Contribution from the ten major emission sectors in Europe and Denmark to the Health-Cost Externalities of Air Pollution using the EVA Model System – an integrated modelling approach, by Brandt et al.

Response to Anonymous Referee #2

GENERAL COMMENTS

Reviewer: The paper of Brandt et al. is dealing with the integrated model system EVA which is used to assess the health-related externalities of air pollution from specific emission source categories based on the SNAP code terminology. Main objective of the work is the identification of those source categories that contribute most to health related external costs. One aim is to support policy-making with respect to emission control. The study is focused on Europe and Denmark. The chemistry and physics within EVA is handled by the evaluated complex nonlinear chemistry transport model DEHM (Danish Eulerian Hemispheric Model) ranging from hemispheric scale with a horizontal grid resolution of 150 km to regional scale for Denmark with a grid resolution of 16.67 km. Europe is treated within an intermediate nest using a grid resolution of 50 km. Therefore the modeling system is able to handle long-range transport on hemispheric as well as regional scale. The modeling system due to its coarse horizontal resolution however is not sufficient for the urban scale and street canyons. Central for the model is the tagging method which allows for a better numerical treatment of the contributions of a specific emission source or category to the overall air pollution (delta contribution). Brandt et al. try to minimize numerical noise (Gibbs phenomenon) by using the tagging method instead of the more simple subtraction method. Brandt et al. state that the delta if using the subtraction method is in the same order as the numerical noise and therefore less accurate than the tagging method. However the tagging method needs more computer resources with respect to storage and time. The concentrations as calculated by DEHM are used as input for the computation of different health impacts of air pollution using exposure-response functions and health cost estimations.

The paper is well written, the model description is detailed and sufficient. The chemistry transport model DEHM takes into account the nonlinearities in the modeling system in particular within the nonlinear chemical reaction scheme. Its limitations are in the quite coarse horizontal resolution which does not allow for the detailed treatment of air pollution on the urban scale and within street canyons where high concentrations of NO2 and PM2.5 are measured. As in most complex modeling systems uncertainties in the large amount of parameters and input data used will lead to uncertainties in the model results. It is difficult to estimate the range of uncertainties in complex modeling systems as EVA but one should have in his mind that the numbers given in the paper, in particular on costs and health impact or air pollution may change considerably due to these uncertainties if increasing scientific knowledge leads to changes/updates in these parameters, e.g. on the toxicity of air pollutants, in particular for atmospheric aerosols (composition, size, form).

Answer: First of all, we would like to thank the reviewer for all the comments and suggestions to improve the paper.

We agree that it is very difficult to estimate the range of uncertainties in such a complex model system. We highly agree to the statement that “numbers given in the paper, in particular on costs and health impact or air pollution may change considerably due to these uncertainties if increasing scientific knowledge leads to changes/updates in these parameters, e.g. on the toxicity of air pollutants, in particular for atmospheric aerosols (composition, size, form)”. The main open question in calculations like the one we have conducted is precisely the toxicity of the particles, which in the case where it is possible on solid scientific grounds to distinguish the toxicity from the different types of particles will influence the results. The EVA system has

already been developed to take into account different toxicity associated with different particle compositions if the information is available (see e.g. the sensitivity analysis where higher toxicity is assigned to primary particles) and will be further developed in the future according to new knowledge in this area.

We also agree that it is the goal that higher resolution should be obtained in order to assess the health impacts and related cost in the urban environment. In fact, we have just developed the EVA system further to include an urban background model with a resolution of 1 km x 1 km to describe the urban increment and to describe the exposure at a much higher resolution in highly populated areas. The results from this work will be published in a future paper.

Reviewer: I have some general recommendations with respect to the tagging methods, which is termed as central for the paper and the uncertainties related to parameterization and horizontal grid resolution.

I recommend illustrating the improvements achieved by the tagging method with concrete numerical examples to give the reader an insight into the orders of magnitudes for the delta in both methods and the numerical noise (which also might depend on the numerical method used to solve the dynamic equations).

