Atmos. Chem. Phys. Discuss., 12, C12204–C12211, 2013
www.atmos-chem-phys-discuss.net/12/C12204/2013/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "The relative importance of impacts from climate change vs. emissions change on air pollution levels in the 21st century" *by* G. B. Hedegaard et al.

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Received and published: 31 January 2013

Answers to Anonymous Referee #2

Reviewer: The study addresses an interesting issue relevant with the compared impact of climate change and emissions on the air pollution changed levels in the 21st century. The scientific tools used to address the issue are well documented and the methodological approach is scientific sound. The paper gives some answer to the question of whether climate change or emissions can drive mostly the pollutant concentration changes in the future. However, in many cases, in-depth explanations of the reasons why climate change or emission changes can be more effective in the determination of

the future air pollution changes are lacking. Under this view, the manuscript can only be published after important additions and more discussion about the physical and chemical mechanisms that contribute to the air quality change either due to climate change or to changes in pollutant emissions.

Answer: We would like to thank the reviewer for the very nice comments about the relevance of the paper and the methodology used. The reason for the lack of more in-depth explanations of the underlying physical and chemical processes that contribute to air quality change, is that we have already published this in two previous papers (given below), where the first is referenced in the paper, whereas the second one was published after the submission of this paper. These papers describe many of the underlying physical and chemical processes, and therefore it was natural for us to take the step forward and focus on an assessment of the impact from climate change vs. emission changes. To include a discussion of the underlying physical chemical processes, would be a repetition and should therefore not be included in this paper, except for the discussion of changes in precipitation and mixing heights, which are included here and relevant for the particles. The underlying chemistry is explained in Hedegaard et al. (2012). However, we have gone through the paper and extended the discussion about the underlying physical and chemical mechanisms that contribute to the air quality change either due to climate change or to changes in pollutant emissions, either as additions to the text or as a reference to the papers below.

Hedegaard, G. B., J. Brandt, J. H. Christensen, L. M. Frohn, C. Geels, K. M. Hansen and M. Stendel, 2008: "Impacts of climate change on air pollution levels in the Northern Hemisphere with special focus on Europe and the Arctic". Atmospheric Chemistry and Physics, 8, 3337-3367, 2008.

Hedegaard, G. B., J. H. Christensen, C. Geels, A. Gross, K. M. Hansen W. May, A. Zare, and J. Brandt, 2012: "Modelling the Impacts of Climate Change on Tropospheric Ozone over three Centuries". Atmospheric and Climate Sciences, 2012, Vol. 2, No. 4, pp. 546-561.

Reviewer: 1) In the manuscript, primary sulfates, nitrates and ammonium are no mentioned. How are they addressed in the analysis?

Answer: In section 4.2 in the paper, the sulfates, nitrates and ammonium are mentioned: "The total PM2.5 consists in the model of the sum of the following species: primary emitted mineral dust, black carbon (fresh and aged), organic carbon, and the secondary formed particles H2SO4, NO3−, NH4NO3, NH4HSO4 and (NH4)2SO4. Secondary formed organic aerosols (SOA) are not included in the current model setup". We do not understand the comment concerning "primary sulfates, nitrates and ammonium", since they are all secondary formed inorganic aerosols and not primary species. The DEHM model, which is documented in detail in Brandt et al. (2012) (see reference in the paper), includes 58 photochemical species and 9 primary particles. We have not focused on their primary components, (sulphur, NOx and ammonia) as atmospheric concentrations, since the focus of the paper has been on the chemical species, which have an important impact on human health (ozone and particles) and on eutrophication of ecosystems (total N deposition). Vi did analyze results for NO2, since this also are related to health impacts, but did not find any significant effects on the hemispheric scale.

Reviewer: 2) Natural emissions are represented mainly by isoprene emissions. Dust emissions are they included in model runs? Which other natural sources are missing (eg sea salt)?

Answer: The focus of this paper has been on the impacts from changes in anthropogenic emissions, however, the ozone chemistry is very important with respect to this matter and therefore the natural isoprene emissions are, of course, also included.

The natural emissions of dust or sea salt are very important for simulating the total particulate matter, however, they are treated as inert tracers in the model and therefore, they do not influence the chemistry of the anthropogenic sources and can just be added to the total PM. Since we have chosen to focus on the impact from anthropogenic

emissions, we have not included dust and sea salt in this study. However, we are right now working on a paper with focus on future climate change impacts on natural emissions in general, including natural dust, sea salt, and the formation of secondary organic aerosols (SOA) from biogenic emissions of mono-terpenes, etc. Furthermore, an update of the isoprene emission will be included in this study.

Besides these types of natural emissions, soil emissions, emissions from lightning and emissions from wild fires are included in the model, but only as emission data included from databases and not modeled within the model. Even though the emissions are included in the model, we have no description of how climate change will affect them and therefore, it is presently impossible to assess the impacts from climate change on these emissions at the moment.

Reviewer: 3) Figure 1 and 2 shows changes in precipitation frequency and mixing height between present and future time. How can these changes be explained?

Answer: The changes in the precipitation and precipitation frequency is a common prediction from many of the climate models, which is a result of warmer temperatures, giving rise to more evaporation and cloud cover in some regions and less in other regions. In some cases the weather systems are simply moving towards north, which is seen e.g. over the Pacific Ocean, where it is most clear, giving rise to the sharp gradients in the difference between the mean values for the two decades. In the European region the paths of the low pressure systems are also moving towards north with similar changes in precipitation patterns. The change in precipitation and especially in the precipitation frequency has an immediate impact on concentration levels of atmospheric particles via increased or decreased wet deposition, which are clearly seen in Figures 4 and 5.

