We would like to thank the reviewers for the careful reading and the pertinent comments that helped improving this manuscript.

Here-below we provide a point-by-point reply to the comments by the reviewers. Text that corresponds to new text in the manuscript is provided in*"quotes and italics"*. At the end of document, a track changes version of the manuscript and of the supplementary material are added.

Reviewer # 1

Comment: A first remark concerns the main message of the paper that I tried to summarise above. Both the abstract and the conclusion sections remain rather vague about what has been exactly done, using rather loose words like "previously thought". I think the main methodology should be clarified in the abstract and conclusion section.

Answer: Both the abstract and the conclusions sections of the manuscript have been rephrased in order to clearly state the methodology and scope of the manuscript.

In the abstract Page 1, line 7: *"...limiting the rapid increase of air pollutants than what is traditionally deduced through comparison of present-day atmospheric composition to that calculated using anthropogenic emissions representative of the pre-industrial period, due to..."*

In the conclusions, Page 11, line 6: *"...significant technology development than that calculated by comparing against constant anthropogenic emissions as is traditionally done."*

Comment: A second remark concerns the results presented in figure 2. One could argue that the selection of stations seems rather arbitrary, and might be driven by the good performance at these stations. However, the appendix shows some extra material.

Answer: The selection of the stations shown in the manuscript and the supplementary material was made based on multi-year data availability and neglecting urban core stations that cannot be represented by our coarse model resolution. In addition, the number of stations shown is limited in order to keep the manuscript and supplementary material in reasonable lengths. However, the performance of the model has been evaluated based on comparison of all available measurements to the model results using the above-mentioned criteria and the derived overall statistics are provided in Table 1, as explained in the second paragraph of section 3.2.

Comment: What I find surprising is that the authors do not spend much time to highlight the clear imprint of emission reductions in the modeled atmospheric concentrations. Both the AE1980 and BA1980 scenarios are clearly not compatible with the observed atmospheric composition in later years. Although this sounds rather trivial I think this should be mentioned more clearly in the paper.

Answer: Indeed, the difference between the scenarios is clearly seen at some of the stations, especially those that are affected the most by anthropogenic sources. This indicates that the scenarios BA1980 and AE1980 that do not account for any legislation applied after 1980 fail to reproduce the observed levels of pollutants. As suggested by the reviewer

this is now discussed in the manuscript at the end of section 3.2, although the effectiveness of applied legislation is analysed further in the document and depicted in Figure 4.

"It is worth mentioning the clear imprint of the effect of emission legislation to the pollutant levels at the stations that are mostly influenced by anthropogenic activities and are depicted in Fig. 2 and supplementary Figures S5-S10. At these stations both the BA1980 and AE1980 simulations clearly fail to capture the pollutant levels observed the recent years."

Comment: I am also not a fan of the equations (with E-formatted numbers) in the plots. I suggest to summarise these in an extra table.

Answer: As for the equations that are printed in Fig.2, we wish to keep them as they are in order to make the direct link between each panel and the corresponding statistics. A separate table containing the equations would add unnecessary complication to the ease of reading and understanding the figure.

Comment: A third and final remark is about a worry I have with the resolution dependence presented in figure 4. CO and to a lesser extend NOX burdens are substantially larger in the 3x2 simulation. We talk here about differences up to 10%! The authors should at least analyse the cause of these differences. Lower global OH in the high resolution simulation seems a likely explanation (maybe driven by lower O3?).

Answer: This points to the spin-up time used for the 30 year simulations, which affects the way we have normalized the simulations to depict trends in Fig. 4. For this study we have applied one year spin-up time using the emissions and meteorology of the year 1980, i.e. by running twice the year 1980. Thus, all emission scenarios simulations started with the same conditions in 1980. In the fine resolution runs, the first years of the simulations have not reached the dynamic equilibrium needed for studying trends. Therefore, we have redrawn Fig. 4 by normalizing concentrations using those of the year 1982 instead of the year 1980 as was done before. This updated figure shows that the differences in the CO and NOx trends mentioned by the reviewer were due to this spin-up issue and the simulations CL and CL-fine produce very similar year-to-year changes.

Therefore, we have updated Fig. 4 in the revised manuscript by normalizing to 1982, i.e considering the extra two years as a part of the spin-up time for the simulations. This is also clarified in the figure caption. The new Fig.4 does not require any change in the discussion of section 3.3.

Appropriate discussion has been added in the manuscript at the end of section 2.1, where we now provide information on the spin-up time of the simulations.

"For this study, one year spin-up time using the emissions and meteorology of the year 1980, i.e. by running twice the year 1980, has been applied. The fine resolution simulation had not reached dynamic equilibrium after one year, as needed for studying the year-to-year changes. Therefore, the year 1982 has been used as reference year to normalize the concentrations in Fig.4 in order to study relative changes in section 3.4."

Comment: For other technical corrections, please see the attached pdf.

Answer: All suggestions of linguistic corrections marked by the reviewer in the pdf, have been addressed in the revised manuscript.

Reviewer # 2

Comment: Review of the paper "The success of emissions control legislation in mitigating air pollution is higher than previously estimated" by Daskalakis et al., acpd, 2016

The paper by Daskalakis et al. (2016) deals with the assessment of emission control measures in improving global air quality through the development of hindcast simulations over the past 30 years. The authors performed simulations using 3 sets of anthropogenic emissions, corresponding to different scenarios:

1) CL: current legislation data based on the ACCMIP database till the year 2000 and then using projections till 2010

Answer: The CL scenario uses the ACCMIP dataset as described in Lamarque et al., 2013. We did not produce a new dataset for performing the simulations.

Indeed, the available on the web ACCMIP data used here as is and contain historical data till 2000 and projections afterwards.

These are monthly global emissions, that are constructed based on the RCP6.0 emissions among others and provide (monthly) data from 1850 to 2100 (Lamarque et al., 2013). Any relevant comparison of these emissions has been performed by the groups that developed them, and is outside the scope of the present manuscript.

To clarify this point, we have modified the text in the manuscript in page 5 lines 10 and onwards, which now reads:

"Monthly anthropogenic and biomass burning emissions for the hindcast current legislation(CL) simulation are from the Atmospheric Chemistry and Climate Model Intercomparison Project (ACCMIP) database (Lamarque et al., 2013) until the year 2000 and RCP6.0 (van Vuuren et al.,2011, Fujino et al., 2006) projections afterwards, also provided by ACCMIP (http://accmip-emis.iek.fz-juelich.de/data/accmip/gridded_netcdf/accmip_interpolated/README.accmip_interpolated.txt). "

Comment: 2) AE1980: anthropogenic emissions of 1980 are assumed constant over the years

3) BA1980: business as 1980 accounts for constant per capita emissions per region. Emissions are then calculated following the observed population changes reported by the World Bank. No technological improvement, mitigation policy and change in per capita energy demand are taken into account.

The authors also show the comparison of their simulations with surface measurements of O3, CO, SO4 and BC obtaining a relatively good agreement.

General comments

Comment: Even though population growth is one of the drivers of emissions, the emissions are not directly scaled with population growth but with human activity. Human activity is more directly scaled with mainly global population growth (not as such with local population growth because of the globalization of industry and trade). Looking into the population growths in different parts of the world and comparing this with the emissions growth, one sees clearly huge differences for different regions. Let us have a

look at the population growth rate and the CO2 emissions growth rate (which excludes the effects of technology and end-of-pipe measures) for different parts of the world:

- Whereas China and the USA show similar population growth rates (of respectively 0.012 /yr and 0.011 /yr), CO2 emissions in China increased much stronger than in USA (0.099/yr and 0.051/yr respectively).

- Moreover China showed an acceleration in emissions increase (more than 10% in average) in 2002-2010, the period with flattening of the population growth rate (only 0.0047 /yr).

- Africa and the Middle East show a fast population growth rate (0.037 /yr) but modest emissions growth rate of 0.047/yr (similar to USA).

- EU showed only a very small population growth rate of in average 0.0030%/yr which is difficult to link with the emissions decrease rate (of -0.0035/yr in average). This exercise illustrates that a constant emissions per capita factor has different meanings for different regions and as such, it is unclear what the meaning of a BA1980 scenario with constant per capita emissions is.

Answer: The numbers and statistics presented by the reviewer (unfortunately without reference) are representative of the current legislation (CL) simulation and cannot be used for the BA1980.

Our hypothetical scenario BA1980 neglects (in purpose) the globalization of industry that happened the last decades, since it is constructed as a business-as-1980 scenario. BA1980 assumes that nothing has changed, other than the population itself (including regional information).

The scenario that we describe in section 2.2 specifically states that no energy demand, industrial growth or technological improvement is taken into account. This is a hypothetical scenario where, as the name states, all human activity, technological status and energy demand were the same as in 1980, but the population increased the way it actually did. The changes take into account the regional population changes, as described in the submitted manuscript in page 5, section 2.2, lines 23-29.

Overall, the scope of BA1980, explained in the manuscript in Section 2.1, page 5, lines 10-13 and Section 2.2 is to provide a picture of what the anthropogenic emissions would have been if per HTAP region the technology, the energy demand per capita and the way this energy was produced remained the same to those of the year 1980.

This was clearly stated in the manuscript. However to further emphasise this hypothesis, we have slightly modified the text in the first paragraph of section 2.2 which now reads:

"Advances in technologies are thus ignored and the energy demand per capita as well as the way energy was produced have been assumed constant with time and per region and equal to those of 1980."

Comment: A compensation with a factor 2 and 3 for China and India to account for increased energy demand seems also quite ad hoc.

Answer: As mentioned in our reply to the previous comment on the Business-As-1980 (BA1980) scenario, we calculated anthropogenic emissions per capita in 1980 per HTAP source region since there was already in 1980 a difference on the human activities for each

one of these source regions, as described by HTAP. These factors were then applied per region to the observed population growth.

The factors of 2 and 3 for India and China are not ad-hoc. They are calculated based on the energy use as equivalent of oil consumption provided by the World Bank (reference in the supplement, Table S2) for the years 2010 and 1980. This information (mean increase in energy use) is provided in Table S2b and explained in the Table caption and note in the supplement of this manuscript.

Comment: The reviewers wonder why the more elaborated retrospective emissions scenarios for 1970-2010 of the PEGASOS FP7 project (acknowledged in this work) are not used instead. Moreover the most important emissions trend, is the trend in China in the last decade 2000-2010, for which the reference case (ACCMIP) uses only a projection of emissions from RCP6.0, although the HTAP paper (cited in this work) provides monthly global emissions gridmaps for 2008 and 2010. At least a comparison of the reference case with more recent bottom-up inventories is needed.

Answer: The emissions of the PEGASOS project as stated in the Crippa et al. (2016) paper (page 3) analyse two different scenarios: "STAG_TECH, assumes after 1970 no further improvements in technologies and abatement measures. The second retrospective and lowest emission scenario (STAG_ENERGY) assumes stagnation of energy consumption since 1970, while the fuel mix, energy efficiency, emission factors and abatements are assumed as in the reference 2010 data".

None of those scenarios represents what we wanted to test, which would have been a combination of STAG_TECH (but global stagnation) and STAG_ENERGY but for 1980 fuels etc. and no increase in energy demand.

Furthermore, the emissions used here for the CL simulation have been developed and published by Lamarque et al. (2013).

The PEGASOS emissions have been constructed and are now published by Crippa et al. (2016), who state in their paper that "Historical global emissions data sets for the past decades or century have been compiled by combining several emission inventories, e.g., Lamarque et al. (2010) for 1850–2000 and Granier et al. (2011) for 1980–2010. However, an analysis of the factors driving these emissions trends is difficult because of the heterogeneity and regional differences of the original data that might show inconsistencies over the full time period and in global coverage and cause artificial variability."

So for the paper specifically:

Comment: 1)In chapter 2.2 (lines 18-20) the authors explain the data preparation for the BA1980 scenario. They say "The BA1980 inventory assumes that land anthropogenic emissions per capita remained constant from 1980 until now in major geographic regions, while population and thus overall human driven emissions changed".

The basic assumption of this scenario is that anthropogenic emissions scale with population. Although it is correct that emissions increase with population growth (e.g. due to higher energy demand, economy growth, etc.), the increasing economic activities are not always in the same place where the population grows. For example African countries have a growing population but the increase in economic activities is not happening there. On the other hand, Chinese population is stabilizing, while Chinese economy is exploding. These are just examples to highlight that population is not a driver for emissions in a

certain location. This is also true for industrialized countries, having a rather stable population but increasing energy consumptions and thus increasing emissions. Therefore the assumption behind the BA1980 scenario is not correct and the authors need to reconsider how to build this scenario. One option could be to focus on certain emission sectors, for example the road transport or the residential ones, where population can possibly be considered as a driver for the emissions (the authors will need to prove it anyway). For other sectors, like the power generation or the industrial ones, population growth in a certain location is not driving the increase of such activities in the same area. So, the population driven scenario is not applicable at all. Due to this major criticism, the authors should define a new methodology for developing hindcast simulations. Then all results should be modified accordingly and possibly the main message of the paper will change.

Answer: We understand the reviewer's reasoning about globalisation of industrial activities that moved a significant amount of industrial and energy demanding activities to regions with cheap labor. Thus the economic development did not follow the population growth.

This is the reason that in India the CL emissions in 2010 are higher than the BA1980 corresponding emissions as shown in the Figure 1 of our paper and discussed in section 3.1 (middle of first paragraph) of the ACPD version of the manuscript (page 6,line 10).

Based on our scenario the human activities in each region remain the same as in 1980- this means we do not account for globalisation of the industrial activities. This is afterwards indirectly accounted when discussing the increase in the energy demand based on Table S2b (see our earlier reply).

Of course, in another scenario, the emissions could increase with human activity and not human population. In our scenario though it is clearly stated (by the name of the scenario – Business-As-1980) that human activity does not change in any way, only number of humans per grid. This results to the population increase being the only driver for emissions increase, which we believe is the best reference scenario for one to use in order to study the impact of emissions control legislation at any region.

To further clarify this approach we have added in section 2.2. where the BA1980 scenario is described the following sentence:

"This also implies that globalisation of industrial activities leading to an increase in energy demand in developing countries disproportional to the population growth is not taken into account in this scenario."

Comment: 2)The authors should also explain the novelty of their work compared to recent literature about hindcast scenarios published within the same FP7 project they acknowledge. For example, Crippa et al. (2015) published a paper about retrospective scenarios assessing the effectiveness air quality legislations at global scale. Turnock et al. (2016) used a composition-climate model to simulate the impacts of European air quality legislation and technology measures implemented between 1970 and 2010. They used 2 scenarios, one with actual emissions in 2010 and the other with emissions that would have occurred in 2010 in the absence of technological improvements and abatement measures.

Answer: The Crippa et al. (2015) paper stagnates specific sectors of the anthropogenic emissions, where in our case all land anthropogenic sectors remain constant per capita to the levels of 1980, so a direct comparison with their results is not possible.

The Turnock et al. (2016) paper, that was published almost a month after our initial submission, discusses the impact of the European legislation only and is a regional study, while our study is a global one not focusing on one specific region and taking into account all legislation globally.

Comment: 3)The authors should compare their CL scenario with other emission datasets since this scenario is based not only on historical data but also on projections (from 2000 to 2010)

Answer: The CL scenario emissions are the well documented ACCMIP emissions and RCP 6.0 projections not developed by us. They are documented in Lamarque et al (2013) and analyzed in Granier et al (2010). These emissions have already been thoroughly analyzed and compared to other datasets by the people that produce them. This was also mentioned in an earlier reply.