Answer: We agree that testing the tagging method for improvements compared to the traditional subtraction method is important and interesting. However, the improvements depends very much on the scenario runs defined – i.e. on the size of the emission reduction change, on the geographical distribution of emissions and whether there are strong gradients in the emissions. A single plot showing a comparison of the two methods is therefore not very illustrative, and we prefer to make a thorough testing and evaluation of the tagging method in another paper. We plan to publish a separate paper on the numerical testing evaluation of the tagging method in another paper, which is in preparation.

Reviewer: Further I recommend discussing the consequences of uncertainties in the parameters used in the EVA system on the numbers given for health impact, mortality and external costs. To my opinion it should be clarified that these numbers are not completely on safe ground in the sense that we really know how many people suffer in Europe from air pollution and what the costs are. Each number is based on model calculation and the underlying empirical assumptions with a lot of uncertainties. There is no direct evaluation of the model output possible; it might be even never possible for the whole of Europe or Denmark. This is not to criticize the study, my opinion is that the study illustrates on a good scientific basis as Brandt et al. stated, the capability of the EVA system to give useful input for the planning of regulation policies. But one should be careful to take the numbers as they are without taking a very critical look on the uncertainties/limitations in the EVA system (parameters, emissions, horizontal resolution) and the empirical relations for the calculation of health effects (exposure-response functions) as well as economic valuation. With respect to the DEHM it should be noticed that horizontal resolution is known to have in particular strong impact on high concentrations of NO2 and to a lesser extent on PM2.5 in source dominated regions. Both are primary emissions leading to high concentrations e.g. in street canyons. These peak concentrations cannot be simulated with a coarse horizontal resolution of 16.67 km.

Answer: We will add a a discussion of the uncertainties and the model systems sensitivity to parameters chosen in the paper. As the reviewer states it is impossible to make a quantification of all the uncertainties in the system. The sensitivity to some of the sources to uncertainty will be examined in future work. For example, the sensitivity of the results to grid resolution. As stated we have just developed a version with very high resolution, which we will use to assess the sensitivity to resolution. We agree that the system might be less sensitive to resolution with respect to $PM_{2.5}$ compared to NO_2. At the moment we have not included concentration response functions for NO_2, since we consider these to be too uncertain presently, even though it is commonly known that NO_2 is associated with health effects. At the moment, it is also unclear what the additional health effects are from short-term and high level exposures in street canyons.

Reviewer: Nevertheless, with these limitations in mind, to my opinion the study by Brandt et al. is an interesting and valuable example for the application of a sophisticated complex chemistry transport model embedded into the EVA system. Brandt et al. have shown in a very impressive way how such a model system can be used as a valuable tool for policy planning and regulatory measures. A complex modeling system like EVA never can be perfect and is – due to its interdisciplinary complexity going from atmospheric dynamics and chemistry to exposure-response and costs of health impacts of air pollution - in a permanent process of further development with respect to increasing scientific knowledge in all the areas. Therefore a permanent update of all parts of the model is always necessary, but nevertheless the EVA system is a good basis for that. I am looking forward to the next steps in the future developments of EVA going perhaps to urban scale and street canyon modeling, improvement of parameters and exposure-response functions. In its current stage the EVA system is a good basis for further improvement and application of complex modeling system which might be useful to optimize air pollution reduction strategies in Europe. Therefore I recommend the paper for publication in ACP.

Answer: We would like to thank the reviewer for the nice comments. We agree that this is the first version of the EVA system, and that it should be continuously updated with new knowledge and data, especially with respect to resolution and toxicity of species. Furthermore, it is important to obtain mass closure for $PM_{2.5}$ e.g. by including secondary organic aerosols.

SPECIFIC COMMENTS

Reviewer: p. 5874, line 5, abstract; p. 5880, section 2.3; tagging method
The tagging method is termed as central and I agree with Brandt et al. that the tagging method might be a central and valuable development to get more sophisticated results for the delta-concentrations. Therefore I would appreciate if this could be illustrated by concrete numerical examples in section 2.3 to give a better insight into the numbers we are talking about if we discuss “numerical noise, Gibbs phenomenon”, delta-concentrations by subtraction method and delta-concentration by tagging-method. Numerical noise also might depend on the method used for the solution of the transport part in the DEHM.

