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ÉCLAIRE

Effects of Climate Change on Air Pollution Impacts and Response Strategies for European Ecosystems

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Effects of Changes in Global Climate, Chemistry, Emissions and Landcover Changes on Air Pollution Metrics

1 Executive Summary

In this report we present results from European-scale chemical transport models (CTMs) which have been used to assess current and future concentrations and depsoitions of key air pollutants. The modelling results show that the main driver of future air pollution (AP) will be emissions changes rather than climate-induced changes. For ozone the impacts of non-European sources are substantial, but their development very uncertain. Hemispheric or global scale methane controls are suggested as a win-win strategy for reducing both greenhouse warming and tropospheric ozone levels. For reactive nitrogen (Nr) compounds, European emissions dominate European deposition levels.

Although emissions changes are likely the main driver for AP changes, there may be significant effects of climate on emissions of some compounds. An important finding of ECLAIRE work is that increased evaporation of ammonia (NH_3) brought about with a warmer climate may be significant, and we show that this has important consequences for critical load exceedances for Nitrogen, and hence for the effectiveness of currently planned emission controls in Europe.

2 Objectives

The objectives of D7.4 are to assess how air pollution metrics (APMs, e.g. concentrations, deposition loads, exceedance statistics) are likely to change in future (up to 2050) using regional chemical transport models, and to compare and contrast the effects of changes in global climate, chemistry, emissions and landcover changes on these APMs.

3 Activities

A series of modelling activities were undertaken and are reported here, involving a number of CTMs. We summarise here the modelling systems that were used, along with highlights from these studies. Further details can be found in the cited publications.

In addition, contributions were made to two review articles which assessed our understanding of the links between climate change and air pollution: Simpson et al. (2014a) and Fowler et al. (2015).

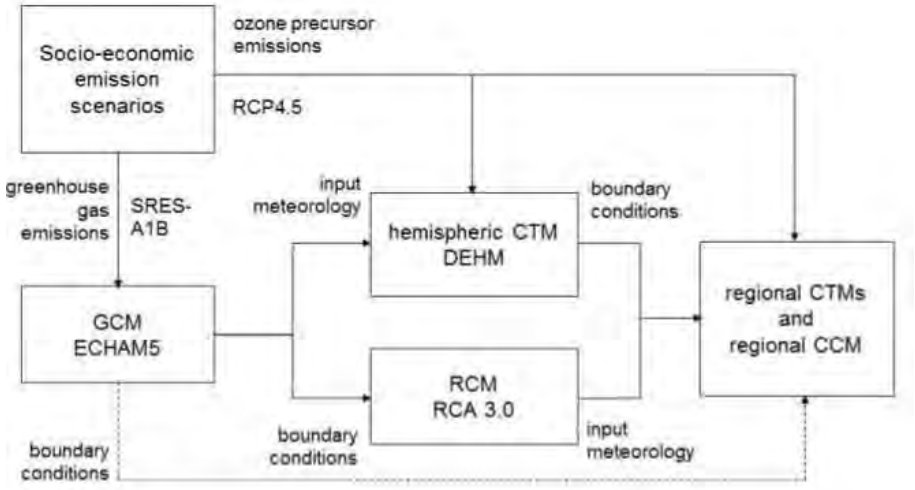


Figure 1: The model chain used in the multi-model simulations for 2005–2050. (From Langner et al. 2012b.)

3.1 Models

A series of modelling activities were undertaken and are reported here, involving three regional (European-scale) CTMs: the EMEP MSC-W model (Simpson et al. 2006, 2012, Fagerli and Aas 2008), the Swedish MATCH model (e.g. Robertson et al. 1999, Langner et al. 1998, Engardt 2000), and Finnish SILAM model (e.g. Sofiev et al. 2008), along with one hemispheric-scale CTM, the Danish DEHM model (e.g. Christensen 1997, Geels et al. 2012). For the ozone modelling study (Langner et al. 2012b) a new online climate-chemistry model (CCM), EnvClimA, developed at the Danish Meteorological Institute, was also used.

Fig. 1 illustrates the flow of information used in the multi-model studies (Langner et al. 2012b, Simpson et al. 2014c). In order to obtain climate-sensitive meteorology, meteorological data from a global climate model (GCM, here ECHAM5) were used in both a regional climate model (RCM; we used SMHI's RCA3 model (Samuelsson et al. 2011, Kjellstrom et al. 2011)) and the offline hemispheric chemical transport model (DEHM). The downscaled meteorology from the RCM was used together with time-varying boundary conditions from the hemispheric DEHM CTM to drive the three European-scale CTMs. The horizontal grid for these CTMs was identical to the RCA3 grid, while the vertical discretization was left free to each model. To ensure consistency, the offline DEHM model was operated with global emissions for 2005 and 2050 from the same system as used for the European-scale CTMs.

Table 1: Emissions for EU28$^+$ used in the calculations for 2005 and 2050. Data from the IIASA/ECLIPSE v.4^e data set; see Simpson et al. (2014c). Unit: $\mathrm{Tg\,yr^{-1}}$ (EU28$^+$ here denotes the 28 EU countries, plus Norway and Switzerland.)

Year	SO_x (as SO_2)	NO_x (as NO_2)	NH_3	NMVOC
2005	8.41	12.5	3.99	10.1
2050	2.10	4.10	4.04	5.94
Change (%)	−75	−67	+1	−41

3.2 Emissions

The models used in this study require emissions of sulfur and nitrogen oxides (SO_x, NO_x), NH_3, non-methane volatile organic compounds (NMVOCs), and CO, and CH_4 for DEHM. The anthropogenic emissions consist of annual, gridded data sets. Ten major types of anthropogenic emissions are used, classified with the so-called "SNAP"-level emission sectors (SNAP stands for Source Nomenclature of Air Pollutants; for example, SNAP-7 is road traffic, SNAP-10 is agriculture, etc.).

In the ECLAIRE regional modelling studies, all models made use of the same emission files (albiet updated for each exercise), which contained gridded SNAP-level data on the RCA3 grid (and a global grid for DEHM). Emission data sets using this procedure were provided for the years 2005 and a 2050 "current legislation" (CLE) scenario. Details of the emissions are given in the respective papers, but the EU totals used for Simpson et al. (2014c), using ECLAIRE-specific emissions from IIASA, are presented in Table 1. For the long-term (1900-2050) historical calculations performed with the EMEP and MATCH models in support of ECLAIRE ecosystem models, these 2005 emissions were extended backwards, first towards EMEP 1990 emissions, then using scaling factors derived from Lamarque et al. (2010).

A number of other emissions sources are typically used in the CTMs. These include so-called natural NO_x emissions from soils; NMVOC from vegetation; and emissions from forest fires, aircraft and lightning. The CTMs have different approaches to these emissions sources, and harmonising these was beyond the scope of our study. Instead, in order to simplify the interpretation of the CTM results, we have adopted the simple policy of setting emissions from soils, forest fires, aircraft and lightning to zero, so that all NO_x emissions in the models stem from the common emission data set discussed above. In contrast to these minor emission sources, emissions of NMVOC from vegetation are too great to ignore (e.g. Simpson et al. 1999), and as in Langner et al. (2012b), each model simply calculates its own emissions at each model time step. Similarly, volcanic emissions are a significant fraction of European S emissions. The official EMEP

estimate of volcanic emissions was used for all models.

3.2.1 Climate-emissions experiments

Estimation of future emissions is of course uncertain for even well understood sources. However, models can be used to explore the importance of different emission scenarios, and even to allow speculation about previously unused emissions. We have explored three sources of emissions which can be considered 'new' in that most have not previously been considered in CTM modelling.

3.2.1(a) Increased NH_3 emissions Two papers arising from the ECLAIRE project drew attention to the possibility of quite significant increases in NH_3 emissions in the future as a result of increasing evaporation from sources such as animal manure (Sutton et al. 2013, Skjøth and Geels 2013). NH_3 emissions are a function of both water availability and temperature with, in principle, a doubling of the emission for each 5 °C increase. Sutton et al. (2013), using empirical models and measurements, estimated a potential 42 % increase in the global NH_3 emissions following a 5 °C increase towards 2100. Skjøth and Geels (2013) used a dynamic NH_3 emission model to study the temporal and geographical variations in ammonia emissions across the northern part of Europe arising from a 'typical' pig-stable. They found increases towards the 2050s of 15–30 % (relative to 2007) in the emissions in central to northern Europe, increasing to ca. 20–40 % by the end of the century.

The projected increase will of course depend heavily on the projected temperature change and hence on the applied climate model, as well as assumptions concerning NH_3 emission factors. However, based on the above studies, Simpson et al. (2014c) explored the potential impact of a 20 and 30 % increase in NH_3 emissions for the future (2050s) scenarios - see Sect. 4.2.4 .

