

1 **A model study on changes of European and Swiss particulate**
2 **matter, ozone and nitrogen deposition between 1990 and**
3 **2020 due to the revised Gothenburg protocol**

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2 **Abstract**

3 We report a study of changes in air quality due to emission reductions using the
4 chemical transport model CAMx. The model domain includes all of Europe with a nested
5 domain over Switzerland. The model simulations were performed **with emissions** for
6 1990 (the reference year for the Gothenburg Protocol), 2005 (the reference year for the
7 revised Gothenburg Protocol), 2006 (for model validation) and 2020 (the target year for
8 the revised Gothenburg Protocol) using three emission scenarios prepared by
9 IIASA/GAINS. Changes in ozone, particulate matter, and nitrogen deposition are the
10 central theme of the study.

11 **The modelled relative changes in the annual average PM_{2.5} concentrations between**
12 **1990 and 2005 look reasonable based on various PM₁₀ and PM_{2.5} observations in the**
13 **past. The results obtained in this study suggest that annual mean concentrations of**
14 **PM_{2.5} decreased by about 20 – 50% in Europe.** Simulations using the baseline scenario
15 (BL 2020) suggest that PM_{2.5} concentrations in 2020 will be about 30% lower than those
16 in 2005. The largest predicted decrease in PM_{2.5}, based on the MTR (Maximum
17 Technically Feasible Reduction) scenario, was about 60% and was located mainly in the
18 eastern part of Europe.

19 In the case of ozone, both model results and measurements show an increase in the
20 mean ozone mixing ratios between 1990 and 2005. The observations, however, suggest
21 a larger increase, indicating the importance of background ozone levels. Although
22 emission reductions caused a decrease in peak ozone values, **average** ozone levels in
23 polluted regions increased due to reduced titration with nitric oxide (NO). This caused a
24 change in the frequency distribution of ozone. Model simulations using emission
25 scenarios for 2020 suggest that annual average ozone mixing ratios will continue to
26 increase. Changes in the levels of the damage indicators AOT40 for forests and
27 SOMO35 are reported as well.

28 The model results suggest that nitrogen deposition decreased by 10 - 30% in the
29 eastern part of Europe since 1990, while it increased by about 20% in the Iberian
30 Peninsula. The decrease is mainly due to the deposition of oxidized nitrogen species,
31 whereas deposition of reduced nitrogen compounds increased. In Switzerland, nitrogen
32 deposition is larger in the northern part of the Alps where ammonia emissions are the
33 highest. Applying the baseline scenario, we found that the deposition of oxidized
34 nitrogen compounds will have decreased by a further 40% by 2020, whereas deposition
35 of reduced species will continue to increase. This will lead to a 10 - 20% decrease in the
36 total nitrogen deposition in most of the model domain, with a 10% increase in the
37 eastern part of Europe.

38 **1 Introduction**

39 One of Europe's main environmental concerns is air pollution. Current policy in this
40 respect focuses mainly on ozone (O₃) and particulate matter (PM₁₀ and PM_{2.5}, particles
41 smaller than 10 and 2.5 μm in aerodynamic diameter, respectively). The policies were
42 especially successful for particulate matter with substantial decreases in the past
43 (Barmopoulos et al., 2012) whereas **observed annual mean** ozone concentrations did
44 not significantly change (Wilson et al., 2012). Ozone and in spite of the improvements
45 also PM₁₀ levels often exceed the ambient air quality standards in Europe, which are:
46 120 μg m⁻³ maximum daily 8-hour mean for O₃ and 50 μg m⁻³ daily mean for PM₁₀
47 (Engler et al., 2012; Hettelingh et al., 2013).

1 In an earlier study, we reported the effects of numerous regulations enforced in Europe
2 since 1985 and predicted the effects of the Gothenburg protocol targets for 2010 on
3 ozone (Andreani-Aksoyoglu et al., 2008). Our results suggested that the decrease in
4 local ozone production due to emission reductions was partly or completely offset by a
5 simultaneous increase in the background ozone, indicating that further development of
6 background ozone concentrations in Europe would be very important for tropospheric
7 ozone levels. The concentration of ozone in Europe is affected by emissions from other
8 continents due to its long atmospheric lifetime. While ozone precursor emissions in
9 Europe and in North America have decreased significantly since the 1980s, NO_x (NO
10 and NO₂) emissions have increased dramatically in Asia in the last decade (Zhang et al.,
11 2010). Also changes of the flux of stratospheric ozone may be important (Ordonez et al.,
12 2007).

13 The major indicators used to characterize ozone damage are AOT40 (Accumulated
14 dose of ozone Over the Threshold of 40 ppb) and SOMO35 (Sum of Ozone Means Over
15 35 ppb). AOT40 is an indicator of damage to vegetation (Ashmore and Wilson, 1992).
16 The UNECE has set the critical level for forest damage at 10000 µg m⁻³ h. SOMO35, on
17 the other hand, was recommended by WHO to be used for health impact assessment
18 (Amann et al., 2008). It is defined as the yearly sum of the daily maximum of 8-hour
19 running average over 35 ppb. It is expected that the strong efforts that have been made
20 to reduce ozone precursor emissions in Europe should decrease the levels of both of
21 these indicators.

22 In 2007, the Convention on Long-Range Transboundary Air Pollution initiated the
23 revision of its Gothenburg multi-pollutant/multi-effect protocol (UNECE, 1999). Fine
24 particulate matter (PM_{2.5}) was included in the revised protocol for which the target year
25 is 2020. In the same context, the EMEP Centre for Integrated Assessment Modelling
26 (CIAM) at IIASA prepared various emission control scenarios for cost-effective
27 improvements to air quality in Europe in 2020 using the GAINS (Greenhouse gas – Air
28 pollution Interactions and Synergies) model.

29 These developments provided the motivation for this study, in which we used the CAMx
30 air quality model to investigate the changes in European and Swiss air quality between
31 1990 and 2005 and to predict the effects of various emission reduction scenarios on air
32 quality in 2020 in Europe and in Switzerland. In this paper, we discuss the changes in
33 annual average concentrations of particulate matter, ozone, ozone damage indicators
34 AOT40 and SOMO35 as well as changes in nitrogen deposition between 1990 and
35 2020.

36 **2 Method**

37 **2.1 Model setup**

38 The models used in this study are the Comprehensive Air quality Model with extensions,
39 CAMx, Version 5.40 (<http://www.camx.com>) and the Weather Research & Forecasting
40 Model (WRF-ARW), Version 3.2.1 (<http://wrf-model.org/index.php>). The coarse model
41 domain covered all of Europe with a horizontal resolution of 0.250° x 0.125°. A second,
42 nested domain with three times higher resolution (0.083° x 0.0417°) covered
43 Switzerland. The meteorological fields were calculated for 2006 and used for all
44 emission scenarios (see Table 1). We used 6-hour ECMWF data (<http://www.ecmwf.int/>)
45 to provide initial and boundary conditions for the WRF model. There were 31 terrain-
46 following σ- layers up to 100 hPa in WRF, of which 14 were used in CAMx. The lowest
47 CAMx layer was 20 m above ground and the model top corresponded to about 7000 m
48 above sea level. The initial and boundary concentrations for the coarse domain were

1 obtained from the MOZART global model data for 2006 (Horowitz et al., 2003). The
2 boundary conditions were kept constant for all future emission scenarios. The choice of
3 background ozone is crucial for air quality simulations and for predicting the effect of
4 emission reductions (Andreani-Aksoyoglu et al., 2008). A recent analysis of various
5 ozone observational data in Europe showed that **annual mean** ozone concentration
6 increased in the 1980s and 1990s (Logan et al., 2012). Summer ozone levels started
7 decreasing slowly in the 2000s, but there were no significant changes in other seasons.
8 Logan et al. (2012) indicated the inconsistencies in various data sets leading to different
9 trends. It is therefore difficult to choose a realistic background ozone values for the
10 model domain and for the period of interest. In view of this, we kept the background
11 ozone levels constant for simulations in the period between 2005 and 2020 (Wilson et
12 al., 2012; Logan et al., 2012). For the 1990 simulation, background ozone mixing ratios
13 were set about 5 ppb lower **in each model layer for all boundaries and for each hour,**
14 **based on the positive trend in the 1990s reported by the long-term measurement studies**
15 **(Cui et al., 2011, Logan et al., 2012).** Seasonal variation was also taken into account.
16 Photolysis rates were calculated using the TUV photolysis pre-processor (Madronich,
17 2002). The required ozone column densities were extracted from TOMS data
18 (NASA/GSFC, 2005). Dry deposition of gases in CAMx is based on the resistance
19 model of Zhang et al. (2003). For surface deposition of particles, CAMx includes
20 diffusion, impaction and/or gravitational settling. CAMx uses separate scavenging
21 models for gases and aerosols to calculate wet deposition. The gas-phase mechanism
22 used in this study was CB05 (Carbon Bond Mechanism 5) (Yarwood et al., 2005).