Comment: 4)The authors state: "Emission factors per species per capita per year for each HTAP source region were calculated for the year 1980 by dividing the 1980 anthropogenic emissions by the population of each region" (page 5, lines 31-32). When dividing emissions by population you do not get emission factors, but population weighted emissions that is a completely different concept since emission factors are calculated from activity data.

Answer: We thank the reviewer for this comment and we correct the manuscript accordingly:

"Population weighted emissions per species per capita per year for each HTAP source region were calculated for the year 1980 by dividing the 1980 anthropogenic emissions by the population of each region. These per capita emissions were then applied on the gridded population maps to construct the database of annually-varying BA1980 anthropogenic emissions."

Comment: 5)The introduction deals mainly with O3 and just in the last few lines the authors introduce their work which seems not to be related with what discussed above. Moreover, the authors state that "Traditionally, air quality assessments are performed by comparing the pollutants concentrations at present with those of a past year". They should provide literature references about this approach since usually modelers make projections for future air quality then looking at past concentrations.

Answer: The introduction of the manuscript is used to state the problem of the atmospheric pollution increase of the near past that is a fact since it has been observed. It is true that more weight was given to O_3 in the submitted manuscript. For that reason we extended the aerosol discussion in the introduction, following also the suggestion by reviewer #3. Page 3, lines 16 and onwards now read:

"The modelling study by Pozzer et al. (2015) also shows globally decreasing AOD trends for the period 2001–2010. Regionally the largest decrease is calculated for easern USA and western Europe, where the eastern Chinese region shows the sharpest increasing trend. Similar results are found in the multi-satellite study by Yoon et al. (2014), where regionally western Europe and eastern USA appear to have the fastest decreasing trends in AOD, while central and east China the fastest increasing trends in AOD. In agreement with that study the analysis of the measurements of surface concentrations of several aerosol species by Leibensperger et al. (2012) shows decreasing trends in the eastern US for the period

1990–2010"

The technique modelers use, irrelevant of present or future, is to compare a time period against another. This is applicable to all aspects of modeling, including air quality. Frequently this involves studies of the future air quality, which is what the reviewer is referring to, and there are hundreds or thousands of studies about it. Also frequently though, studies involve the historical period, and how air quality is right now, compared to the past. There are also hundreds or thousands of studies about that as well. In this study we compare the method currently used to evaluate the already applied legislation to the present day conditions (comparison of what the levels of atmospheric pollutants would be if the emissions where quantitatively the same as those of a past year to the current levels) with a newly proposed method that, in our opinion, provides a better baseline scenario for comparison. Indeed, most future studies assume a business-as-usual scenario, which is a different way to say business-as-2000, if the year 2000 is the one considered as present day. Our BA1980 scenario is exactly that, for a historical analysis.

Comment: 6)it is not completely clear the scope of this paper. From the title the authors claim to have shown the success of emission control legislation in mitigating air pollution. However, most of their results focus on the quality assessment of their simulations through the comparison with measured concentrations (paragraphs 3.2 and 3.3). Only in paragraph 3.4 they want to address the legislation impacts on air quality, but they actually do not show the impact of any specific legislation, or the impact of certain abatement measures or technological advancement. Therefore the objective of the paper should be reconsidered.

Answer: The scope of this paper is to prove that in order to evaluate the effect of applied emission control legislation, the comparison of the pollutant levels of present years to pollutant levels based on a scenario that just keeps the anthropogenic emissions constant to the levels of a past year is not correct. Scenarios that take into account population increase and migration (shown here) and energy demand (not simulated but calculated and discussed here) should be used instead. This is already stated in the title, abstract and conclusions of the manuscript.

In order to extract any results by a model simulation, a thorough validation of the model results needs to be done. Otherwise, any results produced by said model would not be credible. This leads to the current structure of the paper, which demonstrates that the model calculates realistic concentrations of pollutants, and then addresses the key point of the paper.

Comment: 7)For a more detailed review new scenarios need to be developed and more in depth analyses will be required in order to assess the role of mitigation policies on emissions.

Answer: Even though lots of different scenarios can be produced giving different results, we believe that the scenario we chose serves the purpose of this paper sufficiently. As already stated above and in the paper, our study recommends which is the best reference scenario to use, it is not proposing mitigation policies or develops new future emissions scenarios.

Specific comments

Comment: 1) In Figure 2, the BA1980 and AE1980 scenarios are not needed since the

aim of this figure is to compare measured concentrations with the simulated ones (CL)

Answer: Figure 2 is also used to point the difference of the different scenarios in a more localized way than in Figure 4, thus the extra simulations (BA1980 and AE1980) are needed. The extra lines have been valorized following reviewer #1 comment 3.

Comment: 2) In figure 4 the authors should say to what they normalized the concentrations

Answer: The caption of the figure now reads:

"Annual mean surface concentrations for CO, NO_x, O_3, OC, BC, and sulphate aerosols (rows), averaged over the globe, Europe, North America, India and China (columns) and normalized to the 1982 concentrations."

Comment: 3) The authors state (page 2, line 7): "Most anthropogenic activity takes place in the northern mid-latitudes with approximately 80% of global population residing there (Kummu and Varis, 2011)". The authors should report the definition of northern-mid latitudes (degrees or countries included) and the year they are referring to, since population distribution might change in the next years (e.g. higher contribution from India?)

Answer: The sentence now reads:

"Most anthropogenic activity takes place in the Northern Hemisphere (N.H.) with 87.5% of the global population residing there, in particular 81.8% was occuring between the equator and 50° North in 2005 (Kummu and Varis, 2011)"

Reviewer # 3

General comments

Comment: The underlying premise to this paper is a good one: when we estimate the impact of emissions controls, it makes much more sense to compare to a projection of where we would be without them, rather than just assuming no emissions change as the baseline. It is important for policymakers to realise the true benefits that air pollution legislation has brought. Of course, the "true benefit" is always going to be a model construct, as we don't have two planets to monitor.

Answer: We thank the reviewer for the positive comment.

Comment: The choice of reference scenario is always going to be a bit contentious. What the authors have done here is probably reasonable (I am not an expert on constructing emissions scenarios) - however they describe the scenario they use as the reference as a "worst-case scenario" - which is patently wrong, as for some regions the emissions in the "current legislation" scenario exceed those in the worst-case (see specific comments below). The phrase "worst-case" should not be used.

Answer: Indeed the scenario described here is incorrectly named worst-case. It was used in two different parts of the manuscript.

The first (Page 4, line 4-6) now reads:

"The third 30-year simulation is performed with anthropogenic emissions for the period 1980-2010 that have been constructed, as further explained, in order to account for population increase but neither for the globalisation of the industrial activities, nor for the legislation applied after 1980."

The second (Page 6, lines 5-6) now reads:

"...As a result, a new anthropogenic emission inventory was constructed..."

Comment: The paper is well organised and reasonably clearly written – but the English should be improved – I make a few suggestions below, but this isn't really the job of a scientific reviewer. For example the first line of the Introduction(!): "The rapid Earth's population increase..." should be "The rapid increase in the Earth's population..."

Answer: Changes were made throughout the manuscript to improve the English, also following suggestions by reviewer 1.

Comment: If these things and the specific points below are rectified, then this paper should be acceptable for publication in ACP.

Specific comments

Comment: P1 L20 (and elsewhere) 80's → 80s (or possibly 1980s, or '80s, but definitely not 80's); also threat → threaten

Answer: We changed **80's** to **80s** throughout the manuscript.

Comment: P2 l13 Do you mean troposphere, rather than atmosphere? I believe the main thing driving the increase in tropospheric O3 burden between 1890 and 1990 is the increase in anthropogenic emissions. Lamarque et al (2005) find a decrease (not increase) in O3 lifetime of 30%.

Answer: We thank the reviewer for pointing out this typing error. The text now reads:

"This is linked to a decrease of O_3 *lifetime in the troposphere by about 30% (Lamarque et al.,2005)"*

Comment: P2 l28 Glacier?

Answer: Typo corrected.

Comment: P2 An obvious omitted reference here is Fiore et al (2012) (Global air quality and climate, Chem. Soc. Rev., 41, 6663, doi:10.1039/c2cs35095e).

Answer: The paper by Fiore et al (2012) focuses on future changes in global air quality and their impact on climate (period covered 2000-2030 and 2050). This is the main reason for not having a reference to that paper in the introduction. However to satisfy the reviewer, we now discuss the findings from the first 10 years of their simulations and the following comment is added in the second paragraph of the introduction:

"Fiore et al (2012) analysing observations and simulations suggested that recent air quality changes and their uncertainty are mainly associated with emissions changes although climate warming degradates air quality."

Comment: The Introduction has rather more material on ozone than other air pollutants– one could argue that since the paper's title contains the words "air pollution" – and the most important air pollutants (at least for human health) are aerosols, this mismatch should perhaps be addressed.

Answer: The following sentences are added in Page 3, line 16:

"The modelling study by Pozzer et al. (2015) also shows globally decreasing AOD trends for the period 2001–2010. Regionally the largest decrease is calculated for easern USA and western Europe, where the eastern Chinese region shows the sharpest increasing trend. Similar results are found in the multi-satellite study by Yoon et al. (2014), where regionally western Europe and eastern USA appear to have the fastest decreasing trends in AOD, while central and east China the fastest increasing trends in AOD. In agreement with that study the analysis of the measurements of surface concentrations of several aerosol species by Leibensperger et al. (2012) shows decreasing trends in the eastern US for the period 1990–2010"

Comment: P3 l24 Do you mean multi-annual, rather than interannual?

Answer: We changed the term interannual to transient to all occurrences in the manuscript where we refer to the simulations (not the calculated results).

The manuscript now reads:

P3 L24 *"For the present study, a set of three different transient global three-dimensional chemistry transport simulations..."*

P5 L4 *"Three global chemistry–transport transient simulations of atmospheric composition changes..."*

Comment: P4 l5 targets the simulation → simulates

Answer: Changed in the manuscript.

Comment: P4 l18 Maybe there is ice-core data for 1979-89, but I think perhaps you mean firn air data?

Answer: Indeed it is not scaled to ice-core data, but to flask data. The sentence now reads: *"For the years between 1979 and 1989, the* CH_4 *surface distribution of the year 1984 is scaled to fit the observed* CH_4 *data of the respective year."*

Comment: P5 l11 Define all acronyms at first usage (HTAP).

Answer: Definition added in text.

Comment: P5 l14 Fix double negative: "...does not account neither..." (also elsewhere)

Answer: Changed to *"...does account neither..."* in all occurrences in the manuscript.

Comment: P5 l18 Clarify what is done with shipping (and aircraft) emissions in these simulations (you refer to land anthropogenic emissions).

Answer: The last sentence of section 2 explains that shipping and aircraft emissions are from the ACCMIP database. We now further clarify in section 2.1 that these two datasets are used in all simulations. The sentence page 5 line 11 is rephrased as follows:

"All anthropogenic emissions with the exception of shipping and aircraft emissions were different between the simulations. Aircraft and shipping emissions were the same for all simulations. "

Comment: P5 l25 worldbank

Answer: Typo corrected.

Comment: P6 l3 The Business As 1980 (BA1980) case is described as a "worst-case" scenario, but this is misleading. For example, in some regions, for some species, the Current Legislation (CL) scenario actually shows larger increases between 1980 and 2010!! So it isn't really the worst case, is it?

Answer: Indeed the BA1980 scenario is not the worst-case in all cases. 'worst-case' was removed and the text was rephrased appropriately (see reply to the general comments).

Comment: P6 l31 2 AND 3, ... respectively (I guess)

Answer: Corrected.

Comment: P6 l33 What are the "emission ratios" referred to? Clarify.

Answer: The emission ratios mentioned here are those presented in Table S2. They indicate how different are CL and BA1980 scenarios (Table S2a) as well as the CL and assumed (not simulated) "worst-case" scenario where the energy demand is taken into account along with the population increase. The sentence has been rephrased as follows:

"However, when comparing CL to BA1980 emissions for India and China by examining the ratio of the emissions in CL to those in BA1980 (Table S2), most ratios are lower than the corresponding increase in energy use per capita."

Comment: P7 l2 Again, I am not sure you mean "interannual".

Answer: Word "interannual" was removed from the sentence. It now reads:

"The CL to BA1980 comparison indicates that some improvements in air quality have been achieved in these countries during the recent years although they can not be seen in the trends of the CL emissions over these regions that show emission increases"

Comment: P7 l16 spatial

Answer: The typo was corrected.

Comment: P8 l9-18 The discussion of simulated vs observed trends does not match up with what I glean from Figures 2 and S5-S10. For example, you say "O3 and CO trends are also [nicely] well simulated, both in direction and in magnitude in most stations". For CO, the ratio of the simulated CL trend to the observed trend at the seven stations shown is 2.3, 0.9, 0.02, 0.91, 0.45, 0.94 and 0.88. For O3, the ratios are: 0.6, 0.01, 0.41, 0.22, -0.46, 2.8, and 0.84. I think the trend magnitudes for O3 cannot really be described as "well simulated".

Answer: The text now reads:

"CO trends are also well simulated, both in direction and in magnitude for most stations. O_3 *trends are generally underestimated by the model. "*

Comment: P9 l4 Do you mean Figure 4 rather than Figure 3?

Answer: We thank the reviewer for noticing. The figure reference and number were corrected in the manuscript.

Comment: P9 l13 I think the climate impact on concentrations may be arising through factors other than simply changes in natural emissions brought about by variations in climate.

Answer: The sentence is changed to clarify that meteorology and climate driven natural emissions induce significant variability in air quality in these regions. It now reads:

"In Fig. 4, it is clearly seen that the climate impact on surface concentrations of O_3*, OC and BC results in increases in their levels by... China, indicating that changes in meteorology and climate-driven emissions induce significant variability in air quality in these regions."*

Comment: P10 l17 are modifying → have modified

Answer: corrected.

Comment: P10 l21 may or may not → do or do not

Answer: corrected.

Comment: P17 Table 1: define R

Answer: R is now defined in the table caption.

Comment: P18 Figure 1: I can't distinguish the different symbols for different regions.

Answer: Figure 1 was replotted with larger symbols.

Comment: Table S2b: As commented on above, some ratios (CL/worst-case) are >1, which indicates that "worst-case" must be an oxymoron.

Answer: Changed to CL/energy-demand which is more descriptive of the case analyzed.

The success of emissions control legislation in mitigating air pollution is higher than previously estimated.

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Abstract.

During the last 30 years significant effort has been made to improve air quality through legislation for emissions reduction. Global three–dimensional chemistry-transport simulations of atmospheric composition changes over the past three decades have been performed to assess the impact of these measures. The simulations are based on assimilated meteorology to account for the year–to–year observed climate variability and on different ~~anthropogenic emissions scenarios of pollutants~~scenarios of anthropogenic emissions of pollutants, which may or may not account for air quality legislation application. The ACCMIP ~~dataset historical emissions are~~ historical emissions dataset is used as the starting point. We show that air quality legislation has been more efficient ~~than thought~~ in limiting the rapid increase of air pollutants than what is traditionally deduced through comparison of present-day atmospheric composition to that calculated using anthropogenic emissions representative of the pre-industrial period, due to significant technology development. The achieved reductions in nitrogen oxides, carbon monoxide, black carbon and sulphate aerosols are found to be significant when comparing to simulations neglecting legislation for the protection of the environment. We also show the large tropospheric air-quality benefit from the development of cleaner technology. These 30–year hindcast simulations demonstrate that the actual benefit in air quality due to air pollution legislation and technological advances is higher than the gain calculated by a simple comparison against a constant anthropogenic emissions simulation, as is usually done. Our results also indicate that over China and India the beneficial technological advances for the air-quality have been masked by the explosive increase in local population and the disproportional increase in energy demand.