3.2.1(b) Changed shipping routes Another potentially dramatic change in emissions due to global warming involves the rapid retreat of the Arctic sea during recent decades (Comiso 2012, Corbett et al. 2010), with the potential for nearly ice-free summers within this century (Wang and Overland 2012). According to Corbett et al. (2010) the NOx emissions from Arctic shipping in high growth scenarios will increase by a factor of 4 to 2050 compared to 2004, or almost a factor of 14 if high global shipping routes are diverted into Arctic areas (along e.g. the Northeast passage past the Norwegian coast). Earlier studies with the large scale MOZART model (Granier et al. 2006) had warned of dramatic changes in ozone levels over the Arctic, but the relatively low resolution (ca. 2.8°) of this early study may have overemphasised the impact of NOx on non-linear ozone formation. Tuovinen et al. (2013) made use of the higher resolution (50 km) EMEP model, combined

with data interpolated from the 5×5 km resolution Corbett data-sets, to estimate the impacts of these shipping emission changes on ozone (actually phyto-toxic ozone dose, POD) and N-deposition over northern Europe. The EMEP model setup used for this study was otherwise identical to that used for the multi-model Simpson et al. (2014c) study reported above.

3.2.1(c) Increased biotic stress Essentially all photochemical transport models handle the emissions and chemistry of biogenic volatile organic compounds (BVOC), which are assumed to be emitted under the control of simple meteorological factors such as heat and light. These emissions, for example isoprene and monoterpenes, are referred to as 'constitutive' emissions. In addition to these constitutive BVOC emissions, vegetation also emits organic compounds in response to various types of stress – so called stress-induced emissions (SIE). Some types of stress lead to very large induced VOC-emissions and many of the emitted compounds are highly reactive and can form large amounts of organic aerosol after oxidation in the atmosphere (Mentel et al. 2013).

Such SIE are often caused by 'biotic stress' – infestation of insects, viruses, fungi, etc. (Arneth and Niinemets 2010, Berg et al. 2013, e.g.), but they are also affected by other stressors like heat or drought.

In the first modelling study of the possible impacts of such stress-induced missions in Europe, we have investigated the potential impact on organic aerosol formation from these biotic SIE for forests in northern Europe ($> 45°$ N) (Bergström et al. 2014). Although necessarily speculative to some extent, estimates were based upon observations as far as possible. Emission estimates for sesquiterpenes (SQT), methyl salicylate (MeSA) and unsaturated C17 compounds, due to different stressors, were based on experiments in the Jüich Plant Atmosphere Chamber (JPAC, Mentel et al. 2013), combined with estimates of the fraction of stressed trees in Europe based on reported observed tree damage.

SIE were introduced into the EMEP model, and secondary organic aerosol (SOA) yields from the SIE were taken from the JPAC experiments (Mentel et al. 2013, Bergström et al. 2014). Some results are reported below. It should be noted that this study was the first of its kind, with the aim to draw attention to a source of SOA that has previously been ignored. Although this study could only be illustrative in nature, BVOC and BSOA from such sources clearly requires further study, especially since biotic stress may well increase in a warmer climate (Jonsson et al. 2009).

Table 2: Comparison of estimated annual isoprene emissions in current and future decade. Results from four CTMs (Langner et al. 2012b)

Model	Emissions (Gg/yr) 2000-2009	Emissions (Gg/yr) 2040-2049
DEHM	8018	9910
EMEP	3405	4114
MATCH	1592	1917
SILAM	4080	5139

4 Results and Discussion

4.1 Ozone

4.1.1 O_3 - impacts of climate - multi-model results

Details of the ECLAIRE multi-model work on regional ozone modelling can be found in Langner et al. (2012b). Briefly, this work made use of four offline regional CTMs, one offline hemispheric CTM, and one online regional integrated CCM as discussed above, driven by the same global projection of future climate under the SRES A1B scenario.

Ensemble mean changes in summer mean ozone and mean of daily maximum ozone due to climate change were found to be small, close to 1 ppb in parts of the land area in southern Europe. Corresponding changes of 95-percentiles of hourly ozone are close to 2 ppb in the same region. In northern Europe ensemble means for mean and daily maximum O_3 show negative changes while there are no negative changes for the higher percentiles indicating that climate impacts on O_3 could be more important in connection with extreme summer events.

The sensitivity of the simulated surface ozone to changes in climate between the periods 2000-2009 and 2040-2049 was found to differ by a factor of two between the models, but the general pattern of change with an increase in southern Europe was similar across different models (Fig. 2). Emissions of isoprene differ substantially between different CTMs ranging from 1.6 to 8.0 Tg/yr for the current climate, partly due to differences in horizontal resolution of meteorological input data. Also the simulated change in total isoprene emissions varies substantially across models (Table 2) explaining part of the different climate response on surface ozone.

4.1.2 O_3 - Impacts of climate versus emissions

In a complementary study (Langner et al. 2012a), the impact of climate change and changes in ozone precursor emission on summer surface ozone in Europe was studied

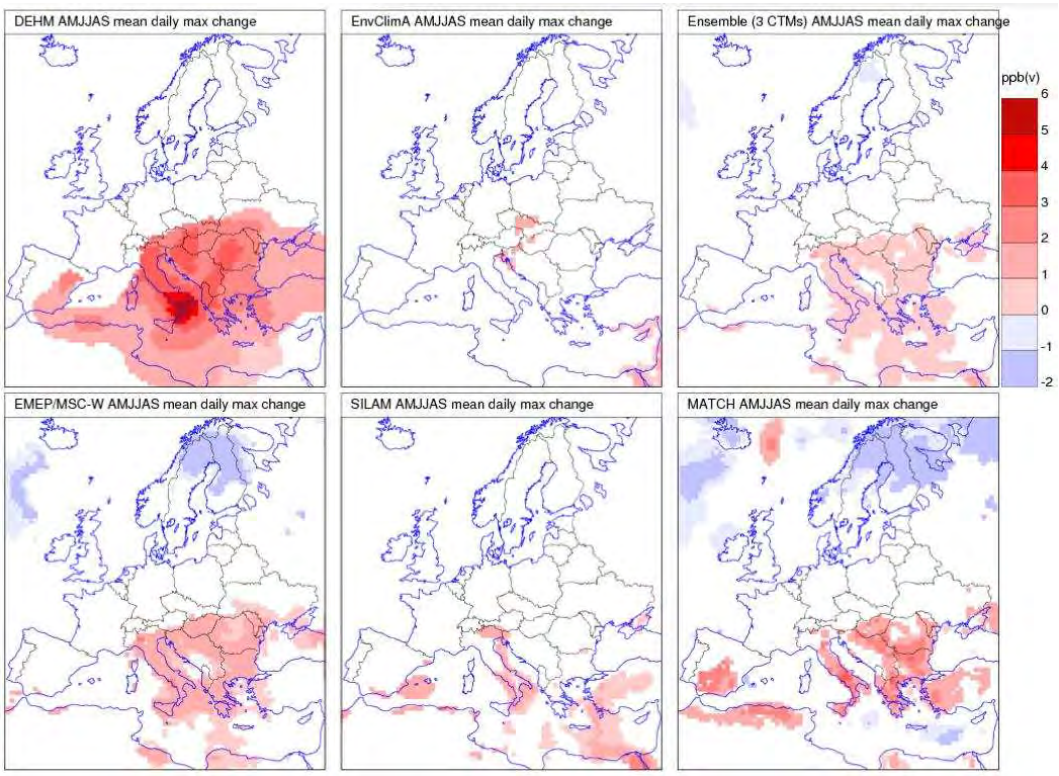


Figure 2: Simulated April–September change 2000-2009 to 2040-2049 in average daily maximum O_3 concentration at the first model level. Only changes that are statistically significant at the 95% level are plotted. Units ppb(v). Results from Langner et al. (2012b).

using the MATCH model over a longer period, 1990 to 2100. Two different climate simulations under the SRES A1B scenario together with ozone precursor emission changes from the RCP4.5 scenario were used as model input.

In southern Europe regional climate change was found to lead to increasing surface ozone concentrations during April-September, but projected emission reductions in Europe had a stronger effect, resulting in net reductions of surface ozone concentrations (Fig. 3). In northern Europe regional climate change decreased surface O_3 and reduced European emissions acts to further strengthen this trend also when including increasing hemispheric background concentrations. The European O_3 precursor emission reductions in RCP4.5 are substantial and it remains to be seen if these reductions can be achieved. There is substantial decadal variability in the simulations forced by climate variability which is important to consider when looking at changes in surface O_3 concentrations, especially until the first half of the 21st century.

4.1.3 O_3 - future developments - remarks

As discussed in more detail in Simpson et al. (2014a) (and refs. therein), tropospheric ozone and reactive nitrogen species are involved in a complex web of interactions with other atmospheric gases and particles, and through ecosystem interactions with the N-cycle and climate change.

Although ozone may have important effects on climate change as discussed above, recent model studies suggest low or modest impact of climate change on future ozone and/or Nr-deposition (Langner et al. 2012b, Colette et al. 2013, Simpson et al. 2014b). The possibility remains however that future climate may be more extreme than used in these studies, which could change O_3 dramatically. The year 2003 provides a clear example, with severe ozone episodes and widespread drought in Central Europe (Solberg et al. 2008). Using regional climate simulations, Beniston (2004) concluded that for 'many purposes the 2003 event can be used as an analogue of future summers in coming decades in climate impacts and policy studies'.

