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## **Response to reviewer #1**

### **General comments**

The paper presents a refinement of the EXPAND model to evaluate the population exposure to air pollutants. It gives a thorough description of an up-to-date approach to evaluate exposure in urban areas keeping into account pollutants concentration and human time activity variability in time and space. The model development proposed consists mainly in the integration of different modules to describe pollutants emission, atmospheric dispersion, population time activities and exposure.

A few details need to be clarified to complete the modelling system description and some aspects of the presented application over Helsinki area need to be discussed in more detail to better identify and possibly quantify the simulation limits.

It would be advisable to enhance the final summary of data and models applied that can be considered local and need to be replaced to enable the application of the proposed methodology in regions characterized by different climatic and social conditions.

- ✓ Yes, we will include a discussion on how to extend the use of this model for other regions.

### **Specific comments**

#### **Section 2.2.1**

Pag. 2340 Lines: 10-11 Are emissions estimated using pollutant dependent emission profiles for each vehicle category? Are those profiles Finland specific or are they generally applicable to the European vehicle fleet in different locations?

- ✓ Yes, we use pollutant dependent emission profiles for each vehicle category. The emission factors are based on European emission factors, but we have taken into account the age distribution of Finnish vehicles fleet. We will describe this more clearly in the revised version.

Lines: 15-22 The meaning of these sentences is not very clear. Does the mentioned evaluation aim to analyse the emission variation from 2005 to 2008? No specific result concerning this item is presented in the rest of the paper.

- ✓ We do not intend to analyse the emission variation, but to obtain more reliable emissions for the years 2008 and 2009. Since the total vehicular exhaust emission values are available for 1005, 2008 and 2009 from LIIPASTO system (Mäkelä, 2002), we assumed that the total emissions in 2008 and 2009 for each traffic link will vary proportionally to the total vehicular exhaust emissions.

Pag. 2341 Lines: 3-5 It would be worth specify if the mentioned resuspension emission model is strictly specific for the Nordic countries or if can be applied even in regions where road sending is episodic and can generally be neglected.

- ✓ The resuspension model used is not specific for the Nordic countries. It can be applied anywhere; however, some local experimental data is needed for its application. We will specify these conditions in more detail in a revised manuscript.

### Section 2.2.3

Pag 2342 Lines 8-10 The impact of the choice to neglect small-scale wood combustion on model results should be better evaluated and possibly quantified. It is not clear if the whole residential heating contribution has been neglected and why. Are public and private buildings located within the city core heated by natural gas boilers or district heating facilities possibly included among point sources?

- ✓ The small-scale wood combustion contributed 18% to PM<sub>2.5</sub> emissions in the Helsinki in the 2009 case study. See figure below.

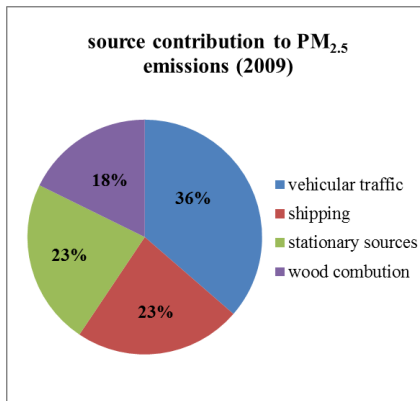


Figure. Source contribution to PM<sub>2.5</sub> emissions in Helsinki, 2009.

- ✓ According to the statistics of Finland, the amount of natural gas boilers in Finland is very low (see figure below). It should be similar for the Helsinki Metropolitan Area. The district heating facilities are indeed the highest contribution to the heating but those are included in the stationary sources inventory.

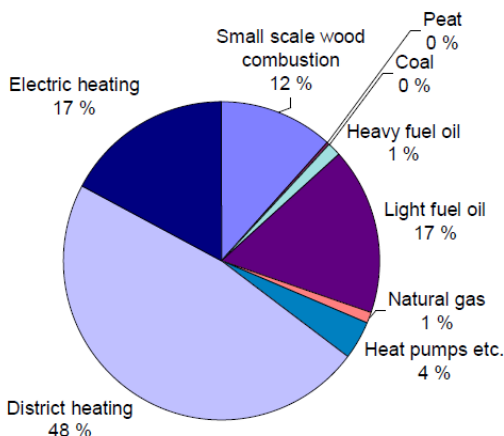


Figure: energy sources in Finland (Statistics Finland, 2004)

### Section 2.3

Pag 2342 Lines: 25-26 Does LOTOS model simulation, used to evaluate long range transport contribution, include Helsinki emissions or have they been excluded? In the former case some emission contribution could be double counted, even if the limited regional model resolution would smooth the effect.

