

Interactive comment on “Modelling the dispersion of particle numbers in five European cities” by J. Kukkonen et al.

Anonymous Referee #2

Received and published: 15 September 2015

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.

Modelling particle concentrations is very challenging, and this paper makes a useful attempt at doing so. The results compare surprisingly well with measurements, but a number of key issues have been given insufficient consideration, which reduces the overall value of the study. The most important issue which is largely ignored is the

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sulphur content of motor fuel. This has a major impact upon the emission factor for particle number, and has been reducing for many years in Europe. The year chosen for modelling in three of the five cities is 2008, which was around the time that the sulphur content of motor fuels was decreasing rapidly in many European countries, from < 50 ppm S to < 10 ppm S. In late 2007, this was associated with a reduction in particle number concentration of around 65% at London, Marylebone Road, and a substantial but lesser decline ~39% at London, North Kensington (Jones et al., 2012). Hence, defining the sulphur content of fuel in each city is essential, but is not currently considered. The corollary to 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, which refer to the higher (< 50 ppm S) fuel sulphur, while the measurement year (2008) is after the transition to low S fuel. The suggestion (p5902, line 23-26) that these emission factors may underestimate those on this congested road is incorrect, as the field measurements were made on Marylebone Road! No doubt also of importance is the canyon nature of the site, which the authors recognise. For Helsinki, emission factors from Gidhagen et al. (2005) are used, which may also overestimate 2008 emissions.

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.

The authors recognise the distinction between the solid particle mode and the nucleation mode particles formed in the exhaust plume by condensation, but give it insufficient attention. The nucleation mode particles comprise semi-volatile organic compounds with a very small solid core. Such particles can evaporate if entering an environment with low concentrations of the associated vapour phase component (Dall'Osto et al., ACP, 6623-6637, 2011). Current knowledge of such processes is insufficient to include a deterministic description or even a meaningful parameterisation in numerical

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models. However, measurements of particle number concentrations in cities include these particles, and one implication is that the measurement method for particle number counts need to be specified. PNC measured by a CPC normally exceeds that measured by an SMPS, even if the greater losses in the latter instrument are accounted for. The usual reason is that the lower size cut of a CPC (depending upon model) will be $\sim 2.5\text{--}7$ nm, whereas most SMPS used in network monitoring have a lower size cut of ~ 15 nm (except Helsinki, which is specified), which cuts off the lower tail of the size distribution, particularly important if size reduction due to evaporation has occurred. Consequently, a modelling paper of this type should specify clearly what PN size range it is seeking to model. The evaporative shrinkage and loss of nucleation mode particles may explain why regional models tend to overestimate concentrations in this size range (p5888, lines 4-8).

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.

Two lesser points: (1) the Hoek et al. (2010) study is not the only source of exposure-response 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 difficult.

Interactive comment on *Geosci. Model Dev. Discuss.*, 8, 5873, 2015.

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