Regardless of climate, the development of ozone in future is critically dependent upon emission changes. Fig. 4 illustrates this with estimates presented by Wild et al. (2012), in which the results of 14 global chemical transport models were parameterised so that surface ozone could be estimated from emissions of NOx, CH_4 and other precursors. The newer and more stringent 'RCP' emissions scenarios produce much smaller increases in O_3 than the older 'SRES' estimates. About 75% of the 5 ppb difference between the outlying RCP 2.6 and RCP 8.5 scenarios could be attributed to differences in methane abundance. There is clearly plenty of scope for emission control to change future ozone.

The importance of ozone as a short-lived climate gas is receiving increasing attention, and mitigation of ozone through precursor control is seen as a promising strategy to help mitigate climate warming (Royal Society 2008, Shindell et al. 2012). Some

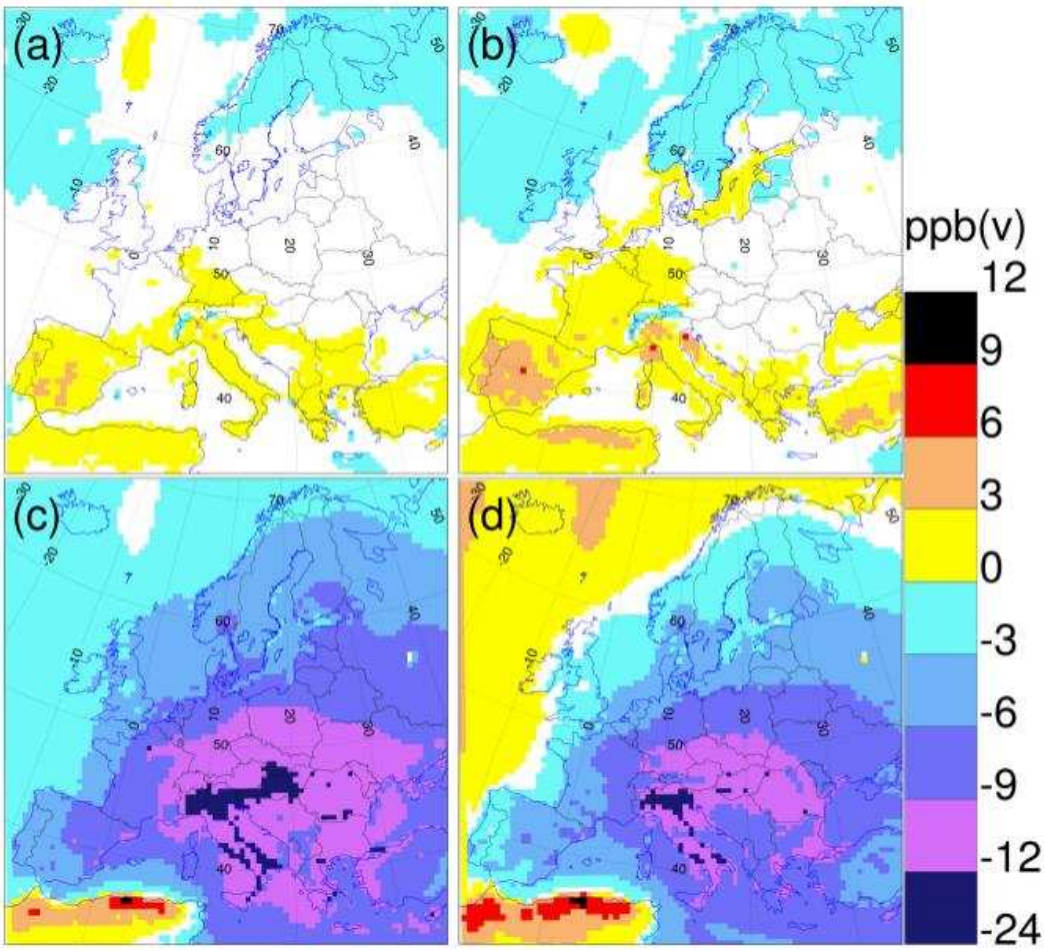


Figure 3: Change in summer (April–September), daily maximum, surface O_3 concentration from 1990–2009 to 2040–2059. (a) and (b) changes due to change in climate only; RCA3 downscaling of ECHAM5 and HadCM3, respectively. (c) ECHAM5 downscaling and change in European O_3 precursor emissions. (d) Increasing boundary case; the change due to combined change in climate, change in O_3 precursor emissions and an increasing hemispheric background of O_3 of 0.1 ppb/yr. Non-significant changes at 95% confidence level are masked white. From Langner et al. (2012a).

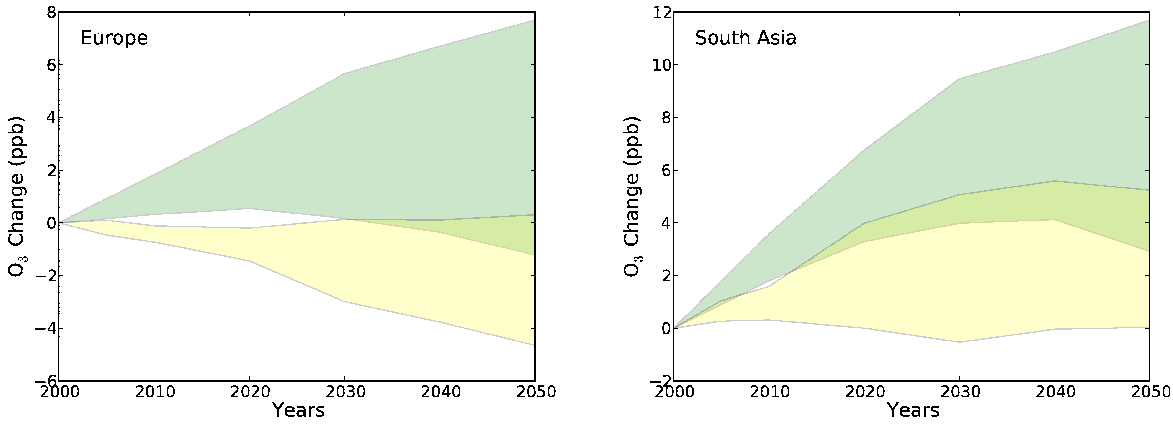


Figure 4: An uncertain future for ozone. Plots show estimates of future surface ozone in Europe and South Asia. The green area shows the range of O_3 predicted from the IPCC 4th Assessment Report (SRES scenarios A2, A1B, B2, B1), and the yellow area gives the updated range using the IPCC 5th AR (RCP8.5,6.0,4.5,2.6). Figure from Simpson et al. 2014a, redrawn from Wild et al. 2012.

measures are complex however, with for example emission control of NOx likely to lead to warming in the short term (ca. 20 years) but cooling in the longer term (Collins et al. 2010). Many studies stress the benefits of CH_4 control on a global scale, since emissions reductions are beneficial for most environmental issues.

4.2 N-deposition

We first illustrate the long-term changes in N-deposition over Europe. As part of the ECLAIRE project, model results for N-deposition were calculated over the period 1900-2050, as an input to various ecosystem models. For 1960-2050, meteorology was taken directly from the same RCM (RCA3) system discussed above for the multi-model calculations. Meteorology prior to 1960 was generated by assigning random years of 1960 to 1969 to represent each of the years 1900 through 1959. Emissions data was generated as discussed in Sect. 3.2 above. Figure 5 illustrates the results of these calculations. The very different development of the oxidised (NOy) and reduced (NHx) nitrogen components is very clear. NOy depositions peak around 1980-1990, but then decline to levels not far above those found at the turn of the 20th century. NOx depositions, on the other hand, remain high from the 1980s onwards, although with a fall from 1990 to 2000 (this was mainly due to structural changes in Eastern Europe). Although comparison with measurements is not the focus of this deliverable, these data have been compared with both current and historical data, and found to reproduce the values remarkably well (Engardt et al. 2015).