- ✓ The emissions used in the LOTOS-EUROS computations account for sources in Helsinki. However, by assuming the same grid-cell where the background station Luukki is located, the influence of local sources is very small. Several previous studies have showed that the influence by local sources at this station is on the average less than 10 %; therefore, no double counting will rarely take place.

### Section 2.4

Pag. 2344 Lines: 4-5 Is the information referred to shops and recreational activities limited to working people or does it include statistics concerning the presence of costumers in shops, etc.?

- ✓ The people included in "other activities" are not working; they are recreational users/costumers. The employees at those shops are included in the work environment. Unfortunately there is no readymade statistics for this information. The data was evaluated by using time use survey data, which includes data about how much time (in minutes) people are spending in shops or restaurants. We have used the number of employees to evaluate how many people visit in shops and restaurants. The more people working in certain shop the more clients the shop will have. We have utilized also statistical information about how many times per week people are shopping. Combining this data we get activity in shops and restaurants.

### Section 3.1

Pag. 2349 Lines: 13-15 157t PM2.5 emissions from wood combustion with respect to 322t from traffic seems quite low with respect to the estimation of house heating contribution to PM emissions in other European areas. Could you provide a little more detail about this evaluation? The referred paper is in Finnish.

- ✓ Currently there is a project ongoing financed by the Finnish entities to improve the estimation of such sources, but results are still uncertain. The inventory available from the Helsinki Region Environmental Services Authority (evaluation only published in Finnish) is based on registration data of small houses (type of heating systems in the city area), questionnaire of wood use in different heating systems (amount of wood used) and emission factors of sauna stoves, masonry heaters and boilers.

What is the fraction of total PM2.5 emissions due to wood combustion? It can provide a first estimate ion of the emission error/underestimation caused by neglecting this contribution.

- ✓ We have mentioned in section 3.1 the contribution of wood combustion to PM2.5 in Helsinki: 23%. The new revised draft we could add the figure presented in 3.1.

### Section 3.4.1

Pag. 2354 Lines: 2-3 The infiltration factor has been previously introduced as  $F_{inf} \leq 0$  (page 2345). Could you explain how can it assume values larger than one?

- ✓ The reviewer is correct, the infiltration factor can never be assumed higher than one. The reason the current version is stating values larger than one is because the authors were discussing indoor to outdoor concentration ratios (i/o); which can be higher than 1. The EXPAND model actually considers i/o instead of  $F_{inf}$  in order to accommodate indoor sources. In this particular study, we do not assume indoor sources and, therefore, the i/o will be the same as  $F_{inf}$ .
- ✓ We will revise: 1) section 5 adding the use of i/o in the model; 2) revise the lines 2-3 accordingly

### Section 3.4.2

Pag. 2354 Lines: 12-14 Can you give an estimate of the uncertainty due to the small scale combustion contribution?

- ✓ At the moment we can only say that we are missing 23% of the emissions, but because the spatial distribution of this data is not known, we cannot estimate the real impact of the wood combustion. What we can foresee is that it would have higher impact than the stationary sources since the emission height is closer to surface levels.

### Section 4

Pag 2355 Lines 1-9 It would be useful the introduction of a table resuming the improvements introduced in the new version of the presented model, e.g. through the comparison with the previous version features.

- ✓ This is a good addition to the paper, we will revise.

Lines:13-14 Stationary sources and shipping have been considered for 2009 simulations only (page 2340, lines 4-5).

- ✓ We will revise

Lines: 15-18 The fraction of PM<sub>2.5</sub> emission neglected should be possibly mentioned.

