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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Impacts of climate change on air pollution levels in the Northern Hemisphere with special focus on Europe and the Arctic" *by* G. B. Hedegaard et al.

G. B. Hedegaard et al.

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First of all, we would like to thank reviewer number one for a very thorough review of our paper, with many relevant comments. We reply to each in turn in the following:

Reviewer: Additional descriptions on emissions used in the model system will better aid the interpretation of the results. In Section 2.2 (pg 1766 line 10-22) emissions are compiled from GEIA, EDGAR and EMEP datasets. Since these are annual emissions, do they differ by year/month throughout the decadal runs? It is obvious that SO2 emission varied throughout the 1990-1999 episode, what about the SO2 emission variations in the future decades?

Answer: The annual emissions from EMEP are provided for every year (see

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www.emep.int).The emissions from GEIA and EDGAR are only provided for the years 1990, 1995 and 2000. As written in the paper, a seasonal, weekly and diurnal variation is prescribed in the model for all the anthropogenic emitted species. For all the results for the future decades the 1990 anthropogenic emissions are used, including the prescribed seasonal and diurnal variation. In the current study only the climate change impacts on the air pollution levels are investigated for the scenario runs. The next step will be to include the effects of changes in anthropogenic emissions; however this was beyond the scope of this paper.

Reviewer: How does the emission variation differ from the sulfur model used in the ECHAM4-OPYC3 model? What about emissions of other species?

Answer: In this paper the emission variation (including SO2) in the climate model and the chemistry-transport models are independent. In the ECHAM4-OPYC3 model, a realistic estimate of the future sulphur concentrations is necessary in order to make a realistic simulation of the future climate change. For the DEHM simulation is was necessary to keep the emissions constant in order to distinguish the climate signal from the emission change signal. Tropospheric ozone is included in ECHAM4, and is allowed to vary as a result of prescribed concentrations of anthropogenic precursor gases (CH4, NOx and CO). In the stratospheric ozone and NOx concentrations are given for the years 1860, 1985 and 2050. Intermediate values are then calculated by linear interpolation and from 2050 and forth the concentrations kept constant at the 2050 level.

Reviewer: "Variable emissions" is mentioned in Section 4.3 (line 15). Is this variability in anthropogenic emissions, or only in biogenic emissions?

Answer: The variable emissions are applied only for the validation of the model system. In this case the variable emissions refer to both the anthropogenic emissions and the biogenic emissions?

Reviewer: Similarly, more descriptions on the biogenic emissions are necessary

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since they vary significantly between the current and future scenarios: what species are being estimated with the BEIS model? How does the emission change with global/regional forcing between scenarios? Does the model account for changes in vegetation/CO2 from the influences of global forcing?

Answer: The only specie estimated by using the current version of the BEIS model is isoprene. This has now been added to the paper in section 2.3. The change between the current and future biogenic isoprene emission is only due to the change in temperature. The present version of the model does not account for changes in vegetation.

Reviewer: If biogenic isoprene is the only specie that changed between scenarios, this should be mentioned in Sec 2.2 with summary of magnitude changes.

Answer: We have now included a sentence in the introduction describing that biogenic emitted isoprene is the only specie that is allowed to vary during the experiments.

Reviewer: Section 3.1 last paragraph: What is the fifth 10-year long simulation? (1) ECMWF-MM5 DEHM 1990-1999 (2) ECHAM4-OPYC3 DEHM 1990-1999 (3) ECHAM4-OPYC3 DEHM 2040-2049 (4) ECHAM4-OPYC3 DEHM 2090-2099

Answer: There are five ten-year simulations, since the 1990-1999 period has been simulated twice with ECHAM4 meteorology – one with variable anthropogenic emissions for the validation case and one with constant 1990 emissions used as the reference for the future scenario simulations.

Reviewer: In addition to temporal comparisons with spatially averaged data (average across all sites), how does the model system perform spatially? Can the (ECHAM4-OPYC3 DEHM) system capture the urban/rural concentration variability similar to that driven by a forecast system (MM5 DEHM)?