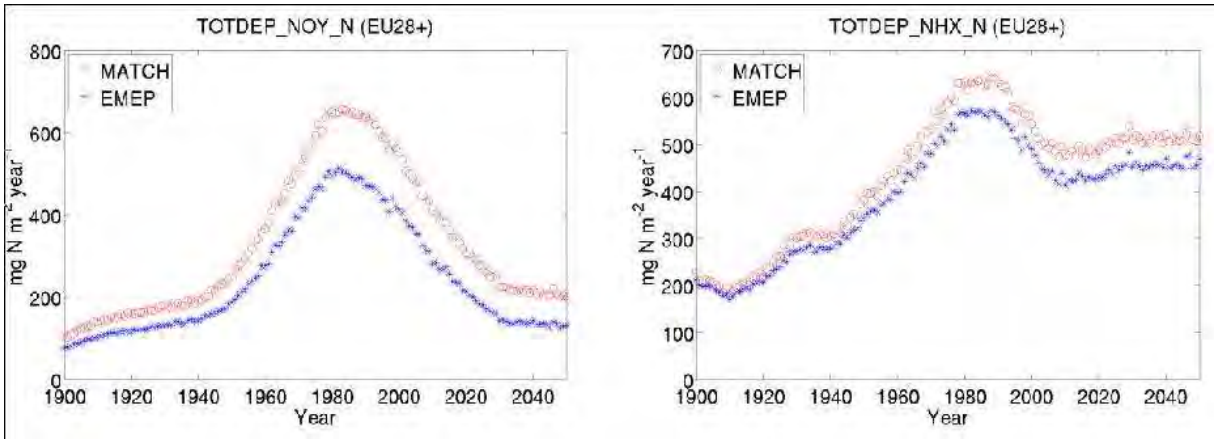
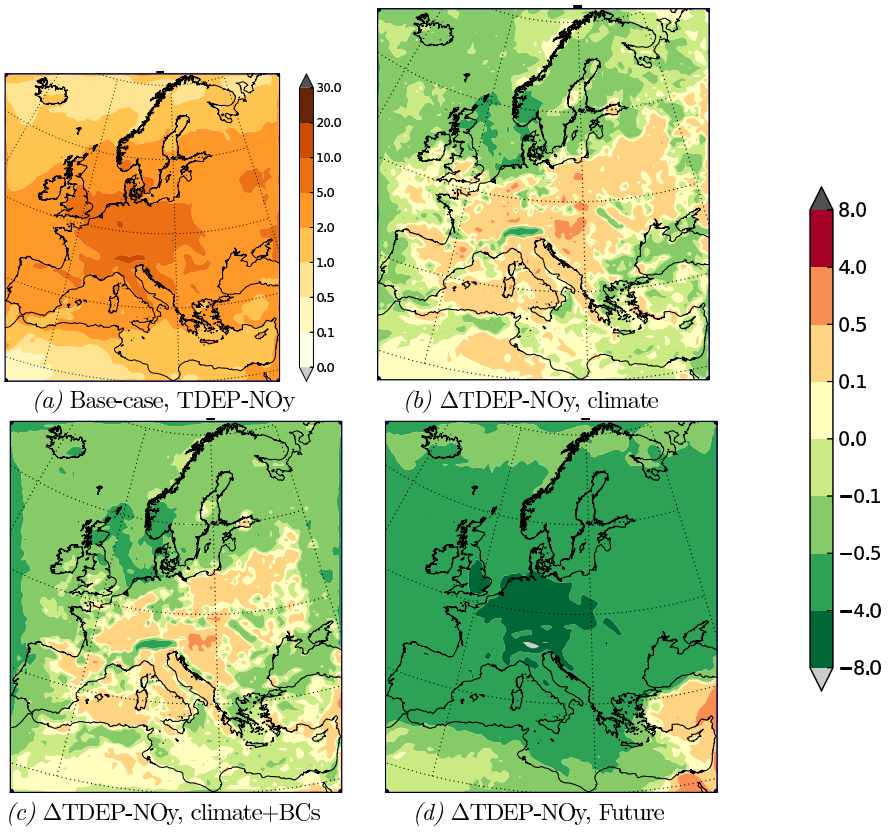


Figure 5: Calculated trends in N-deposition over EU28 (+Switzerland + Norway) using the EMEP and MATCH regional CTMs, 1900-2050

4.2.1 N-dep. - multi-model comparisons of climate versus emission changes

The results of this multi-model ensemble for the base-year, including comparison with measurements and comparison of concentration and deposition fields have been presented in ECLAIRE deliverable D7.1, and in Simpson et al. (2014c). Here we summarise the results of this study pertinent to the D7.4 aims, evaluating the relative impact of emissions versus climate-change on N-deposition across Europe.
Figure 6a shows the ensemble mean NOy deposition from the European-scale CTMs, with levels of around 5–10 kg (N) ha $^{-1}$ in central Europe, declining to less than 2 kg (N) ha^{-1} in northern areas. Figure 6b shows the changes in NOy deposition arising from climate change only. Levels of NOy deposition increase in central Europe to some extent (ca. 0.1–0.5 kg (N) ha^{-1}), but decrease in, for example, the Nordic area by a similar amount. Figure 6c shows the corresponding changes brought about by scenario in which boundary conditions are also allowed to change to 2050 levels, but the picture is little changed from the effects of climate change alone. Figure 6d shows much more dramatic changes in the case where European emissions are set to the 2050 levels. NOy deposition is reduced by more than 0.5 kg (N) ha^{-1} over almost all of Europe, and more than 4 kg (N) ha−1 in central areas.

Figure 7 provides similar results for NH_x deposition. The results of the climate and climate+boundary-conditions simulations are rather similar in magnitude to the equivalent results for NOy species, although climate change seems to increase NHx deposition in northern and eastern regions to a greater extent than NO_y. In broad terms, these climate-related runs seem to reflect the pattern of rainfall change to some extent. The most dramatic difference, though, is with Fig. 7d, which shows that future emissions will substantially increase NH_x deposition in large parts of Europe.



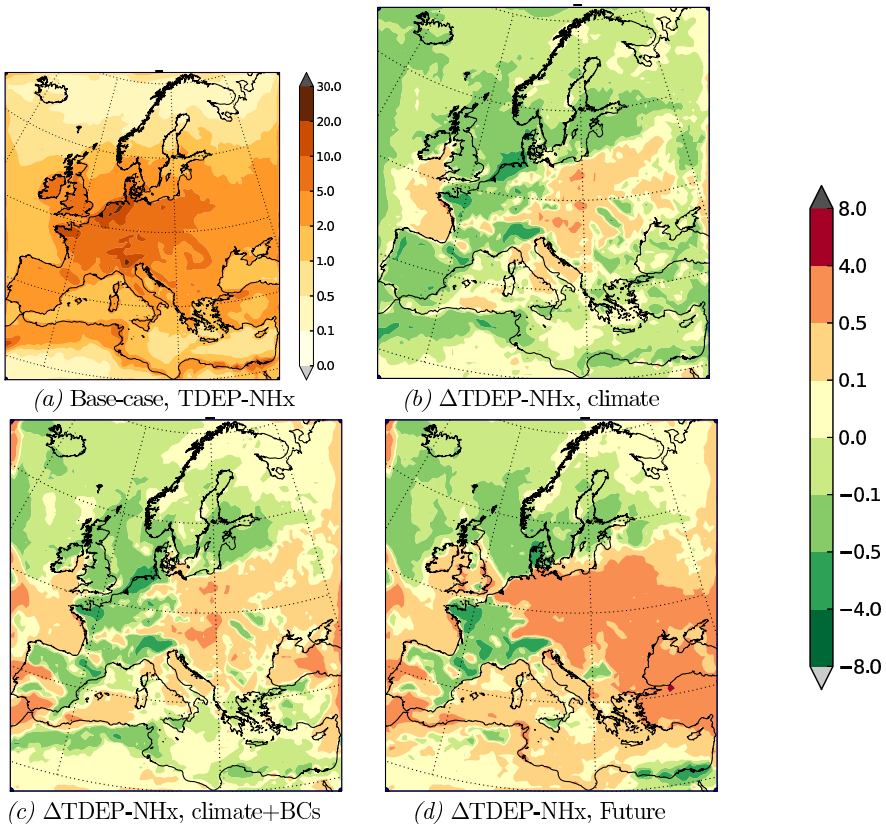
(a) Base-case, TDEP-NOy

(b) ΔTDEP-NOy, climate

(c) ΔTDEP-NOy, climate+BCs

(d) ΔTDEP-NOy, Future

Figure 6: Results from the 4-model ensemble, for (a) base-case deposition of NOy (TDEP-NOy); and changes in TDEP-NOy (rightmost legend) resutling from (b) 2050s climate (E05-M50-BC2), (c) 2050s climate and boundary conditions (E05-M50-BC3), (d) 2050s emissions, climate and boundary conditions (E50-M50-BC3). Units kg (N) ha^{-1}.



(a) Base-case, TDEP-NHx *(b)* ΔTDEP-NHx, climate

(c) ΔTDEP-NHx, climate+BCs *(d)* ΔTDEP-NHx, Future

Figure 7: Same as Fig. 6 but for reduced nitrogen, NH_x.

4.2.2 N-dep - sensitivity to meteorology and emissions

The study of Simpson et al. (2014c) used three different regional CTMs, all driven by one regional climate model. In Engardt and Langner (2013), one of these CTMs, MATCH, was also used to study the impact of using three different climate models. This study used emissions from the RCP4.5 scenario, but again, the modelling systems were evaluated by comparing average modelled precipitation, deposition and concentrations over a 20-year period with observations collected around the year 2000.

It can be noted that the magnitude and distribution of changes in Nr deposition over Europe is sensitive to the climate projection that is used. Engardt and Langner (2013) compared three different climate projections (including the one used above) using the MATCH model and found changes due to climate change by 2050 of less than ± 1 kg(N) ha for both NH_y and NH_x. These changes are comparable to the ensemble mean changes presented here. Hedegaard et al. (2013) reported a general reduction in the Nr deposition over Europe above 0.2 kg(N) ha due to climate change in the period 1990 to 2090 using the hemispheric DEHM model. This could be compared to the case with changing BCs and changing climate in this study, which gives an increase in central/southern Europe for NH_y and a more widespread increase for NH_x. These differences in results are, however, small enough to be explained by differences in the climate projection used. Engardt and Langner (2013) also reported changes in Nr deposition due to emission changes until 2050 using the RCP4.5 scenario. The reductions in deposition are comparable to those reported here for NH_y, but for NH_x the distribution of the changes are different, primarily due to differences in the emission data.

