Atmos. Chem. Phys. Discuss., 10, C14215–C14222, 2011
www.atmos-chem-phys-discuss.net/10/C14215/2011/

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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Received and published: 28 February 2011

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 #2 Received and published: 5 November 2010 General: The Author's present a very interesting data set of aerosol flux measurements at a tower in Stockholm. The topic is relevant and such data are still sparse in the literature and definitely worth publishing. The paper has a number of issues that need to be addressed in detail before further consideration. This regards some of the analysis, presentation and discussion of the data as well as the language of the manuscript that need to be carefully checked preferably by a native speaker.

The manuscript language has been corrected by a native speaker (New Zeelander Dr. Hamish Struthers).

Specific comments:

-Abstract: needs a bit more focus on the results, some introduction and general statements could be shortened or removed.

Author's response

Abstract has been shortened and should be more focused now.

-Page 21524, line 5-10: What about other sources to both CO2 and particles like house warming, industry and others? Can they be excluded for the case of Stockholm? Some numbers/facts documenting the dominance of the traffic source both for CO2 and particles maybe from an emission inventory would be needed.

Author's response

In Agreement with referee 1 Emission statistics can be found in (Johansson & Eneroth, 2007). For CO2 emissions within the footprint area energy contributes 8 %, road traffic = 80 %, sea traffic= 9 %, industry = 1 %, other = 2 %. So the statistic shows clearly that road traffic dominates.

-Page 21524, line 26-27 and Figure 1: The location of the tower and the discussed Söndra Länken should be clearly marked in the figure.

Author's response

The figure has been changed according to the referee's suggestions

-Page 21526, line 21: How do the (negative) CO2 fluxes for south easterly direction depend on time of the day and season? Photosynthetic activity should depend on light and temperature. Showing this in a graph or at least giving the results in the text would strengthen the argumentation very much.

Author's response

The season and diurnal variation of the CO2 flux is discussed in a different paper which is currently under revision in Tellus B. Since that review is not online, we added a figure (fig. 1) from that paper to show the CO2 and H2O fluxes. In the current manuscript, we have added a reference to the Tellus paper.

-Page 21527, line 2 and Figure 2b. The long range transport versus local urban contribution argument is not 100% convincing just looking at the plot. What about showing a day and a night average curve in Figure 2b? Local contributions to concentrations should be very different day/night (as for fluxes) and long range transport more constant.

Author's response

This is not a new phenomenon. It has been seen before (Areskoug et al., 2000; Tunved et al., 2005). Text and references have been added to in the updated manuscript.

-Page 21528, line 13-14 and Figure 4: Was this plot done for wind directions around 40-80 only? If yes, add in the text and figure caption. If not, why not, should this be better changed?

Author's response

The correlation has been made for 0-90. Some additional text in section 3.3 has been added to clarify the approach.

-Page 21528, line 25, Figure 5: The discussed "exponential increase" for the mass emission factor is probably the beginning of a nearly log-normal-distributed mode at a size larger than 2 μm. A similar mode can be assumed for number distribution below 0.25μm. This reveals actually to an important limitation of the study that should be discussed and argued for in the manuscript. The cut-off of the OPC it at the maximum of the size distribution both for mass as for number and the measured values will be very sensitive to the cut-off diameter of the instrument. How stable and exact is this

cut-off e.g. with changes in ambient conditions or operating conditions? Have you checked for influence of relative humidity on the data? This is also critical in relation to the reported number and mass emission factors. How well does the OPC- "PM2.5" emission factor compare with the e.g. TEOM-PM2.5 emission factor when measured at the same site (e.g. Hornsgatan)? Is there a systematic bias when you mix the different methods in Table 2? Page 21529, line 1-9: Include the TEOM PM2.5 data at Hornsgatan in order to compare the optical and the mass based methods.

Author's response

The reviewers concerns about the limitation of the study, is absolutely valid. Nevertheless this study is one of the few if not the only one where urban size resolved aerosol fluxes are measured. After long discussion with the manufacturer Grimm and their science and technical staff it became quite clear that there shouldn't be any concerns about the cut off diameters. The cut off does not change on the Grimm OPC with any operational or ambient conditions. It is true that one has to consider hygroscopic growth when measuring size-resolved particle number fluxes. However, in this study hygroscopicity seems to have a negligible impact on the results. The most important reason for this is that we have dried the aerosol before sampling. Our drying procedure is described in the manuscript. Hence, effects of hygroscopic growth on cuts are negligible in this study. Unfortunately we cannot calculate OPC-PM2.5 emission factors for Hornsgatan in the same way as is done for TEOM-PM2.5, since we do not have an OPC at roof-top(and the method relies on calculating the difference between street and roof). But the agreement of the two methods depends on what density we assume. Earlier calculations (Norman et al-., Helsinki proceedings) of particle densities based on measured volume concentration using OPC (assuming spherical particles) and measured mass concentrations using TEOM resulted in densities typical of mineral particles as expected, which indicates that the two different methods gives consistent results. But some systematic bias may not be ruled out since the comparison relies on several assumptions.