Answer: The DEHM model has previously been run on realistic emissions and meteorology for a 20 year period and compared to measurements from e.g. the EMEP network. The model performs well both with respect to temporal as well as spatial

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distribution. The whole validation material is huge and it is impossible to include more validation plots in this paper which has focus on impacts from climate change. The model is, however, able to capture the urban/rural concentration variability similar to that driven by a forecast system (MM5-DEHM). However, due to the uncertain nature of such scenario studies where we in the best case are only guessing about the future directions in meteorological and air pollution conditions, we choose to run the model on a relatively coarse resolution (150 km x 150 km). For other investigations related to the urban/rural gradients, we use a two-way nested version of the model with a resolution of 16.67 km x 16.67 km.

Reviewer: Pg 1774: What is the likely cause for over predicting SO4/SO4 WD? The over prediction is more obvious in MM5 driven DEHM and there is a clear seasonal trend in the positive bias.

Answer: The dry deposition module used in the model has a tendency to under-predict the dry deposition, which can explain the higher levels of SO2, SO4 and wet deposition SO4. Since these calculations were carried out, the dry deposition module has been improved and this under-prediction reduced. The difference between the two simulations must originate from the difference in meteorology.

Reviewer: The unit label for SO4 in Figure 2 and 3 are [mgN/m2] and not [mgS/m2]?

Answer: The unit label is actually totally wrong. The unit should be [ppb] and not [mgN/m2], since it is the SO4 concentration in air that is displayed.

Reviewer: There are repeated descriptions of the statistical methods. Section 4 and Sections 5.1 and 5.2.

Answer: The repeated description of the statistical methods has been removed in section 5.2.

Reviewer: Table 1 and Table 3 are not "color coded" as mentioned in the manuscript and the table captions.

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Answer: The text about color coding has been replaced with an explanation of the results.

Reviewer: Table 2 and Table 3 the entry for "O3"; is repeated. What is the difference between "O3", "O3 H", and "O3 DM"?

Answer: The double entry for O3 has been removed in the tables. Explanations for the difference between "O3", "O3 H" and "O3 DM" has been inserted in the table text O3 is diurnal mean values of O3, O3 H is the hourly mean values and O3 DM is the diurnal maximum values of O3.

Reviewer: If I understand correctly, in Section 5.2 and 5.3 the t-tests are carried out with annual averaged concentrations (10 samples and 18 degrees of freedom). Since most species have large seasonal concentration variations, how would the conclusions differ with monthly/seasonal averaged concentration comparisons?

Answer: When making statistical tests it is very important that the values in the time series are independent - meaning that you should not be able by physical arguments to identify where a number is located in a time series. This would not be the case if a monthly or seasonal variation is present in the time series.

Reviewer: It is not entirely clear how FB in Table 2 and Table 3 are calculated. In Table 2, is a positive FB means MM5 driven DEHM is overestimated compared to the ECHAM4-OPYC3 driven DEHM?

Answer: The reviewer is correct that it is not explained how the FB was calculated in the tables. This information has now been added to the table text. In Table 2, a positive FB does actually mean that MM5 driven DEHM is overestimated compared to the ECHAM4-OPYC3 driven DEHM?

Reviewer: Following the analyses in Section 5.3 Pg 1780, one can see consistent reductions in NH3/NH4 WD and increases of SO2 from 1990 to 2040 and 2090. If the emissions are constant for the three scenarios, what cause these species to vary?

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Answer: The consistent reductions in NH3/NH4 WD should be seen together with the increase in NH4 concentrations. There are especially two processes which can explain this: 1) the typical life time of NH3 and reactions into NH4 is decreased due to increased OH in the atmosphere in these simulations, and 2) the wet deposition of NH4 is decreased due to changes in precipitation patterns.

Reviewer: Does the DEHM model have inorganic thermodynamic equilibrium algorithm to account for the inorganic chemistry of the sulfate-ammonium-nitrate system?

Answer: Yes

Reviewer: – Pg 1781 line 1: The conclusion: "Generally the concentration of particles and the wet depositions are predicted very well with respect to their mean values." is slightly overstated. Only results for sulfate PM were presented and discussed in Section 5. The t-test results in Table 2 showed significant differences in the annual mean concentrations for nitrate and ammonium between the two model systems.