Answer: As stated above, we agree that testing the tagging method for improvements compared to the traditional subtraction method is important and interesting. However, the improvements depends very much on the scenario runs defined – i.e. on the size of the emission reduction change, on the geographical distribution of emissions and whether there are strong gradients in the emissions. A single plot showing a comparison of the two methods is therefore not very illustrative, and we prefer to make a thorough testing and evaluation of the tagging method in another paper. We also agree that the numerical noise depend on the advection algorithm chosen, however all modern higher order advection schemes include numerical noise. We have previously made great efforts to choose the best performing advection scheme (see Frohn et al. 2002).

Reviewer: Different methods might show different numerical noise and therefore might influence the improvement which is achieved by using the tagging method instead the subtraction method. It might be helpful to have a discussion on this issue according to the following questions just to emphasize the importance of the tagging method:

What is the order of magnitude of numerical noise e.g. for a certain scenario for the different air pollutants calculated by the model? What is the value of the delta as calculated by the subtraction method in comparison with the delta calculated with the tagging method? Is the difference of the two methods in the order of some per cent or is it up to 100% or even more? Does the difference between the methods

depend on scenario or/and constituents which is considered? How is the numerical noise influenced by the numerical method selected to solve the transport equations in the DEHM-CTM?

Answer: We will include a discussion of the difference of the two methods in a qualitative manner in the paper as requested. The order of magnitude of the signal to noise ratio can be very large using the subtraction method, when tested for strong single point sources. However, the magnitude of noise comparing the two methods depends very much on the chemical species and the emission scenarios under investigation. We initially decide to develop the tagging methods while trying to assess some emission reduction scenarios, where the noise was much bigger than the deltas, and with the tagging method, the signal was very clear and the noise was not a problem anymore.

Reviewer: p. 5874, line 15 "When quantifying emissions, more than ten major emission sectors are defined . . ." My recommendation: Just define the term "SNAP category" here, it is done later in the text, but the ten emission sectors are mentioned here for the first time and perhaps a hint to SNAP code terminology and table 2 might be helpful for the reader even in this early place in the introduction.

Answer: Done

Reviewer: p.5878, section 2.2: The Danish Eulerian Hemispheric Model What is the height of the upper boundary of the model (layer 20)?

Answer: The top of the model is at 100 hPa corresponding to 15-18 km height. This information is now included in the model description.

Reviewer: How is ozone treated, in particular for the upper part of the model (upper troposphere)? Is the exchange between stratosphere and troposphere taken into account?

Answer: For ozone, the initial and boundary conditions are based on ozonesonde measurements, interpolated to global monthly 3D values with a resolution of 4^{o} x 5^{o} (Logan, 1999). The exchange between the stratosphere and troposphere is taken into account. This information is now included in the model description.

Reference:
Logan, J. A., 1999: An analysis of ozonesonde data for the troposphere: Recommendations for testing 3-D models, and development of a gridded climatology for tropospheric ozone, J. Geophys. Res., 104, 16, 115-16, 149, 1999.

Reviewer: What is the thickness of the lowest layer?

Answer: The thickness of the lowest layer is 15-20 meters. This information is now included in the paper.

Reviewer: Are natural NO-emissions included in the model?

Answer: With respect to emissions from natural sources, emissions from retrospective wildfires are included based on Schultz et al. (2008). The current version of DEHM includes the temporal allocation of emissions from the IGAC-GEIA biogenic emission model for biogenic isoprene (International Global Atmospheric Chemistry – Global Emission Inventory Activity; Guenther et al., 1995) as an in-line module. This module is presently being replaced by the Model of Emissions of Gases and Aerosols from Nature (MEGAN) (Guenther et al., 2006, Zare et al., 2012), also including a module for the formation of secondary organic aerosols (SOA) (Zare et al., 2013), however, these implementation were not available for the

present study. Natural emissions of NOx from lightning and soil as well as emissions of NH_3 from soil/vegetation based on GEIA are also implemented in the model.

This information is now included in the text.

