel modifications, such as low-sulphur fuels and
- 16 particulate matter filters (e.g. Ristovski et al. (2006); Morawska et al. 2008; Fiebig et al., 2014).
- The EU 15 emissions are estimated to decline strongly in future years, due to implementation of
- 18 new emission standards in road transport, and the phase-out of the older vehicles that have less
- 19 stringent emission limits.
- 20



21

Figs. 3a-b. (a) Total anthropogenic particle number emissions and total particle number emissions in the particle size range of 10-100 nm for UNECE-Europe for 2005, 2020 and 2030 and (b) the same emissions exclusively for road transport, segmented by country group.

25 26

To facilitate the modelling of PN on a regional scale, the PN emissions were spatially distributed using available proxy data (Denier van der Gon et al. 2010b; Pouliot et al, 2012.) Examples of such proxy data are maps of population density, road networks, shipping tracks, land use, and port capacities. The spatial distribution of the PN emissions has been presented in

31 Fig. 4.



Fig. 4. Spatial distribution of anthropogenic PN emissions in Europe in 2005, on a longitude vs. latitude grid, on a resolution of $1/8^{\circ}$ x $1/16^{\circ}$. The unit of the legend is 10^{24} particles per computational cell per annum.

7 8 The estimates for PN emissions are associated with a relatively high uncertainty, compared with 9 the emissions of the commonly regulated pollutants. This uncertainty varies substantially in 10 terms of the different source categories. Vehicle-originated PNC's can change on a short 11 timescale after the emissions exit the tailpipe, due to both rapid dilution and microphysical 12 processes. The latter depend on ambient temperature and other environmental conditions, as well 13 as on secondary particle formation. Due to such transformations, the PN concentration flux is not 14 conserved. For some source categories, no PN emission factors were available. In such cases, the 15 PN emission was calculated based on PM measurements and estimated particle size distributions. 16

For the road transport emission factors reported here, an uncertainty analysis for the particle mass -based emission has been carried out. This analysis shows an uncertainty between 10 and

19 20 %, depending on the quality of the country's statistics (Kioutsioukis et al., 2010). Particulate

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number emission factors were not included in the uncertainty evaluation of the above mentioned
 study. However, it is possible to indirectly estimate also the uncertainties of the PN emissions,

- based on the correlations between PN emission factors derived in this study with the COPERT
 PM emission factors (Vouitsis, 2014).
- 5

6 Solid particles can be measured more accurately than semi-volatile ones; the emission standards 7 for road transport are therefore currently based on the solid fraction of PN. The PN emissions are 8 influenced by numerous factors, such as, e.g., vehicle category, PN measuring equipment and 9 environmental conditions. The overall uncertainty of vehicular PN emissions can therefore be 10 evaluated to have high uncertainties: (i) 81-144 %, when after-treatment device effects are not 11 included and (ii) 144-169 %, when these effects are included (UNECE, 2010).

12

13 Road transport is the most intensively studied source category for PN emissions. It can therefore 14 be expected that the uncertainties for other source categories are at least of the same magnitude. 15 For example, the total PN emission factor is dependent on the set-ups of the measurements. In 16 particular, the measurement can (i) include only solid PN, or solid and volatile PN, and (ii) the lower particle size cut-off used in the measurements can vary, as this is dependent on the 17 instrumental method. Sometimes a lower cut-off of 3 nm is used, but frequently also only PN's 18 19 for sizes larger than 20 or 30 nm are reported. This definition of lower size cut-off can have 20 substantial effects on the estimates of the total PN emissions. For a more detailed discussion of 21 the various techniques used to measure PN, we refer to McMurry (2000) and Morawska et al. (2008).

22 23

Another important uncertainty is caused by the sulphur content in shipping fuels. It is known what the regulatory limit values for the fuel sulphur content are, and in some cases also what the average fuel sulphur content is; however, it is not commonly known what the actual values are. Therefore, for all transport modes the uncertainty is expected to be at least equal to the previously listed uncertainty estimate for road transport; this is in the range of 100-170%.