23 We performed CAMx simulations for 1990 (the reference year for the Gothenburg
24 Protocol), 2005 (the reference year for the revised Gothenburg Protocol), 2006 (for
25 model validation) and 2020 (the target year for the revised Gothenburg Protocol) with
26 different emission scenarios as described in the next section. For all of these
27 simulations, however, the 2006 meteorology was used.

28 In order to determine the changes in pollutant concentrations in the past (since 1990)
29 and in the future (until 2020), the annual average ozone and PM_{2.5} for each scenario
30 were compared with those in the reference year 2005. Dry and wet deposition of
31 nitrogen species were summed over the entire year for each scenario and compared
32 with 2005. AOT40 for forests was calculated for the daytime hours (8:00 AM – 8:00 PM)
33 from the beginning of April until the end of September in all scenarios. SOMO35
34 was calculated by summing the daily maximum of the 8-hour running average over 35 ppb
35 for the whole year.

36 **2.2 Emissions**

37 We prepared six emission scenarios (see Table 1). The gridded (0.125° x 0.0625°)
38 TNO/MACC data (<http://www.gmes-atmosphere.eu/>) for 2006 were used as the basic
39 anthropogenic emission inventory (Denier van der Gon et al., 2010). The European
40 values **in both domains** were replaced by the high-resolution Swiss emission data for
41 grid cells located within the Swiss national boundary (INFRAS, 2010; Heldstab and
42 Wuethrich, 2006; Kropf, 2001; Heldstab et al., 2003; Schneider, 2007; Kupper et al.,
43 2010). The output of the CAMx simulation using the meteorological data and emissions
44 for 2006 was used for model evaluation.

45 The TNO/MACC emission inventory was scaled with the annual data from The Centre
46 for Emission Inventories and Projections (CEIP) of the European Monitoring and
47 Evaluation Programme (EMEP), and The International Institute for Applied Systems
48 Analysis/Greenhouse Gas and Air Pollution Interactions and Synergies Model
49 (IIASA/GAINS) was used to prepare gridded, hourly emissions for 1990, 2005 and 2020.

1 CEIP manages a database of annual emissions before 2009 based on data submitted
2 by participating countries ([http://www.ceip.at/webdab-emission-database/emissions-as-](http://www.ceip.at/webdab-emission-database/emissions-as-used-in-emep-models/)
3 [used-in-emep-models/](http://www.ceip.at/webdab-emission-database/emissions-as-used-in-emep-models/)). IIASA uses the GAINS model to predict national emission
4 projections until 2020 on the basis of the assumed economic development of each
5 country (<http://gains.iiasa.ac.at/gains/EUN/index.login>). The emissions for a given
6 emission scenario were calculated by scaling the raw data using annual emission totals
7 for each country, species and SNAP (Selected Nomenclature for Air Pollution) category.
8 For scenarios 1990 and 2005, the annual emissions for each SNAP category were
9 extracted from the EMEP/CEIP database, which contains the historic emissions
10 submitted by the EMEP member states. Data for PM_{2.5} and PM₁₀ are only available for
11 2000 and later, so the 1990 data were calculated from the 2005 data, using GAINS
12 simulations. For the 2020 scenarios, the 2005 data were scaled to 2020 using GAINS
13 CIAM4 / 2011 simulations. The baseline scenario (BL) assumes that emissions will
14 continue to be regulated by the current legislation. The MTRF (maximum technically
15 feasible reductions) scenario uses the lowest expected emissions for most of the source
16 categories. The MID scenario uses moderate emission reductions that are between
17 those of BL and MTRF.

18 The relative changes in emissions between 2005 and 2020 for various scenarios in
19 Switzerland (CH) and an average of 27 European countries (EU) are shown in Fig. 1.
20 The emissions for the revised Gothenburg Protocol (2020 rev) are included in the figure,
21 although there was no GAINS scenario available at the time of this work. After its
22 publication, however, the reductions specified by the revised Gothenburg Protocol were
23 found to be very close to those for the baseline (2020 BL). In general, emission
24 reductions increase with increasing ambition, i.e. they are lowest in BL and highest in
25 MTRF. The relative changes for Switzerland are usually lower than those for the EU
26 countries (due to the larger emission reductions that had previously been imposed in
27 Switzerland) except for PM_{2.5} for which all reductions are comparable.

28 The biogenic emissions were calculated using the method described in Andreani-
29 Aksoyoglu and Keller (1995) for each CAMx domain using the temperature and
30 shortwave irradiance from the WRF output, the global USGS land use data and the
31 GlobCover 2006 inventory. For each European country the deciduous and coniferous
32 forest fractions were split into tree species according to the method reported in Simpson
33 et al. (1999). Inside the Swiss border the global data were replaced by data based on
34 land use statistics (100 m resolution) and by forest data (1 km resolution) taken from the
35 national forest inventory (Mahrer and Vollenweider, 1983). Currently this biogenic
36 emission inventory is being improved by extending the number of species and trees,
37 using the best available land use data and including updated temperature and irradiance
38 dependencies (Oderbolz et al., 2013).

39 **3 Results and discussion**

40 **3.1 Model evaluation**

41 The results from the lowest layer of both model domains were compared with various
42 observations in 2006. The comparison of modelled meteorological parameters with
43 observations is given in Fig. S1 of the Supplement. In general the agreement between
44 measurements and model results was good with high correlation coefficients (0.76-0.98)
45 and low mean bias error, MBE (0.00023 for specific humidity, -1.13 for air temperature,
46 0.57 for wind speed). These values fulfil the desired accuracy suggested by Cox et al.
47 (1998) which is 2 °C for temperature, 1 m s⁻¹ and 2.5 m s⁻¹ for wind speeds < 10 m s⁻¹
48 and > 10 m s⁻¹, respectively.

1 The predicted concentrations of ozone and PM_{2.5} in the European domain were
2 compared with measurements at the rural background stations of the European Air
3 quality database AirBase (<http://acm.eionet.europa.eu/databases/airbase/>). Table 2
4 gives the overall statistical parameters for all of the year 2006 (only those stations below
5 500 m asl and with 80% of data available were used for the statistical analysis and these
6 sites cover a large part of Europe between -8.8 and 27.7 degrees from west to east, and
7 between 37.3 and 60.5 from south to north). Mean annual O₃ and PM_{2.5} are slightly
8 over- and under-estimated, respectively. In the case of ozone, although the temporal
9 variation is captured, the maximum concentrations in summer are underestimated as
10 reported by other studies (Fig. 2, upper panel). For instance, evaluation of several air
11 quality models for 2006 within the Air Quality Model Evaluation International Initiative
12 (AQMEII) showed that the models have a predominant tendency to underestimate (in
13 some cases significantly) the peak daily mixing ratio in summertime as well as to
14 overestimate night time mixing ratios with the exception of central Europe (Solazzo et
15 al., 2012a). Time series show that the model reproduced the temporal variation of PM_{2.5}
16 quite well, except for January-February when unusually high concentrations were
17 recorded in Europe (Fig. 2, lower panel). The underestimation of PM_{2.5} is partly due to
18 the severe meteorological conditions prevailing during that exceptionally cold inversion
19 period. It is also possible, however, that the contribution of wood burning to emissions
20 was underestimated. As reported by Solazzo et al (2012b) none of the models used in
21 AQMEII study consistently matched PM_{2.5} observations for all locations throughout the
22 entire year. Results of the AQMEII study suggest that while the models do relatively well
23 in simulating the inorganic aerosol species, large uncertainty remains in the simulation
24 of other components such as secondary organic aerosols and unspiciated PM_{2.5}.
25 Elimination of the sources of PM bias in the models is still challenging.

26 The frequency distributions of modelled and measured ozone and PM₁₀ values in 2006
27 are shown in Fig. 3. Comparison of the model results from the nested domain with
28 measurements in Switzerland suggests that the model performance is better at rural
29 sites. At the rural site, Chaumont, for example, the shape of the measured and modelled
30 distributions of O₃ is similar: both have the highest number of points approximately in the
31 middle of the graph. At the urban site, Zurich, on the other hand, the discrepancy
32 between the measurements and model results at low concentrations are clearly seen.
33 Because of the finite model resolution, NO_x concentrations are usually underestimated
34 at urban sites, where local emissions are relatively high and variable. This leads to
35 overestimation of ozone at night and in the morning. In addition to the model horizontal
36 resolution, its representation of the inversion layer at night and the mixing layer during
37 the day also plays an important role in the prediction of pollutant concentrations. In the
38 case of PM₁₀, the measured and modelled concentrations also show a very similar
39 distribution at the rural site Chaumont, indicating very good model performance,
40 whereas the high concentrations at the urban background site, Zurich, were
41 underestimated.