1 Introduction

The rapid increase in Earth's population ~~increase~~ that took place the last 60 years and the changes in human practices towards a society with larger energy consumption resulted in intensifying the atmospheric pollutant emission (Lamarque et al., 2013; Hand et al., 2012; Wang et al., 2014; Steffen et al., 2007). Air pollutants like ozone (O_3), carbon monoxide (CO), nitrogen oxides (NO_x), nitric acid (HNO_3) and particulate black carbon (BC), organic carbon (OC), sulphate (SO_4^{2-}) and nitrate (NO_3^-)

aerosol components have been observed to increase, reaching levels in the early ~~80's~~ 80s that threat ecosystems, leading for instance to agricultural efficiency decrease (Avnery et al., 2011) and human health inducing mortality increase (West et al., 2013), increase atmospheric acidity (Likens et al., 1996) and affect climate (West et al., 2013). To limit the negative impacts of air pollution, aggressive measures have been taken in the ~~80's~~ 80s (Lamarque et al., 2013) to reduce human-induced emissions. In parallel, the growing number and accuracy of air quality observations enabled monitoring of the air quality changes and statistical association of such changes with health issues and other environmental impacts.

Pollutant levels and seasonal variation are closely connected to emissions of the pollutant or its precursors, and meteorology. Tropospheric ozone and aerosols have largely increased since pre-industrial times as a result of intense anthropogenic activity (Volz and Kley, 1988; Staehelin et al., 2001). Fiore et al. (2012) analysing observations and simulations suggested that recent air quality changes and their uncertainty are mainly associated with emissions changes although climate warming degradates air quality. Most anthropogenic activity takes place in the ~~northern mid-latitudes with approximately 80% of~~ Northern Hemisphere (N.H.) with 87.5% of the global population residing there, in particular 81.8% was occuring between the equator and 50°North in 2005 (Kummu and Varis, 2011). Thus, the anthropogenically induced change in pollutant levels, hence in air quality, since pre-industrial times is expected to be larger in this area than in the southern hemisphere. Anthropogenic activity emitting NO_x into the atmosphere can influence the quantity of tropospheric ozone but because of ozone's non-linear dependency on the NO_x levels (Finlayson-Pitts and Pitts, 2000) the net effect on O_3 levels requires careful evaluation. Previous studies have shown that there ~~is~~ was a 30% increase in the tropospheric ozone burden, that corresponds to 71 Tg, between 1890 and 1990 (Lamarque et al., 2005). This is linked to ~~an increase of~~ a decrease in O_3 lifetime into the ~~atmosphere~~ troposphere by about 30% (Lamarque et al., 2005). Parrish et al. (2013) report a shift of 3–6 days in the seasonal cycle per decade for the mid–northern latitudes, where most of the anthropogenic activity takes place, based on long continuous data records from monitoring sites in Europe. They attributed this shift to changes in the relative contribution of the different tropospheric O_3 sources, from the stratosphere, changes in large scale circulation and changes in O_3 precursor emissions and subsequent photochemical production within the troposphere. The springtime maximum in ozone concentrations in the ~~Northern Hemisphere (~~N.H. ~~) reflects the combination~~ reflects the combined effect of increased free troposphere photochemical activity and stratospheric input (Vingarzan, 2004). In that work an increase of background ozone levels in the N.H. of 0.5–2% per year for the last 3 decades is reported, resulting to a mean concentration of 20–45 ppb for the N.H. for the year 2000. At Finokalia, Greece, Gerasopoulos et al. (2005) presented the changes in O_3 seasonality due to changes in meteorology and found a decreasing trend in ozone for the period 1997–2004 of about 1.64 ppb y^{-1} (3.1% y^{-1}). At the Mauna Loa observatory in Hawaii the 40–year timeseries of O_3 measurements shows that there is little change during spring but there is a rise during autumn, attributed to the weakening of the airflow from Eurasia in spring and strengthening in autumn (Lin et al., 2014). Oltmans et al. (2013) performed an extended analysis on long term (20–40 years) time series of global surface and ozonesonde observations. They found that the substantial tropospheric ozone increases observed in the early 1990s, the flattening, or even the decrease in ozone levels observed at several locations (e.g. ~~Galcier~~ Glacier NP, Minamitorishima) during the past 10 years are the results of the restrictions on precursor emissions. In the Southern Hemisphere subtropics a significant increase has also been observed (Oltmans et al., 2013). Similar findings were reported by Logan et al. (2012) by analysing measurements from sondes, aircraft and surface sites. These observations support

the fact that O_3 variability depends on the geographical location ~~(Chin, 2012)~~ (Oltmans et al., 2013) and the extent of regional anthropogenic influence (Vingarzan, 2004). The enhancements have ~~levelled–off~~ levelled off in the most recent decades, most probably due to O_3 precursors emissions control (Oltmans et al., 2013).

The global modelling study performed by Horowitz (2006) using the MOZART–2 model showed a 50% increase in the ozone burden calculated since pre-industrial times and a -6% to +43% change projected for 2100 using different emission scenarios. In agreement with that study, the Stevenson et al. (2006) analysis of multi model simulations of the future atmosphere has shown that, depending on the anthropogenic emissions, ~~and in particular~~ particularly of NO_x, the change in the tropospheric O_3 burden in 2030 compared to the 2000 levels can range between -5% (most optimistic scenario) and +15% (most pessimistic scenario).

Observations also show statistically significant trends in surface levels of atmospheric pollutants like CO (Yoon and Pozzer, 2014)~~,~~. CO surface levels were observed to increase before 1990s (Khalil and Rasmussen, 1988) and decrease in recent years, due to a decrease in CO anthropogenic emissions (Novelli et al., 1994). MOPITT, AIRS, TES and IASI satellite instruments have recorded a decreasing trend in the total column of CO of about -1% per year in the N.H. and less than that in the southern hemisphere from 2000 to 2011 (Worden et al., 2013).Similarly trends have been reported for the surface levels of sulphate (Hand et al., 2012) as well as the aerosol optical depth (AOD) that provides a measure of the interaction of aerosol atmospheric column content with radiation (Zhang and Reid, 2010; Karnieli et al., 2009). Global AOD over the ocean has been recorded by the Advanced Very High Resolution Radiometer (AVHRR) satellite instrument to increase from 1985 to 1990 and to decrease from 1994 to 2006 (Li et al., 2014). ~~surface levels were observed to increase before 1990s (Khalil and Rasmussen, 1988) and decrease in recent years, due to a decrease in anthropogenic emissions (Novelli et al., 1994). MOPITT, AIRS, TES and IASI satellite instruments have recorded a decreasing trend in the total column of of about -1% per year in the northern hemisphere and less than that in the southern hemisphere from 2000 to 2011 (Worden et al., 2013)~~The modelling study by Pozzer et al. (2015) also shows globally decreasing AOD trends for the period 2001–2010. Regionally the largest decrease is calculated for easern USA and western Europe, where the eastern Chinese region shows the sharpest increasing trend. Similar results are found in the multi-satellite study by Yoon et al. (2014), where regionally western Europe and eastern USA appear to have the fastest decreasing trends in AOD, while central and east China the fastest increasing trends in AOD. In agreement with that study the analysis of the measurements of surface concentrations of several aerosol species by Leibensperger et al. (2012) shows decreasing trends in the eastern US for the period 1990–2010.

In the troposphere all major air pollutants have sufficiently long lifetimes to be transported (Textor et al., 2006; Oltmans et al., 2013; Worden et al., 2013; Daskalakis et al., 2015). Thus, in addition to their precursor emissions and chemistry, atmospheric circulation is controlling air pollutant levels. Changes in transport patterns and spatial and temporal changes in anthropogenic emissions of O_3 precursors were suggested as the main causes of the observed shifts in seasonal maxima of O_3 in the northern hemisphere (Lin et al., 2014; Parrish et al., 2013) and the increase in wintertime levels of O_3 compared to earlier years (Parrish et al., 2013). Simulations suggest that the weakening of monsoon circulation in past decades contributed to the observed high aerosol levels in China (Chin, 2012), while a 6–year analysis of the Moderate Resolution Imaging Spectroradiometer

(MODIS) AOD over the Mediterranean attributed ~~to~~ observed AOD changes to anthropogenic emissions during summertime and to changes in precipitation during wintertime (Papadimas et al., 2008).

Traditionally, air quality assessments are performed by comparing the pollutants concentrations at present with those of a past year ~~. However,~~ (Lin et al., 2014), however, in order to evaluate the effectiveness of the applied ~~legislation in air quality~~ air quality legislation, we need to account for the changes induced by meteorology and the increase in anthropogenic emissions due to ~~increase~~ increases in population and energy demand.

For the present study, a set of three different ~~interannual~~ transient global three–dimensional chemistry transport simulations of atmospheric composition changes over the past three decades has been performed and analysed to assess the effectiveness of measures taken in the past years to limit air quality deterioration. The first simulation is performed using historical anthropogenic emissions and the second one using anthropogenic emissions as those of the year 1980. The third 30-year simulation ~~represents the worst case scenario and~~ is performed with anthropogenic emissions for the period 1980-2010 that have been constructed, as further explained, in order to account for population increase but ~~not for the applied legislation since~~ neither for the globalisation of the industrial activities, nor for the legislation applied after 1980.

2 Methodology – The global model setup

The model used for this work is the global 3D chemistry–transport model (CTM) TM4–ECPL (Kanakidou et al., 2012; Daskalakis et al., 2015) that takes into account gas and multiphase chemistry (Myriokefalitakis et al., 2011), gas–particle partitioning of semi volatile organics (Tsigaridis et al., 2006), and ~~computed~~ computes the gas–to–particle partitioning of inorganic components and the aerosol water using the ISORROPIA II aerosol thermodynamic model (Fountoukis and Nenes, 2007) in which the dust components are neglected for the present study. TM4–ECPL low horizontal resolution of 4°latitude $\times$6°~~latitude by~~ longitude and 34 hybrid vertical layers to 0.1 hPa was used here driven by ECMWF ERA–Interim meteorology from 1980 to 2010 (Dee et al., 2011).

TM4–ECPL ~~model targets the simulation of~~ simulates the tropospheric composition and chemistry. To be computationally efficient, the model has low vertical resolution in the stratosphere, and a very primitive representation of the full set of stratospheric chemistry. For this reason TM4–ECPL forces the O_3 concentrations in the top ~~3 model~~ layers (50hPa – 10hPa) based on the monthly mean observations by the Microwave Scanning Radiometer (MSR) satellite for the years 1980–2008 and Global Ozone Monitoring Experiment (GOME2) for the years 2009–2010. These data have been interpolated to the model layers by the Royal Netherlands Meteorological Institute (KNMI) (van der A et al., 2010). TM4–ECPL also calculates the stratospheric nitric acid concentration based on O_3 levels using a ratio derived from Upper Atmosphere Research Satellite (UARS) at 10 hPa. To account for CH_4 oxidation in the stratosphere, the CH_4 concentrations in the top eight layers of the model (roughly above 17 km height) are forced to the HALOE CH_4 climatology (Huijnen et al., 2010).

Since TM4–ECPL, like most CTM's, does not explicitly consider methane emissions, it forces the surface CH_4 distribution to observations using the latitudinal monthly varying surface levels of CH_4 calculated by Dentener et al. (2003) for the year 1984, which correspond to a global mean surface concentration of 1.69 ppm. This surface concentration changes depending

on the simulated year and following the measured increase of CH_4 in the atmosphere. For the years between 1979 and 1989, the CH_4 surface distribution of the year 1984 is scaled to fit the observed ~~ice core~~ CH_4 data of the respective year. For the years between 1990 and 2010 prescribed CH_4 surface concentration files based on National Oceanic and Atmospheric Administration (NOAA) observations are used (M. van Weele, personal communication, 2013).

The oceanic emissions of aerosols and isoprene are calculated by the model based on the 3–h varying meteorology and on monthly varying chlorophyll (Myriokefalitakis et al., 2011; Vignati et al., 2010). Other VOC emissions from the ocean have been taken into account in the model using the POET database (Granier et al., 2003) and have been kept constant from year to year (Myriokefalitakis et al., 2010). Interannually and daily varying dust emissions are from AeroCom (Aerosol comparisons between observations and models Dentener et al., 2006, extended by E. Vignati, personal communication, 2012) for years from 2000 to 2010 and have been also applied to earlier decades assuming the same interannual variability. Even though dust emissions are not representative for the first years of the simulation, none of the pollutants examined here ~~are~~ is significantly influenced by dust.Therefore, this model deficiency does not affect the present study. Monthly varying isoprene, terpenes and other biogenic volatile organic compounds for the years 1980–2010 are from the global emissions calculated by the Model of Emissions of Gases and Aerosols from Nature (MEGANv.1) (Sindelarova et al., 2014). Lightning NO_x emissions are calculated online (Meijer et al., 2001) and soil emissions are climatological emissions from the POET database (Granier et al., 2003).

Monthly anthropogenic and biomass burning emissions for the hindcast ~~historical~~ current legislation (CL) simulation are from the Atmospheric Chemistry and Climate Model Intercomparison Project (ACCMIP) database (Lamarque et al., 2013) until the year 2000 and RCP6.0 (van Vuuren et al., 2011; Fujino et al., 2006) projections afterwards, also provided by ACCMIP (http://accmip-emis.iek.fz-juelich.de/data/accmip/gridded_netcdf/accmip_interpolated/README.accmip_interpolated.txt). As anthropogenic emissions in this paper we consider the sectors included in the anthropogenic emissions of the ACCMIP database (Lamarque et al., 2013). International shipping and aircraft emissions are also from the ACCMIP database in all simulations.

2.1 Simulations performed

Three global chemistry–transport ~~interannual~~ transient simulations of atmospheric composition changes during the past 3 decades (1980–2010) were here performed using assimilated meteorology, natural emissions and biomass burning emissions specific of the simulated year. ~~Only the anthropogenic emissions of pollutants~~ All anthropogenic emissions with the exception of shipping and aircraft emissions were different between the simulations~~:~~. Aircraft and shipping emissions were the same for all simulations. The Current Legislation (CL) simulation uses the historical emissions of the ACCMIP database where legislation is applied to limit air pollution (Lamarque et al., 2013). The Anthropogenic Emissions 1980 (AE1980) simulation uses anthropogenic emissions that are constant throughout the years and equal to those of the year 1980. This corresponds to simulations that are typically used for comparison when evaluating the efficiency of emission control scenarios. The ~~Business As 1980~~ Business-As-1980 (BA1980) simulation accounts for constant anthropogenic emissions per capita and per HTAP ~~region~~(Hemispheric Transport of Air Pollution) region (supplementary Figure S1), as of the year 1980, resulting in anthropogenic emissions which follow the observed population changes (World Development Indicators, The World Bank; http://www.worldbank.org/). In this simulation the anthropogenic emissions neglect the air pollution legislation applied for

emission mitigation after 1980 and hence show increases due to population growth since 1980. ~~It does not~~ BAU1980 does account neither for the technological improvements achieved since 1980 nor for the per capita energy demand changes and thus no geographic shift in industrial activities due to globalisation of production (Table S2). Finally, an extra simulation was performed identical to the CL simulation but using the fine resolution of the model (3°lon × 2°lat) in order to investigate the effect of the model resolution on the results.