Engardt and Langner (2013) concluded that if future emissions follow the pathway of the RCP4.5 scenario, Europe can expect significantly lower deposition of sulphur and oxidised nitrogen in 2050 compared to 2000. For reduced nitrogen, large areas of western Europe will receive considerably more deposition in 2050 than in 2000, due to feedback of decreased sulphur concentrations on the atmospheric turnover time of reduced nitrogen. Domain averaged reductions of total deposition from 2000 to 2050 are 63, 41 and 0.9% for sulphur, oxidised- and reduced nitrogen, respectively. Climate change results in decreased wet deposition of sulphur and reduced nitrogen leading to increased atmospheric turnover time of these species. Climate and emission changes lead to decreased atmospheric turnover times of reduced nitrogen but increased atmospheric turnover times of sulphur and oxidised nitrogen.

4.2.3 N-dep. - changes in NH_x partitioning

Results presented so far have dealt with groups of either oxidised or reduced depositions of Nr compounds. Figure 12 illustrates changes for particular compounds, from one model (EMEP). The oxidised compounds NO, NO_2 and nitrate all show relatively straightforward reductions, as expected from the emissions change. PAN is also re-

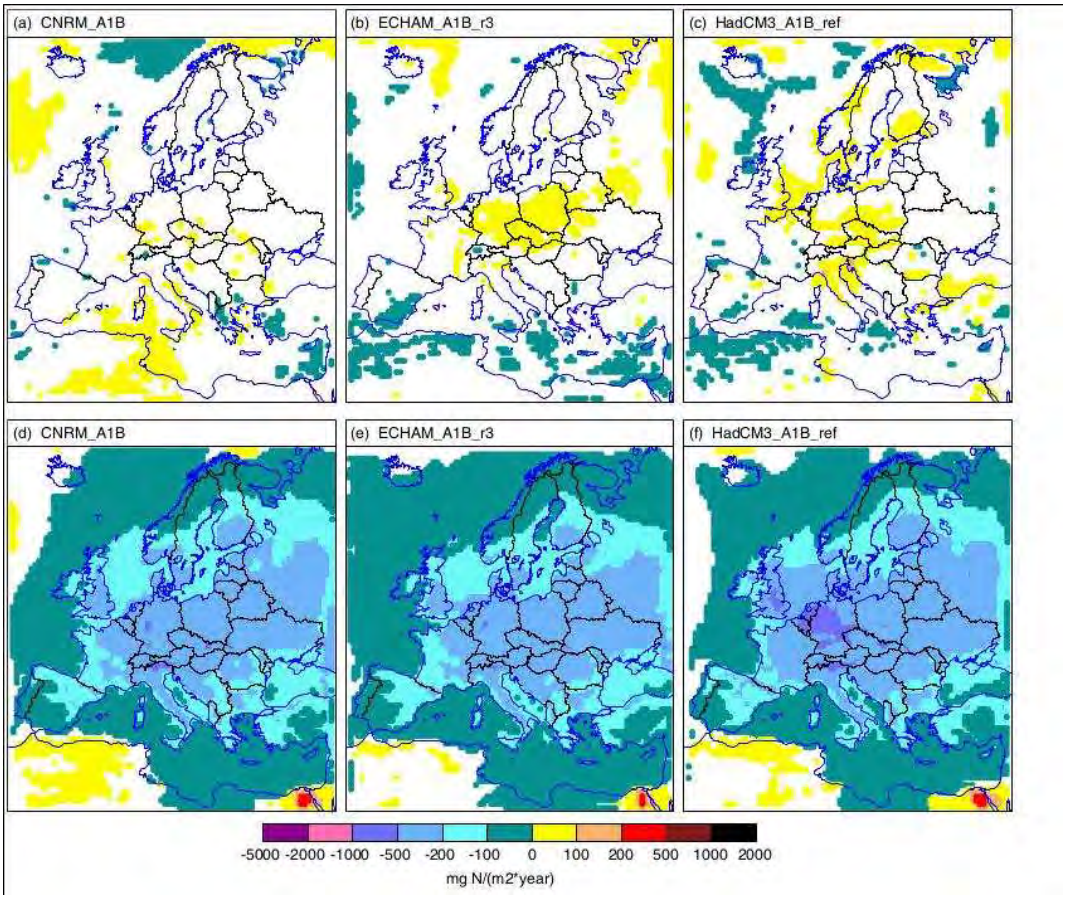


Figure 8: Change in total deposition of oxidised nitrogen from 1990—2009 to 2040—2059. Top: Only changing climate. Bottom: Changing climate and changing emissions. Only changes significant at the $p = 0.05$ level are shown. Engardt and Langner (2013).

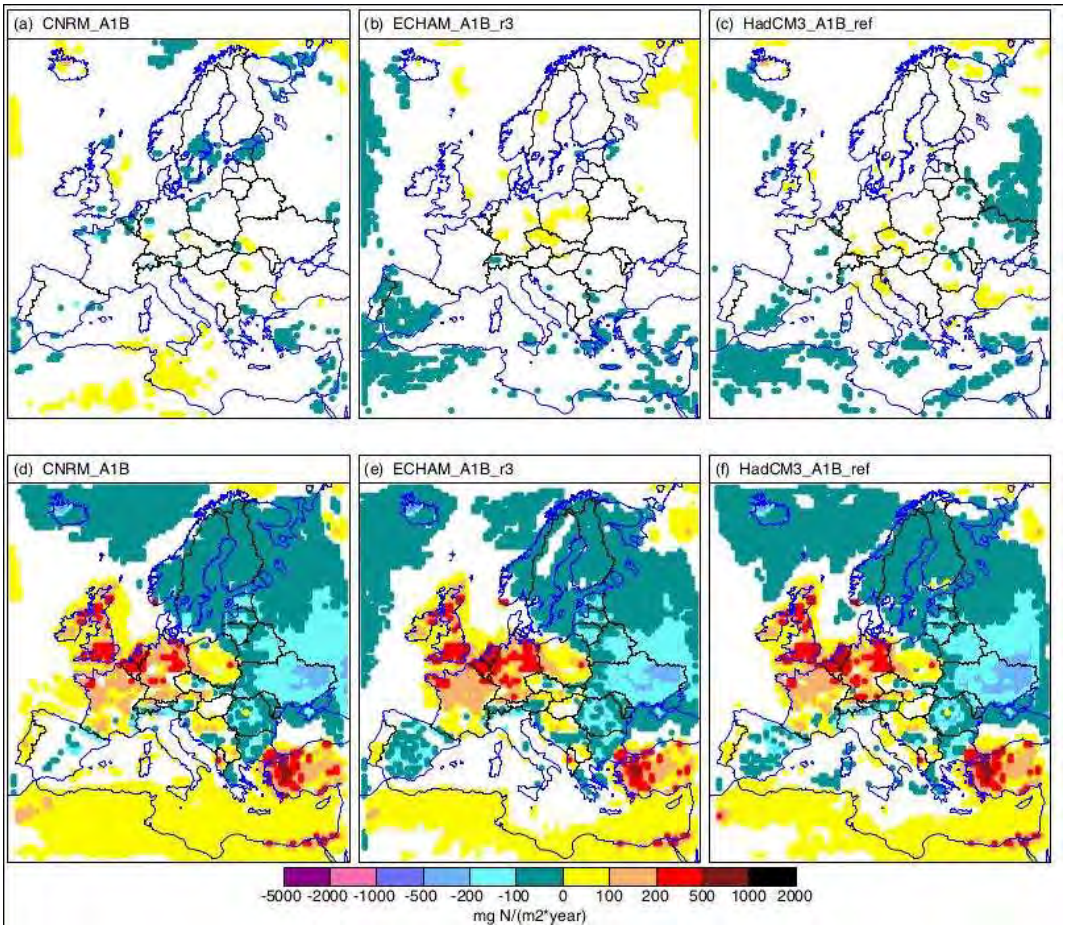


Figure 9: As Fig. 8, but for changes in total deposition of reduced nitrogen. Engardt and Langner (2013).

duced, but not to the same extent, and PAN also shows more sensitivity to the climate and boundary condition changes than other NO_y species. The most interesting changes are seen for the reduced compounds – with substantial increases in gaseous NH_3 and substantial decreases in particulate ammonium. This effect was also noted by Engardt and Langner (2013) and is caused by the fact that, in the year 2050 scenarios, there is too little sulfate and even too little HNO_3 to react with NH_3. This effect is discussed in more detail in Simpson et al. (2014c) and Engardt and Langner (2013).

4.2.4 Climate experiment - increased NH_3 emissions

As noted above, the Simpson et al. (2014c) ECLAIRE study made a first estimate of the impact of possible NH_3 emission increases over Europe for 'year 2050s' simulations, arising from climate-induced increases in evaporation. They explored the impact of 20% and 30% increases in NH_3, and how these changes affected the exceedance of the critical loads (CL) for nutrient nitrogen across the EU28 area. (Actually, 20 meteorological years were used, centred on 2050, along with emissions for the year 2050; we refer to this as the 2050s scenario below. Similarly we have a 2000s scenario with meteorology centred around the year 2000, but emissions from 2005.)

