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HIGH RESOLUTION MODELLING OF ELEMENTAL CARBON FOR DENMARK

*Matthias Ketzel¹, Madeleine Burman^{1,2}, Jacob K. Nøjgaard¹, Jesper H. Christensen¹, Ulas Im¹ and
Jørgen Brandt¹*

¹Department of Environmental Science, Aarhus University, Roskilde, Denmark

²Department of Physics, Lund University, Sweden

Abstract: Elemental Carbon (EC) has been in focus for a long time due to its importance for climate effects and human health (WHO / IPCC). The pollutant is released by incomplete combustion of carbon fuels, making road traffic, off-road machinery, and wood stove burning the largest sources of EC emissions. In this work, we investigate the distribution of EC over Denmark using a combination of three models with increasing resolution with a final resolution calculating EC concentrations at the street level. Results have been validated against measurements from four locations in and near Copenhagen.

Key words: *Elemental Carbon, DEHM, UBM, OSPM, High resolution modelling, Urban and street scale*

INTRODUCTION

The motivation for including EC as new pollutant in our modelling system is coming from the large concerns about the possible health effects caused by EC. EC has also been discussed as new pollutant in the EU air quality directive and many places long term monitoring of EC has been established. With this paper we would like to discuss EC modelling in the scientific community and contribute to model intercomparison and harmonisation of the modelling approaches.

There exist different similar terms or definitions for EC that are basically related to different measuring techniques as Black Carbon or Soot. In the modelling terms we do not distinguish between these terms and the modelled values should represent all the different terms in equal manner.

Both PM_{2.5} and PM₁₀ are released as primary particles (directly at source) originating from both natural and anthropogenic activity. Natural primary PM are, e.g., sea salt, wind dust, pollen, and wildfire particles (which can be argued to also be anthropogenic to the intended extent of forest fires). Anthropogenic primary PM are particles from combustion of wood and fossil fuel, agricultural processes, road abrasion, etc. EC is exclusively released from incomplete combustion of primarily biomass and fossil fuel or in connection with extraction and distribution of fossil fuel.

According to the Danish emission inventory from year 2014 (see Figure 1, Nielsen et al. 2015), the largest sources of PM_{2.5} and EC are road transport (including incomplete combustion of fossil fuel as well as road and tire abrasion) and non-industrial combustion (including residential woodstove-burning). This implies that concentrations are expected to be higher in locations where these sources are placed such as residential and urban environments.

In this work we introduce EC as new pollutant into our modelling system and validate the results with available measurements.

METHODOLOGY

A combination of three chemistry transport models (called the THOR system, www.au.dk/thor/) is used to calculate the air pollution concentration, recently including EC as pollutant, at different domain and scales:

- the Danish Eulerian Hemispheric Model (DEHM) to calculate the air pollution levels in the Northern Hemisphere with a resolution down to 5.6 km x 5.6 km over Denmark (Brandt et al. 2012),
- the Urban Background Model (UBM) to further calculate the air pollution in Denmark at 1 km x 1 km resolution using results from DEHM as boundary conditions (Berkowicz 2000a, Brandt et al. 2001), and
- the Operational Street Pollution Model (OSPM[®]) is applied to calculate the local contribution from road traffic in urban street locations (Berkowicz 2000b, Ketzel et al. 2012).

Emission factors for EC have been established as fraction of PM emissions based on COPERT methodology (Jensen & Ketzel 2013).

Modelled results are compared with the four available EC measurements in Denmark operated in the frame of the Danish Air Quality Monitoring Programme (Ellermann et al. 2016) and representing different concentration levels, covering rural background, sub urban, urban background and kerbside, see Table 1.

PM concentrations used here are measured using a low-volume sampler (LVS) that is a reference method according to EU First Daughter Directive 1999/30/EC. PM is collected on filters and weighted before and after exposure. EC is also collected on filters and analysed through a thermal-optical analysis method. The EC measurements used in this work are using the NIOSH thermal-optical transmission method (Birach, 2003).