For this study, one year spin-up time using the emissions and meteorology of the year 1980, i.e. by running twice the year 1980, has been applied. The fine resolution simulation had not reached dynamic equilibrium after one year, as needed for studying the year-to-year changes. Therefore, the year 1982 has been used as reference year to normalize the concentrations in Fig.4 order to study relative changes in section 3.4.

2.2 Construction of the BA1980 anthropogenic emissions

The BA1980 inventory assumes that land anthropogenic emissions per capita remained constant from 1980 until now in major geographic regions, while population and thus overall human driven emissions changed. Advances in technologies ~~as well as increases in~~ are thus ignored and the energy demand per capita as well as the way energy was produced have been assumed ~~to have remained unchanged with time .~~ constant with time and per region and equal to those of 1980. To construct this anthropogenic emissions database the ACCMIP anthropogenic emissions of the year 1980 together with global population maps were used.

The global population for the year 1980 (not gridded) and global gridded population maps for the period 1990–2010, available at five–year increments were obtained from the United Nations (http://www.un.org/) and The World Bank (http://www.worldbank.org/). Using a fine resolution of 1°× 1°, linear interpolation between the 5–year steps was applied per grid to produce global gridded population density maps for each year for the period 1990–2010. The gridded population maps for the years 1980–1989 were subsequently constructed based on the gridded population distribution of the year 1990 and a backwards extrapolation of the year–to–year change of the total global population using a polynomial fit, thus assuming uniform population change in all regions.

Figure S1 provides a visual representation of the 16 regions considered in this study ~~which correspond~~ and corresponding to the HTAP source regions (Janssens-Maenhout et al., 2015). ~~Emission factors~~ Population weighted emissions per species per capita per year for each HTAP source region were calculated for the year 1980 by dividing the 1980 anthropogenic emissions by the 1980 population of each region. These ~~factors~~ per capita emissions were then applied on the gridded population maps to construct the database of annually–varying BA1980 anthropogenic emissions. Based on the population density, the anthropogenic land emissions of 1980 and the source regions as socio-economic regions, an emission inventory has been created that takes into account the per capita anthropogenic emissions of 1980, the population density increase per grid for the period 1980-2010 and the population relocation since 1990. As a result, a ~~worst-case~~ new anthropogenic emission inventory was constructed, which assumes non-mitigation for improvement of air quality after 1980 and does ~~not~~ account neither for technological developments since then nor for increases in energy demand per capita but accounts for population increase. Increase in energy demand per capita is almost negligible in Europe and N. America but is significant in the fast developing

countries, India and China (a factor of 2 and 3 respectively, Table S2b). Taking into account the energy demand would result in even higher emissions in higher emissions globally. This also implies that globalisation of industrial activities leading to an increase in energy demand in developing countries disproportional to the population growth is not taken into account in this scenario.

3 Results and discussion

3.1 Emission Trends

In Fig. 1, the annual mean anthropogenic emissions for the CL (historical changes) and the BA1980 emission scenarios for CO, NO_x, NH_3, OC, BC and SO_2 are presented, normalized to the respective emissions of the year 1980 (fluxes for the year 1980 are shown in the histograms in the same figure) for the considered regions: globe, Europe, N. America, China and India. During the entire 1980-2010 period the CL anthropogenic emissions of primary pollutants are lower than the BA1980 on the global scale and regionally over North America and Europe (Fig. 1 showing global and regional totals, and Fig. S2 and S3 showing gridded changes of the emissions). This demonstrates that effective emission controls and new technologies (CL emissions) have contributed to the reduction of air pollutants emissions, in a per-capita basis compared to those in 1980, resulting to overall reductions in anthropogenic emissions by 18% for BC to 44% for SO_x (sum of SO_2 and sulphate) globally and between 25% for NH_3 over N. America and 75% for SO_x over Europe (Table S2a) where most development of cleaner technologies was implemented. Energy demand per capita remained constant in the European Union while that over North America decreased by about 10% according to World Bank statistics (Table S2). On the other hand, regions which experience fast population and economic growth in the last three decades (such as China and India) are calculated to have higher anthropogenic emissions under the CL scenario than under the BA1980 scenario (by 60% and 55% for NO_x, respectively, and by 25% and 135% for SO_x, respectively, Table S2a). This means that their anthropogenic emissions per capita have increased in the last 3 decades, resulting in a "dirtier" case from what one might ~~had~~ have expected. It is obvious though that ~~efforts have been made to reduce pollution over India~~ the past 10 years over India, where anthropogenic emissions of CO, BC and OC show stability, or even decreasing trends, efforts have been made to reduce pollution,for instance use of renewable energy.

During the last 30 years the industrial sector in Asia has experienced an explosive growth, resulting in a disproportional increase compared to the local population growth. Globalization and cheap labour led a large fraction of the world's industrial production to occur over the greater India and China regions, in response to the global population and energy demand growth, not just the local one, which explains the increased per-capita emissions over these regions (CL compared to BA1980). Indeed, as reported by the World Bank, the energy demand per capita has increased by factors of about 2 ~~to~~ and 3 for India and China, respectively, derived as the ratio of the energy demand per capita in 2010 to that in 1980. However, when comparing CL to BA1980 emissions for India and China ~~most emission~~ by examining the ratio of the emissions in CL to those in BA1980 (Table S2), most ratios are lower than the corresponding increase in ~~the~~ energy use per capita. The CL to BA1980 comparison indicates that some improvements in air quality have been achieved in these countries during the recent years although they can not be seen in the ~~interannual~~ trends of the CL emissions over these regions that show emission increases. Emission mitigation

can be detected in the CL scenario by the 25% – 70% reduction of all major pollutant emissions over India and China, except SO_x over India (Fig. 1f), compared to regional emissions estimates that account for mean increase in energy demand (Table S2b). Ammonia emissions present a more complicated pattern (Fig. 1c), as a result of the absence of any legislation on NH_3 emissions (Lamarque et al., 2013) and high uncertainty on ammonia emissions from India (Sharma et al., 2008).

3.2 Comparison against surface measurements

TM4–ECPL simulations were performed with the different emissions scenarios. The accuracy of the model is evaluated by comparison with available observations around the globe (see locations in Fig. S4). Surface observations for O_3 and CO were obtained from the World Data Centre for Greenhouse Gases (WDCGG; http://ds.data.jma.go.jp/gmd/wdcgg/introduction.html). Surface observations of O_3, BC and SO_4^{2-} over Europe and the U.S.A. were obtained from the European Monitoring and Evaluation Programme (EMEP; http://www.emep.int) and the Interagency Monitoring of Protected Visual Environments (IMPROVE; http://vista.cira.colostate.edu/improve/) respectively. The OC measurements are from the AeroCom Phase II database (Tsigaridis et al., 2014). Monthly mean and standard deviation are calculated from the original datasets available at various temporal resolutions. These datasets provide adequate coverage over Europe and N. America. For the rest of the globe, the temporal and ~~spacial~~ spatial variability of the measurements is scattered, resulting in a model evaluation which is highly biased ~~by~~ towards N. America and Europe.

Surface concentrations and trends were calculated for each of the three scenarios to compare the computed atmospheric composition changes during the studied period. CL simulations have been compared to global observations of O_3, CO, SO_4^{2-}, BC and OC (Fig. 2, Fig. 3 and supplementary figures S5 to S10). These comparisons are performed on monthly mean basis for all stations and all years from 1980 to 2010 where data are available. Statistical analysis of the results was conducted per model grid. For this, monthly mean of the measurements were grouped per model grid (6°lon. $\times$ 4°lat. or 3°lon. $\times 2^\circ$lat., depending on model resolution) and they are then averaged to derive the annual mean concentration. Model results are sampled at the time and location of the observations, then annual means in each grid box are computed for model results and compared to those derived from observations. The statistics of these comparisons are presented in Table 1.

In the following we focus on the normalized mean bias (NMB) to evaluate the over/underestimation of the observations by the model as well as the correlation coefficient that provides the strength and direction of the relationship between the model results and the observed levels of air pollutants, while the coefficient of determination (R^2) provides the fraction of the observed air pollutant variance that is reproduced by the model. In Table 1 that summarizes the statistics of Fig. 3, it can be seen that at the studied locations where observations are available the model reproduces very well the mean observed surface levels of CO (NMB between -1% and 2% for fine and coarse model resolution, respectively) and of BC (NMB between 1% and -3% for fine and coarse model resolution, respectively). It also satisfactorily reproduces the surface O_3 levels (NMB between 17% and 13% for fine and coarse model resolution, respectively). For sulfate the NMB is of the order of 61% and 52% respectively, indicating a model overestimate of the observations, while for particulate OC, the negative NMB between -57% and -62% respectively indicates a model underestimate of the surface OC observations as also pointed out for most AeroCom models by Tsigaridis et al. (2014). The model performance is not as good for NH_4^+ aerosol which is overestimated by the model by

a factor of about 1.4 to 1.5. The model results show relatively weak correlations with O_3 and CO observations (0.4<R<0.5) and very good correlations (R>0.5) with the other air pollutants (Table 1). However, when focusing on the model's ability to reproduce the measured patterns as determined by the coefficient of determination (R^2), the model seems to capture nicely the observed variance of sulfates and OC with R^2>0.5, i.e. more than 50% of the observed variability is reproduced by the model. It also reproduces more than 40% of the observed NH_4^+ variability, 50% of that of BC , 25% of that of O_3 and about 20% of that of CO.

When further analyzing the model results to evaluate the success of applied legislation for improving air quality, we need to take into account the above outlined model's ability in reproducing observed air pollutant trends. For that, we used available monitoring stations with long timeseries of measurements (Fig. 2 and supplementary Figures S5-S10). For each of these stations the trends from the observations and from the corresponding values calculated by each simulation were derived. The model reproduces the sign of the observed trend for most species and locations. Specifically, SO_4^{2-} slope direction is always captured by the model, although the magnitude is not always well reproduced. ~~and~~ CO trends are also ~~nicely~~ well simulated, both in direction and in magnitude ~~in~~ for most stations. O_3 trends are generally underestimated by the model. OC, BC and NH_4^+ calculations appear to have the largest deviations from the trends derived ~~form~~ from observations, which likely reflects the difficulties both in measuring carbonaceous and ammonium aerosols and in simulating their sources and fate in the atmosphere, in particular the semi volatile character of a large fraction of organics and of ammonium nitrate. It is worth mentioning the clear imprint of the effect of emission legislation to the pollutant levels at the stations that are mostly influenced by anthropogenic activities and are depicted in Fig. 2 and supplementary Figures S5-S10. At these stations both the BA1980 and AE1980 simulations clearly fail to capture the pollutant levels observed the recent years.

3.3 Impact of the model resolution on the calculated results

To determine the impact of the model resolution to the calculated results, an extra 30–year long simulation was performed identical to the CL simulation but using a finer model resolution of 3°lon × 2°lat. The calculated normalized annual mean concentrations of this simulation are depicted by the yellow line in Fig. 4. Also, statistical analysis identical to the one of the CL simulation was performed. The results of this analysis are also provided in Table 1 and shown in Fig. S11.

The statistical analysis of the comparison of the CL-fine simulation against measured values shows a global performance very similar to that of the CL simulation, indicating that, for the studied period and pollutants, the model resolution has only minor impact on the results. The largest differences in performance between the fine and coarse grid CL simulations are found for O_3, where the calculated model mean value for the CL-fine simulation is about 4% higher than that of the CL simulation. Note that the differences in the model resolution ~~reflect also in~~ also lead to differences in the number of observational sites per grid and in the mean of the observed concentrations that are compared to the model results. These differences also reflect the inhomogeneity in the spatial coverage of the observations. When using finer resolution grids, the largest differences in the mean concentrations (Table 1), have been computed for the aerosol components and in particular for OC (2.92 $\mu g\,m^{-3}$ compared to 3.24 $\mu g\,m^{-3}$, i.e. 10% lower compared to the coarse resolution grid). These differences are in accordance with the changes in the normalized mean simulated concentrations that are shown in Fig. 4.

3.4 Air quality changes

Because the CL, BA1980 and AE1980 simulations have been performed using the same meteorological fields, natural and biomass burning emissions, and differ only in the anthropogenic emissions over land, the effectiveness of the applied mitigation policies can be evaluated by comparison of the computed annual mean surface concentrations of air pollutants (Fig. 2, ~~3~~ 4 and supplementary figures S5 to S10 and Table S3).

The results are analysed by examining the changes in the computed concentrations in 2010 compared to the concentrations in 1980 as well as the percent changes between the different simulations in 2010. Focus is put on developed areas where mitigation legislation was applied (North America and Europe) and contrasting these with India and China that experienced rapid growth during the last years. The AE1980 simulation shows a small variability (maximum 20%, usually about 5%) in the normalized mean concentrations of pollutants (Fig. 4) that can be attributed to the climate variability in meteorology and the natural and biomass burning emissions. In Fig. 4, it is clearly seen that the climate impact on surface concentrations of O_3, OC and BC results in increases in their levels by about 5%, 15% and 5% respectively, in 2010 compared to 1980 on the global scale and also over Europe, North America and China, indicating that ~~natural emissions are also significant there~~changes in meteorology and climate-driven emissions induce significant variability in air quality in these regions. The other pollutants in Fig. 4 show less sensitivity to meteorological and biomass burning changes.

Focusing on the CL simulation, small global decreases are computed for CO and SO_4^{2-} in 2010 (about -5% and -15%, respectively) since 1980, while for the other pollutants global increases are calculated (Fig. 4 left column). Regionally the picture is quite different, with significant reductions in primary pollutants over Europe and North America (except NO_x) and increases over China and India. For surface CO the environmental gain is substantial for Europe and for North America, with simulated reductions reaching -40% and -20% compared to the AE1980 levels (Fig. 4 green lines compared to blue lines), while the large increase in energy use in China resulted in about 20% increase in surface CO.

The present study shows that the effectiveness of air pollution abatement is larger than what is deduced by comparing CL to AE1980 (Table S3a), as is usually done since comparisons of the CL simulation with the BA1980 one (that takes into account the population changes but neglects any technological developments since 1980, Table S3b) reveal a higher gain in air quality. Globally the computed surface CO levels under the CL simulation are lower than the BA1980 ones by 22%. Regionally the reductions achieved in 2010 are even higher (Europe -69%, N. America -44%, China -19%, India -24%). This proves that technological advances of the past years have contributed to reduce the CO levels even in fast developing countries. These agree with changes in the total CO columns retrieved from satellite observations where reductions of the total column of CO are observed for all the studied regions for the period from 2000–2010 (Worden et al., 2013).