29

30 On a regional to city-scale, Kalafut-Pettibone et al. (2011) determined average size-resolved and 31 total number- and volume-based emission factors for combustion. They estimated that the 32 uncertainty of the PN emission factor is approximately plus or minus 50 %. This uncertainty 33 value is based on longer term temporal averages.

34 35

36 **3.1.2 Emissions in the target cities**

37

38 All of the emission inventories in the target cities included vehicular traffic. However, the details 39 of the treatments for other source categories varied substantially from city to city. The urban 40 inventories for Helsinki, Oslo, Rotterdam and Athens included also the primary particulate 41 matter emissions from shipping. In the case of London, the importance of shipping emissions 42 was found to be negligible, compared with that of other urban emissions. The stationary sources 43 were included at varying levels of detail for Helsinki, Oslo, London and Athens. In the case of 44 Rotterdam, the airports, refineries and other major sources were included in the regional 45 background. For Helsinki, the influence of shipping and major stationary sources was estimated 46 indirectly, but the actual PN emission values for these source categories were not included in the

urban emission inventory. The influence of small-scale combustion was explicitly evaluated for
 Oslo, and its importance was evaluated for Helsinki.

3

The sulphur content of vehicular motor fuel is an important factor for selecting the emission factors of PN's. There has been a decreasing trend in the fuel sulphur (S) contents in Europe. During the later part of 2000's, the S content of motor fuels was decreasing rapidly in many European countries, commonly from < 50 ppm to < 10 ppm S. One should therefore use the vehicular emission factors (EF) that were determined for the same S content as for the target year of modelling (in this study 2008). For all the target cities, we used the best available locally applicable EF's.

11

12 For Helsinki, calculations were based on EF's by Gidhagen et al. (2005) for Stockholm. The 13 measurements that were the basis for these EF's were made in Stockholm for heavy duty vehicles (HDV) in 1999 and for light duty vehicles (LDV) in 2003. Sweden introduced its 14 15 Environmental Class 1 (EC1) diesel fuel in 1991, with maximum sulphur content of 10 ppm 16 (weight). The EC1 grade reached nearly complete market coverage in Sweden already in the nineties, due to a strongly supportive tax policy. The EF's used for Helsinki therefore refer to 17 fuel with lower than 10 ppm sulphur content. As also Finland used the lower S content vehicular 18 19 fuel in 2008, the EF's used in the manuscript are appropriate in this respect. Also in Oslo and 20 Rotterdam both the modelled and actual S contents of the vehicular fuel were lower than 10 ppm 21 in 2008.

22

For London, the emission factors from Jones and Harrison (2006) were used, which refer to the higher (< 50 ppm) fuel S, while the target year for modelling (2008) was after the transition to lower S fuel. For Athens, the situation was the opposite: EF's correspond to the lower fuel S content, whereas a higher S content fuel was actually used. The applied EF's are therefore expected to somewhat overestimate the measured concentrations in London, and underestimate those in Athens.

29

The most detailed emission inventory was compiled for Oslo. The proportions of total emissions in Oslo in 2008 have been presented in Fig. 5. The sector denoted 'heating' includes all heating, of which domestic heating is the largest part, 95%. Traffic exhaust emissions were responsible for about ³/₄ of the total emissions; the contributions from shipping, heating and other mobile sources are also notable.

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- 39



Fig. 5. The contributions of various source categories on the total emissions of particulate number in Oslo in 2008. The total amount of emissions was $1.1 \ 10^{24}$ particles /a.

3.2 Modelled concentrations

3.2.1 Concentrations in Europe

The LOTOS-EUROS model, including the M7 module, was used together with the above
 mentioned new PN emission inventory, to evaluate the PNC's in Europe.