42 The modelled concentrations of particulate species in the nested domain were
43 compared with AMS (Aerosol Mass Spectrometer) measurements of particulate nitrate,
44 sulphate, ammonium, and organic aerosols (Lanz et al., 2010) in June 2006 at Payerne
45 (Fig. 4). Although the model calculates PM_{2.5} and the AMS measures only particles
46 smaller than 1 μm, the results may be compared, because the difference between PM₁
47 and PM_{2.5} measurements is very small as shown in Aksoyoglu et al. (2011). Elemental
48 carbon (EC) data are obtained from Aethalometer equivalent black carbon (BC)
49 measurements. The model performance for aerosol components in this study is
50 significantly better than that in our previous study, which used the MM5 meteorological
51 model with an earlier CAMx version (Aksoyoglu et al., 2011). The modelling of organic

1 aerosols, however, is still quite challenging, mainly due to limited knowledge about the
2 processes involved in secondary organic aerosol (SOA) formation. The CAMx model
3 used in this study includes an SOA model based on a theory of the gas-particle
4 partitioning of various precursors, such as anthropogenic and biogenic VOC species.
5 The oligomerization process, which leads to an increase in aerosol concentrations, is
6 also included. The model performance for organic aerosols is reasonably good for
7 relatively low concentrations. It becomes worse, however, when the formation of
8 secondary organic aerosols increases. The total modelled PM_{2.5} (sum of inorganic and
9 organic species) concentrations match the observations quite well, with one exception
10 on 14-16 June, which was due to underestimation of increased levels of organic
11 aerosols. Models that take into account the volatility distribution and atmospheric aging
12 of OA might give more realistic results (Bergström et al., 2012).

13 **3.2 Particulate matter**

14 The modelled annual average PM_{2.5} concentrations vary between 5 and 40 µg m⁻³ for
15 the reference year 2005 in Europe (Fig. S2 of the Supplement). Our results suggest that
16 PM_{2.5} concentrations decreased significantly in Europe between 1990 and 2005. The
17 relative changes range from -20% in Scandinavia to more than -60% in the eastern part
18 of the domain; they are between -40 and -45% in central Europe (Fig. 5). There have
19 been long-term measurements of PM₁₀ throughout Europe since the late nineties, but
20 measurements of PM_{2.5} at some European sites are available only after 2000 (Tørseth
21 et al., 2012). The available data, however, show average changes between 2000 and
22 2009 of -18% and -27% for PM₁₀ and PM_{2.5}, respectively. Recently Cusack et al. (2012)
23 reported that PM_{2.5} concentrations in various parts of Europe decreased by 7-49%
24 between 2002 and 2010. The average trends of -0.4 µg m⁻³ y⁻¹ for PM₁₀ and PM_{2.5} at
25 several European sites reported by Barmpadimos et al. (2012) correspond to a
26 decrease of about 40-45% between 1998 and 2010. The PM₁₀ measurements at various
27 sites in Switzerland indicate a large decrease (20-56%) between 1991 and 2008
28 (Barmpadimos et al., 2011). This supports our model results (see Fig. 5), because most
29 of the change in PM₁₀ was in the PM_{2.5} fraction (Barmpadimos et al., 2012). **Combining**
30 **the PM10 trends from Barmpadimos et al. (2011) with the modelled PM10 for 1990,**
31 **2005 and 2006 in this study at four stations shows the interannual variability in the**
32 **observed trends together with the emission-induced changes modelled in this study (Fig.**
33 **S3 of the Supplement).**

34 Changes in particulate matter concentrations result not only from changes in primary PM
35 emissions, but also from changes in precursor emissions such as nitrogen oxides (NO_x),
36 non-methane volatile organic compounds (NMVOC), sulphur dioxide (SO₂) and
37 ammonia (NH₃). As seen in Fig. 1, the European emission reduction of NH₃ for the 2020
38 scenarios is much smaller than the reduction of other precursor emissions. We
39 compared the predicted annual mean PM_{2.5} concentration for 2020 with that of the
40 reference year (2005) and found that a considerable reduction in PM_{2.5} would be
41 obtained in Europe under the BL scenario (Fig. 6). The decrease in PM_{2.5} would vary
42 from 30-40% in central Europe, up to 50% at some local polluted areas in Eastern
43 Europe. The predicted reductions using the MID and MTRF scenarios are about 50-
44 60%, with the largest changes being predicted in the Balkan countries (only MTRF is
45 shown in the lower panel of Fig. 6.) In Switzerland the predicted reductions in PM_{2.5} are
46 30 and 40%, using BL and MTRF scenarios, respectively.

47 **3.3 Ozone**

48 The average ozone mixing ratios for the reference year (2005) are shown in Fig. S4 of
49 the Supplement. The model results - **based on the assumption that the background**

1 ozone levels increased by 5 ppb between 1990 and 2005 - suggest that the average
2 annual ozone increased between 1990 and 2005 in a large part of Europe in spite of the
3 large reductions of precursor emissions (Fig. 7). The increase in ozone was predicted
4 especially for England, the Benelux countries and around Ukraine. In an earlier
5 sensitivity study, we reported that these areas have VOC-limited regimes for ozone
6 production (Aksoyoglu et al., 2012); a reduction of precursor emissions leads to an
7 increase in ozone levels in such regions.

8 The impact of the choice of boundary conditions for 1990 on the results shown in Fig. 7
9 was investigated by a sensitivity test in which we increased the background ozone by 5
10 ppb. The results suggested that the change in the annual mean ozone (Fig. 7) would be
11 1-2 ppb lower in central Europe. Long-term observations however, show a significant
12 positive trend in the annual mean ozone especially between 1990 and 2000 (Cui et al.,
13 2011, Logan et al., 2012). A positive trend of 0.32 ppb/y reported by Cui et al. (2011) for
14 annual mean ozone measured at Jungfrauoch between 1990 and 2008 supports our
15 choice of 5 ppb increase in the background ozone between 1990 and 2005.

16 The predicted O₃ increase is about 1-2 ppb (3-9%) over the Swiss Plateau, whereas
17 observations indicate larger changes between 10% at rural areas and 40-50% at urban
18 sites (Table 3). On the other hand, modelled peak ozone values are lower in 2005 than
19 in 1990 (see Table 4). Measurements also show a decrease in peak ozone levels except
20 in Basel (suburban) and Zurich (urban). The simulation of ozone trends is quite
21 challenging, as has been shown in other model studies (Colette et al., 2011; Wilson et
22 al., 2012). As seen in the example for Zurich, the frequency distribution of ozone mixing
23 ratios in 1990 and 2005 is clearly different (Fig. 8). The most frequent ozone levels are
24 shifted toward higher levels in 2005 and the change is larger in the measurements.

25 The relative change in annual average ozone mixing ratios between 2005 and 2020 is
26 shown in Figs. 9a and 9b for the European and Swiss domains, respectively. For both
27 BL and MTRF scenarios, the predicted decrease is small (< 4 ppb, < 10%) in central
28 Europe, whereas ozone is expected to increase further in England and the Netherlands,
29 due to reduced titration with NO. On the other hand, no further increase is expected
30 around Ukraine between 2005 and 2020 as predicted for the period between 1990 and
31 2005 (see Fig. 7, upper panel). A decrease of about 5-7% is predicted over the Alpine
32 regions and the southern part of the Alps, while ozone is predicted to increase by about
33 1 ppb (3%) at urban sites (Fig. 9b). One has to keep in mind, however, that the
34 background ozone levels in these simulations were assumed to stay constant between
35 2005 and 2020, based on the study of Logan et al. (2012).

36 3.4 AOT40 and SOMO35

37 The modelled AOT40 and SOMO35 results for the reference year (2005) are shown in
38 Fig. 10. AOT40 values range between 5–30 ppm h, with elevated levels in southern
39 Europe. The SOMO35 values show a similar spatial distribution, lying between 1000 -
40 5000 ppb d. In Switzerland, the modelled AOT40 is 10-15 ppm h and 20-30 ppm h in the
41 north and south, respectively (Fig. 11, upper panel). We predicted SOMO35 values
42 between 2400-2800 ppb d for northern Switzerland and 4000-4800 ppb d for the
43 southern part of the Alps (Fig. 11, lower panel). These results match very well the
44 AOT40 and SOMO35 values derived from measurements in 2005 (Table 3). Compared
45 to an EMEP model study which reported average AOT40 and SOMO35 of 35.1 ppm h
46 and 5303 ppb d, respectively for Switzerland in 2005 (Gauss et al., 2012), our results
47 are lower and in better agreement with the measurements.