Another successful story is that of SO_4^{2-}. Compared to the BA1980, SO_4^{2-} levels computed by the CL simulation are lower by 54% globally, and more than a factor of 3 over Europe and almost a factor of 2 over N. America. In contrast, in the fast developing economies of China and India, computed SO_4^{2-} levels are higher by about 10%. However, if we account for the increase in energy demand per capita in these regions (by factors of 3 for China and 2 for India) that is not taken into consideration when constructing the BA1980 emissions database, then technological advances seem to have limited the air

pollution even in these regions (Table S2b). The computed increase in surface SO_4^{2-} levels over China until about 2007 followed by a stabilization and even a decrease in their levels, is in general agreement with the SO_2 column satellite observations (GOME/SCIAMACHY) that show increases between 1996 and 2007 and then a decrease in SO_2 tropospheric column over China (SCIAMACHY Product Handbook).

For OC and BC, the global gain computed is 10% and 22% respectively. Regionally, Europe shows the highest gain with a computed reduction of OC of 54% and BC of 66%. N. America also shows high computed air quality gains concerning OC and BC, with a 43% and 51% reduction respectively. For India a gain of 20% is computed for both pollutants, where for China the changes are not significant (less than 5% for both).

Ozone levels show the least sensitivity to the reduction policies of its precursors' emissions, with computed global/regional changes of less than 10%. These model results are also supported by the small zonal changes (between -0.2±0.4% y^{-1}, 0.3±0.4% y^{-1}) in tropospheric ozone column retrieved from SCIAMACHY observations between 2003 and 2011 (Ebojie et al., 2015). Surface NO_x concentrations show increases by 13% and 28% over India and China respectively in the CL compared to the BA1980 simulation while over Europe a reduction of 63% and over N. America of 29% is computed, where the global gain is 21%. These results are in general agreement with the satellite observations by Richter et al. (2005), according to which the tropospheric columns of NO_x are found to be rather stable or with a decreasing trend over Europe and the U.S.A., while they show an increasing trend over the Asian regions they studied during the period between 1996 and 2002.

4 Conclusions

Over the last century increasing population and technology development ~~are modifying~~ have modified the Earth's landscape, depleting resources and changing the atmospheric composition, thus, deteriorating the quality of life on Earth. During the last 30 years significant effort has been made to improve air quality through legislation for emissions reduction. The extent of success of the legislation applied to improve air quality has been here assessed based on three global three–dimensional chemistry transport simulations of atmospheric composition changes which were performed using different anthropogenic emissions of pollutants which ~~may or may~~ do or do not account for air quality legislation application. Otherwise identical, the simulations account for the year–to–year observed climate and natural emissions variability. Air quality legislation has been shown to be more efficient ~~than thought~~ in limiting the rapid increase of air pollutants due to significant technology development ~~. The~~ than that calculated by comparing against constant anthropogenic emissions as is traditionally done. The comparison of simulations between them and with observations show the clear imprint of the effect of anthropogenic emission legislation to the pollutant levels. The achieved reductions in nitrogen oxides (21%), carbon monoxide (22%), black carbon (22%) and sulphate aerosols (54%) are found to be significant when compared to simulations that neglect legislation for the protection of the environment and when taking into account the limits of the model to reproduce the variance of the observations and the observed trends as earlier discussed. We also show the large environmental benefit from the development of cleaner technology. Our 30–year hindcast simulations demonstrate that the actual benefit in air quality due to air pollution legislation and technological advances is higher than the gain calculated by a simple comparison against a constant anthropogenic emissions simulation~~, as is usually~~

done. Our results also indicate that over China and India the beneficial technological advances for the environment have been masked by the explosive increase in local population and the disproportional increase in energy demand.

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Table 1. Statistics of comparison of model (CL simulation) vs. observations and fine resolution simulation (CL-fine) vs. observations (corresponding to pollutant concentration scatter plots shown in Fig. 3 and Fig. S2). #pairs shows the number of pairs used for the comparison, meas and model are mean of all gridded data that are used for the comparisons (as described in section 3.2). R is the calculated correlation between the measured and the modelled data. NMB stands for normalized mean bias of the model against the measurements. C stands for the CL simulation ($6^\circ \times 4^\circ$) and F stands for the CL-fine simulation ($3^\circ \times 2^\circ$). The units are ppb for gases (O_3, CO) and μg m^{-3} for aerosols (SO_4^{2-}, OC, BC, NH_4^+).

	#pairs[&]		meas		model		R		NMB* (%)	
	C	**F**	**C**	**F**	**C**	**F**	**C**	**F**	**C**	**F**
O_3	1555	2417	30.86	31.08	34.89	36.32	0.46	0.48	13	17
CO	211	229	149.24	149.87	151.64	148.62	0.42	0.41	2	-1
SO_4^{2-}	2008	3592	2.57	2.51	3.91	4.04	0.74	0.71	52	61
OC	1037	1825	3.24	2.92	1.25	1.22	0.59	0.71	-62	-57
BC	931	1728	0.29	0.28	0.28	0.29	0.59	0.50	-3	1
NH_4^+	626	777	0.94	0.92	2.24	2.30	0.65	0.64	140	149

[&] The number of pairs corresponds to the number of gridboxes that contain measurements. Hence the larger number of pairs in the finer resolution of the model

*Normalized mean bias is calculated as: $NMB = \frac{\sum(M_i - O_i)}{\sum O}$ where M stands for Model and O for Observations

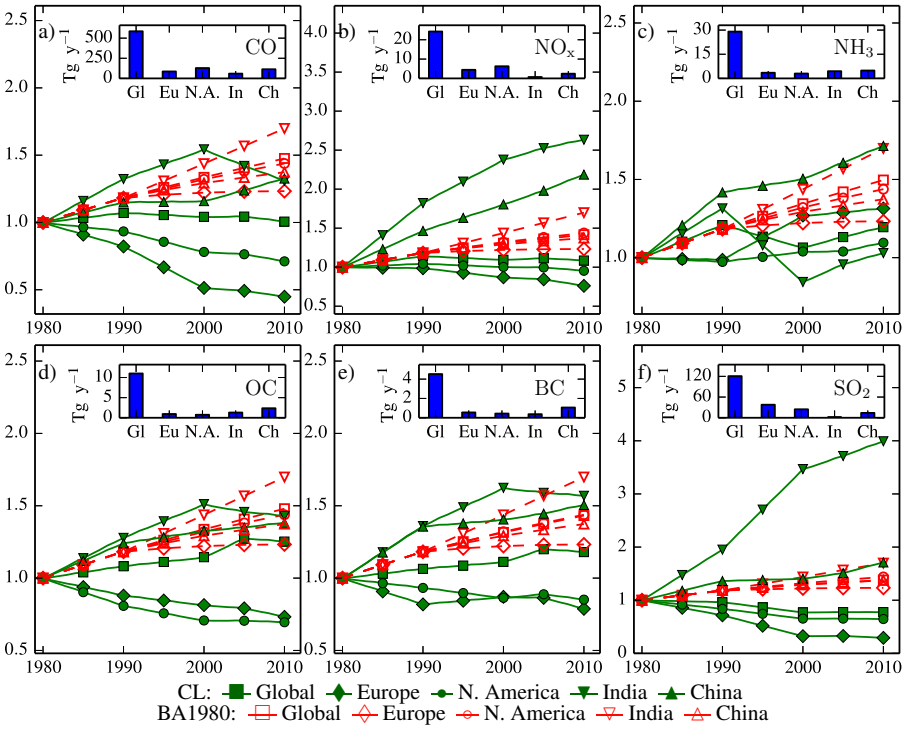

Figure 1. Normalized (to 1980 levels) annual mean anthropogenic emissions of CO, NO_x, NH_3, OC, BC and SO_2 for the CL (green/solid) and BA1980 (red/dashed) simulations as a function of time. Different regions appear with different symbols: squares for the globe (Gl), diamonds for Europe (EU), triangles for China (Ch), inverted triangles for India (In), and circles for North America (N.A.). The histograms at the top of each panel show the absolute emissions in 1980 (used to normalize the emissions) for these regions that are used as reference to normalize emissions (also shown in Table S1).

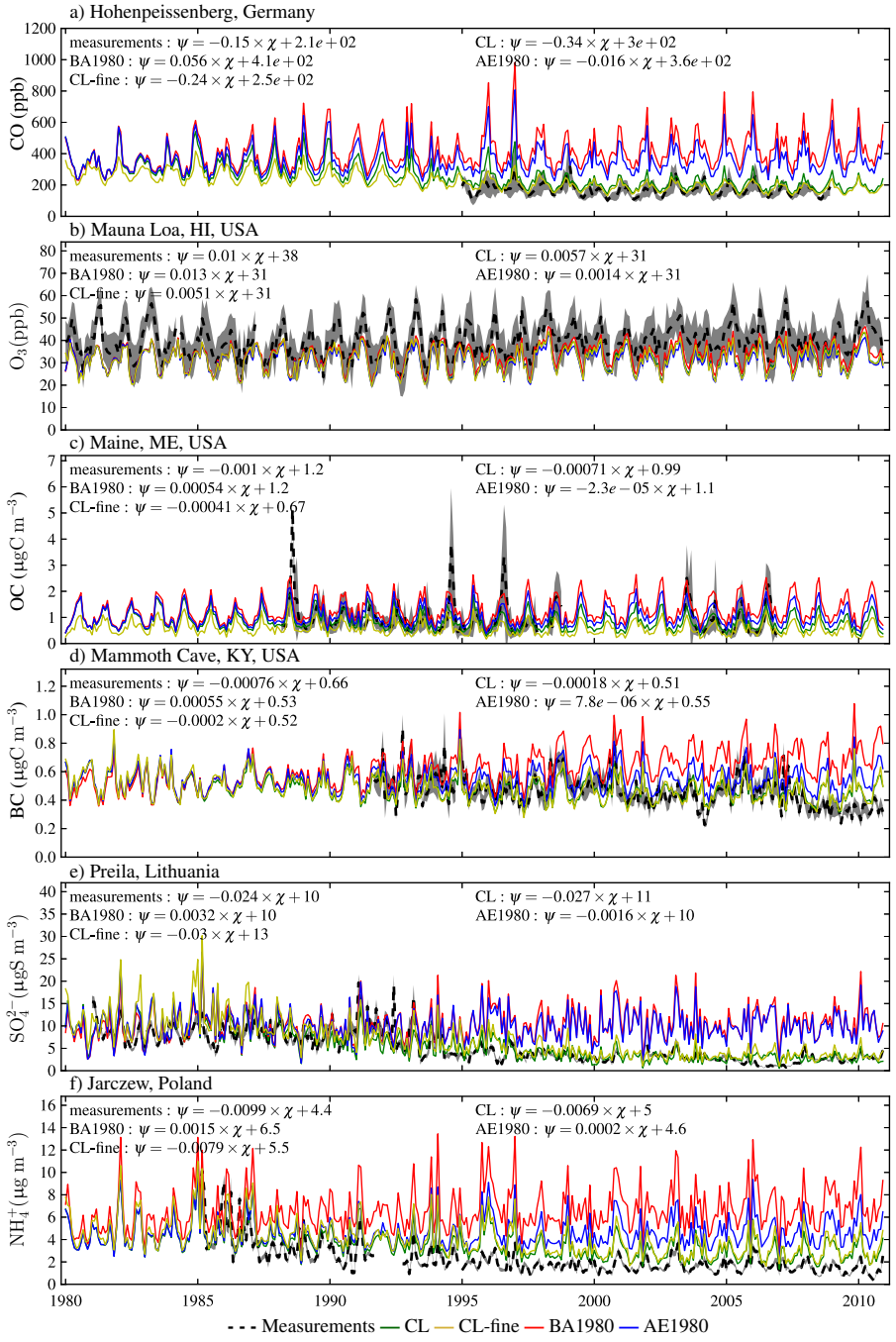

Figure 2. Comparison of the four simulations against observations. The dashed line and shadowed areas indicate monthly mean surface observations and one standard deviation. Simulations are CL: current legislation (green); CL-fine: current legislation in the fine resolution of the model; BA1980: Business As in 1980, with constant anthropogenic emission rates per capita as in 1980 (red); AE1980: constant anthropogenic emissions as in 1980 (blue). Trends derived from the concentrations (ψ) as a function of the year (χ) are provided for the measurements and the four simulations inside the frames.

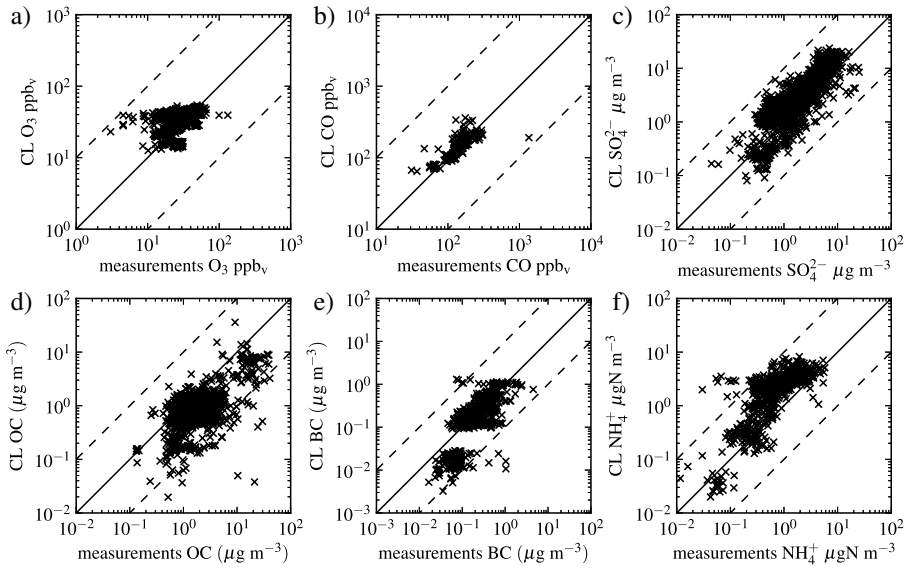

Figure 3. Comparisons of annual average surface model results versus observations per model grid(see Fig. S5 for station locations) a) for O_3, b) for CO, c) for SO_4^{2-}, d) for OC, e) for BC, f) NH_4^+. The continuous line denotes the 1:1 slope and the dashed lines the 10:1 and 1:10 slopes