References:
Guenther, A., Hewitt, C., Erickson, D., Fall, R., Geron, C., Graedel, T., Harley, P., Klinger, L., Lerdau, M., McKay, W., Pierce, T., Scholes, B., Steinbrecher, R., Tallamraju, R., Taylor, J., and Zimmerman, P., 1995. A global-model of natural volatile organic-compound emissions. Journal of Geophysical Research, 100 - Atmosphere:8873–8892.

Guenther, A., Karl, T., Harley, P., Wiedinmyer, C., Palmer, P. I., Geron, C., 2006. Estimates of global terrestrial isoprene emissions using MEGAN (Model of Emissions of Gases and Aerosols from Nature). Atmos. Chem. Phys. *6*, 3181–3210.

Schultz, M. G., A. Heil, J. J. Hoelzemann, A. Spessa, K. Thonicke, J. Goldammer, A. C. Held, and J. M. Pereira, 2008. Global emissions from wildland fires from 1960 to 2000. Global Biogeochemical Cycles, 22:B2002, April 2008.

Zare, A., J. H. Christensen, P. Irannejad and J. Brandt, 2012: "Evaluation of two isoprene emission models for using in a long-range air pollution model". *Atmospheric Chemistry and Physics*, 12, pp. 7399-7412, 2012. doi:10.5194/acp-12-7399-2012.

Zare, A., J. H. Christensen, A. Gross, P. Irannejad, M. Glasius and J. Brandt, 2013: Quantifying the contributions of natural emissions to ozone and total fine PM concentrations in the Northern Hemisphere. *Atmospheric Chemistry and Physics Discussions*, Vol. 13, pp. 16775-16830, 2013.

Reviewer: It is not clear to me how the different classes of the particulate matter interact? Is coagulation, condensation, nucleation included? Is a modal or sectional approach used?

Answer: The DEHM model does unfortunately not yet include a particle module for coagulation, condensation, or nucleation. We have until now prioritized to obtain mass closure in the model and is presently working on a module for SOA as explained above. However, particle classes are divided into $PM_{2.5}$, PM_{10} and TSP.

Reviewer: Is the formation of secondary organic particles considered? Is there an interaction between clouds (cloud water, aqeous pahse) and particles? How is the interaction of gases and particles treated?

Answer: As mentioned above a module for formation of secondary organic particles is presently being included and evaluated (Zare et al. 2013) but SOA is not included in the present calculations. Aqueous phase chemistry is included, but not directly for particles. The interaction of gases and particles is a longer story. The model includes 122 chemical reactions, where many of them concern the formation of secondary inorganic aerosols. The total $PM_{2.5}$ in the model consists of the sum of the species: primary emitted mineral dust, black carbon (fresh and aged), organic carbon, and the secondary formed particles included in SIA: SO_4^{2-}, NO_3^-, NH_4NO^3, NH_4HSO_4 and $(NH_4)_2SO_4$. Sea salt has been implemented after the submission of this paper.

Reviewer: p. 5879, section 2.3: The tagging method as mentioned above: Did Brandt et al. perform a model run for the same case a) using the tagging method and b) using the subtraction method? What are the differences with respect to the constituents and the source category considered?

Different constituents might show different performance in the delta, also for the different source categories? Is the subtraction method, which is commonly used, completely insufficient in all cases? How is the delta influenced by different numerical solution methods of the transport equation?

The tagging method is one of the central issues in this paper as claimed by Brandt et al., I would appreciate if there will be one example which underline the improvement in calculating the delta by the use of the tagging method.