Figures 10-11 summarises the results of these runs from the EMEP model against those involving scenarios for the 2000s and 2050s. Comparing first the 'traditional' emissions options, with prescribed emissions from the IIASA/GAINS model, the area of exceedance of CLs for the 2050s is substantially lower than for the 2000s situation. Comparison of the runs with 20% and 30% increased NH_3 due to a climate effects shows that even a 30% increase in NH_3 will not bring exceedances back to 2000s levels, but such climate-induced increases cause CL exceedances that are substantially larger than those of the standard 2050 emission scenario. Policy studies and emission inventories in Europe and elsewhere have not taken account of this hidden potential for increases in NH_3 emissions. As noted in Sutton et al. (2013), the approaches used to calculate and report NH_3 emissions for both CTM modelling and policy assessments need complete revision to cope with this new paradigm.

4.2.5 N-deposition - issues and challenges

There are of course many issues with the modelling of ammonia exchange, with clear model limitations associated with the lack of bidirectional exchange in these CTMs (Bash et al. 2013, Flechard et al. 2013, Wichink-Kruit et al. 2010).

With regard to emissions control strategies, the increased NH_3 deposition noted above implies that local control measures might become more effective. On the other hand, Engardt and Langner (2013) also estimated longer lifetimes of S and NH_y compounds in the future, thus increasing the international transport of some particles. Wichink-Kruit et al. (2012) also showed that inclusion of bidirectional exchange increases the

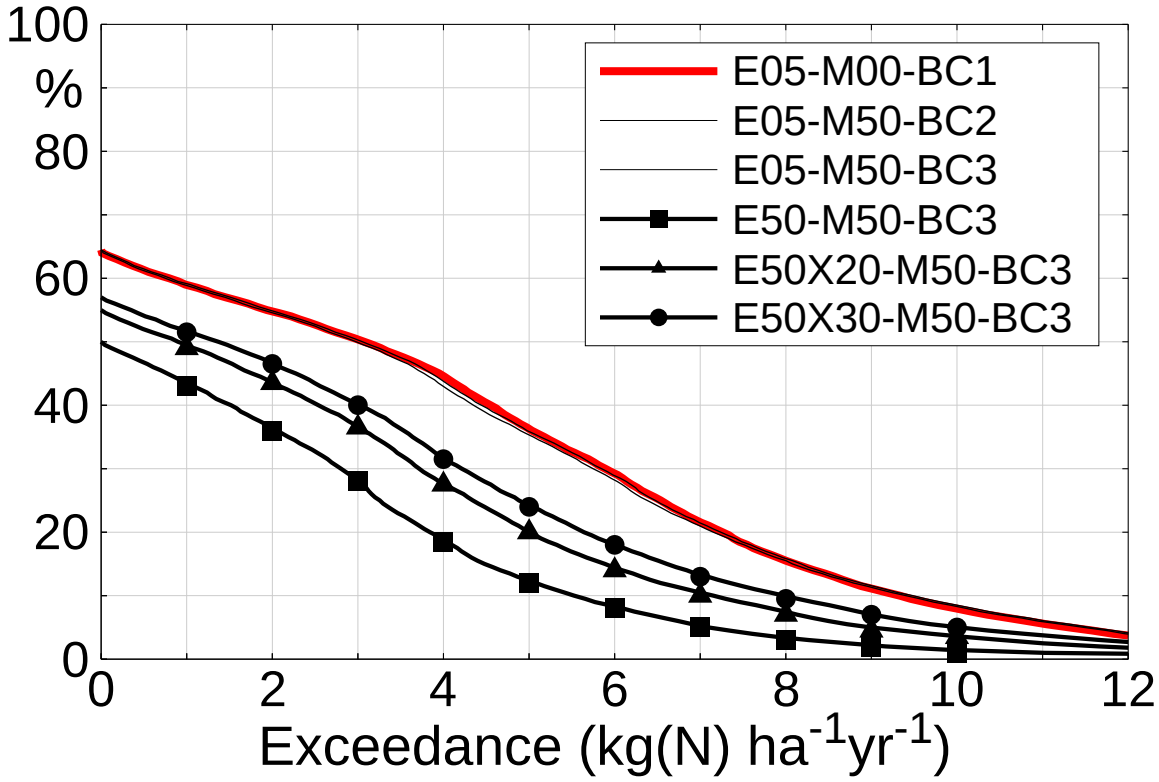


Figure 10: Percentage of $EU28^{+}$ area exceeding the Critical Levels for eutrophying Nitrogen. The red line (E05-M00-BC1) represents a year 2000 base-case and the E50-M50-BC3 scenario represents current estimates of 2050 emissions with 2050s meteorology and boundary conditions. The E50X20 and E50X30 scenarios illustrate calculations with 20% and 30% extra NH_3 emission due to climate-induced evaporation. From Simpson et al. (2014c), where details of the calculations can be found.

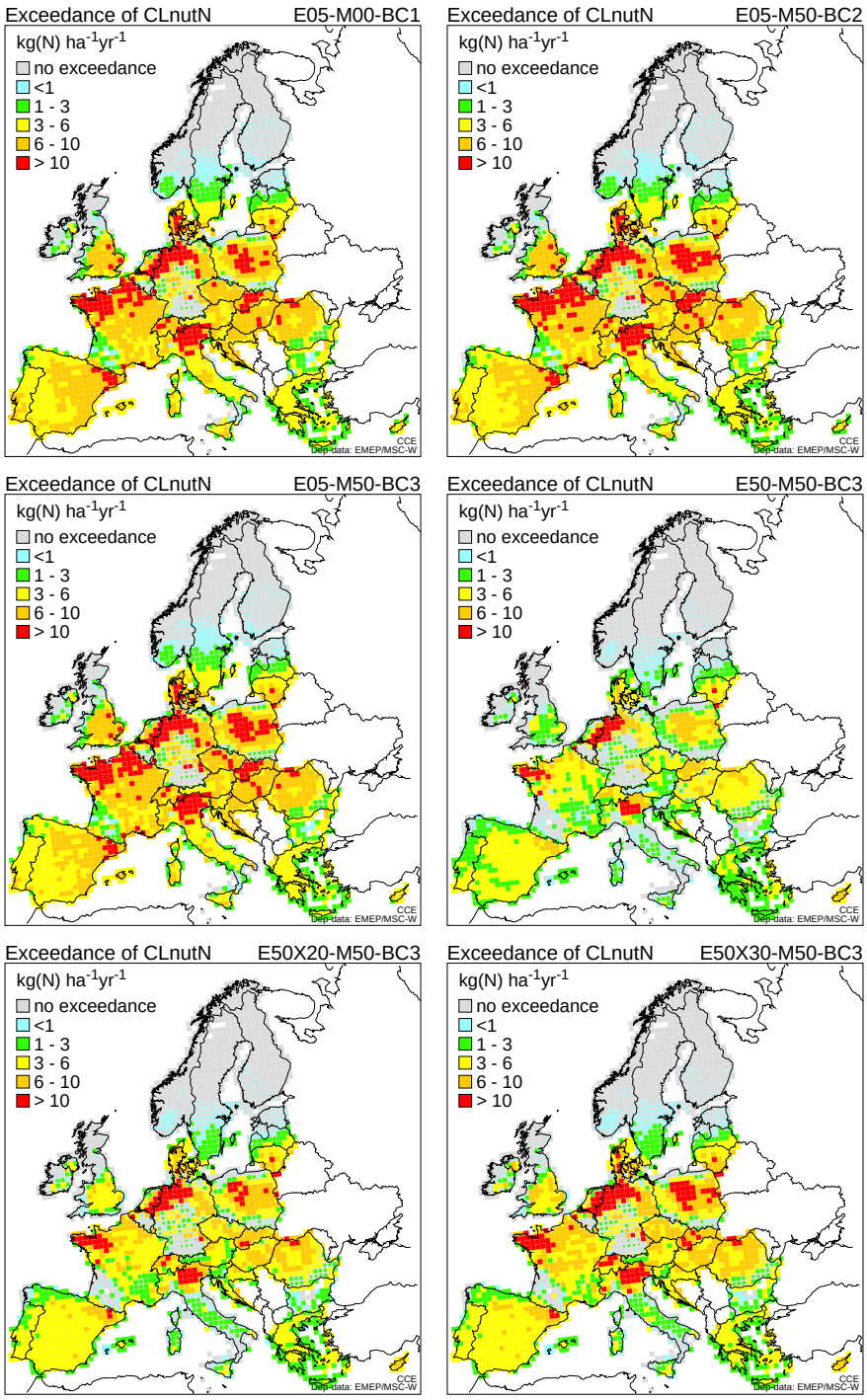


Figure 11: Exceedances of the critical loads for nutrient nitrogen ($CL_{nut}(N)$) in the $EU28^{+}$ region, EMEP MSC-W model, for the six scenarios. (See Fig. 6 for explanation of scenario labels,and Simpson et al. 2014c for details)

