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Atmospheric Chemistry and Physics Discussions

Interactive comment on "The relationship between 0.25–2.5 µm aerosol and CO_2 emissions over a city" *by* M. Vogt et al.

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First we would like to thank the referee #1 for several useful comments and fair criticism that have helped improve the manuscript.

Interactive comment on "The relationship between 0.25–2.5μm aerosol and CO2 emissions over a city" by M. Vogt et al. Anonymous Referee #1 Received and published: 13 October 2010 General Impression Few datasets of direct flux measurements of pollutant emissions at the city scale exit, which could help shed light on the quality of emission factors or derive emission factors for metrics that are not currently treated by national bottom up emission inventories. The present study uses a long-term dataset of size-segregated particle flux measurements in an attempt to derive emission factors for traffic sources relative to CO2. This is a rather specific use of the data and in my

mind this manuscript and the one currently under review with Tellus-B would both have been strengthened from combining the two. The paper reads generally well, although the text should be carefully checked for linguistic issues, some of which are pointed out below. These should really have been ironed out before publication in ACPD. In addition, there are a number of scientific issues that need to be addressed before the paper can be accepted for publication in Atmos. Chem. Phys. C8630 The manuscript language has been corrected by a native speaker (New Zeelander Dr. Hamish Struthers).

Major Scientific Comments 1. The measurements were made 105 m above the ground. Because sub-micron particles start to experience a significant gravitational settling velocity, some of the material emitted from the city will re-deposit before reaching the measurement height. Thus, the fluxes derived here are representative of the net emission from the city, relevant for atmospheric transport models. By contrast, they are less representative of the amount that is actually emitted / lifted off the roads and therefore contributes to human exposure at street level. This needs to be clarified throughout the manuscript.

1)Author's response

Some text in section 3.7 has been added to clarify this issue.

2. The analysis is based on the fact that CO2 and the particles (in the size range measured) originate from the same source (which is already stated in the Abstract). The analysis, however, shows that this is clearly not the case:

a) The CO2 flux in some wind sectors is negative demonstrating that the CO2 flux is also affected by terrestrial sources and sinks. In addition, there other urban sources (gas central heating, cooking, ...), which are not considered here at all. How important are these in the flux footprint, according to the bottom-up emission inventories? Some indication on this in the manuscript would help. The relative effect from terrestrial sources/sinks is probably smallest in the North wind sector. While Fig. 3 is clearly limited to this sector, it is not clear whether the analysis for Figs. 4 and 5 was also limited to this sector. It probably should have been to minimize the effect.

2a)Author's response

The analysis for figure 4 and 5 has been made only for the northern sector which should limit the effect of other terrestrial sources like cooking etc to a minimum. This is a valid concern and we clarified it in the manuscript that we did not consider other sources than traffic. Anyhow, this approach should be valid considering the low number of houses with residential heating within the area. The emissions from residential heating are low and can be found in (Johansson & Eneroth, 2007).Some additional text in section 3.3 has been added to make our point. In the emission database for the footprint area the relative contribution for different sources is listed which are the following: For CO2 emissions within the footprint area energy contributes 8 %, road traffic = 80 %, sea traffic= 9 %, industry = 1 %, other = 2 %. So the emission inventory shows clearly that road traffic dominates.

b) Even if both CO2 and particles come from traffic sources, the process of emission is different as demonstrated by the wind speed dependence of the emission ratio for super-micron particles. If the super-micron particle flux is dominated by traffic-induced and wind-driven resuspension, it should be linked to km driven (and presumably traffic speed) and wind speed, respectively, rather than fuel combustion, as implicitly assumed by ratioing the emissions to CO2. It is also possible that even vehicle-induced resuspension needs to be supported by atmospheric turbulence for efficient transport of particles out of the street canyon. I would therefore urge the Author's to attempt an analysis in which they attempt to parameterise the super-micron flux through a two parametric parameterisation, using both traffic counts and wind speed.

2b)Author's response

An attempt to parameterize with wind speed and traffic counts has been made for particles sizes above 0.8 μm. It has been made for light and heavy vehicles and can be found in table 3.

3. There are many assumptions that go into the Ef derived by the NOx scaling method. Do the traffic counts on Honrsgatan not include information on light vs heavy duty vehicles? A further sentence or two on how the method works would help support the manuscript. Author's response

Some text in section in 3.4 has been added to clarify the method.