15 Evaluation of predicted concentrations with measured values on a European scale

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The predicted PNC's were compared with the EUCAARI measurements (Asmi et al 2011), with 17 18 a focus on eight selected stations: Cabauw, Melpitz, Vavihill, Harwell, SMEAR, Ispra, Kosetice 19 and Kpuszta. Cabauw is a rural site in an agricultural area in the Netherlands, with influence 20 from the nearby city of Rotterdam; this is the type of region, for which the model is well suitable. 21 Melpitz is a rural site in Germany, the concentrations of which are dominated by long-range 22 transport and biogenic emissions. The site of Vavihill in Sweden is close to the sea; this site is 23 representative for fairly clean background conditions, with occasional influence from shipping 24 and nearby cities. The station of Harwell (UK) is a regional background site that can 25 occasionally be influenced by the urban plume originated in London. The SMEAR II site (in 26 Hyytiälä, southern Finland) is a high-latitude regional station, exposed alternatively to both clean 27 and fairly polluted air masses. The site of Ispra (Italy) is in the vicinity of the Alps; it

experiences the influences of the polluted air from Po Valley. The site of Kosetice (Czech 2 Republic) is located in the agricultural countryside. Kpuszta represents the central European 3 regional background, relatively far from local sources.

4

1

5 Modelled PNC's in the Aitken and accumulation mode were compared with the observed PNC's 6 in size bins 30-50 nm, > 50 nm and > 100 nm (the latter two bins are partly overlapping). 7 Measured and modelled monthly average PNC's for the three sites have been presented in Figs. 8 6a-d. The nucleation mode was excluded. The values correspond to the size fraction 30-250 nm 9 for the observations, and the sum of Aitken and accumulation mode for the LOTOS-EUROS 10 computations (defined as the interval 10-1000 nm).

- 11
- 12



14 15

16 Figs. 6a-d. A comparison of the seasonal variation of the monthly averaged model predictions and observations of the particle number concentrations (particles/cm³, panels a-b), and the 17 18 correlation coefficients of the hourly predicted and measured concentration values within each 19 month, at eight selected measurement sites in 2008. In the upper panels (a-b), the solid lines are 20 model predictions, and the dotted lines are measurements. The modelled values are the 21 predictions of the LOTOS-EUROS model. The nucleation mode has been excluded; the values 22 correspond to the size fractions 30-250 nm and 10-1000 nm for the observations and the model 23 computations, respectively.

24 25

26 At Vavihill, the modelled and observed monthly average concentrations match well for the whole year. At Cabauw, since May, the overall measured and predicted levels of the PNC's were 27

28 in agreement; however, the observed concentrations from January to April were not comparable

29 with the predictions, due to different settings of the measurements. The modelled monthly

1 average concentrations at Melpitz were clearly lower than the corresponding measured values. 2 These relatively high measured values have probably been caused by the substantial contribution 3 of particles formed from biogenic emissions, which were not accounted for in the present model 4 version. In winter, when the biogenic emissions are smaller, the model and observations match 5 relatively better at Melpitz. The predicted monthly concentrations at Harwell agree well with the 6 measurements; the model is well suited for this type of environment. For the site of SMEAR, the 7 model under-predicts; contributions from biogenic emissions in summer are not taken into 8 account in the model. For the site of Ispra, the concentrations in winter were the highest observed 9 amongst the stations considered here, and the model substantially under-predicted. This location 10 is, together with Cabauw, most strongly affected by anthropogenic emissions. In particular in winter, high concentrations are expected due to wood burning, in combination with stagnant 11 12 conditions in the Po valley. For the site of Kosetice, the model shows a smaller underestimation 13 in winter than in summer. For K-Puszta statistics before September are based on a small set of 14 measurements and are therefore only indicative.

15

16 The correlation coefficients were reasonable, ranging on the average from 0.3 to 0.6 for the 17 stations in Fig. 6c, and from 0.3 - 0.5 for the stations in Fig. 6d. A higher correlation was not 18 always related to an accurate estimate of total particle number.

19

Modelled values were on the average within a factor of two of the measured values for the Aitken mode, compared to observed particle modes in the range 30-100 nm (results not shown here). However, the number of particles with diameter > 100 nm was under-predicted, whereas the number of particles < 100 nm was in most cases over-predicted. Fountoukis et al. (2012) previously reported a similar result; a systematic under-prediction of the number of particles larger than 100 nm, using the original EUCAARI emission inventory and another chemical transport model.

27

These model evaluation studies indicate that the applied regional scale modelling provides reasonably accurate results for PNC's in the size range larger than 30 nm, in the presence of dominating anthropogenic emissions. In case of substantial biogenic contribution, the predicted PNC's will probably be underestimates. Clearly, the prediction of particle size distributions is a more challenging task, compared with the prediction of the PNC's integrated over all particle sizes.