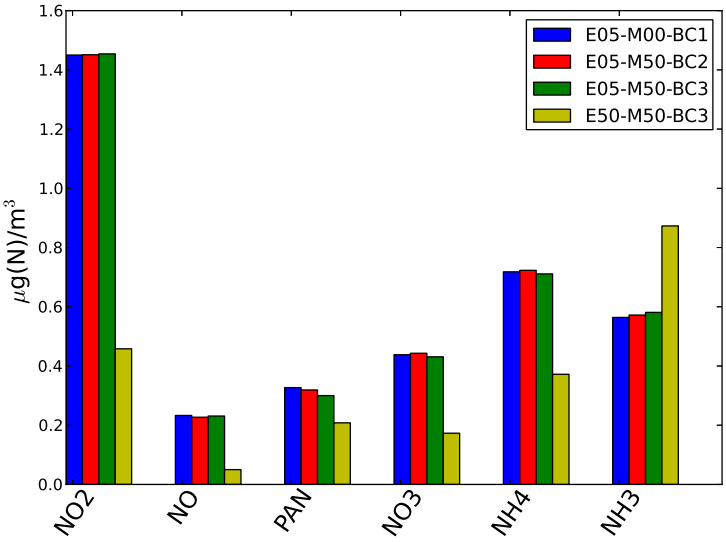


Figure 12: Calculated concentrations of major Nr species from the EMEP MSC-W model for four scenarios. EU28^{+} region. See Fig. 6 for explanation of scenario labels.

transport distance of NH_x, which would affect any predictions of Nr deposition and CL exceedance. Indeed, the complexities of bidirectional exchange have been noted in many papers (e.g. Sutton et al. 1995, Nemitz and Sutton 2004, Fowler et al. 2009, Massad et al. 2010, Flechard et al. 2013), and some CTMs have attempted to include such exchange (e.g. Wichink-Kruit et al. 2010, Bash et al. 2013). However, such modelling is limited by many factors, including process uncertainties (Massad et al. 2010, Flechard et al. 2013), problems of sub-grid heterogeneity (e.g. Loubet et al. 2001, 2009) and lack of necessary and accurate input data.

Still, the overriding conclusion of these papers is probably robust: reducing future deposition of Nr in Europe is mainly dependent upon the way in which future NH_3 emissions develop. The new recognition that climate change may influence emissions much more than currently accounted for in official inventories makes it even more important that methods to deal with NH_3 emissions are improved.

4.3 Other climate impacts

Climate change will have numerous impacts that are difficult to quantify. Here we summarise the results of model studies and/or literature reviews conducted in connection with the ECLAIRE project.

4.3.1 Biogenic volatile organic compounds

Globally, emissions of biogenic volatile organic compounds (BVOC) far exceed anthropogenic emissions (Guenther et al. 2012). In Europe, BVOC emissions still play an important role for ozone production (Simpson 1995, Simpson et al. 1999), and for secondary organic aerosol (Kanakidou et al. 2000, Simpson et al. 2007, Bergström et al. 2012, Zhang et al. 2013). The roles of BVOC and climate for future O_3 and SOA formation are in clear need of thorough evaluation.

The roles of BVOC and climate for future O_3 and biogenic secondary organic aerosol (BSOA) formation are still unclear though. Climate change may well increase vegetation and foliar growth in many areas, especially in the boreal and temperate regions (e.g. Lathiere et al. 2005, Morales et al. 2007, Ahlstrom et al. 2012). These changes, and the direct temperature effects, may promote increases in BVOC emissions in future (Lathiere et al. 2005, Arneth et al. 2011), and this increase has indeed been assumed in most modelling studies to date. However, increasing CO_2 can inhibit isoprene metabolism, and some studies suggest that higher CO_2 levels will reduce BVOC emission rates (e.g. Arneth et al. 2007, Wilkinson et al. 2009, Possell and Hewit 2011). The overall effect of increased CO_2 on BVOC emission rates is still unclear, even with regard to the sign of such changes.

One further problem concerns the chemistry of isoprene degradation. In a recent study, Squire et al. (2015) compared a number of chemical mechanisms, and concluded that the choice of reduced isoprene mechanism may alter both the magnitude and sign of the ozone response. Here again, more measurements and laboratory studies are needed to validate these reduced mechanisms especially under high-volatile-organic-compound, low-NOx conditions (*ibid*). There is a strong need for new experimental studies before we can reliably predict the sign of changes in BVOC emissions and hence changes in ozone and BSOA. For further discussion and some recent references on these issues, see Simpson et al. (2014a).

4.3.2 Impacts of land-cover changes

There is considerable scope for future BVOC emissions levels to be further modified, up or down, by land use changes, and in this respect it is agricultural policy or practice that will be the main determinant of future BVOC changes, not climate-induced biophysical changes in emission rates. For example, tropical land cover changes may affect global background O_3 and SOA as well as local conditions elsewhere due to long-range transport (Arneth et al. 2007, Ashworth et al. 2012, Lathiere et al. 2010, Pyle et al. 2011, Fowler et al. 2011).

Other responses are also complex. For example, the length of growing seasons has already changed significantly over the last 30 years, generally longer and with an earlier start (Menzel and Fabian 1999) (although Hogda et al. (2013) suggested that

in Fennoscandinavia these trends have stopped in the last decade). Such changes are promoted by increasing temperatures, with strong links to changes in atmospheric circulation patterns such as the North Atlantic Oscillation (Cook et al. 2005). These longer growing seasons may have benefits for agriculture in some areas, although limited by drought in others. Longer growing seasons will tend to give increased BVOC emissions, but greater uptake and thus possible damage from ozone over the season.

Finally, O_3 changes might in themselves affect BVOC emissions. Some BVOC species seem to to play a role in protecting vegetation from the toxic effects of O_3, so it might be speculated that BVOC would increase with increasing O_3. However, while some studies do find increased BVOC in response to O_3 (Fares et al. 2006) and Jardine et al. (2012) identified products of within-plant isoprene oxidation, other studies find decreased BVOC emissions in response to O_3 (Velikova et al. 2005, Fares et al. 2006, Calfapietra et al. 2009).

4.3.3 Changes in Arctic shipping

As discussed in Sect. 3.2.1(b) above, we have explored the impact of changes in shipping routes brought about by the retreat of the Arctic ice-sheets. Fig. 13 illustrates the impact of shipping emissions on the important metric, phyto-toxic ozone dose (POD). Changes are concentrated along north-west European coasts, with the largest relative changes seen in northern Norway. Although the changes are not large, e.g. 1 mmole O_3 m^{-2} yr^{-1}, these values are a significant percentage of the base-case POD_1 values calculated for the Arctic ecosystems. These results, and also the results found for nitrogen-deposition, are discussed further in Tuovinen et al. (2013).

4.3.4 Changes in stress-induced emissions

As described in Sect. 3.2.1, and in detail in Bergström et al. (2014), we have investigated the possible impacts of stress-induced emissions (SIE) of BVOC in Europe, based upon a combination of experimental data from the Jülich plant chamber (JPAC), forest statistics (for insect infestation) and the EMEP model. Model calculated $OM_{2.5}$ (organic matter in $PM_{2.5}$) and the relative fraction of BSOA, from the reference case model simulation without explicit SIE from vegetation (Case 0, Bergström et al. 2014), for the summer half-year (Apr–Sep) 2007, are shown in Fig. 14. The modelled BSOA is low in most of Europe. The relative contribution of BSOA to modelled regional background $OM_{2.5}$ is below 20 % except in parts of Northern Europe (parts of Sweden, Finland, the Baltic states, Russia, Belarus etc.) and some BVOC-emission hotspots. (The simulated absolute BSOA concentrations are below 0.6 μg m^{-3}, except in the south-eastern Mediterranean region, parts of Russia, and some smaller high-BVOC-emission areas.)

Based on estimates of current levels of infestation and the JPAC aerosol yields we have estimated the potential increase of $OM_{2.5}$ due to the much higher degree of infes-