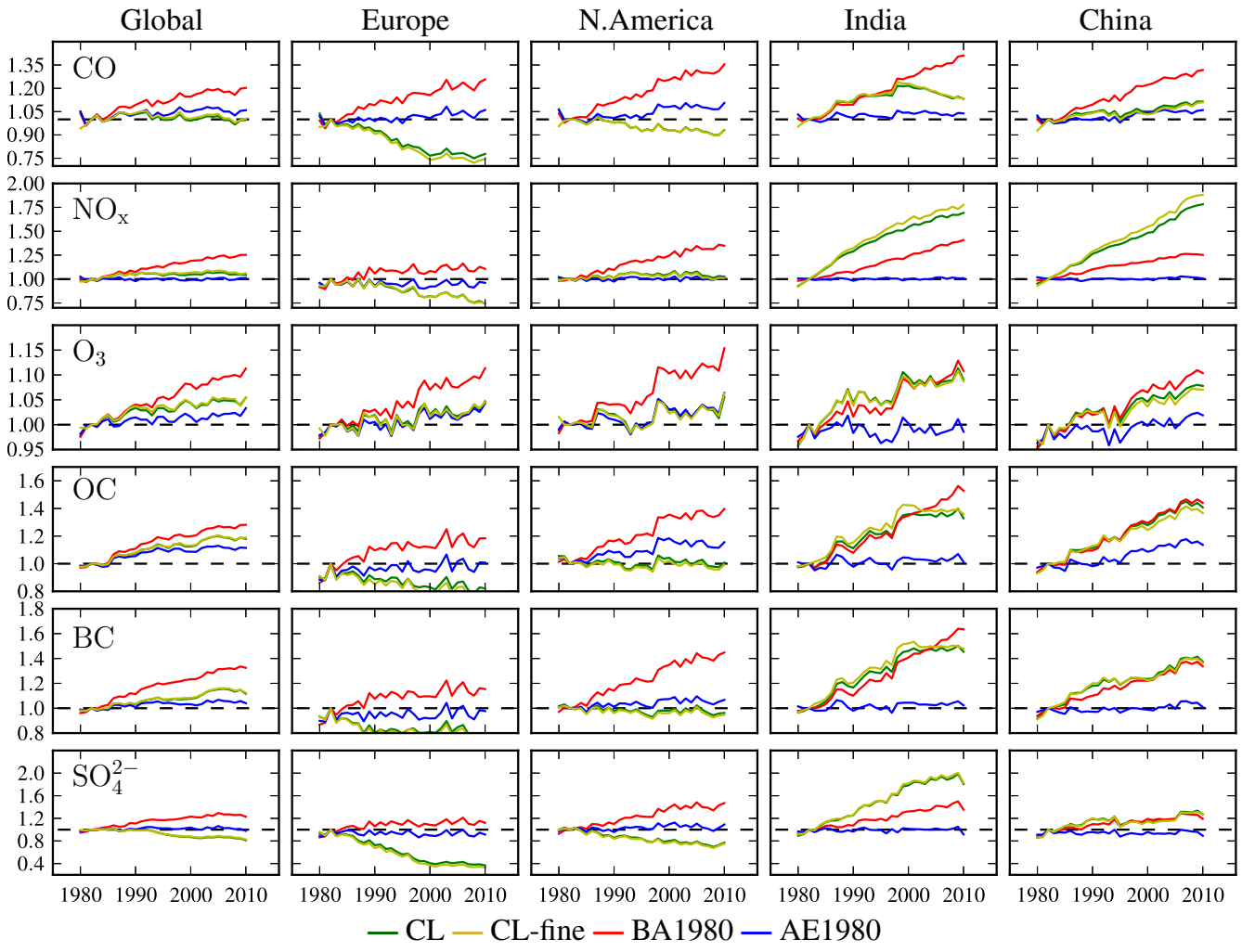

Figure 4. ~~Normalized annual~~ Annual mean surface concentrations for CO, NO_x, O_3, OC, BC, and sulphate aerosols (rows), averaged over the globe, Europe, North America, India and China (columns) and normalized to the 1982 concentrations. The difference between the blue (AE1980) and green (CL) lines is the anticipated change in concentrations when assuming 1980 anthropogenic emissions at any given year, while the difference between the red (BA1980) and green (CL) lines is the calculated change in concentrations at any given year when taking into account the increased anthropogenic emissions due to the population growth. The yellow lines depict the annual mean surface concentrations of the CL-fine simulation. Note the difference in scales between species.

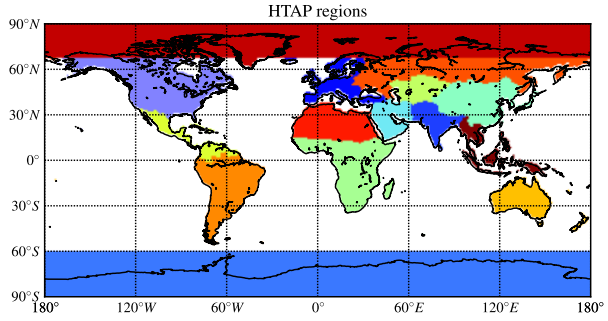

Figure S1. The HTAP source regions. Each region is represented by a different color see text for explanation of regions 2-17.

The globe was split in 16 socioeconomic regions according to the Hemispheric Transport of Air Pollution (HTAP) source regions. These regions are:

1. World (this region represents the whole globe).
2. Non-arctic/Antarctic Ocean.
3. U.S. and Canada (up to 66°North).
4. Western and Eastern E.U. and Turkey (up to 66°; polar circle).
5. South Asia: India, Nepal, Pakistan, Afghanistan, Bangladesh, Sri Lanka.
6. East Asia: China, Korea, Japan.
7. South East Asia.
8. Pacific, Australia and New Zealand.
9. Northern Africa, Sahara and Sahel.
10. Sub Saharan/ sub Sahel Africa.
11. Middle East: S. Arabia, Oman etc., Iran, Iraq.
12. Mexico, Central America, Caribbean, Guyana, Venezuela, Colombia.
13. South America.
14. Russia, Belarus, Ukraine.
15. Central Asia.
16. Arctic Circle (North of 66°N) and Greenland.
17. Antarctic.

Table S1. Global anthropogenic emissions of pollutants for the year 1980 in Tgy^{-1} used in TM4-ECPL model for CL simulation ($\mathrm{NO_x}$ is reported as N)

	Global	**Europe**	**N. America**	**India**	**China**
CO	585.16	86.15	128.56	59.40	113.56
$\mathbf{NO_x}$	24.26	4.45	6.27	0.72	2.44
$\mathbf{NH_3}$	29.06	3.51	3.02	4.48	4.89
OC	11.00	0.93	0.74	1.30	2.37
BC	4.52	0.55	0.44	0.37	1.06
$\mathbf{SO_2}$	120.67	37.84	24.56	1.74	14.08

Table S2. a) Anthropogenic emission reductions (CL) compared to BA1980 accounting for increase in population and assuming constant per capita emissions as those of the year 1980. b) Emission reductions (CL) when in addition to the population increase (BA1980) the increase in energy use per capita is also taken into account per region (based on the World Bank statistics of 2010 versus 1980) and all energy is assumed to be produced from oil consumption.

a. Achieved emission reductions compared to no policy accounting for increased population								
CL/BA1980	**N. America**	**Europe**	**India**	**China**	**S.E. Asia**	**Africa**	**Asia**	**Global**
OC	0.48	0.59	0.84	1.01	1.52	0.81	0.93	0.85
BC	0.59	0.64	0.92	1.10	1.67	0.81	0.89	0.82
NO_x	0.66	0.62	1.55	1.59	1.91	0.45	1.00	0.77
CO	0.49	0.36	0.77	0.97	1.01	0.75	0.87	0.68
NH_3	0.76	1.07	0.61	1.25	1.30	0.46	0.87	0.80
SO_x	0.45	0.24	2.35	1.25	2.09	0.40	1.03	0.56
b. Achieved emission reductions compared to no policy accounting for increased population and increased energy demand								
mean increase in energy use**	0.90	1.01	2.04	3.08	2.30	1.00[&]	2.50[&]	1.30
CL/~~worst-case~~energy-demand	**N. America**	**Europe**	**India**	**China**	**S.E. Asia**	**Africa**	**Asia**	**Global**
OC	0.54	0.59	0.41	0.33	0.66	0.81	0.37	0.65
BC	0.66	0.63	0.45	0.36	0.73	0.81	0.35	0.63
NO_x	0.74	0.61	0.76	0.52	0.83	0.45	0.40	0.59
CO	0.55	0.36	0.38	0.31	0.44	0.75	0.35	0.52
NH_3	0.85	1.06	0.30	0.41	0.57	0.46	0.35	0.61
SO_x	0.50	0.23	1.15	0.40	0.91	0.40	0.41	0.43

** assuming increase in energy use per capita and that all is based on oil consumption, i.e. no renewable energy; http://data.worldbank.org/indicator/EG.USE.PCAP.KG.OE/countries/1W-B8-CN-IN?display=default energy use as equivalent of oil consumption
[&] rough estimate

Table S3. Fractional changes in regional mean surface concentrations in 2010 for AE1980 and BA1980 compared to the CL simulation.

a. Anthropogenic Emissions fixed to 1980 (AE1980)								
AE1980/CL	**Global**	**Europe**	**China**	**India**	**N. America**	**Africa**	**S.E. Asia**	**S. America**
NO_x	0.95	1.30	0.53	0.55	0.98	1.02	0.60	0.99
SO_2	1.19	2.82	0.60	0.30	1.49	0.97	0.50	1.65
O_3	0.98	0.99	0.93	0.89	1.00	0.99	0.85	0.98
CO	1.06	1.39	0.94	0.89	1.19	0.99	0.94	1.00
SO_4^{2-}	1.19	2.54	0.65	0.47	1.43	1.22	0.57	1.43
OC	0.94	1.25	0.78	0.74	1.17	0.93	0.78	0.98
BC	0.93	1.33	0.69	0.67	1.12	0.88	0.62	0.88
b. Business As 1980 (BA1980) increase in population and constant per capita emissions as in 1980								
BA1980/CL	**Global**	**Europe**	**China**	**India**	**N. America**	**Africa**	**S.E. Asia**	**S. America**
NO_x	1.21	1.63	0.72	0.87	1.29	1.30	0.95	1.17
SO_2	1.55	3.41	0.84	0.72	1.98	1.62	1.03	1.89
O_3	1.05	1.07	1.03	1.03	1.08	1.10	1.00	1.05
CO	1.22	1.69	1.19	1.24	1.44	1.14	1.09	1.06
SO_4^{2-}	1.54	3.22	0.91	0.92	1.87	1.72	0.94	1.60
OC	1.10	1.54	1.01	1.19	1.43	1.07	0.90	1.06
BC	1.22	1.66	0.95	1.19	1.51	1.13	0.84	1.05

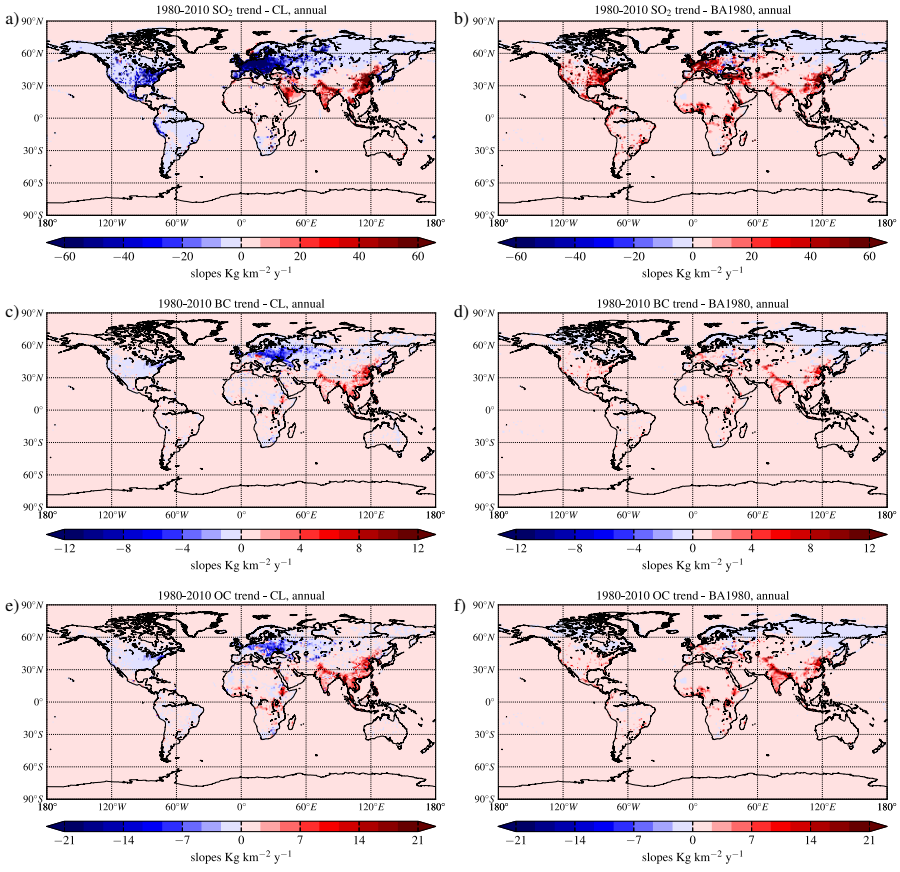

Figure S2. Distribution of annual mean anthropogenic emission trends for the period 1980-2010. Trends are calculated per grid box as mean over the period. Units are $\mathrm{Kgkm^{-2}y^{-1}}$ for $\mathrm{NO_x}$ in $\mathrm{Kg(N)km^{-2}y^{-1}}$. Left column for CL scenario, right column BA1980 scenario. Top: CO, middle: $\mathrm{NH_3}$, bottom: $\mathrm{NO_x}$.

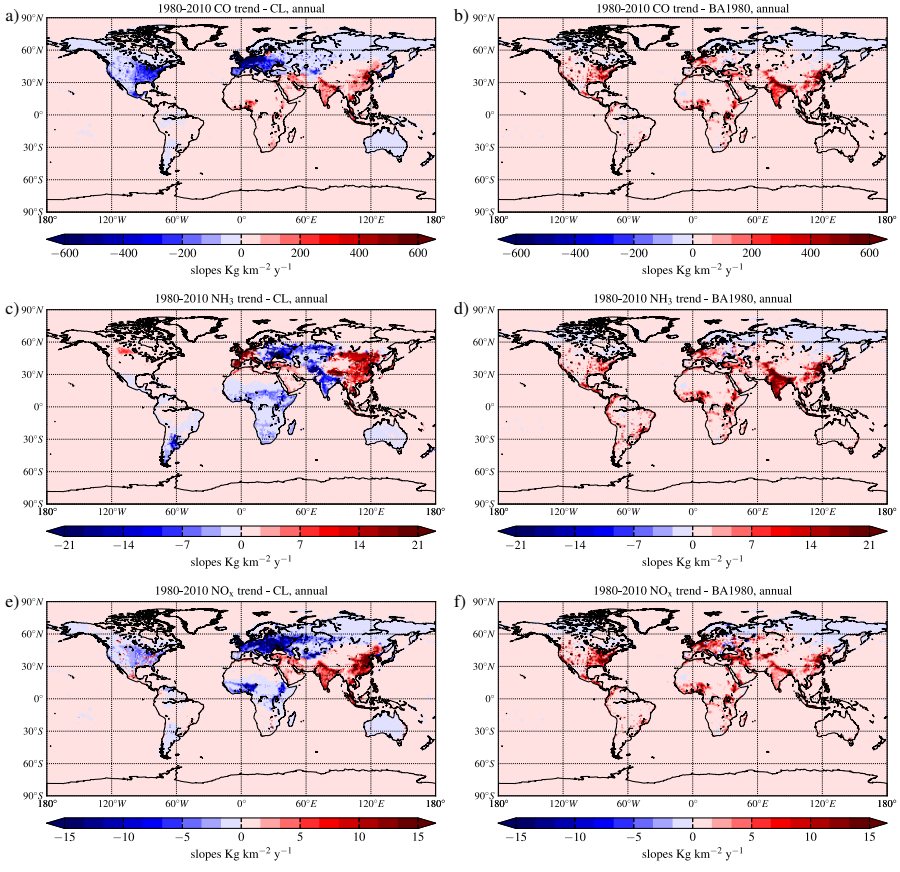

Figure S3. Distribution of annual mean anthropogenic emission trends for the period 1980-2010. Trends are calculated per grid box as mean over the period. Units are $\mathrm{Kgkm^{-2}y^{-1}}$. Left column for CL scenario, right column BA1980 scenario. Top: SO_2, middle: BC, bottom: OC.