- ✓ We will revise

Pag 2357 Lines: 12-17 Data and models that need to be provided to make the proposed methodology applicable elsewhere should be better specified to make easier to understand the possible need of local investigations concerning e.g. emission details, infiltration factors,...

- ✓ We will revise

## Response to reviewer #2

General Comments It is good to see further refinement of detailed exposure models such as EXPAND, which are a critical element of air pollution research. This paper presents useful insights into the location of most of the exposure, and some insight into the sources responsible.

Since the revised model can handle population subgroups, it would be interesting to see the exposure results summarised by age groupings, given the differing sensitivity of various age groups to air pollution. Overall the paper presents the material in a credible way, by including caveats as appropriate ( e.g. the discussion about the fact that traffic congestion was not accounted for). In a few places some critical issues were not adequately covered (see Specific Comments section below).

- ✓ Thank you for these positive and encouraging comments about the manuscript. We will address the specific comments below.
- ✓ The results could indeed be summarized by age groups, or by some socio-economic criteria, such as by salary or prosperity of the population. However, this would mean a substantial amount of further computations and analysis; we would like to present such an analysis in a future manuscript.

It's not clear why the two regions studied (Helsinki Metropolitan Area, and the City of Helsinki) were analysed with different years. The paper would have presented a much clearer narrative by sticking with a single year, or by using both years together (i.e. 24 months of simulation), which would have allowed more rigorous comparisons of the city area with the broader metropolitan area.

- ✓ The computations of this study required a collection of a huge amount of data from different sources, and various time-consuming computations. The reason for selecting these two years and two domains is that the data and computational results were available from two (separate) research projects, for these specific years and domains. We admit that using results for a single year, or for a continuous two-year period would have been simpler and probably 'a more clear narrative', as stated by the reviewer.
- ✓ However, we feel that the main conclusions of this article are still valid, despite using these separate years and domains, especially those regarding the distribution of exposure spatially and to various micro-environments, and due to various source categories. Clearly, the concentrations, and therefore exposures, computed for the two years cannot be directly compared with each other. We suggest clarifying these limitations in a revised manuscript.

## Specific Comments

Page 2341, line 17 (section 2.2.2)

I note that a detailed model was used for computing shipping emissions, but were the shipping emissions treated as coming from ground level (sea level), or were they treated as stacks with a specific height and exhaust temperature & velocity? Ship exhaust stacks can be ~40m above sea level, with hot exhausts that give rise to significant buoyancy – this can significantly affect the predicted concentrations at ground level in populated areas.

- ✓ The STEAM shipping emission model includes a detailed database that contains technical properties of all major ships that travel in the Baltic Sea (as described in the references of articles by Jalkanen et al.). These data include also stack heights. So, the stack heights are allowed for, and the plume rises are also

treated in a simplified way. We suggest adding some description of these effects to a revised manuscript.

Page 2342, lines 17-20 (section 2.3)

This brief section is the only part of the paper that mentions meteorology, which is a critical input to air pollution modelling. For transparency and reproducibility, it is important for a modelling paper to describe or summarise all the key input datasets, including meteorological data.

I suggest a few key parameters be presented, such as example temperatures, wind speeds and mixing heights. This information can also be very useful for presenting exposure results in a weather context, for example, exposures can be summarised by month of year, or by temperature strata.

- ✓ We have used a meteorological pre-processing model for obtaining the meteorological input data for dispersion computations (named MPP-FMI). This model has used the data from three synoptic stations and one sounding station, to evaluate an hourly meteorological time series for the dispersion calculations. We should add a description of the details of the sources of met data to a revised manuscript.
- ✓ The dispersion models CAR-FMI & UDM-FMI are used to compute all the statistics based on hourly values. These models therefore generate 8760 cases /year, and the final results (yearly averages) are based on these “as-realistic-as-possible” separate cases. These data could be segmented in principle according to any met or other variable included in the input data or computations. Maybe we should clarify this in a revised manuscript.
- ✓ The (hourly) exposure results could indeed be segmented based on, e.g., ambient temperature, mixing height, wind speed, etc., or any combination of these, possibly with focus on cold spells or heat waves, which would provide for interesting new results. However, conducting such an exposure vs. meteorology assessment properly would be a fairly extensive study. While we agree with the reviewer that this is an interesting idea, we would like to suggest such an analysis to be presented in a future manuscript.