48 A comparison of simulations for 1990 and 2005 suggests that AOT40 and SOMO35
49 have decreased in Switzerland since 1990 (Fig. 12), although average annual ozone

1 mixing ratios increased (Fig. 7). This indicates that peak ozone values decreased due to
2 emission reductions, as shown in Table 4. Although measurements also show a
3 decrease at rural sites, they suggest that AOT40 and SOMO35 increased significantly at
4 urban sites (Table 3). This discrepancy between the model results and observations
5 indicates the sensitivity of these indicator parameters to threshold values.
6 Overestimation of ozone concentrations by regional models at night in polluted urban
7 areas is a common problem. This alone however, cannot be responsible for the
8 discrepancy between measured and modelled AOT40, because AOT40 is the sum of
9 ozone concentrations above 40 ppb and is calculated only during the daytime. The
10 difference between the modelled and measured frequency distributions of ozone mixing
11 ratios above 30-40 ppb is relevant to an understanding of the changes in AOT40 and
12 SOMO35 (Fig. 8). The discrepancy between the modelled and measured relative
13 change in damage indicators is most likely due to the background ozone levels, but this
14 needs further analysis.

15 Assuming constant background ozone after 2005, AOT40 and SOMO35 were predicted
16 to decrease substantially by 2020 (Figs. S5-S8 of the Supplement). One must keep in
17 mind, however, that these indicators depend strongly on the threshold values, which
18 might be affected by the background ozone and its evolution in the future.

19 **3.5 Nitrogen deposition**

20 The atmospheric deposition of pollutants raises serious concerns for ecosystems. In
21 Switzerland, emissions of air pollutants such as sulphur dioxide and nitrogen oxides
22 have been substantially reduced in the last couple of decades. While sulphur emissions
23 are now stabilized at lower levels than in the past, nitrogen oxide emissions are still
24 rather high. In this section, therefore, we focus on nitrogen deposition.

25 In general, the main nitrogen sources are emissions of nitrogen oxides from combustion
26 processes and ammonia from agricultural activities. The deposition of atmospheric
27 nitrogen species constitutes a major nutrient input to the biosphere, which enhances
28 forest growth. Despite this, increased nitrogen input into terrestrial ecosystems
29 represents a potential threat to forests. Enhanced nitrogen deposition can cause soil
30 acidification, eutrophication and nutrient imbalances, causing a reduction in biodiversity.
31 The deposition of atmospheric nitrogen compounds occurs via dry and wet processes.
32 NO₂, NH₃, nitric acid (HNO₃), and nitrous acid (HONO) are the most important
33 contributors to nitrogen dry deposition. Nitrogen wet deposition results from the
34 scavenging of atmospheric N constituents.

35 The predicted annual deposition of total nitrogen in Europe varies between 5 and 45 kg
36 N ha⁻¹ y⁻¹ in 2006 (Fig. 13, upper panel) and it is mainly dominated by dry deposition
37 (Fig. S9 of the Supplement). Dry deposition is generally largest over regions with large
38 ambient NH₃ concentrations over the Netherlands and Belgium as also reported in the
39 literature (Flechard et al., 2011). We also predict high nitrogen dry deposition around the
40 Po Valley in northern Italy. The modelled total nitrogen deposition varies between 10
41 and 45 kg N ha⁻¹ y⁻¹ in northern Switzerland (Fig. 13, lower panel). Elevated levels can
42 also be seen in the south (10-20 kg N ha⁻¹ y⁻¹). On the other hand, they are lower at
43 high-altitude sites (about 5 kg N ha⁻¹ y⁻¹). These numbers are in the same range as
44 those based on measurements at various locations in Switzerland (Schmitt et al., 2005).
45 In a recent study, Roth et al. (2013) reported an average N deposition on 122 plots in
46 Switzerland of 18.3 kg N ha⁻¹ y⁻¹ for the year 2007.

47 Deposition of oxidized and reduced nitrogen species for 2006 is shown in Figs. 14 and
48 15 for the European and Swiss domains, respectively. The calculated deposition of
49 reduced nitrogen compounds is higher than that of oxidized species. Deposition of

1 reduced N species - especially NH_3 dry deposition - is high in central Switzerland, where
2 the ammonia emissions are the highest in the country. The combination of high
3 ammonia concentrations and land use favourable for dry deposition leads to the highest
4 deposition of ammonia in the nested domain in a few grid cells in central Switzerland.

5 A comparison of the simulations for 1990 and 2005 suggests that nitrogen deposition
6 decreased mainly in the eastern part of the European domain, while it increased in the
7 Iberian Peninsula (Fig. 16, upper panel). In Switzerland, the decrease in nitrogen
8 deposition was mainly over the Alpine regions and the southern part of the country (Fig.
9 16, lower panel). The decrease in nitrogen deposition is mainly related to the oxidized
10 fraction, due to large reductions in NO_x emissions that occurred in the past.

11 The future simulations assuming the BL 2020 scenario suggest that the oxidized
12 nitrogen deposition will decrease further by about 40% in all of Europe until 2020,
13 whereas deposition of reduced nitrogen compounds will continue to increase by about
14 20% especially in the southern and eastern part of Europe (Fig. 17). This would lead to
15 a 10 - 20% decrease in the total nitrogen deposition in most of the model domain, with a
16 10% increase in the eastern part of Europe.

17 4 Conclusions

18 The results presented in this study give an overview on predicted nitrogen deposition
19 and the concentrations of ozone and particulate matter in Europe for the past, the
20 present, and different emission scenarios for 2020. They also indicate the importance of
21 the background ozone concentrations in Europe for use in calculating AOT40 and
22 SOMO35 trends.

23 The modelled relative decreases of the annual average $\text{PM}_{2.5}$ concentrations between
24 1990 and 2005 were 20% and 50% in Europe. Although $\text{PM}_{2.5}$ observations were not
25 available for the entire time period, PM_{10} and more recent $\text{PM}_{2.5}$ measurements support
26 the modelled trends. In order to have a quantitative evaluation however, it would be
27 advantageous to run and evaluate the model for several years when significant
28 measurement data is available. Among the three Gothenburg scenarios for 2020 (BL,
29 MID and MTRF), the BL scenario is the closest to the recently revised Gothenburg
30 Protocol. Our results show that the application of emission reductions according to the
31 BL scenario would lead to a significant decrease of $\text{PM}_{2.5}$ (~30%) in 2020 compared to
32 2005. The largest predicted decrease in $\text{PM}_{2.5}$ based on the MTRF scenario was about
33 50%-60%, especially in the eastern part of Europe; although its implementation before
34 2020 is unlikely.

35 Observations show that mean ozone concentrations did not decrease, but actually
36 increased further in certain areas, in spite of large reductions in European emissions
37 between 1990 and 2005. The model predictions also suggest a similar trend, but the
38 predicted increase is lower than the measured one. This indicates the importance of
39 background ozone. We showed that peak ozone values decreased due to emission
40 reductions whereas ozone levels in polluted regions increased due to reduced titration
41 with NO. The modelled damage indicators AOT40 and SOMO35 for 2005 are in the
42 same range as the measurements. The change in these indicator values between 1990
43 and 2005, however, did not match the observations. The model results suggest a
44 significant decrease in the indicator levels since 1990. The observations, on the other
45 hand, indicate a decrease at rural sites, but an increase at urban sites. Since the AOT40
46 and SOMO35 values are very sensitive to the threshold values, the background ozone
47 concentrations might affect the results. We conclude that even though the change in the
48 background ozone used in the model between 1990 and 2005 were adjusted to the
49 recent observations, they might need further revision.

1 We predicted that the annual average ozone values will continue to increase in the
2 future, by applying the three emission scenarios (BL, MID, MTRF) for 2020. Assuming a
3 constant background ozone levels after 2005, AOT40 and SOMO35 were predicted to
4 decrease by large amounts until 2020 with respect to the reference year 2005. These
5 results, however, have high uncertainty.