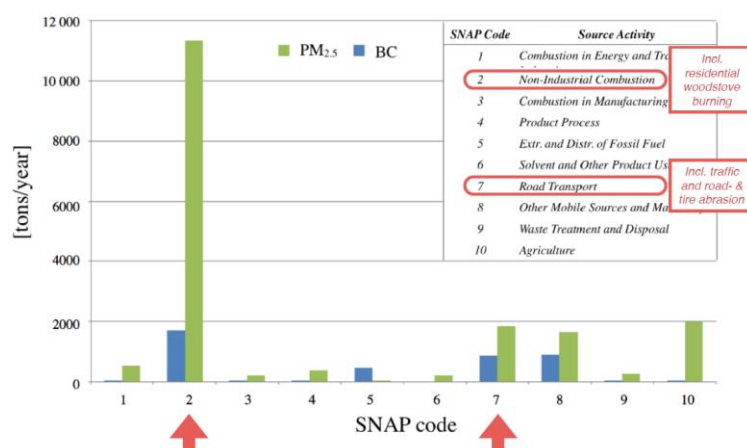


Figure 1. Sum of PM_{2.5} and EC emissions in Denmark divided into ten emission source categories. Main sources are marked and SNAP codes explained in the legend. (based on Nielsen et al. 2015)

Table 1. List of stations with EC observations in Denmark

Nr	Station Name	Short Name	Description	Location	Time Period
1	Risø	RISOE	Rural Background	Lille Valby	2010 - 2016
2	Hvidovre	HVID	Suburb	Copenhagen	2015 - 2016
3	H.C. Ørsted Institute	HCOE	Urban Background	Copenhagen	2014 - 2016
4	H.C. Andersens Boulevard	HCAB	Street (kerbside)	Copenhagen	2010 - 2016

The models were validated by comparing their output to measurement values using the programming language R (R; RStudio Team, 2017) including the R package 'openair' (Carslaw, D.C and K. Ropkins, 2012). All model outputs are in hourly time resolution, and the measurements are daily means. Routines are programmed in R to create different validation plots of the pollutants (Burman, 2017). Mainly three different type of plots for each pollutant and station were generated in order to extract different information:

1. Time series plots for the monthly mean values over the time period 2010-2016 showing all model outputs as well as the measurements. This type of representation of data is suitable for detecting

concentration differences between years as well as between models and measurements, finding outliers, and finding errors in the measurements or data handling.

2. Scatter diagrams and linear regression between model output and measurements. This plot type does not give information on temporal concentration patterns, but it provides a good method of comparing measurements and model output.
3. Time variation plots which, as the first plot type, also show all model outputs and the measurements but summarise the entire time series and provides monthly (or weekly) variations of concentrations. This representation type is suitable for detecting agreement between modelled and observed concentration differences between months e.g. seasonal variations due to changes in emissions or meteorological parameters as well as between weekdays due to changes in emissions.

RESULTS AND DISCUSSIONS

Figure 2 shows the monthly averages for observed and modelled EC ($\mu\text{g}/\text{m}^3$) at stations HCAB, HCOE and RISOE. Values at HVID station are very close to HCOE and only available for the latest 1½ years and were therefore omitted here for better readability of the plot.

By comparing concentration levels between stations in Fig. 2, it is evident that levels are higher in urban environments (HCOE) and substantially higher at street level (HCAB) compared to rural background (RISOE). This is expected as there are more emission sources (dominantly traffic) in urban environments and higher concentrations in street canyons. It also is apparent that concentrations are lower in the middle of each year (summer season in Denmark).

Studying the trend of the full time series, EC concentrations at HCAB decrease each year. This is also apparent for HCOE and RISOE but with less amplitude. Because traffic is the main contributor to EC concentrations at the street level, the decrease of EC levels with the years can be the result of implementation of stricter Euro emission standards for petrol and diesel vehicles. Another possible reason for the reduced concentrations is the use of cleaner use of woodstoves. The Danish EPA has campaigned at several occasions for the public to use cleaner methods for wood burning. In 2015, they launched a campaign that will give the public the change to receive funding from the government (up to EUR 290) if they eliminate wood stoves older than from 1990 from their homes (EPA 2014).