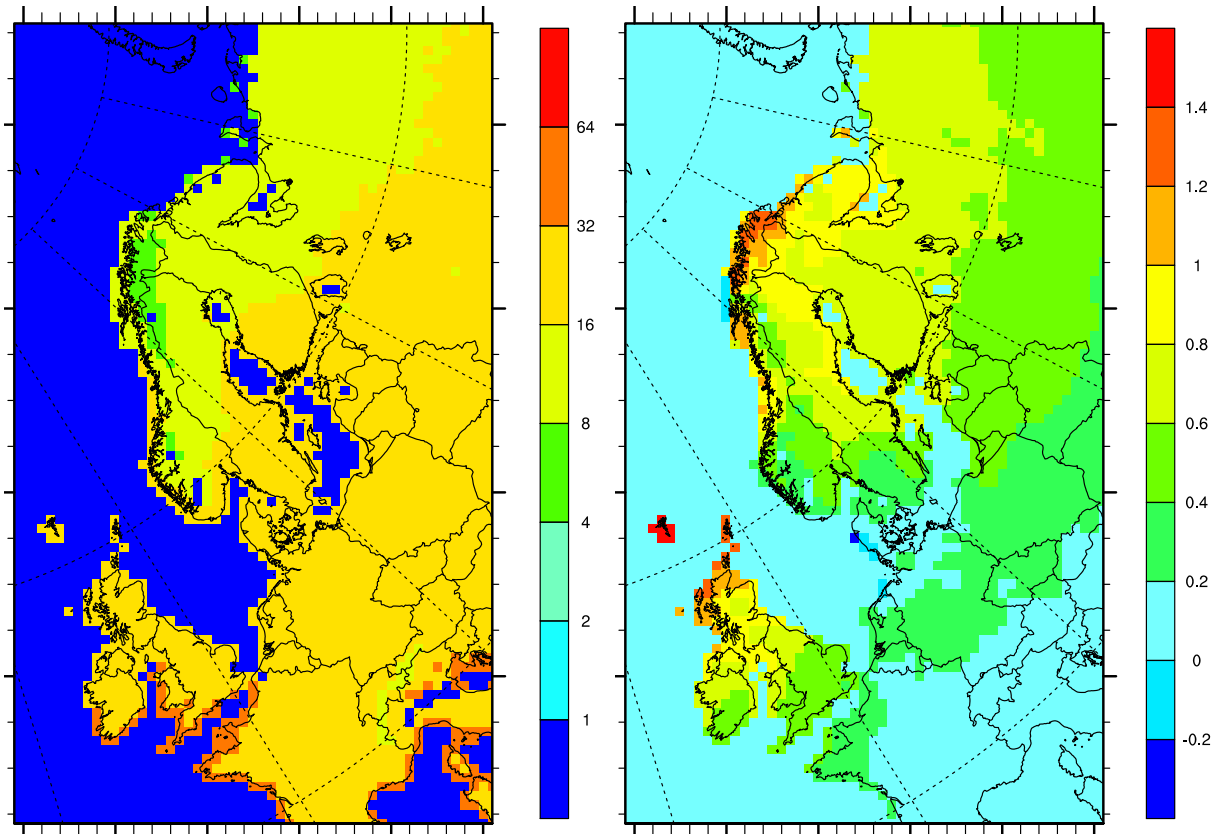
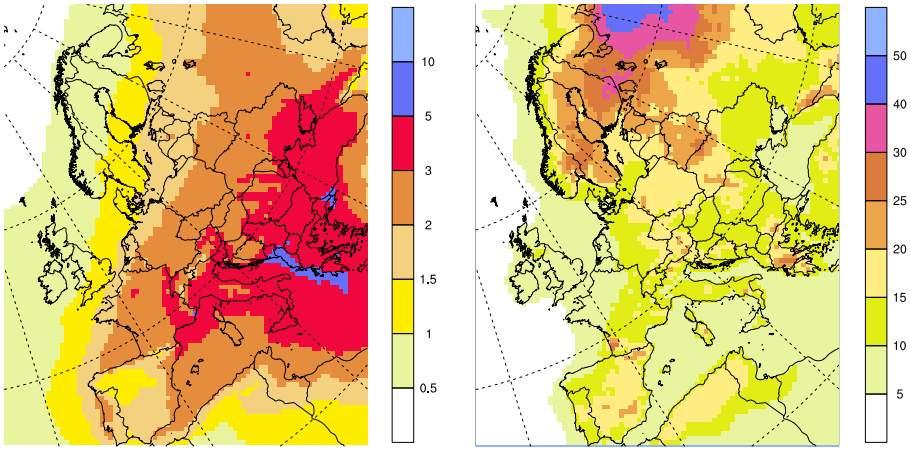


Figure 13: Calculated POD_1 for coniferous forest in 2030 (left), and changes brought about by increased Arctic shipping emissions, including diversion routes (right), units: mmole O_3 m^{-2} yr^{-1}. Calculations with EMEP MSC-W model, see Tuovinen et al. (2013).



(a) Total $OM_{2.5}$ (b) BSOA-fraction (%)

Figure 14: Model calculated 6-month mean (Apr–Sep) concentrations of **(a)** total Organic Matter in $PM_{2.5}$ ($OM_{2.5}$) (Unit: $\mu g\ m^{-3}$). **(b)** Fraction of biogenic secondary organic aerosol (BSOA) (% of total $OM_{2.5}$) for the reference scenario without explicit biotic stress emissions (Case 0).

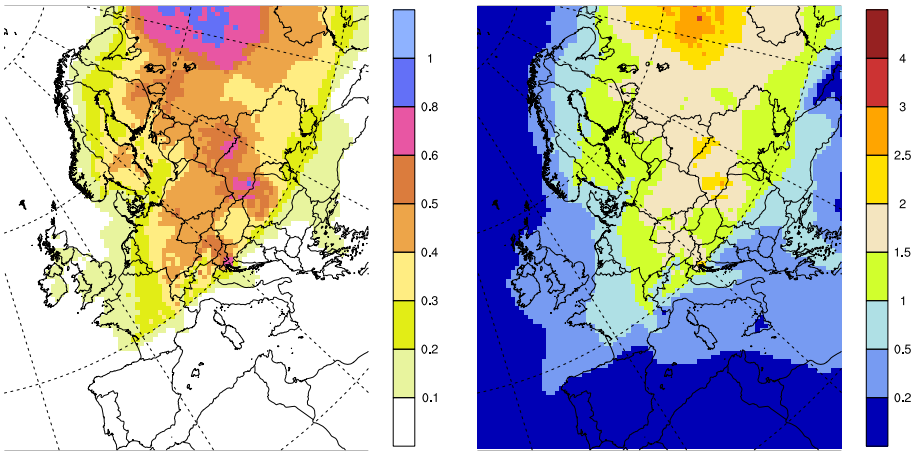
tation assumed in the two future scenarios (denoted Case 1F and 2F in Bergström et al. 2014).

Figure 15 shows the potential increase of $OM_{2.5}$ for these future, high infestation, scenarios. As seen here, increases in SOA can be rather dramatic for these 'maximum impact' scenarios. Such results indicate that SIE-SOA is or could potentially become an important source of regional background $PM_{2.5}$ in large parts of central/eastern Europe; at least if the scenarios with high SIE emissions and large SOA yields are found to be realistic. It has to be stressed that although we have used the best available laboratory data on biotic SIE applicable to northern and central European forests, the magnitude of the impact even for present-day conditions is very uncertain and needs to be constrained by further laboratory, field and modelling studies. Still, this study shows that stress-induced emissions should not be ignored, and Bergström et al. (2014) make suggestions for future field work to help quantify their impacts.

4.4 Final remarks

The ECLAIRE model studies have generated a large number of useful results on the impacts of future changes in emissions and climate for air pollution metrics across the EU. Key results have been presented here, but the publications listed in Sect. 7 provide many more details.

A general conclusion arising from many of these subject-areas is the need for more experimental data to underpin the assumed levels of emissions and for understanding



(a) Case 1F - $OM_{2.5}$ increase (b) Case 2F - $OM_{2.5}$ increase

Figure 15: Potential increase of $OM_{2.5}$ (6-month mean, Apr–Sep) from increased biotic stress-induced emissions (SIE) in two "maximum" impact scenarios, compared to the corresponding current-situation model calculated concentrations. The concentration-difference fields illustrate potential effects of a changed climate that the northern/central European forests have not had time to adapt to; **(a)** Difference in $OM_{2.5}$ between Case 1F and Case 1 (SIE with only sesquiterpenes (SQT)), **(b)** Case 2F – Case 2 (SIE of both SQT and methyl salicylate). Unit: μg m^{-3}. Note: Different colour scales.

the relevant processes. Emissions from biogenic sources (BVOC, NO from soils, etc.) in particular are very uncertain, even for current simulations (e.g. Simpson et al. 1999, Langner et al. 2012b).

Recent model studies suggest low or modest impact of climate change meteorological factors on future ozone and/or deposition of reactive nitrogen over Europe (Langner et al. 2012a,b, Colette et al. 2013, Simpson et al. 2014c), with the most dramatic exception being the hitherto neglected possibility of increased NH_3 emissions (Sect. 3.2.1). One important meteorological caveat to this conclusion is that future climate may be more extreme than used in these studies. As discussed in e.g. Schär et al. (2004) and Solberg et al. (2008), summers with extreme drought and severe ozone episodes such as those seen the year 2003 might well become more frequent in future, with potentially very dramatic consequences for ozone levels. Future developments are also critically dependent on hemispheric background concentration changes (Wild et al. 2012, Simpson et al. 2014a).

For reactive nitrogen (Nr), a number of studies (including those mentioned in this document) have now shown that, on the European scale at least, and for time scales up to 2050, the main driver of future Nr deposition changes is the specified emission change, rather than any effect of climate change on chemistry or deposition patterns. (Engardt and Langner 2013, Simpson et al. 2014c). These studies suggest significant reductions in oxidised N concentrations and deposition, but slightly increasing levels of reduced N deposition, consistent with the prescribed emission changes.

With regard to both O_3 and Nr (especially NH_3) changes, it is increasingly apparent that the underlying emissions data are uncertain in ways that can significantly impact assessments of future air quality and ecosystem-impacts. Still, some main lines of emission control seem clear; reduction of methane and other O_3 precursors at hemispheric scale is needed to manage future O_3 levels, and further action on NH_3 emissions will be needed in order to reduce harmful levels of N-deposition in Europe.

5 Milestones achieved

MS29 Initial ensemble runs for current conditions

MS31 Future scenario data-sets ready

6 Deviations and reasons

None

7 Publications

Bergström, R.B., Carbonaceous Aerosol in Europe. Out of the woods and into the blue?, PhD Thesis, Dept. Chemistry % Molec. Biology, University of Gothenburg, Sweden, ISBN 978-91-628- 9505-1, Sept. 2015

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8 Meetings

Participation in ECLAIRE annual meetings, plus several WP7 meetings to organise and analyse the model ensemble results.

9 List of Documents/Annexes:

n.a.

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