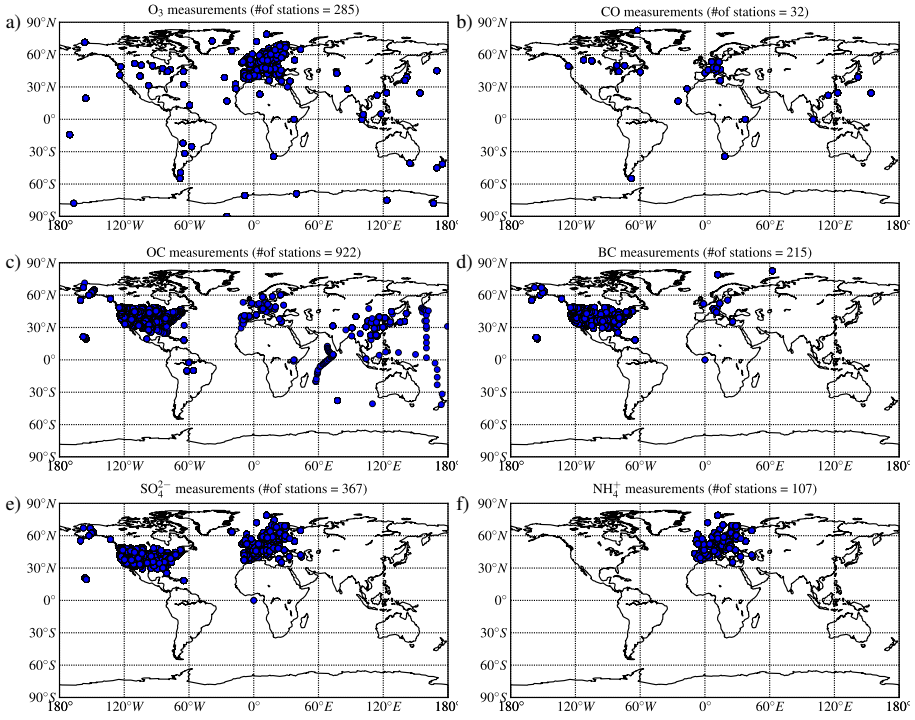

Figure S4. Location of surface stations used for model evaluation: a) for O_3, b) for CO, c) for OC, d) for BC, e) for SO_4^{2-}, f) NH_4^+. Number of stations used is provided in the title.

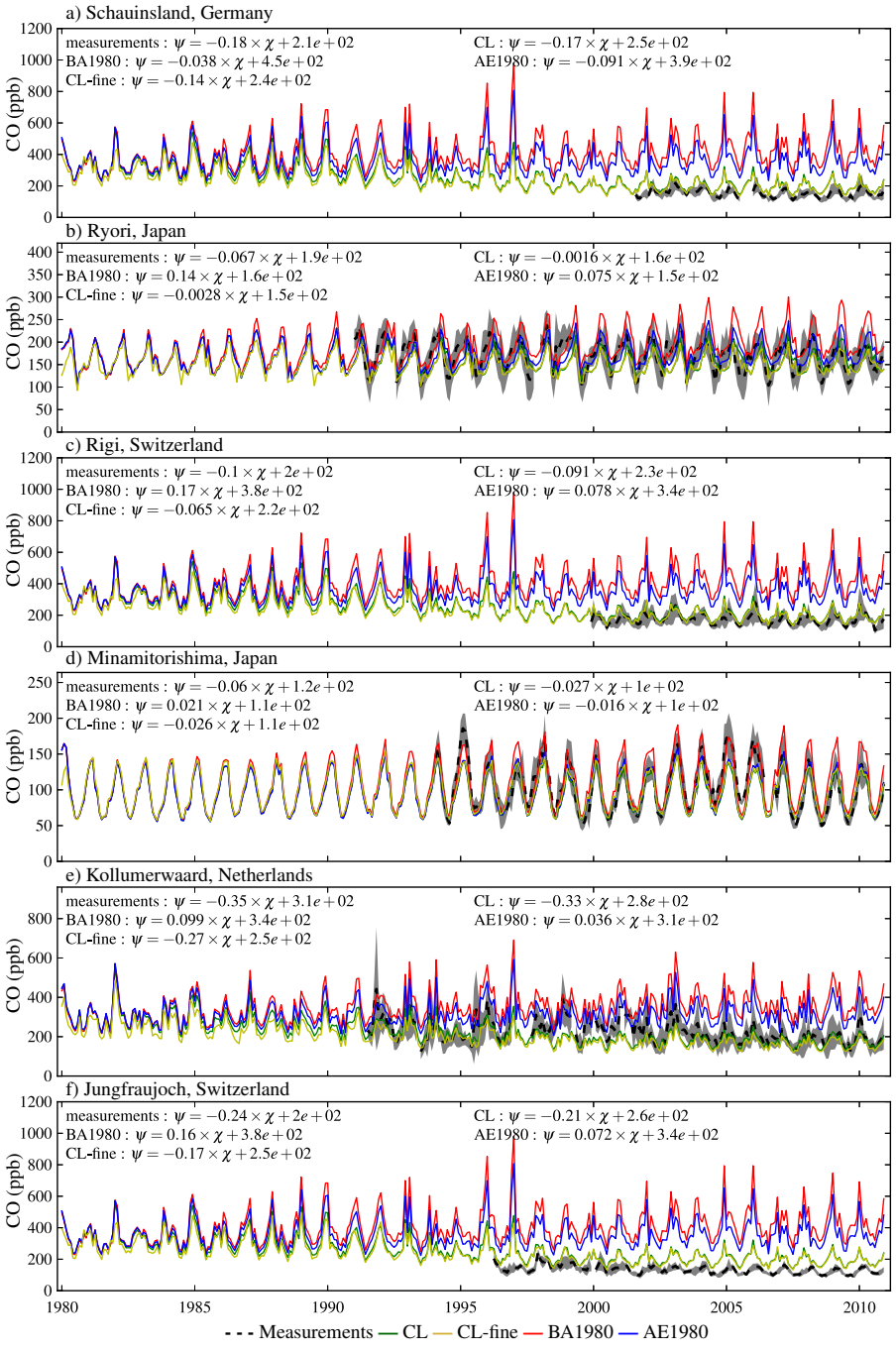

Figure S5. Comparison of the four simulations against CO observations. The dashed line and shadowed areas indicate monthly mean surface observations and one standard deviation. Simulations are CL: current legislation (green); CL-fine: current legislation in the fine resolution of the model; BA1980: Business As in 1980, with constant anthropogenic emission rates per capita as in 1980 (red); AE1980: constant anthropogenic emissions as in 1980 (blue). Trends derived from the concentrations (ψ) as a function of the year (χ) are provided for the measurements and the four simulations inside the frames.

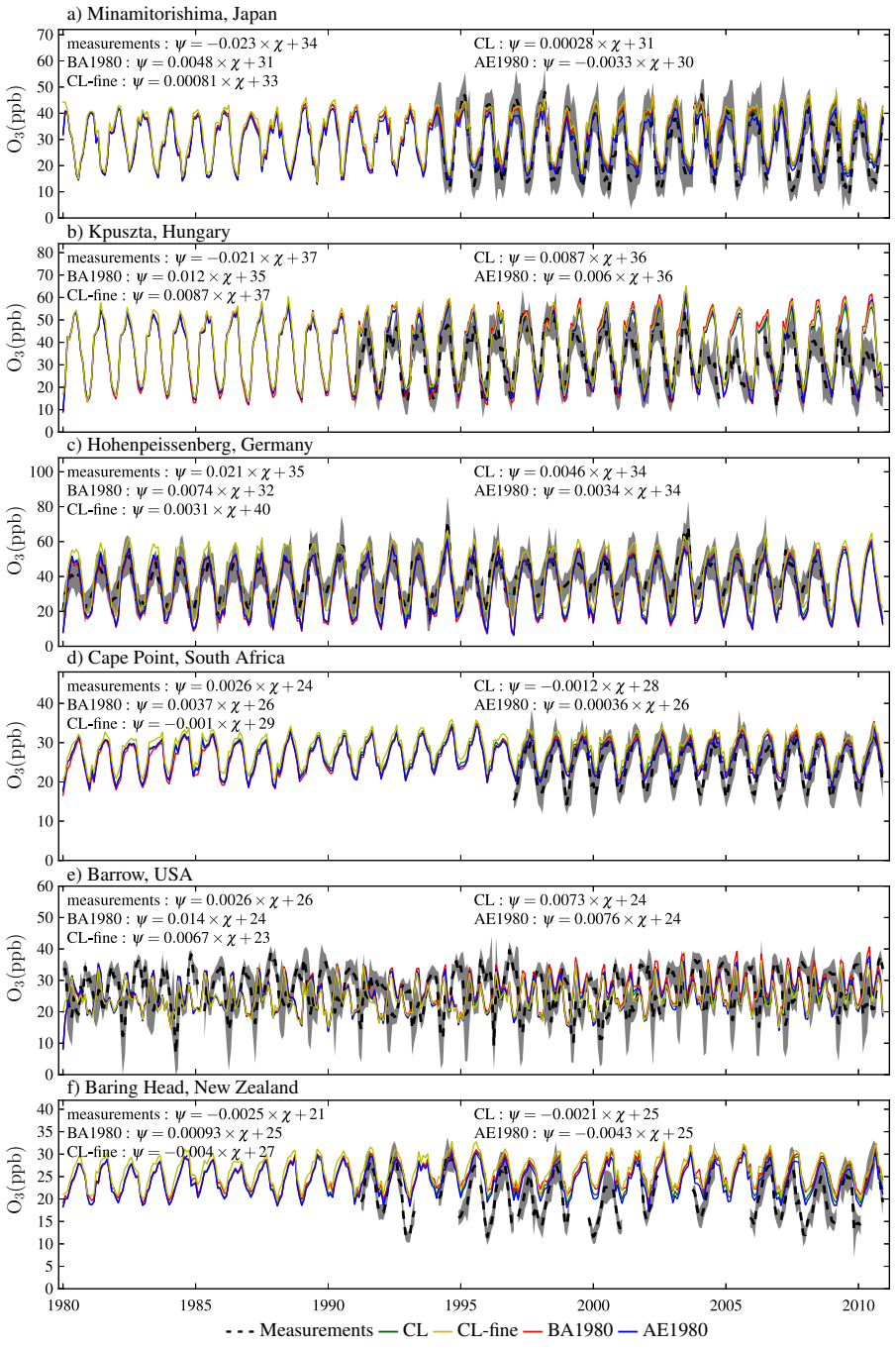

Figure S6. Comparison of the four simulations against O_3 observations. The dashed line and shadowed areas indicate monthly mean surface observations and one standard deviation. Simulations are CL: current legislation (green); CL-fine: current legislation in the fine resolution of the model; BA1980: Business As in 1980, with constant anthropogenic emission rates per capita as in 1980 (red); AE1980: constant anthropogenic emissions as in 1980 (blue). Trends derived from the concentrations (ψ) as a function of the year (χ) are provided for the measurements and the four simulations inside the frames.

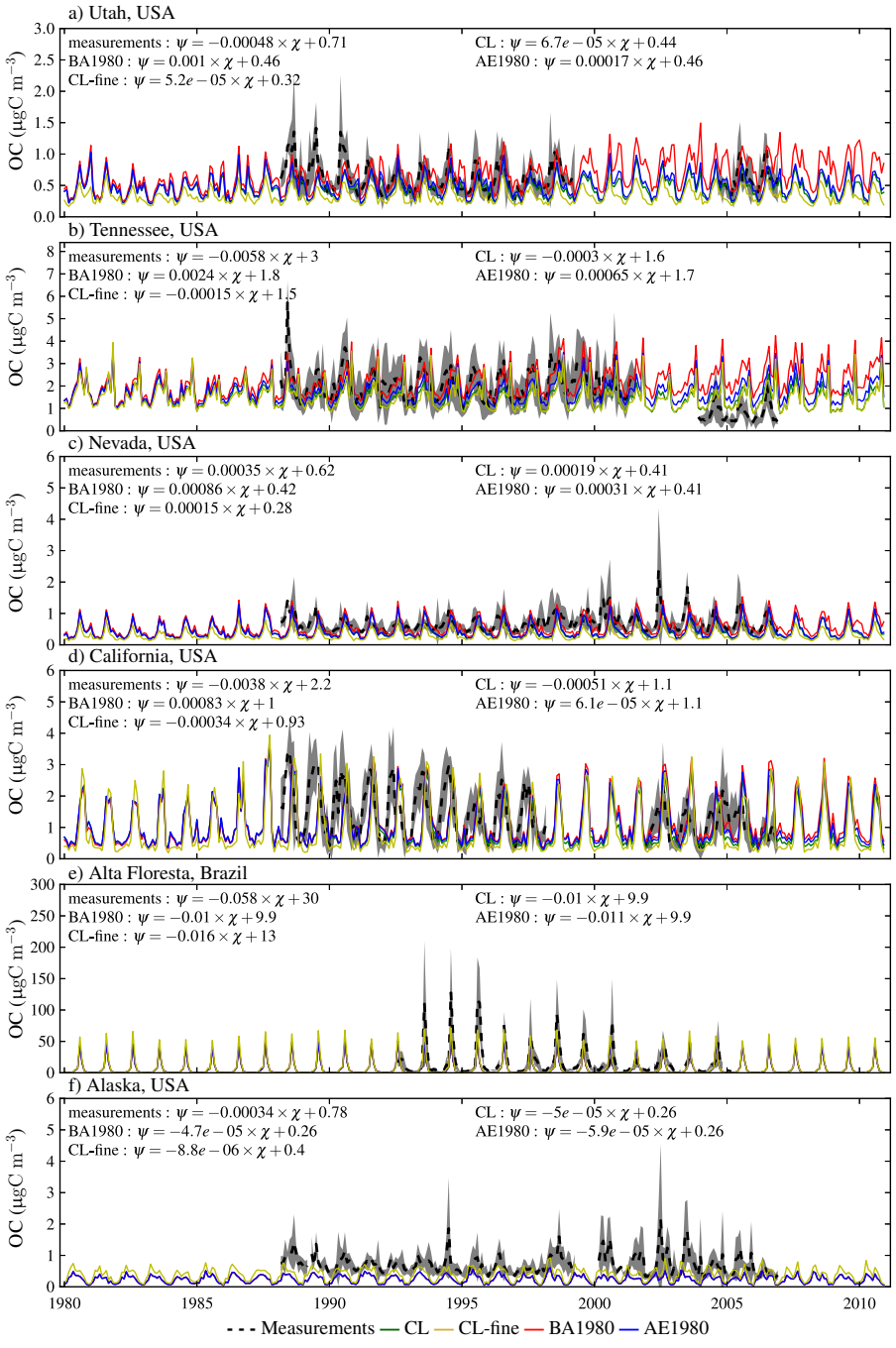

Figure S7. Comparison of the four simulations against OC observations. The dashed line and shadowed areas indicate monthly mean surface observations and one standard deviation. Simulations are CL: current legislation (green); CL-fine: current legislation in the fine resolution of the model; BA1980: Business As in 1980, with constant anthropogenic emission rates per capita as in 1980 (red); AE1980: constant anthropogenic emissions as in 1980 (blue). Trends derived from the concentrations (ψ) as a function of the year (χ) are provided for the measurements and the four simulations inside the frames.