34 35

36 The spatial concentration distributions in Europe in 2005 and 2008

37

The modelled European scale PNC's for 2005 and for 2008 are presented in Figs. 7a-b. The
 differences of the concentrations between these two years have been presented in Figs. c-d.

40

41 The anthropogenic emissions of PN were assumed to be the same for these two years. However,

42 for the emissions of gases, we used the MACC project emissions for the different years. The

43 meteorological conditions and the natural emissions (which were influenced by meteorology)

- 44 were also assumed to be different. During both years, the highest concentrations occurred at
- 45 urban and industrialized areas, and along the most densely trafficked shipping lanes. Annual

- 2 were higher for 2005, compared with those for 2008.





The largest concentration differences between the two target years were approximately 25 %.
The fairly large differences of the concentrations near the western boundary of the domain are
caused by the natural emissions, which were determined by the meteorological conditions. At
other locations, differences are due to the combined effect of meteorology and decreased SO₂
emissions; the emissions were lower for 2008.

3.2.2 The influence of aerosol processes on an urban scale

We did not include a treatment of aerosol processes to all of the urban scale modelling systems used in this study. Instead, their influence was examined in a numerical study performed for Oslo in 2008. We have used a simplified aerosol process parametrization based on the more complex MAFOR aerosol process model and some experimental results. The numerical accuracy of the simplified model, as compared with the more complex model, was evaluated to be approximately 10 %.

8 The model needs as input values an initial size distribution, which was based on experimental 9 data in Oslo, Rotterdam and Helsinki. An initial size distribution ratio was defined as the initial 10 fraction of the total PN concentration in each size bin (PNC₁, PNC₂ and PNC₃). These model 11 input values have been presented in Table 2.

- 12
- 13

Table 2. Data and coefficients required for the implementation of the PNC parameterization used in Oslo. Typical predicted time scales associated with deposition and coagulation are also presented.

- 17
- 18

Size class	Size range (nm)	Initial size distribution ratio	Dry deposition velocity v_d (cm s ⁻¹)	MAFOR derived K _{c,i} (cm ³ # ⁻¹ s ⁻¹)	Typical deposition time scale (h)	Typical coagulation time scale (h)
PNC ₁	8.5 - 25	0.79	0.904	6.31x10 ⁻⁹	0.6	0.9
PNC ₂	25 -100	0.20	0.202	5.58 x10 ⁻⁹	2.7	2.9
PNC ₃	100 - 400	0.01	0.032	$8.82 \text{ x}10^{-10}$	17	292

19

20

The impact of this parametrisation was tested in comparison with the measured data in Oslo for a three-monthly period from January to April, 2008. In these computations, the upper limit values were used both for the coagulation coefficient and the dry deposition velocity, in order to evaluate the maximum possible effects due to these processes.

25

26 Use of the parametrisation resulted in lower PNC levels further from sources. At the urban 27 background station in Oslo (Sofienbergparken), the above mentioned parametrisation resulted in 28 a maximum reduction of PN concentrations by approximately 45%, compared to treating PN as a 29 tracer. The range of this percentage value, allowing for the uncertainty of the simplified aerosol 30 process modelling, can be considered to be approximately from 40 to 50 %. The impact of 31 deposition was larger than that caused by coagulation; however, the influences of both processes 32 were significant. The model-derived deposition and coagulation rates in the selected three size 33 classes, and the relevant time scales are presented in Table 2.

34 35

36 **3.2.3 Predicted concentration distributions in the target cities**

1 The predicted annually averaged spatial concentration distributions in the target cities are 2 presented in Figs. 8 a-f. The same concentration legend is used for all the cities. The 3 concentrations in various cities can therefore be inter-compared, allowing for the differences in 4 the computational methods. The central area of London has been separately presented, by using a 5 more closely spaced concentration legend.