Answer: The reviewer is correct that the statement should only concern the results shown in the paper namely SO4 and not particles in general. The comment about the wet deposition results has been removed.

Reviewer: Section 6.2.2: The changes of SO4 WD (Fig 7) are insignificant everywhere, however in the t-test (Table 3) there are significant reductions in SO4 WD between the current and future decades. What cause the inconsistent results? The reverse is seen for ozone, where Fig 10 showed significant changes in most areas but Table 3 showed no significant changes for O3, O3H and O3 DM between current and future scenarios.

Answer: The difference between the results in table 3 and the figures 4-14 is the way the time series are made. In table 3, a time series has been constructed as a spatial mean over the locations of the EMEP measurement stations. In this way the spatial variability has been - averaged out - over Europe. In the figures 4-14, the results are displayed for every grid point - meaning no spatial averaging over measurement

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stations. As statistical testing is very sensitive to the way one constructs the time series, this explains the different results.

Reviewer: Section 6.2.4: Besides the Caribbean, portions of Alaska also have higher ozone and NO2. There are evidences of ship emissions causing higher NO2 in the future (compare Fig 7 with Fig 11). If emissions for the scenarios are the same, what caused the increases in SO2/NO2 on these ship routes given the higher OH/O3 in the future?

Answer: The anthropogenic emissions from the scenarios are the same. Therefore, the only other explanations for increases in NO2 are changes in chemical production, life times and reaction rates. To find the precise explanations requires a lot more sensitivity studies which have been beyond the scope of this paper.

Reviewer: Section 6.2.5: "Since the model do not posses any memory of this specie [OH], but is only estimated via a production term and a loss term." - OH is known to form and deplete via many different chemical pathways (eg R1). Does the statement mean that the model does not assume a background concentration of OH? Or does it mean that because of the fast reactivity/short lifetime, the model does not account for the transport of OH?

Answer: The statement means that the model does not assume a background concentration of OH?

Reviewer: Section 6.2.5: What are the likely causes of different OH spatial distribution with elevation? Besides Greenland, what caused the large gradient differences between land and ocean?

Answer: The large gradient difference between land and ocean is due to the increase in biogenic isoprene emissions and concentration over land that acts as a sink for OH. This isoprene emission is a surface emission and therefore the largest influence is found in the lowest layers of the model and over the land areas.

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Interactive Comment

Reviewer: Section 6.3 pg 1791: The SO4 reductions in Norilsk surrounding areas correspond better to reductions in SO2 (Fig 2) than to increases in SO4 wet deposition emphasized in the manuscript (Fig 9). The significant increases in SO4 in the northeastern portion of North America also correspond with increases in SO2 than to changes in SO4 wet deposition.

Answer: The SO4 reductions within the Norilsk area cannot be compared to the results in Fig 2, representing European monitoring stations. In Fig 2, only the variations during the 1990ties are displayed in contrast to the results around Norilsk where the future concentration changes are evaluated. The increase in SO4 in the northeastern portion of North America probably is due to a shorter lifetime of SO2 because of increased OH concentrations (away from the surface layer).

Reviewer: - Section 6.3 pg 1791: The authors argued that increase in sulfate (SO4) over Norilsk is due to increases in OH which caused more oxidation of SO2 - (thus decrease SO2 lifetime and increase SO4 concentrations). This does not seem to be supported by the predicted OH changes for the region. In Fig 12 (bottom right subplot) there is no significant difference in the future OH concentrations for the areas representing Norilsk (the red hotspot in the center of Fig 8 bottom right subplot).

Answer: Fig 12 displays the surface concentrations of OH. Near the surface over land the isoprene emissions will act as a sink for OH. This is why we have also plotted the OH concentrations higher up in the atmosphere (layer 5), where an increase are seen. This increase is due to an increase in ozone concentrations, which again is due to the increase in biogenic isoprene emissions as well as an increase in H2O. In general the OH concentrations increases in the atmosphere and this will influence the life time.

Interactive comment on Atmos. Chem. Phys. Discuss., 8, 1757, 2008.

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