4. The relative emission factors shown on Fig. 5 and in Table 1 are not presented in a form in which they could be used by others. Firstly, like size-distributions they should be presented as a distribution function, i.e. normalised by bin width (dEf/dlogDp), because at the moment the values are specific to the bin width of the OPC used. Secondly, could these functions be parameterised, maybe as a composite of two log-normal modes? This would increase the chance of them being used by modellers compared with Table 1. The sentence at the end of page 21528 should be rephrased accordingly.

4)Author's response

It was not possible to parameterize with a two log normal mode without accepting large errors. This data set would require at least four modes, which does not reduce the number of parameters. Therefore it would probably not increase the possibility to be used by modelers. Table 1 shows the values which are specific to the bin width and Figure 5 has been removed.

5. Was the particle density validated by gravimetric analysis of the GRIMM filters?

5)Author's response

The particle density was not validated by the OPC that we used in the tower. However in agreement with referee 2 the particle densities have been calculated from Norman et al (Helsinki proceedings). The results showed densities typical of mineral particles as expected.

6. Was the Webb correction applied to the OPC? Does it apply?

6)Author's response

Our drying procedure, described in the manuscript the air entering the inlet is mixed one to one with completely dry and aerosol free air before the measurements in the OPC). This means that the absolute humidity before sampling decreases to half of its atmospheric value, and the relative humidity is correspondingly decreased. Therefore Webb correction does not apply. See Ahlm et al. (2010).

7. What is the overall uncertainty (rather than variability, as presented) in the emission factor, given the uncertainties in the CO2 flux from non-traffic sources, uncertainties in the flux measurements, uncertainties in the shape and density of the particles etc.?

7) Author's response

To determine the total uncertainty is complicated and not always possible. The overall uncertainty in CO2 fluxes was probably dominated by the uncertainty in sources. The non traffic CO2 sources where limited to 20% of the total CO2 emission, probably even less for the northern sector. The overall uncertainty in aerosol fluxes was probably dominated by the counting error, between 15 and 35% on average depending on size. The uncertainties in the flux might be quite different on a daily base, as it is really hard to determine the storage term in a heterogeneous area like a city. To assume some value here would bias the total uncertainty more than a difference in shape or density of particles. Therefore the variability is the most reasonable way to present uncertainty in the resulting emission factors. Note that section 2.2 includes information on the average errors and corrections of the data set.

Technical Corrections

Abstract, line 18: 'influences'

Page 21522, line 25: 'This is despite ...'

Page 21524, line 19: Either 'enables' or, better, 'enabled'.

Page 21525, line 3: 'of the communication tower'

Page 21525, line 21: 'been corrected for the limited ...' C8632 Page 21526, lines 9 & 16. 'data' is plural, i.e.: 'the data have been ...'

Page 21528, line 20. 'this has not been taken into account ...'

Page 21529, line 21: 'less brake wear production.'

Page 21530, lines 12 & 17: 'Nemitz et al.'

Page 21530, line 23: 'have a significant impact'

Page 21530, line 27: redundant 'were'

Page 21531, line 5: u* should have * as a subscript like elsewhere in the manuscript.

Reference list: format sub- and super-scripts throughout.

Caption Fig. 2: '(a) Average aerosol number ...'

Caption Fig. 4: for all wind sectors or N sector only? See above.

Caption Fig. 7: 'emission factor for 1 - 2.5 um particles ...' (In the air quality community

'coarse' typically relates to the fraction between 2.5 and 10 um, not to the fraction between 1 and 2.5 um.

Author's response:

Technical corrections have been made to the referee's suggestions.

Additional references not used in manuscript

Johansson, C. and Eneroth, K., 2007. TESS - Traffic Emissions, Socioeconomic valuation and Socioeconomic measures. PART 1. Emissions and exposure of particles and NOx. Environment and Health Administration of Stockholm, SLB-report 2007:2. Box 8136 104 20 Stockholm, Sweden. http://www.slb.nu/slb/rapporter/pdf/lvf2007_2.pdf

Zhang, L., Gong, S., Padro, J. and Barrie, L.: A size-segregated particle dry deposition scheme for an atmospheric aerosol model, Atmos. Environ., 35, 549–560, 2001.

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Ahlm, L., Krejci, R., Nilsson, E. D., Mårtensson, E. M., Vogt, M., and Artaxo, P.: Emission and dry deposition of accumulation mode particles in the Amazon Basin, Atmos. Chem. Phys., 10, 10237-10253, doi:10.5194/acp-10-10237-2010, 2010.

Interactive comment on Atmos. Chem. Phys. Discuss., 10, 21521, 2010.