Figs. 8a-f. The predicted spatial distributions of particle number concentrations in the target cities in 2008. The cities in the top row are Helsinki and Oslo, in the middle row Rotterdam and London and in the bottom row Athens and the centre of London (the location of which is shown in panel 8d as a rectangle). The concentration unit in all the legends is particles per cm^3 . The legends are identical for the panels a-e, but different for the panel f (the center of London). The 7 8 water areas are presented in blue grey.

1 2 The differences of the numerical results in the various cities are mainly due to the differences in 3 the spatial distribution and strengths of emissions, the regional background contributions, 4 meteorological conditions and other specific characteristics of the cities. Clearly, these 5 differences are also partly caused by the inaccuracies and deficiencies of the methods. In 6 particular, the concentration distribution for Athens was evaluated on a spatial resolution of 500 7 m, which is coarser that the corresponding resolution used for the other cities; this tends to 8 smooth out the maximum concentrations on finer spatial scales. Further, the modelling in this 9 study did not explicitly allow for the influence of street canyons in all the target cities, except for

10 the semi-empirical modelling of the effects of street canyons in Rotterdam. The predicted PNC's at street canyon locations, and more generally in the vicinity of locations that are influenced by 11 12 high buildings tend therefore to be under-predictions in this respect.

13

14 The maximum values of annual average PNC's were approximately 20 000 in Helsinki, 30 000 15 in Oslo, 30 000 in Athens, and 50 000 in Rotterdam and London. These values were relatively 16 higher in London, due mainly to very high traffic flows along the most trafficked roads, and in

17 Rotterdam, due to both high regional background and intensive urban traffic. 18

19 The locations of harbours and airports in the target cities have been presented in Fig. A1 in 20 Annex 1. Also tunnel entrances have been presented for Oslo. In all cities, the most important 21 emission category that influenced the spatial distributions of PNC's was vehicular traffic; the 22 major traffic networks are clearly visible in all the target cities. E.g., the main ring road or ring 23 roads (for Helsinki, Oslo, London and Athens) or the main highways (for Rotterdam) 24 surrounding the city centers are clearly visible. The concentrations were also elevated in the 25 central areas of the cities. In Helsinki, Oslo, Rotterdam and London, the highest concentrations 26 occurred in the vicinity of the most densely trafficked ring roads, and near the junctions of such 27 ring roads and other major roads. In Athens, the highest predicted concentrations of PN's occurred in the vicinity of the Athens International Airport.

- 28 29
- 30 The second most important urban source category was shipping and harbours. Their influences on the PNC's over land areas can be distinctly detected in the case of Oslo and Athens, and to a 31 32 smaller extent also in Rotterdam. In Oslo, the higher concentrations in the vicinity of the 33 harbours are also partly caused by the traffic tunnel entrances. It was assumed that there was no 34 deposition of particles within the tunnels; therefore all traffic-originated PN's within the tunnels 35 were treated as emitted at these entrances. In Athens, there were substantially elevated PNC's 36 near the main harbour regions (Piraeus and Rafina). For Helsinki, the shipping emissions have 37 not been included in the PNC map shown in Fig 8a; however, it was separately evaluated that 38 their influences can be notable near the main harbour areas (Soares et al. 2014). For London, 39 shipping along the River Thames, and the related harbour activities cause only a negligible 40 impact on the overall PNC's.
- 41

42 Although the harbours in the vicinity of Rotterdam are amongst the largest in Europe, the

43 influence of harbour activities was only modestly detectable in Fig. 8c. The main reason for this

- 44 was the fact that the most densely trafficked harbours in that region are located outside the city
- 45 of Rotterdam. The harbors within the city of Rotterdam are located to the south and north of the
- river Nieuwe Maas, which flows through the centre of Rotterdam. These urban harbours serve 46

mainly inland shipping. The larger harbours serving sea going vessels are located at a distance of
5-10 km to the west of the centre of Rotterdam, near the coast of the North Sea. The harbours
within the city of Rotterdam are also dispersed on a relatively wide region on both sides of the
river; this tends to spatially smooth out concentration hotspots.