The model results reproduce the general decreasing trends and seasonal variation especially for RISOE and HCOE, discrepancies are seen for HCAB where the model underestimates as further discussed below.

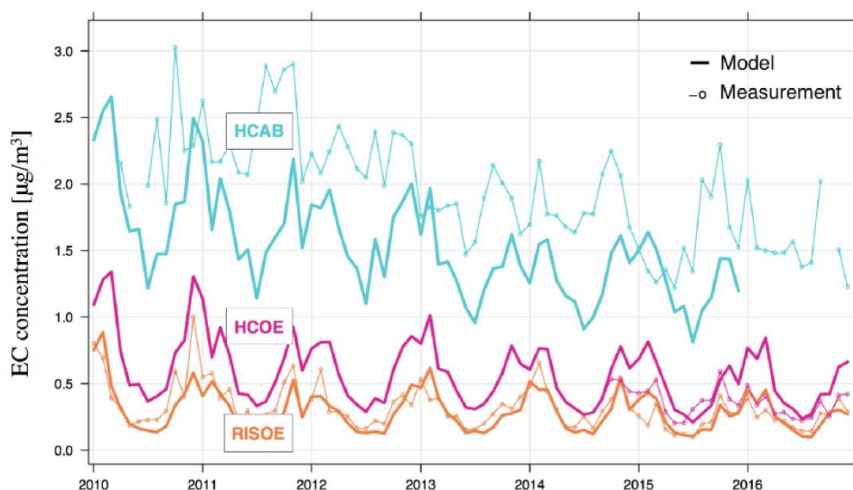


Figure 2. Trends of monthly averages for observed and modelled EC ($\mu\text{g}/\text{m}^3$) at HCAB, HCOE and RISOE (2010 – 2016). The model concentrations for RISOE are from DEHM, for HCOE from UBM, and for HCAB they are from OSPM.

Figure 3 shows as an example the monthly and weekly time variation for HCAB and RISOE. The largest gap between modelled and measured values at HCAB is found in the summer and autumn months, where all models estimate a decrease in EC as compared to the other months but the observed decrease is marginal. One possible reason for this is that the increase in tourism during summer and could lead to an increase in traffic possibly mostly tourist buses and taxis, that is not represented in the applied emission inventory for

HCAB. Another possible explanation is that during summer months more restaurants cook outside (street food, Tivoli nearby) resulting in an increase in combustion emissions. These reasons, however, do not explain the gap being so large between measured and OSPM levels. Since the OSPM model is based on input concentrations from UBM, the underestimation might partly lie in the UBM concentrations and not necessarily the OSPM.

For RISOE (Lower plots in Fig. 3 and Fig. 2), the DEHM output fits accurately to the measured concentration levels, but including UBM at RISOE leads to a clear overestimation of the levels. The same overestimation is, seen in all pollutants for RISOE which reflects a general problem in the model. Most probably the mixing height is too small in the UBM due to an exaggeration of the influence of the Roskilde Fjord, and hence it models for too high concentrations.