6 We also analysed the model results for both dry and wet deposition of all oxidized and
7 reduced nitrogen species. The annual deposition of total nitrogen in Europe was
8 predicted to vary between 5 - 45 kg N ha⁻¹ y⁻¹ in 2006 and it was mainly dominated by
9 dry deposition. Dry deposition was generally largest over regions with large ambient NH₃
10 concentrations over the Netherlands, Belgium and the Po Valley. The modelled annual
11 nitrogen deposition is in the same range as those based on measurements. The
12 predicted annual nitrogen deposition in northern Switzerland varied between 10 - 45 kg
13 N ha⁻¹ y⁻¹. Deposition of reduced N species - especially NH₃ dry deposition - is high in
14 central Switzerland, where the ammonia emissions are the highest in the country. The
15 combination of high ammonia concentrations and land use favourable for dry deposition
16 leads to the highest deposition of ammonia in central Switzerland.

17 Our model results suggest that the nitrogen deposition decreased by 10-30% in the
18 eastern part of Europe between 1990 and 2005, whereas it increased in the Iberian
19 Peninsula. Further reductions in emissions until 2020, according to the baseline
20 scenario, would lead to about 40% lower oxidized nitrogen deposition - mainly due to a
21 reduction in the oxidized fraction - while deposition of reduced nitrogen compounds
22 would continue to increase in most of Europe.

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Table 1: Description of emission scenarios

| scenario | Description |
|------------|---|
| 1990 | retrospective analysis |
| 2005 | reference year |
| 2006 | model validation |
| 2020 BL* | baseline scenario |
| 2020 MID* | mid scenario |
| 2020 MTFR* | maximum technically feasible reduction scenario |

- from IIASA/GAINS

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Table 2: Statistical quantities for O₃ and PM_{2.5} using rural background stations in the AirBase dataset for 2006 (model output from the European domain).

| | O ₃ (ppb) | | PM _{2.5} (µg m ⁻³) | |
|-------------------------|----------------------|-------|---|-------|
| | Obs. | model | Obs. | model |
| Mean | 29.5 | 33.5 | 13.5 | 11.6 |
| Standard deviation | 11.2 | 7.9 | 6.4 | 4.9 |
| Minimum | 10.0 | 15.1 | 4.1 | 2.6 |
| Maximum | 70.4 | 53.9 | 63.4 | 41.2 |
| Mean bias | 4.0 | | -1.9 | |
| Correlation coefficient | 0.84 | | 0.43 | |
| Number of stations | 300 | | 19 | |

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2 Table 3: Mean O₃, AOT40 and SOMO35 from measurements at NABEL stations in Switzerland (FOEN).

| Station | type | O ₃ (µg m ⁻³) | | | AOT40 (ppm h) | | | SOMO35 (ppb d) | | |
|-------------|-----------------|--------------------------------------|------|----------|---------------|------|----------|----------------|------|----------|
| | | 1990 | 2005 | % change | 1990 | 2005 | % change | 1990 | 2005 | % change |
| Basel | suburban | 34.0 | 47.4 | +39 | 13.6 | 14.9 | +10 | 2164 | 2752 | +27 |
| Davos | rural, elevated | 65.6 | 70.8 | +8 | 19.8 | 19.4 | -2 | 3817 | 4519 | +18 |
| Duebendorf | suburban | 32.4 | 43.9 | +36 | 15.0 | 15.2 | +1 | 2261 | 2700 | +19 |
| Jungfrauoch | mountain | 70.2 | 74.8 | +7 | 39.8 | 41.6 | +5 | 7014 | 7969 | +14 |
| Lugano | urban | 38.1 | 52.8 | +39 | 23.8 | 30.0 | +26 | 3740 | 4672 | +25 |
| Lägeren | rural | 63.9 | 69.3 | +9 | 22.4 | 17.7 | -21 | 4690 | 3962 | -16 |
| Payerne | rural | 51.6 | 56.1 | +9 | 24.8 | 18.2 | -27 | 4044 | 3350 | -17 |
| Sion | highway | 30.0 | 41.0 | +37 | 12.7 | 11.7 | -8 | 2504 | 2516 | +1 |
| Tänikon | rural | 46.2 | 55.9 | +21 | 19.8 | 16.1 | -19 | 3740 | 3099 | -17 |
| Zurich | urban | 29.3 | 44.8 | +53 | 7.5 | 14.0 | +87 | 1959 | 2469 | +26 |

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 2 Table 4: Measured and modelled peak ozone concentrations at NABEL stations in 1990
 3 and 2005

| Station | type | Measured max O ₃ (µg m ⁻³) | | Modelled max O ₃ (µg m ⁻³) | |
|-------------|-----------------|---|------|---|------|
| | | 1990 | 2005 | 1990 | 2005 |
| Basel | suburban | 200 | 224 | 180 | 145 |
| Davos | rural, elevated | 142 | 136 | 147 | 123 |
| Duebendorf | suburban | 216 | 212 | 214 | 163 |
| Jungfrauoch | mountain | 131 | 130 | 144 | 121 |
| Lägeren | rural | 217 | 205 | 213 | 160 |
| Lugano | urban | 269 | 255 | 235 | 185 |
| Payerne | rural | 196 | 184 | 175 | 133 |
| Sion | highway | 174 | 170 | 138 | 115 |
| Tänikon | rural | 212 | 199 | 210 | 158 |
| Zurich | urban | 190 | 210 | 213 | 161 |

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1 **Figure Captions:**

2 Figure 1: Relative changes (%) in annual emissions of SO₂, NO_x, NMVOC, NH₃ and
3 PM_{2.5} with respect to reference year (2005) for various scenarios in Switzerland (CH)
4 and the European countries (EU) (for definition of scenarios see text).

5 Figure 2: Time series of modelled (CAMx) and measured (AirBase) hourly average O₃
6 (ppb) (upper panel) and daily average PM_{2.5} (µg m⁻³) (lower panel) in 2006 (European
7 domain). Number of measurement sites: 300 for O₃, 19 for PM_{2.5}.

8 Figure 3: Frequency distributions of ozone (upper panel) and PM₁₀ (lower panel) at
9 Chaumont (rural) and Zurich (urban background) in 2006.

10 Figure 4: Measured (red) and modelled (black) hourly concentrations of particulate
11 nitrate, sulphate, ammonium, organic aerosols (OA), elemental carbon (EC) and PM_{2.5}
12 (sum of all species shown above) at Payerne in June 2006. EC was measured by an
13 Aethalometer, the other components by an AMS.

14 Figure 5: Relative changes in annual average PM_{2.5} concentrations over the European
15 (upper panel) and Swiss (lower panel) domains, 2005-1990.

16 Figure 6: Relative changes in annual average PM_{2.5} concentrations over the European
17 domain for two scenarios: BL 2020 - 2005 (upper panel) and MTFR 2020 - 2005 (lower
18 panel).

19 Figure 7: Changes (ppb) in annual average ozone mixing ratios over the European
20 (upper panel) and Swiss (lower panel) domains, 2005 – 1990.

21 Figure 8: Changes in frequency distributions of measured (left) and modelled (right)
22 ozone between 1990 and 2005 in Zurich.

23 Figure 9a: Changes in annual average ozone mixing ratios (ppb) over the European
24 domain, BL 2020- 2005 (upper panel), MTFR 2020- 2005 (lower panel).

25 Figure 9b: Changes in annual average ozone mixing ratios (ppb) over the Swiss domain,
26 BL 2020- 2005 (upper panel), MTFR 2020- 2005 (lower panel).

27 Figure 10: Modelled AOT40 (ppm h) (upper panel) and SOMO35 (ppb d) (lower panel)
28 over the European domain for the reference year (2005).

29 Figure 11: Modelled AOT40 (ppm h) (upper panel) and SOMO35 (ppb d) (lower panel)
30 over the Swiss domain for the reference year (2005).

31 Figure 12: Relative changes in AOT40 (upper panel) and in SOMO35 (lower panel) over
32 the Swiss domain, 2005 – 1990.

33 Figure 13: Total N deposition (kg N ha⁻¹ y⁻¹) over the European (upper panel) and the
34 Swiss (lower panel) domains (2006).