5

6 A potentially important source is also vehicular traffic to the airports and aviation. In Athens, 7 there were substantially elevated PNC's near the Athens International Airport, located to the east 8 from the centre of the city (it is clearly visible in Fig. 8e). Detailed computations showed that 9 aviation emissions were responsible for the largest share of the concentrations within this airport 10 and in its immediate vicinity. 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 11 12 concentrations were caused by the congested roads leading to the Heathrow airport; the 13 emissions originated from aviation were not taken to account in the computations for London. The Helsinki-Vantaa airport is only slightly detectable (to the north of the outer ringroad, in the 14 15 northern part of the metropolitan area). The airport in Oslo is outside the modelled domain. The 16 influence of the Airport Rotterdam is not visible; it is a fairly small airport.

17

18 There are also some other significant source categories, such as major refineries in the vicinity of 19 Rotterdam; however, these were not located within the modelled urban domain. Especially in 20 Oslo, the small-scale combustion in households can also be an important source in residential 21 regions in winter

21 regions in winter.

22 23

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25

3.3 Evaluation of model predictions against measured data in the target cities

The model predictions were compared with the available PNC measurements in the target cities.Such measured data were available in four of the cities, as presented in Table 3.

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29

30

- 1 Table 3. The comparison of measured and predicted PNC's in four target cities. IA is the index
- of agreement and FB is the fractional bias. NA refers to data or evaluation measures that werenot available.
- 4

City	Name of station	Classification of station	Period	Mean of the observed values $(10^3$ particles/ cm ³)	Mean of the predicted values $(10^3$ particles/ cm ³)	IA (based on the hourly means)	FB
Helsinki	SMEAR III, Kumpula	Urban background	Whole year 2012	7.1	NA	NA	NA
	Ring Road I	Urban traffic	Whole year 2012	19.5	20.0	0.75	+ 0.02
Oslo	Sofienber gparken	Urban background	Three months, Jan-Mar 2008	9.3	10.8	0.77	+ 0.15
	Smestad	Urban Traffic	Three months, Jan-Mar 2008	24.0	19.8	0.79	- 0.19
Rotterdam	Zwartewa alstraat	Urban background	Whole year 2011	14.5	10.1	NA	- 0.22
	Rotterda m, Bentinckp lein	Urban traffic	Whole year 2011	17.7	20.1	NA	+ 0.20
London	North Kensingto n	Urban background	Whole year 2008	14.0	10.8	NA	- 0.26
	Marylebo ne Road	Urban traffic	Whole year 2008	36.7	15.7	NA	- 0.81

7 The predictions and measurements were compared at two stations, representing urban 8 background and urban traffic environments, in three cities, viz. Oslo, Rotterdam and London. In 9 the case of Helsinki, such comparisons were performed only at one station (Ring Road 1, Malmi, 10 urban traffic) for 2012. The comparisons were performed for different years in Rotterdam (2011) 11 and in Helsinki (2012), as the relevant measured data was not available for those cities in 2008.

12

13 The comparison in the case of annual averages is also presented graphically, in Fig. 9. The 14 predicted concentrations consist of the regional background and the local urban contributions. 15 The regional background values presented in the figure are the predictions of the LOTOS-





6 7 8



11 contributions. The names of the stations have been specified in Table 3.

12

13 The regional background concentrations were clearly lower than the contributions originated 14 from urban sources in Helsinki and Oslo, and lower (for traffic site) or almost equal (urban 15 background) in London. However, for Rotterdam the regional background was the largest contributor (for urban background) or responsible for almost half of the total concentration 16 17 (urban traffic site). This result was to be expected, as Rotterdam is surrounded by a high 18 population density and several intensive emission sources (such as other major cities, refineries 19 and major harbours). The uncertainties caused by the regional scale modelling have therefore a 20 relatively smaller effect in Helsinki, Oslo and London (but vice versa for Rotterdam), compared 21 with the uncertainties associated with the urban scale modelling.