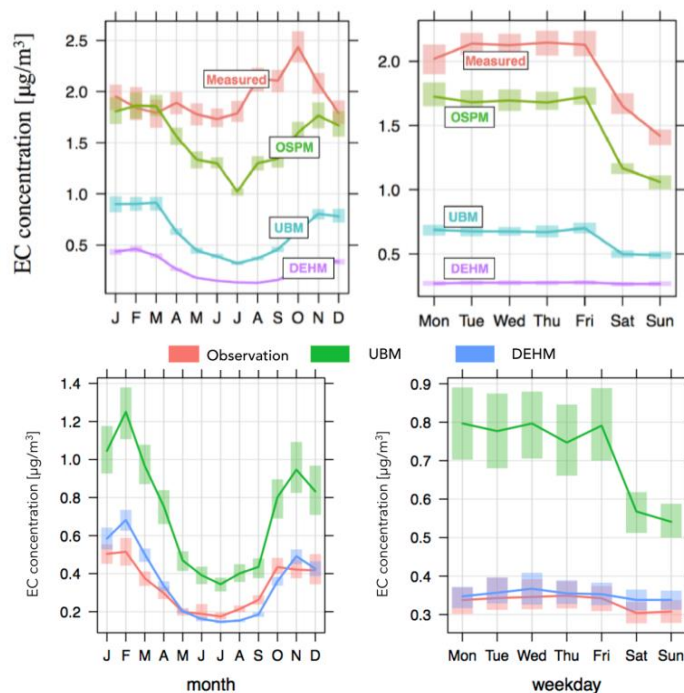


Figure 3. Monthly and weekly time variation in measured and modelled EC concentrations (in $\mu\text{g}/\text{m}^3$).
Top plots: for street station HCAB. Lower plots: for rural background station RISOE.

Figure 4 shows the 2015 mean concentrations from DEHM and UBM output. The improved resolution of EC modelling by moving from $5.6 \times 5.6 \text{ km}^2$ to $1 \times 1 \text{ km}^2$ leads to more visible details in the concentration patterns and as expected to higher maximum EC concentrations close to urban sources throughout Denmark.

CONCLUSION

For the first time we are able to present modelled EC concentrations over the whole of Denmark modelled in a fine spatial resolution of $1 \text{ km} \times 1 \text{ km}$. The three models DEHM, UBM and OSPM have been evaluated against measurements from 4 stations.

The different types of plotting (time series plots, scatter plots, average time variation) made it possible to not only evaluate trends in emission activity, but moreover to find areas where the models need to be improved.

In general, the measured EC concentration levels are well reproduced by the models at all 4 stations as well as the annual and weekly variation in concentrations with a few exceptions. For HCAB (street location) the model under-predicts the EC concentrations especially in the summer months, here the annual variation in the emissions need to be verified. For RISOE (rural station), the regional model shows a good agreement with the measurements, while the additional contribution from the UBM model gives too high concentrations, possibly attributed to unrealistic low mixing heights in the UBM model. More EC measurements are needed for a more sound validation of the model.

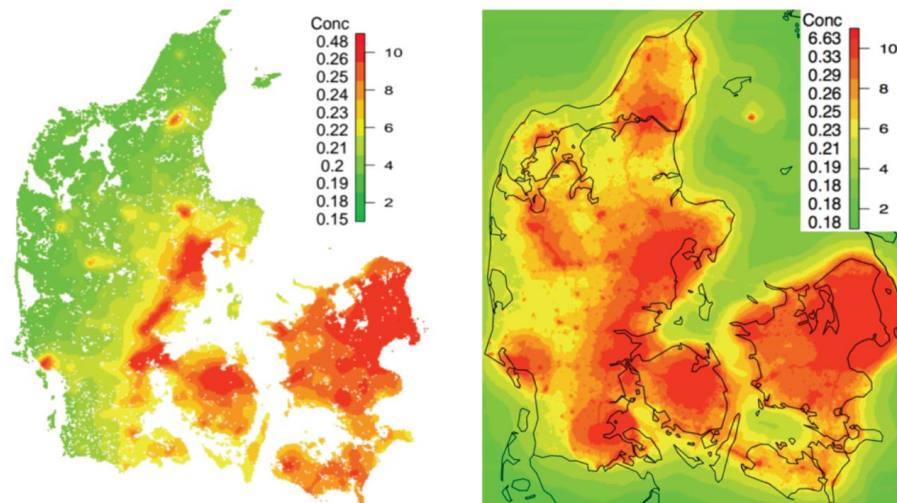


Figure 4. Annual mean concentrations of EC in μm^3 for year 2015 estimated with DEHM 5.6x5.6 km^2 resolution (left) and UBM in 1x1 km^2 (right).

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