Answer: See the answer concerning the tagging method above. We plan to publish the numerical test and evaluation of the tagging method in another paper. We understand the thinking of the reviewer that just one example illustrating the performance of the tagging method would be desirable. However, as we see it, it is not so simple. The performance of the tagging method compared to the subtraction methods is very varying from essential to nearly no difference, depending on the chemical species and emission scenario under consideration. The tagging is most important when considering strong gradients in the emission fields, especially strong point sources, which results in Gibbs phenomenon. Of cause, the noise also depends very much on the advection scheme. Nearly all advection schemes result in unwanted oscillations, and is together with accuracy used together with a chemical scheme a part of the testing when advection schemes are chosen. The challenge is to find the right balance between all the desirable properties of the advection schemes. We have chosen a higher order scheme based on research over more than two decade (see Frohn et al., 2002 for a discussion and documentation of the advection scheme chosen in DEHM). Some advection schemes have less problems with unwanted oscillations, e.g. the flux schemes, however they are not always mass conserving, which is indeed also a desirable property. Since the advantage of using the tagging methods depends on all these aspects, we would sincerely like to document this properly in a paper especially focused on this issue instead of showing a selected case where the tagging scheme totally outperforms the subtraction method.

Reviewer: p.5882, section 2.5: Exposure-response functions and monetary values Compounds included in the EVA system are: O3, CO, SO2, SO2-4, NO-3, primary emitted PM2.5.

Can Brandt et al. comment on NH4- ? Is it part of DEHM or EVA? Is benzene or other hydrocarbons considered? What about PAH?

Answer: NH4+ is a part of DEHM and is included in the chemistry part of the model emitted as ammonia. In this case it is especially interesting with respect to the formation of ammonium-nitrate and ammonium-sulphates. Benzene and a long list of other hydrocarbons are included in the model as well as PAH – a total of 58 chemical species and 9 primary particulates.

Reviewer: How is primary PM2.5 separated from secondary PM2.5 which might be formed by gaseous precursors during transport?

Answer: The primary part of $PM_{2.5}$ is handled for alone in the model and only involves primary emissions and advection (including 9 particles). The secondary particles are emitted as gases in the model and are formed via chemistry and also advected.

Reviewer: Maybe that I missed that somewhere, but what is WTP (p. 5885, line 21) p. 5896,

Answer: WTP is the “Willingness To Pay” concept for valuating impacts. This is now more clearly explained in the paper.

Reviewer: section 5: Discussion and overall conclusions:
As mentioned also by Brandt et al., there is a lot of uncertainties in parameters (e.g. exposure-response functions) or input data (as emissions) used in the DEHM as well as in the exposure-response functions and the numbers used for the calculation of the costs. I recommend discussing the uncertainties of the model outcome (e.g. number of deaths attributed to certain species and source categories with respect to changes in parameters, input data (emissions), horizontal resolution (which cannot account for the urban scale and street canyon concentrations) with the aim to make clear that the current application is a starting point for further improvements and development of the EVA system.

Answer: We agree with the reviewer and the following is now included in the discussions: The current application on regional background scale is a starting point for further improvements and development of the EVA system. Besides the above mentioned uncertainties used in the DEHM, the results can be sensitive to spatial resolution in the model system. Presently, the system describes the health impacts down to 16.67 km^2, however, this resolution is not optimal for describing the condition in the urban background or even in street canyons. Presently an urban background model is being implemented in the model system which can describe the conditions in the urban background with a 1 km^2 resolution. The difference between using a relatively course and a relatively fine resolution will be evaluated in the future.

The model outcome in the form of health impacts and economic valuation is, of cause, very much depending on the concentration-response functions and economic unit prices chosen. However, as both functions are considered linear in the system, another choice of concentration-response functions and economic valuation will have a linear response in the system, and sensitivity to these functions can be applied afterwards. E.g. applying a relative risk of 1.03-1.12 instead of 1.06 presently used would imply similar linear responses in the health impacts and economic valuation of half or double the number of premature deaths and equally for the external costs. Furthermore, the response to assigning different toxicity to primary particles and secondary inorganic particles as carried out in the paper by factors of 1.3 and 0.7, respectively, can also be done post to the calculations as impacts and costs are assigned to the individual species before adding everything up to the total impacts and cost. However, many of these uncertainties would not change the relative contribution from the different emission sectors. The main uncertainty is associated with the equal toxicity assigned to all the particles, and the relative contributions would be very sensitive this the choices of different toxicity for different types of particles (e,g. assigning much less toxicity to SIA and much more to primary particles would decrease the relative contributions of e.g. the agricultural sector and increase the relative contribution of e.g. road traffic and domestic heating.