Page 2342, line 21 (section 2.3)

PM<sub>2.5</sub> was treated as a tracer contaminant in this study, however it is well known that PM<sub>2.5</sub> has a strong secondary component (both organic and inorganic aerosols). This is confirmed by the data in Table 2, which show that the concentrations in the shipping affected areas are not that much different from the urban background level – the urban background PM<sub>2.5</sub> being strongly affected by secondary production of aerosols from various sources, with the amount of secondary production depending on meteorology. There are also other factors that affect PM<sub>2.5</sub> - including sea salt and wind-blown dust, which do not appear to have been modelled in this study. These sources also depend on meteorology, and geographic factors such as proximity to the sea.

- ✓ For the computations in 2008, we have used the regional background concentration values computed using the LOTOS-EUROS model. This model includes formation of secondary aerosol, including sulphates, nitrates and ammonia (but not secondary organic aerosol). The contributions from sea salt, wild-land fires and elemental carbon are also included. The secondary PM<sub>2.5</sub> is therefore included with a reasonable accuracy in the regional background concentration values.

- ✓ However, the urban scale modelling does not include secondary aerosol formation. This is a common (state-of-the-art) assumption in urban scale dispersion models; although we admit that it is not totally accurate. We would like to clarify which sources and components were included, which were not, and the associated sources of uncertainty in a revised manuscript.
- ✓ For the 2009 computations, we have used the measured regional background concentration values from a regional background station. These values therefore, by definition, include all possible regional background contributions.
- ✓ Sea salt is not a major source of PM<sub>2.5</sub> for the Helsinki Metropolitan area. Sofiev et al (2011) has shown than in average, a concentration of sea-salt in Helsinki is on average < 0.2 ugPM/m<sup>3</sup>; the low value is mainly due to the low salinity of the Baltic Sea. The regional background station at Virolahti contains measurements of Na<sup>+</sup> in Eastern Finland, the values were of the order of 0.2 ugNa<sup>+</sup> in PM<sub>2.5</sub>. The wind-blown dust concentrations are also low on an annual average level, compared with e.g. the values in southern Europe.

Therefore it is likely that there would be some meteorological conditions (and locations) for which the predicted local contribution to PM<sub>2.5</sub> would vary significantly from the true contribution, due to the lack of PM<sub>2.5</sub> chemistry and treatment of natural sources. Given that population exposures were computed for each hour, the temporal variability in PM<sub>2.5</sub> is important.

Consequently, some assessment of model performance at shorter time scales than annual (Table 2) should be presented. I note (page 2350) that a verification study is to be presented in "Aarnio et al. (2014)", but this paper is still in preparation and is not accessible. There is a summary of findings in terms of an index of agreement and bias, but a graphical representation would be better in the current paper, e.g. a quantile-quantile plot, or a scatter plot. As part of this, some discussion of likely causes of disagreement between modelled and measured values (e.g. sources that are not modelled, or absence of chemistry, etc.) should be provided.

- ✓ We agree that this is a reasonable request. We will add some graphical presentations on the agreement of model predictions and measured data to a revised manuscript, with associated discussion on the reasons for disagreement.

Page 2345, line 4 (section 2.5)

The definition of the infiltration factor ( $F_{inf}$ ) appears to be a steady-state definition. In reality, there is differential equation that describes the dynamics of how outdoor air enters a building (accounting for air exchange rate, and other factors such as chemical transformation and destruction within the building).

This study has computed hourly PM<sub>2.5</sub> values and used these to derive exposure estimates in various microenvironments, with different infiltration factors. The use of steady-state infiltration factors probably means that the true indoor variation in concentrations may have been overestimated, since the outdoor PM<sub>2.5</sub> that enters a building will take some time to do so, resulting in a smoothing out of the variation in the outdoor signal.

The paper should note the impact of using steady state infiltration factors on estimated hourly exposure values.