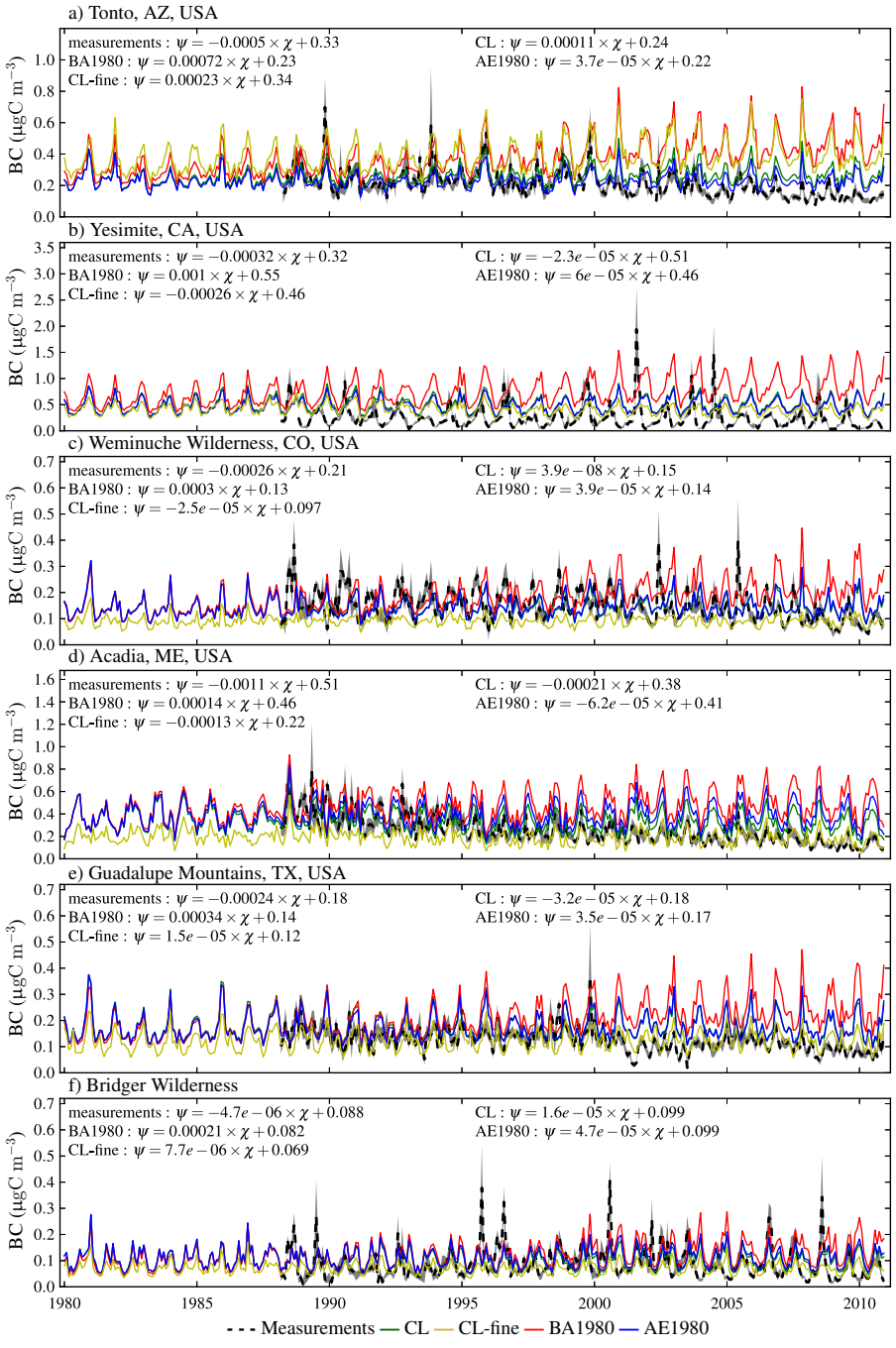

Figure S8. Comparison of the four simulations against BC observations. The dashed line and shadowed areas indicate monthly mean surface observations and one standard deviation. Simulations are CL: current legislation (green); CL-fine: current legislation in the fine resolution of the model; BA1980: Business As in 1980, with constant anthropogenic emission rates per capita as in 1980 (red); AE1980: constant anthropogenic emissions as in 1980 (blue). Trends derived from the concentrations (ψ) as a function of the year (χ) are provided for the measurements and the four simulations inside the frames.

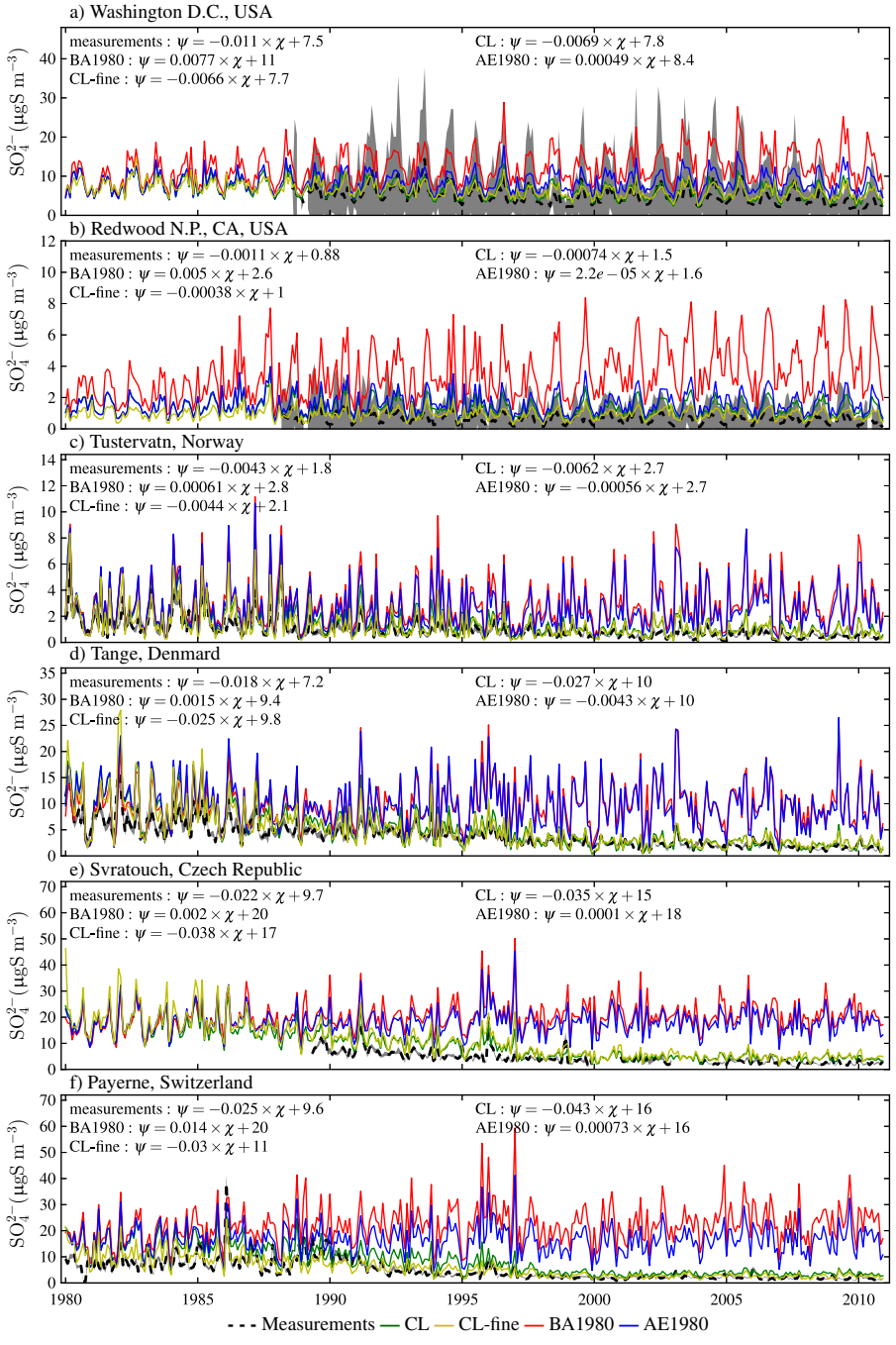

Figure S9. Comparison of the four simulations against SO_4^{2-} observations. The dashed line and shadowed areas indicate monthly mean surface observations and one standard deviation. Simulations are CL: current legislation (green); CL-fine: current legislation in the fine resolution of the model; BA1980: Business As in 1980, with constant anthropogenic emission rates per capita as in 1980 (red); AE1980: constant anthropogenic emissions as in 1980 (blue). Trends derived from the concentrations (ψ) as a function of the year (χ) are provided for the measurements and the four simulations inside the frames.

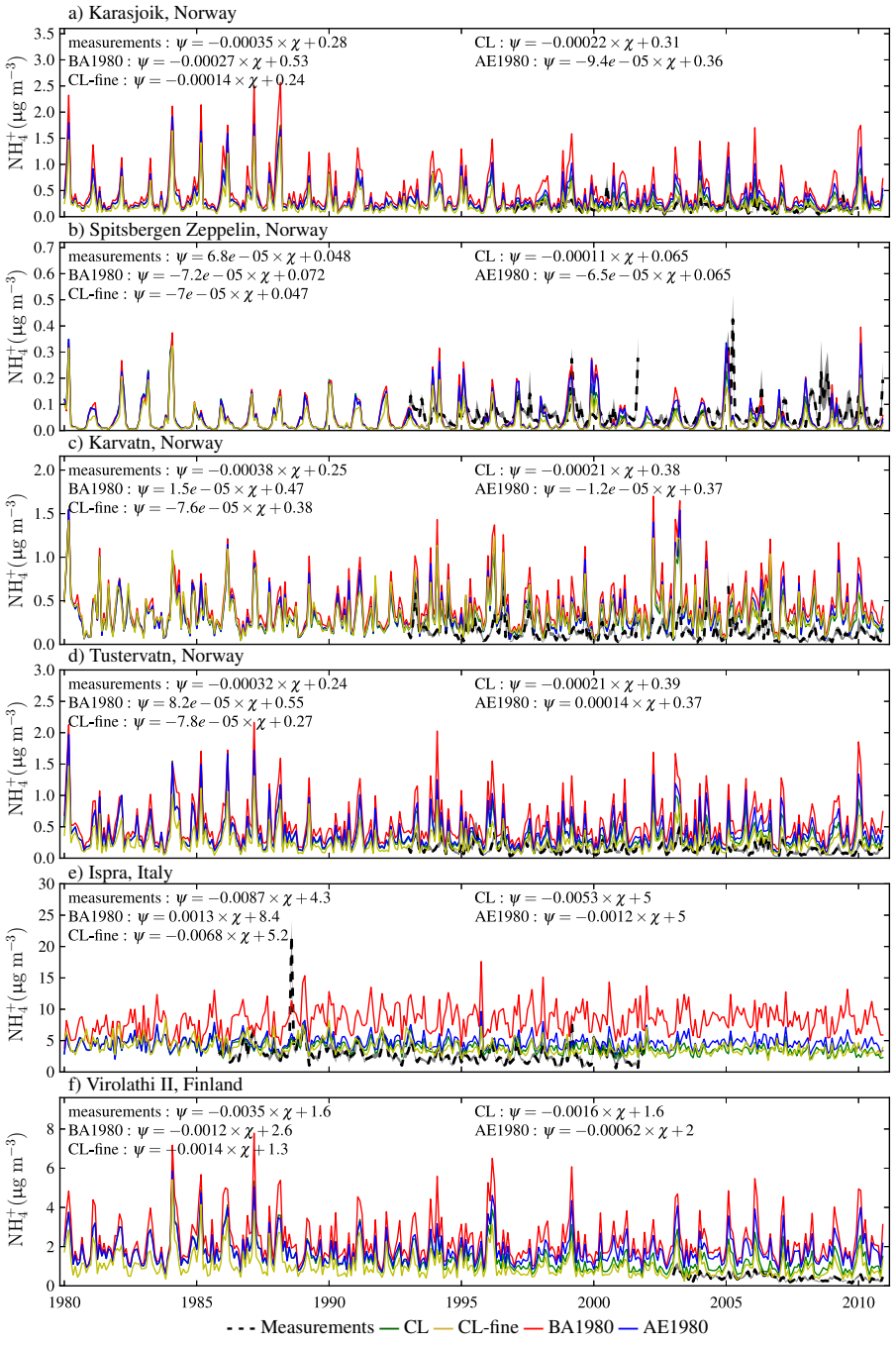

Figure S10. Comparison of the four simulations against NH_4^+ observations. The dashed line and shadowed areas indicate monthly mean surface observations and one standard deviation. Simulations are CL: current legislation (green); CL-fine: current legislation in the fine resolution of the model; BA1980: Business As in 1980, with constant anthropogenic emission rates per capita as in 1980 (red); AE1980: constant anthropogenic emissions as in 1980 (blue). Trends derived from the concentrations (ψ) as a function of the year (χ) are provided for the measurements and the four simulations inside the frames.

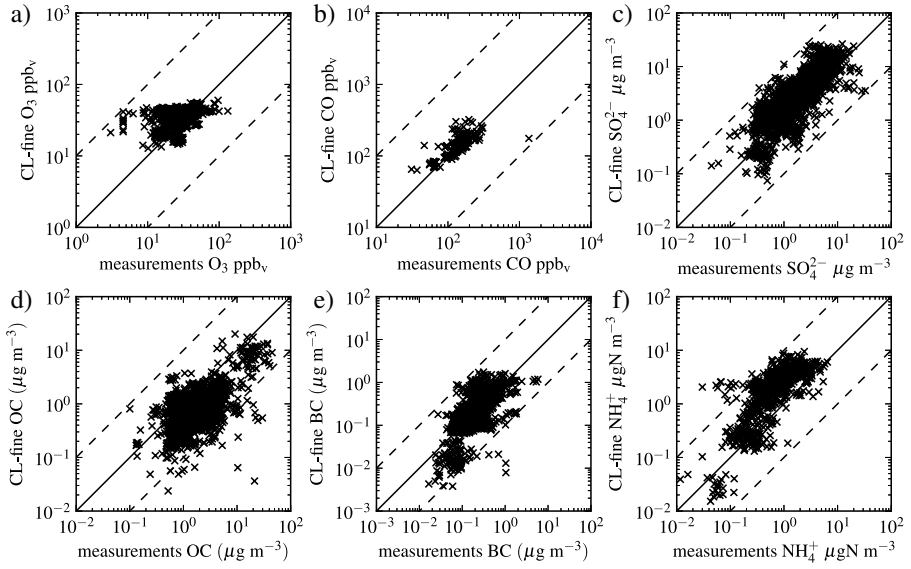

Figure S11. Comparisons of annually average surface model results ($3^\circ \times 2^\circ$version) with observations (see Fig. S4 for station locations) a) for O_3, b) for CO, c) for SO_4^{2-}, d) for OC, e) for BC, f) NH_4^+. The continuous line shows the 1:1 slope and the dashed lines the 10:1 and 1:10 slopes