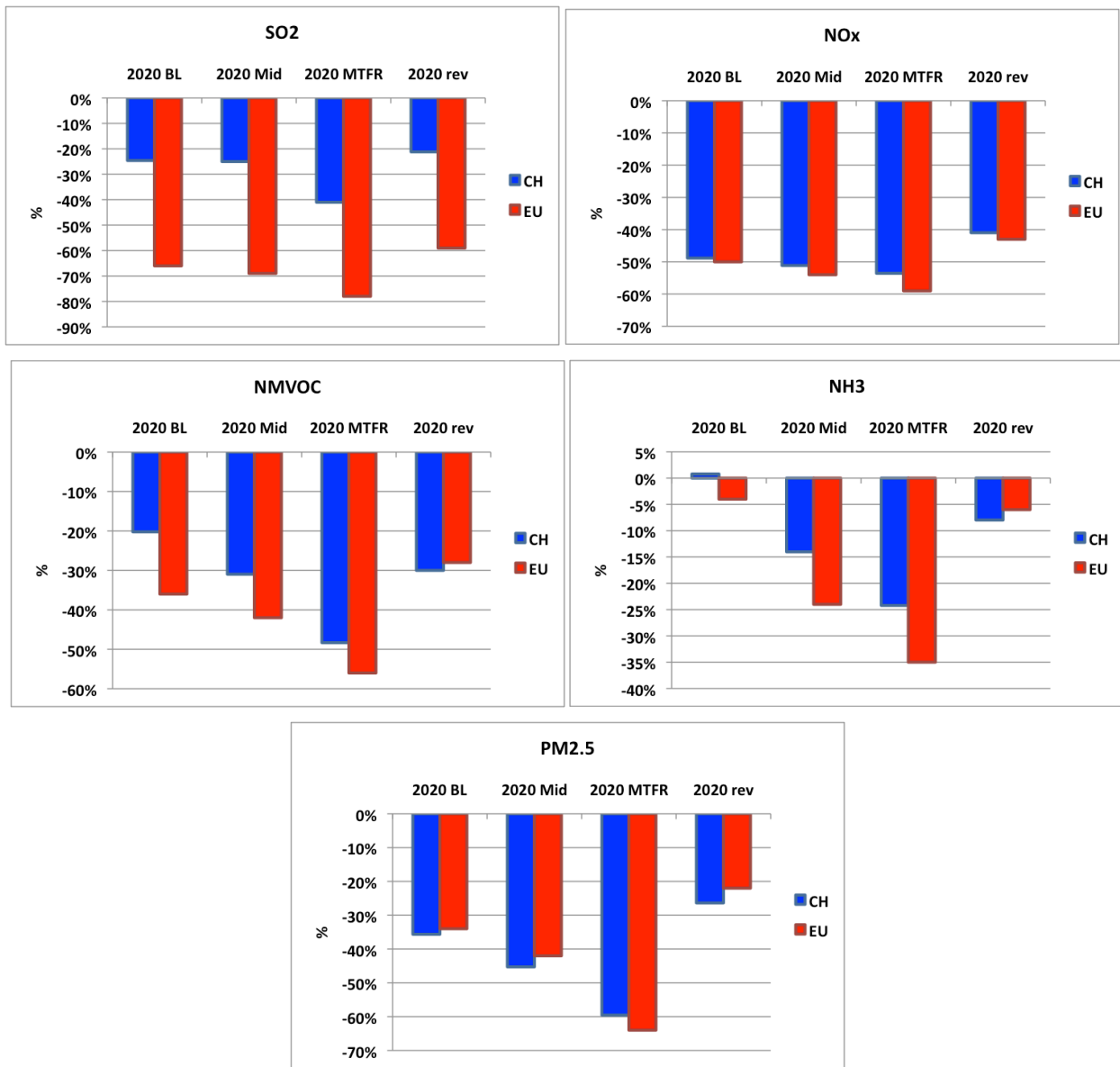
35 Figure 14: Deposition (kg N ha⁻¹y⁻¹) of oxidized (upper panel) and reduced (lower panel)
36 nitrogen compounds over the European domain (2006).

37 Figure 15: Deposition (kg N ha⁻¹y⁻¹) of oxidized (upper panel) and reduced (lower panel)
38 nitrogen compounds over the Swiss domain (2006).

39 Figure 16: Relative changes in nitrogen deposition over the European (upper panel) and
40 the Swiss (lower panel) domains, 2005 – 1990.

41 Figure 17: Relative changes in deposition of oxidized (upper panel) and reduced (lower
42 panel) nitrogen species over the European domain, BL 2020 – 2005.

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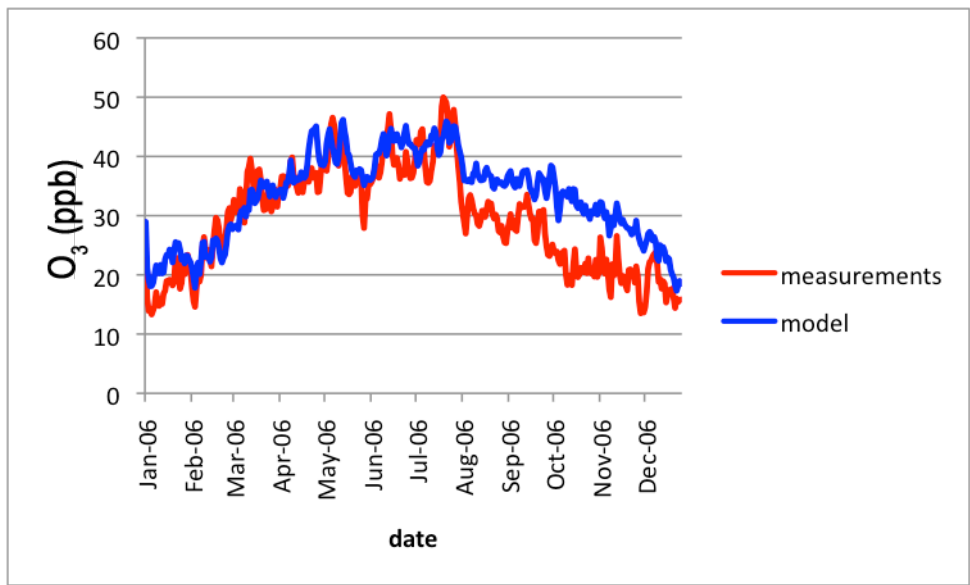
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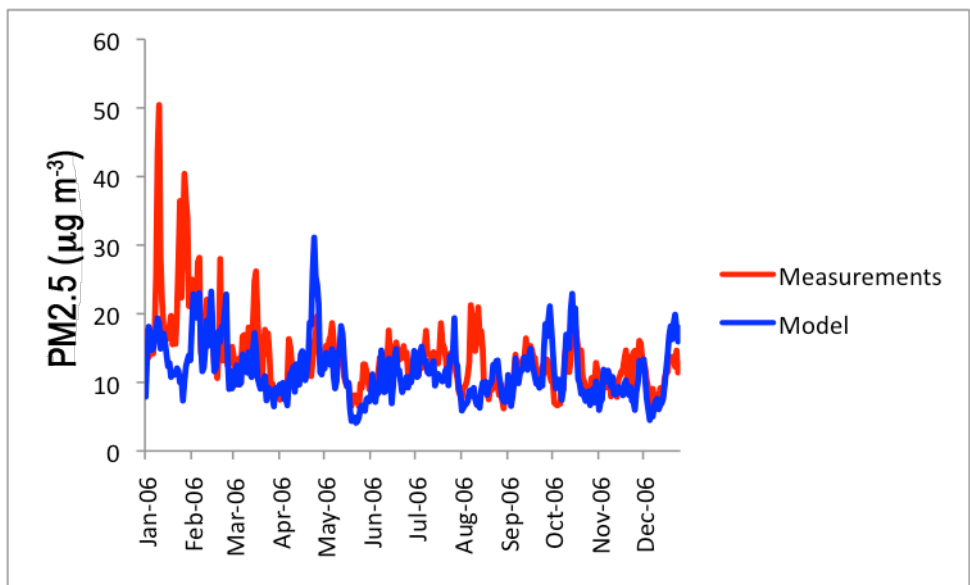
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4 Figure 1: Relative changes (%) in annual emissions of SO₂, NO_x, NMVOC, NH₃ and
 5 PM_{2.5} with respect to reference year (2005) for various scenarios in Switzerland (CH)
 6 and the European countries (EU) (for definition of scenarios see text).