- 22
- 23 The corresponding results for Athens are not presented in Fig. 9, as the experimental data was
- 24 not available for 2008. The representative annual average of the urban background of PNC in
- 25 Athens, predicted at the station of Nea Smyrni, was $6.8 \ 10^3 \text{ cm}^{-3}$. A characteristic annual average
- 26 PNC predicted at an urban traffic station, Athinas, was $12.2 \ 10^3 \ cm^{-3}$. The measured regional
- 27 background of PNC was 1.8 10³ cm⁻³. However, the predicted values at specific point locations

- in Athens are not directly comparable with those in the other cities, due to the more coarse
 resolution of the computations. The air quality stations in traffic environments in the Greater
 Athens Area are also not located in the immediate vicinity of the major highways.
- 4

5 The predicted and measured annual averages agreed within approximately ≤ 26 % (measured as 6 fractional biases), except for the traffic station in London. As expected, the agreement of annual 7 average concentration values was better at urban background stations compared with urban 8 traffic stations in Oslo and London; however, these agreements were not substantially different 9 in Rotterdam. The urban traffic station in London is Marylebone Road, which is located in a 10 street canyon and has continuously severe traffic congestion. The measured concentration at the Marylebone Road station is substantially higher than the predicted value. The lower predicted 11 12 concentration values are probably mainly caused by the fact that the computations in this study 13 did not allow for the effects due to street canyons for London.

14

15 It was possible to evaluate the agreement of the measured and predicted hourly time series of 16 PNC's at three stations located in two cities, Oslo and Helsinki (cf. Table 3). The period of these 17 comparisons was one year in the case of Helsinki, and three months in the case of Oslo. The 18 indexes of agreement (IA) for these comparisons were 0.75 for the annual time period in 19 Helsinki, and 0.77 and 0.79 for the three-monthly periods in Oslo; these values indicate a fairly 20 good agreement of measurements and predictions. However, the computational methods also 21 influence the values of the IA's. In the case of Oslo, the regional background values and local 22 urban contributions were separately modelled, whereas for Helsinki, the predicted values contain 23 measured urban background PNC values and the predicted local contributions. The measured 24 annual average of the urban background of the PNC (at the site of Kumpula) was 7.1 10³ cm⁻³ and the modelled contribution originated from urban vehicular sources was $12.9 \ 10^3 \ cm^{-3}$. 25

26 In general, we evaluate that for Helsinki, Oslo and London, the largest contributors to the 27 differences of predictions and measurements are (i) the uncertainties of the urban scale emission 28 inventories, and (ii) the uncertainties associated with the urban dispersion modelling systems. 29 For cities located in highly urbanized regions, such as Rotterdam, the uncertainties of evaluating 30 regional background can be even more important. Clearly, sources or source categories that are 31 missing from the computations can also have a significant effect. For instance, we performed 32 computations for Rotterdam, neglecting the contributions from shipping and harbours. The FB's 33 were -0.36 (including shipping that was -0.22) and +0.13 (including shipping +0.20).

34 35

36 **3 Conclusions**

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38 We have presented the results of the modelling of PNC's in five European cities in 2008. Novel 39 emission inventories of particle numbers have been compiled both on urban and European scales 40 (the latter is called the TRANSPHORM inventory). It has not previously been possible to 41 conduct such computations on a European scale, due to the deficiencies of the previously 42 available emission inventories. The TRANSPHORM PN emission inventory was based on a 43 previous inventory that was compiled in the EUCAARI project (Kulmala et al., 2011). The new 44 inventory focused on improving the representation of the emissions of the transport sector; major 45 improvements were made to the previous inventory in this respect. The previous emission 46 inventory was also substantially re-structured and improved for particulate matter emissions.

2 However, there are still unresolved issues on PN emissions. The semi-volatile particulate matter 3 should also be allowed for, in addition to solid state particles. Another challenge is to allow for 4 the short-term temporal transformations of particulate matter, after the exhaust of pollutants from 5 an engine or an industrial process. PNC is not a conserved quantity, and the emission values are 6 therefore dependent on the detailed definition of emissions; especially on the assumed spatial 7 distance from the emission source. Clearly, the transformation is dependent on ambient 8 conditions, especially on the ambient air temperature. The values of measured PN emissions are 9 also dependent on the selected lower particulate matter limit; this is commonly determined by the 10 capabilities of the experimental techniques. The impacts of fuel quality and the sulphur content of fuels on PN emissions are also not currently sufficiently understood. 11

12

1

We have also compiled detailed and extensive urban scale emission inventories in the five target cities. However, the present knowledge is not sufficiently accurate regarding the variation of PN emission factors in terms of the various source categories, especially for shipping and small-scale combustion, and for various environmental conditions. In future work, an in-depth intercomparison of such urban emission inventories would be valuable, in terms of both the physical assumptions and the numerical emission values.