The change is mixing height is driven by two processes: Firstly, the future changes in cloud cover have an immediate impact on the global radiation and therefore on the energy balance at the surface, which has a direct effect on the mixing height. Secondly,

future changes in surface wind speeds, due to changes in the general atmospheric circulation, have a direct impact on the turbulence near the surface and therefore the mixing height.

Reviewer: 4) In the paper there are no figures to show the changes between present and future time emissions for chemical species like ozone precursors (NOx, VOCs), BC, PM2.5. These figures could better help the reader to understand the air pollution changes linked to emission changes (figures 3b, 4b, 6b and 7b).

Answer: We will consider to include these figures in the paper.

Reviewer: 5) The negative values in the figures 3d, 4d, 6d and 7d should be presented with separate bins in the arithmetic scale. Provide explanation why they appear.

Answer: The figures 3d, 4d, 6d, and 7d are explained in the end of section 2 as follows:

"For CS/ES= 1: the climate and emission signal is of equal size and sign and the sign can be determined from Eqs. (1) and (2).

For CS/ES> 1: the size of climate signal if larger than the size of the emission signal and both effects are either both positive (increasing) or both negative (decreasing) and therefore results in an amplified effect on a given concentration or deposition (x).

For CS/ES< 1: the emission signal dominates and either the climate signal or the emission signal is negative. The sign of the two signals can again be determined from Eqs. (1) and (2) (see e.g. Fig. 3a and b in the case of ozone concentration)."

The values are therefore around 1 and with no negative values, since we are only examining the absolute signals from climate change (CS) vs. emission change (ES) in these figures. The sign of the values can be found in the a and b figures. There is therefore no need for separate bins for negative values in the arithmetic scale.

Reviewer: 6) The changes in physical and chemical processes that cause the PM2.5 concentration changes and the climate to emission signals ratio shown in figure 6 are

not thoroughly explained in section 4.2 (as it is done for BC). Probably a figure could be presented also for PM2.5 that is similar with the figure 5.

Answer: We found that the results for PM2.5 were very similar to those for BC, and therefore we chose not to include a figure for PM2.5 similar to figure 5 in order to keep the length of the paper reasonable. Once formed in the atmosphere, the physical processed influencing PM2.5 concentrations behave similar to BC and therefore, we did not duplicate the explanations from the BC.

Concerning the chemical processes involved in forming the secondary inorganic aerosols (SIA), which are different from the inert tracers of e.g. BC, the most important process is the climate change impacts on OH concentrations. OH is very important for the life time of the primary emitted species; i.e. the speed they transform into SIA components. At scale covered in this study, the Northern Hemisphere, we found that the differences in the patterns for BC and PM2,5 including SIA was so small, that the climate change impacts on the life time of the primary species forming SIA was insignificant. Therefore, the discussion of this was not included in the paper. The chemistry involving climate change impacts on OH concentrations is given in detail in:

Hedegaard, G. B., J. H. Christensen, C. Geels, A. Gross, K. M. Hansen W. May, A. Zare, and J. Brandt, 2012: "Modelling the Impacts of Climate Change on Tropospheric Ozone over three Centuries". Atmospheric and Climate Sciences, 2012, Vol. 2, No. 4, pp. 546-561.

We have included the discussion above in the results for PM2.5, including the reference.

Reviewer: 7) In section 4.3, there should be more discussion on the scientific reasons for the signals and impacts shown in figures 7 and 8.

Answer: The main interesting feature of the total N is the change in deposition, which is dominated by changes in wet deposition. The impacts from climate change on the

chemistry involving nitrogen species is diminished in this study, since we are assessing the total N. The chemistry only changes the concentrations from some N-species to other N-species and therefore the total N is constant including the effects from chemistry alone. The main process is therefore the atmospheric transport of the N species and the atmospheric life time. One important process can have influence on the life time of the different chemical nitrogen species in the atmosphere, and that is the dry deposition process. The dry deposition velocity is higher for e.g. for the gaseous compound HNO3 than for the nitrate particle, which is formed from HNO3. The same is true for e.g. NH3 and NH4. A detailed study of the climate change impacts on the nitrogen chemistry and change in the removal rate by dry deposition depending on changes in the chemical balance is a full paper in itself and cannot be included in this study. We have included the discussion above in section 4.3.

Reviewer: 8) Please give in the manuscript the definition of NOy (=NOx + HONO + PAN + HNO3 organic nitrates + other N-species??) and NHx?

Answer: We have done that.

Reviewer: 9) Conclusions should be shortened because they repeat what is written in the previous sections.

Answer: We have gone through the section and tried to concise it. Our purpose with the section is, however, to draw up the main conclusions from the results and discussion section. We think that it is important in this paper to summarize the main findings to help the reader to acquire the final overview of the main results and as such, we believe that a section summarizing the results and making overall conclusions of little more than 2 pages is not too bad.

Reviewer: 10) It is clear that in figures 3,4, 6 and 7 the relative impacts from (a) climate change, (b) emission change and (c) total change, (d) the climate signal relative to emission signal are shown. No need to repeat it many times in the manuscript.

Answer: We have gone through the manuscript and removed the repetitions.

Interactive comment on Atmos. Chem. Phys. Discuss., 12, 24501, 2012.