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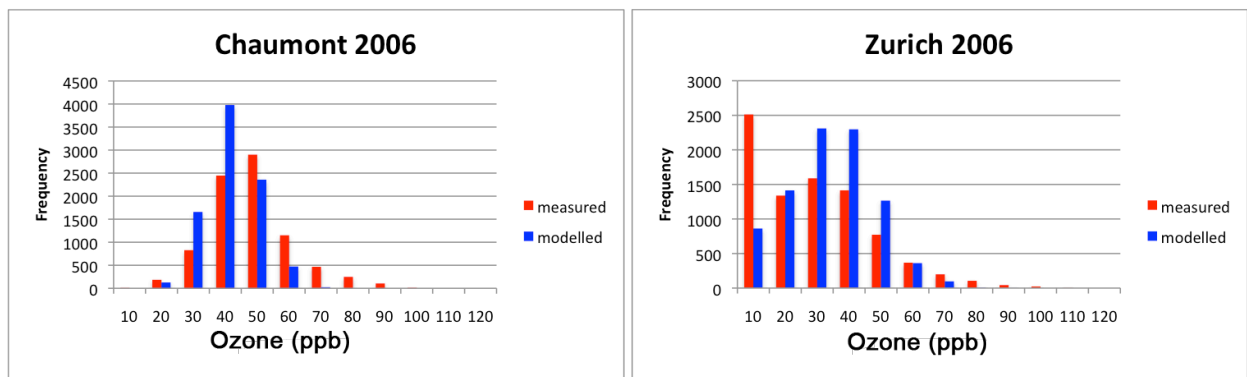
4

5 Figure 2: Time series of modelled (CAMx) and measured (AirBase) hourly average O₃
 6 (ppb) (upper panel) and daily average PM_{2.5} (µg m⁻³) (lower panel) in 2006 (European
 7 domain). Number of measurement sites: 300 for O₃, 19 for PM_{2.5}.

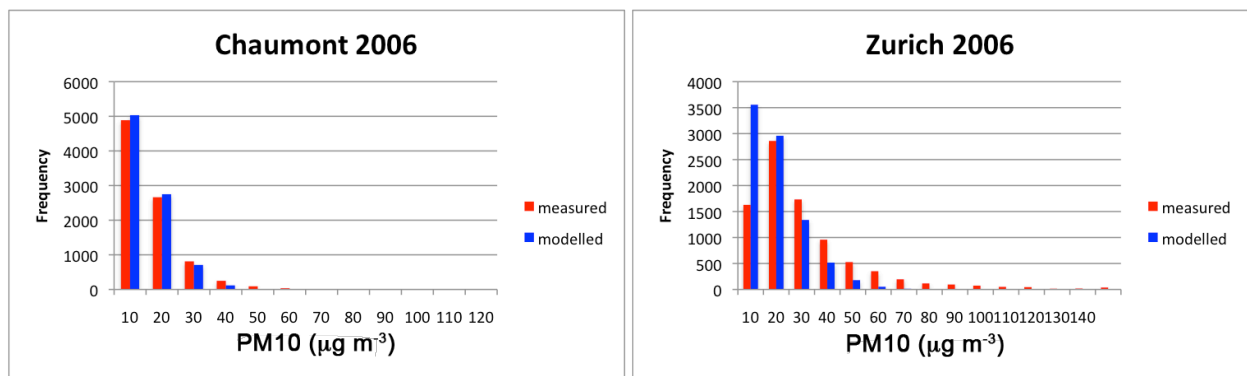
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5 Figure 3: Frequency distributions of ozone (upper panel) and PM₁₀ (lower panel) at
6 Chaumont (rural) and Zurich (urban background) in 2006.

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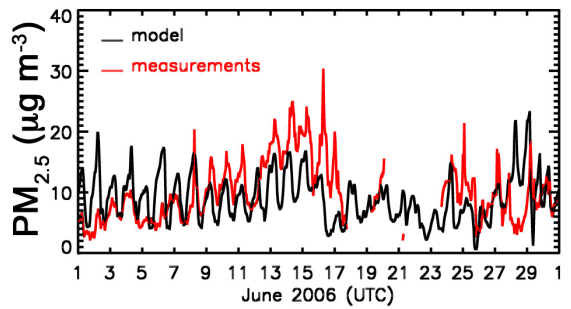
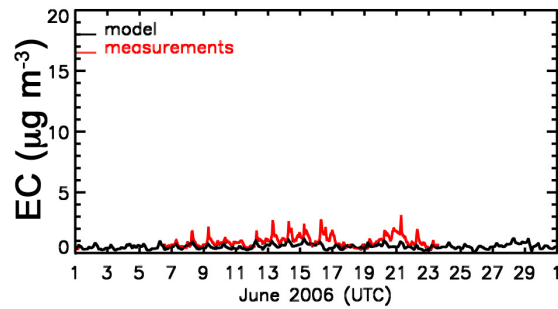
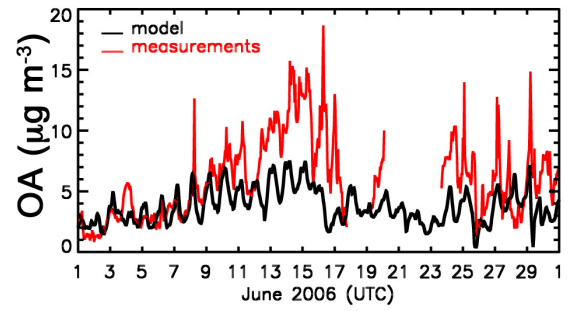
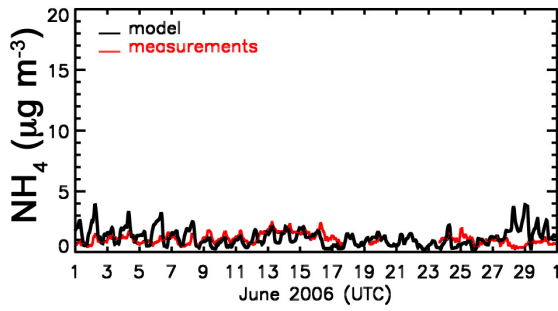
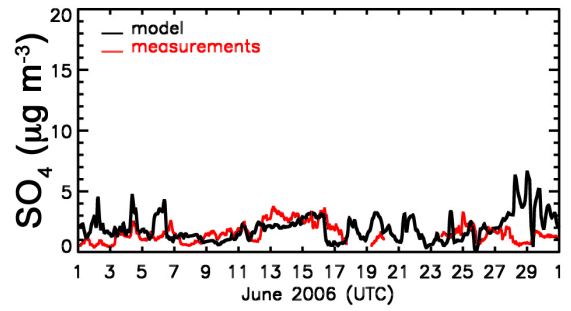
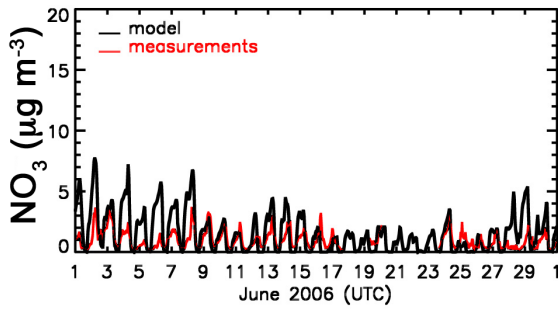
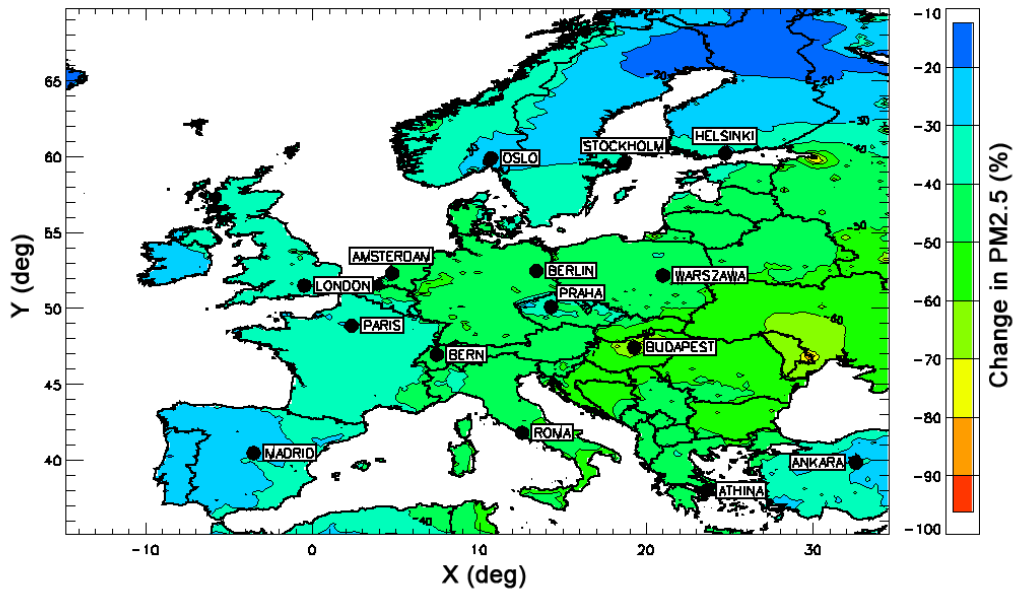
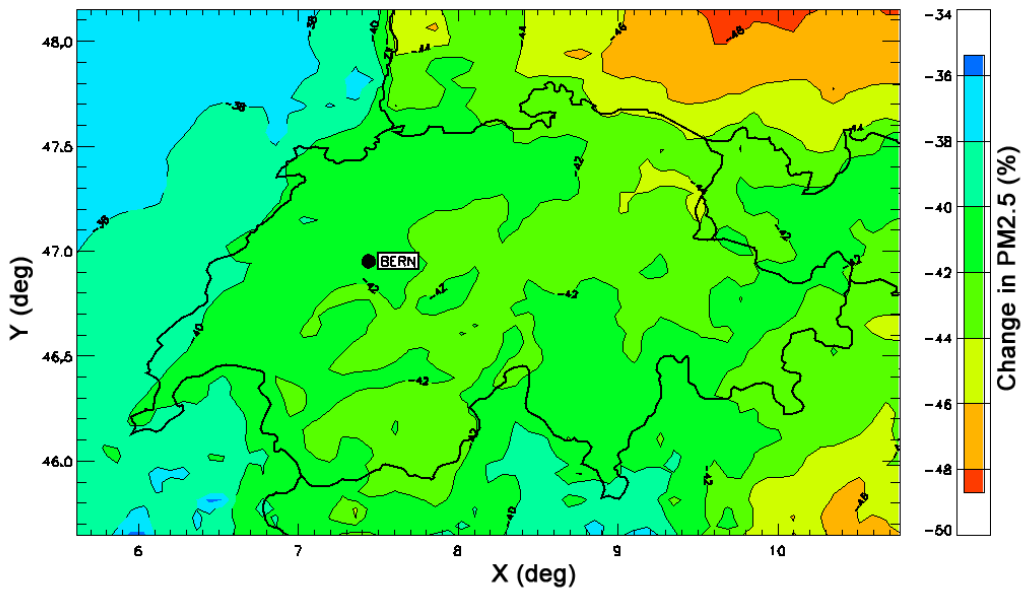