19

20 We have conducted dispersion modelling on both European and urban scales. The European 21 scale computations included aerosol process modelling; however, it was not possible to include a 22 detailed treatment of aerosol processes to all of the urban scale modelling systems. Instead, the 23 influence of coagulation and deposition was examined numerically for the background air 24 pollution in Oslo in 2008. These processes were estimated to reduce the background air PNC's 25 maximally by approximately 40 - 50 % in the considered environmental conditions. The urban 26 scale modelling also did not explicitly allow for the influence of urban buildings and other 27 structures.

28

29 In all of the target cities, the highest concentrations occurred in the vicinity of the most densely 30 trafficked roads, and near the junctions of such roads and other major roads. The concentrations were also elevated in the city centers. The influence of shipping and harbours was also 31 32 significant for all the target cities, except for London. Three of the target cities are located on the 33 seaside (Helsinki, Oslo and Athens), and two are situated along major rivers (Rotterdam and 34 London). The regional background concentrations were an important factor for London, and the 35 largest factor for Rotterdam. In Oslo, the PNC's were also enhanced near the road tunnel 36 entrances.

37

38 The predicted and measured annual average PNC's in four cities agreed within approximately \leq 39 26 %, except for one traffic station in London. We consider this agreement to be reasonable, 40 considering the many potential uncertainties associated with the PNC modelling. The indexes of 41 agreement (IA) for the comparisons of hourly measured and predicted time-series in Oslo and Helsinki ranged from 0.75 to 0.79, indicating a fairly good agreement. However, the amount of 42 experimental data that could be used for model evaluation was modest: only one or two stations 43 44 for each city, and no relevant data were available for Athens. More long-term hourly 45 measurements of PNC's would therefore be valuable for a more thorough model evaluation in various urban locations. 46

4 Code availability

The computer code of the LOTOS-EUROS model can be made available upon request (contact:
Astrid Manders on email astrid.manders@tno.nl). The code is written in FORTRAN 90 and uses
NetCDF libraries and python scripts.

9 The access to the CAR-FMI model for educational and non-commercial research use can be 10 granted after signing a collaborative agreement with the Finnish Meteorological Institute 11 (contact: Jaakko Kukkonen on email jaakko.kukkonen@fmi.fi). The code is written in 12 FORTRAN 77.

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The computer code of the EPISODE model can be made available upon request (contact: Leonor
 Tarrason on email leonor.tarrason@nilu.no). The code is written in FORTRAN 90.

16

17 The OSCAR model can be configured for any urban area in collaboration with the Centre for 18 Atmospheric and Instrumentation Research (CAIR) at the University of Hertfordshire, UK. 19 Access to the model for educational and non-commercial research use can be granted after 20 signing a collaborative agreement with the University of Hertfordshire. The code has been 21 developed to assess air quality and exposure to air pollution at local scales across cities (contact: 22 Ranjeet S Sokhi on email r.s.sokhi@herts.ac.uk). The model code is written in FORTRAN 90, 23 except for emission model, which is written in Matlab.

24

The MEMO and MARS-aero models can be obtained for educational and non-commercial
research use, after signing an end-user license agreement from the Aristotle University of
Thessaloniki (contact: George Tsegas on email gtseg@aix.meng.auth.gr). The code is written in
FORTRAN 95 and uses OpenMP and MPI directives.

29 30

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32

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43 44

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Figs. A1 a-e. The locations of major harbours, airports and tunnel entrances in the target cities.
The harbour areas have been marked with oval shapes. The airports have been marked with
rectangles, except for Oslo, for which the rectangles correspond to the locations of the tunnel
entrances. The geographical areas denoted by the shapes in the panels are approximates. Panels
a-e correspond to Helsinki, Oslo, Rotterdam, London and Athens, respectively.

1 Annex 1. The locations of major harbours, airports and tunnel entrances in the target cities.