-Page 21530 and Figure 7: One would expect a very high correlation between u* and wind speed especially for the high values where thermal convections is less influencing. How well is the correlation in your data set? Are not both measures very similar and figure 7a and 7b are essentially showing the same effect? If this is the case just give one of the figures and also shorten the discussion. In case both figures contain essentially different information, some justification should be given in the text.

Author's response

It is true that the measured wind speed correlates very well with the friction velocity (r2=0.76). Therefore the friction velocity part has been removed from the text and the figure adjusted. In agreement with reviewer 1 a parameterization was made for particle fluxes larger than 1 μm with heavy and light traffic and wind speed. Note that the wind speed is measured at the top of the tower not at the street canyon.

-Page 21531 sections 3.6: The whole section need substantial rewriting Line 11-15, sentences not clear. Line 15: catalysts do normally not alter PM emissions (solid particles), do you mean particle filter? Since no differences in the contributions in exhaust/ non-exhaust emissions are given in Table 2 the discussion can not be followed by the reader. Consider to add the division in exhaust / non-exhaust if available in the reference in Table 2 or rewrite discussion.

Author's response

Section has been rewritten and exhaust/ non exhaust contribution illustrated in the table if the data was available.

-Have you estimated the range and importance of the various removal processed, deposition, gravimetric settling? Do rain days have significant influence, e.g. due to wet deposition?

Author's response

No estimation has been made to estimate removal processes. The measured flux is

the sum of emission and deposition. It would be desirable to be able to correct for dry deposition, or gravimetric settling in order to obtain the "real" emission. However, from our experience of using deposition models (Slinn (1981) or Zhang et al. (2001)) in the past, we know that they are very sensitive to parameters like roughness length, displacement height, sizes of collectors, wind speeds at reference heights and drag coefficients. All these parameters are highly uncertain for the site in this study. In addition, the OPC size range largely coincide with the accumulation mode, where aerosol dry deposition has a minimum Therefore, we are afraid that an attempt to correct for dry deposition would likely increase the uncertainty in the emission rather than decrease the uncertainty. Rain days on the other hand might have an influence but more in the case that the emission from wet streets is lower than on dry streets especially in spring time (Norman et al. 2006). We were thinking of removing data with wet streets, but this would bias the emission factor as well and therefore we kept the original approach.

Technical corrections:

Page 21523, line 3+4: Should this not be the other way around? measurements are needed to do the source apportionment? or do you mean AQ management??

Page 21524, line 19 . . .enables. . .

Page 21528, line 2-5 needs rewriting, not ". . .amount of fuel burned [g/l]. . ." but ". . . amount of emitted pollutant per amount of fuel burned [g/l]. . ."

C9476

Page 21528, line 8: Do you mean "15 CO2 flux intervals" not "concentration intervals"?

Figure 4, caption: remove one time " . . .the linear fit. . . "

Page 21529, line 21: Why only break wear is mentioned, not the other non-exhaust emissions (road and tyre wear, re-suspension)?

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.

Slinn, W. G. N.: Predictions for particle deposition to vegetative canopies, Atmospheric Environment, 16, 1785-1794, 1982.

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.

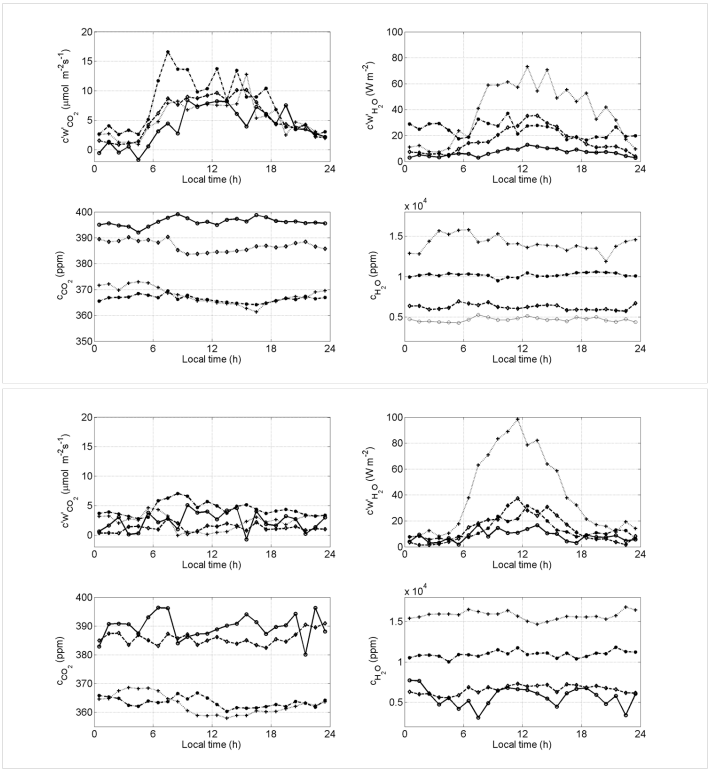

Fig. 1. Figure 8: Median diurnal cycles of seasonal fluxes for the Northern sector: (a) CO2 b) H2O and seasonal concentrations of (c) CO2, (d) H2O, in the Northern sector, (spring=diamond, plus=summer, star=a