- ✓ Yes, infiltration factor is a steady state concept in this study. This is a simplification of course, and we could allow neither for (i) the indoor sources and sinks of pollution, nor (ii) the temporal variation of

infiltration. The temporal variation of indoor concentrations would therefore be smoother, compared with our assessments, due to the delay associated with infiltration to indoors (as the reviewer commented). However, indoor sources such as tobacco smoking, cooking, heating, cleaning etc. can cause additional short-term concentration maxima. We should clarify these effects in a revised manuscript.

Page 2349, line 13 (section 3.1)

This text (under the Results & Discussion section) refers to small-scale wood combustion, and notes that this is a significant source, but then indicates that dispersion modelling was not possible due to a lack of knowledge about the spatial distribution of the source. It is standard practice in regional airshed modelling to spatially allocate the distribution of domestic emissions to a surrogate such as population density, in the absence of any more detailed information. This would have been more realistic than simply neglecting the source altogether.

A related question arises which is about all other domestic and small business emissions. In typical urban environments these sources can be significant, not only in terms of total emissions but also because they often emit pollutants near to ground level (unlike large industry which typically uses tall stacks). In policy terms, such sources are even more important, because (compared with industry and motor vehicles) they tend to be poorly regulated, and can assume a greater relative importance over time as vehicle sources become more tightly controlled. An overview of what is known about domestic and small business emissions in this region would be useful.

- ✓ Yes, we agree that an overview of this topic should be added to the manuscript. A mapping of small-scale sources is within this metropolitan region complicated by the fact that there are a lot of different small-scale heating systems (main heating system or a supplementary one), fireplaces, saunas, and a lot of different fuels, such as various wood products (dry, partially dry or partially wet), and a lot of different ovens (new efficient ones and very old and inefficient ones) and burning habits; all of these influence the amount of emissions. Anyway, we will compile a review of the scientific evidence on this topic.

#### Technical Corrections

Page 2366, line 12 (ABSTRACT) The revised model can also be used for evaluating intake fractions for various pollutants, source categories and population subgroups”

I suggest replacing “evaluating” with “estimating”, since this is a computer simulation of intake fractions.

- ✓ OK

Page 2336, line 17 (ABSTRACT) “The population exposure originated from the long range transported background concentrations was responsible for...”

This appears to be incorrect grammar. Should this have said “The population exposure originating from long range transported background concentrations ..” ?

- ✓ OK

Page 2337, lines 3-5 (INTRODUCTION)



Since the urban population spends typically 80–95% of their time indoors (Hänninen et al., 2005; Schweizer et al., 2007), the exposure to outdoor particles is dominated by exposure in indoor environments.

Delete the word “outdoor” here.

✓ OK

Page 2337, lines 5-6 (INTRODUCTION) The most simplistic approaches ignore indoor and outdoor conditions.

Presumably this should read “ignore the differences between indoor and outdoor air”.

✓ yes

Page 2347, line 6 (section 2.6)

The units presented for  $E_i$  are incorrect.  $\mu\text{g m}^{-3} \text{ s}^{-1}$  should be  $\mu\text{g m}^{-3} \text{ s}$

✓ Yes

Page 2368, Figure 2

In the figure caption, the greek letter “ $\mu$ ” should be used instead of “u” to represent the prefix micro (for micrograms per cubic metre). Also, if these are annual averages, this should be clearly stated.

✓ Yes; yes.

Page 2373, Figure 6

Population exposure is presented here in units of concentration x persons “( $\mu\text{g m}^{-3} \times \text{no. people}$ )”. However a true exposure metric must include the duration of exposure (i.e. a time element). I expect what is being presented here is annual average concentration x population density, which has the time unit removed by computing the annual average concentration. This needs to be clarified, and perhaps some appropriate terminology introduced, e.g. “population exposure per year.”

✓ Yes, it should be population exposure per year.

## **Relevant changes to manuscript**

The relevant changes to the manuscript are:

- ✓ Clearer description of the emission estimation
- ✓ Estimation of the contribution of small-combustion emission to the total emissions.
- ✓ Clearer description of the dispersion computations
- ✓ Assessment of the urban-scale dispersion model performance: modelled vs measured at two different measurement sites (scatter plot, new figure)
- ✓ Clarification of how to apply the proposed methodology to other regions.