Figure 4: Measured (red) and modelled (black) hourly concentrations of particulate nitrate, sulphate, ammonium, organic aerosols (OA), elemental carbon (EC) and PM_{2.5} (sum of all species shown above) at Payerne in June 2006. EC was derived from Aethalometer measurements, the other components were measured by an AMS.

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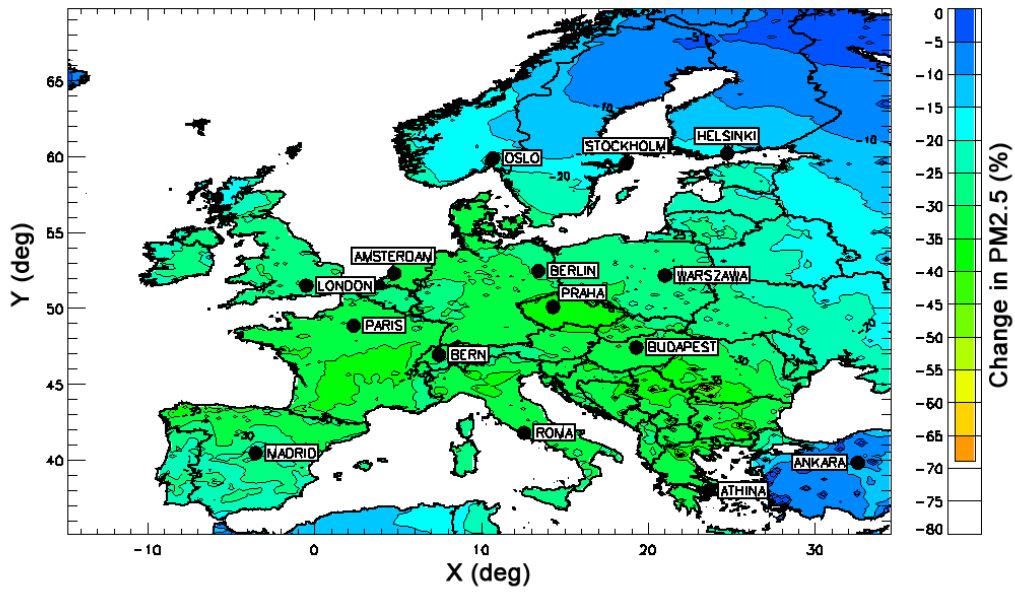


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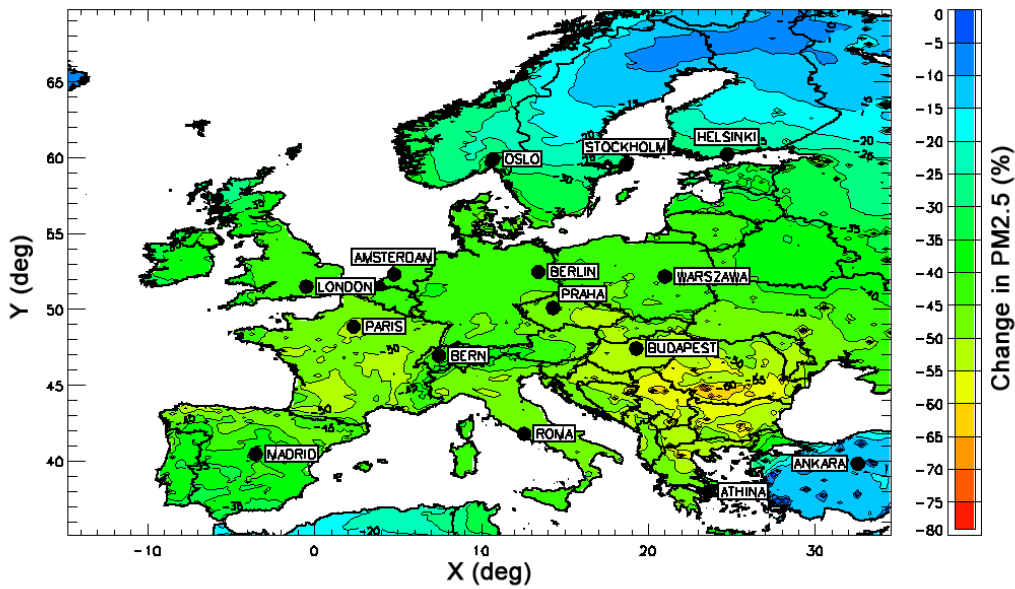
4 Figure 5: Relative changes in annual average $PM_{2.5}$ concentrations over the European
5 (upper panel) and Swiss (lower panel) domains, 2005-1990.

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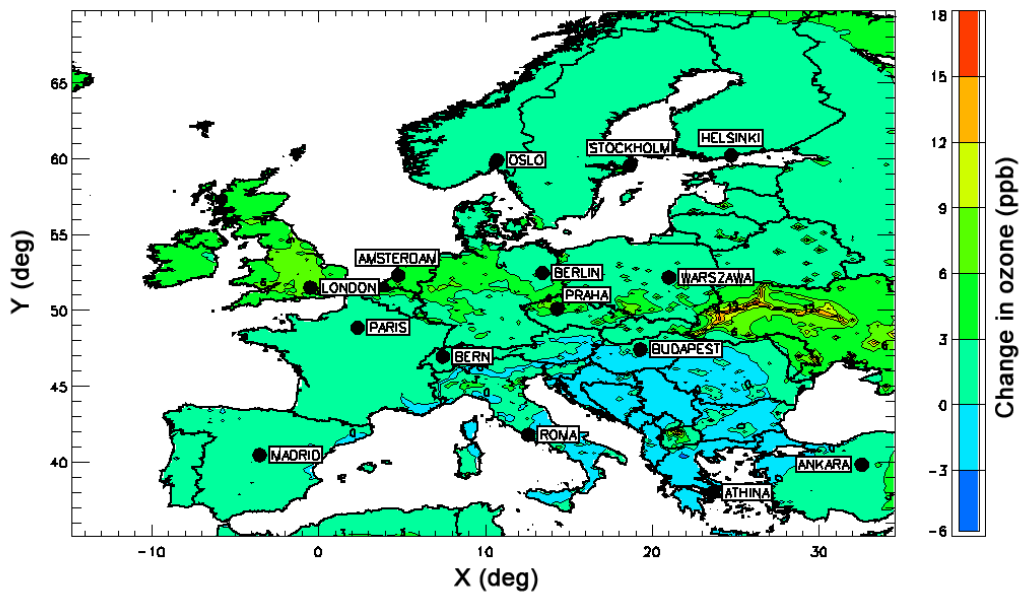


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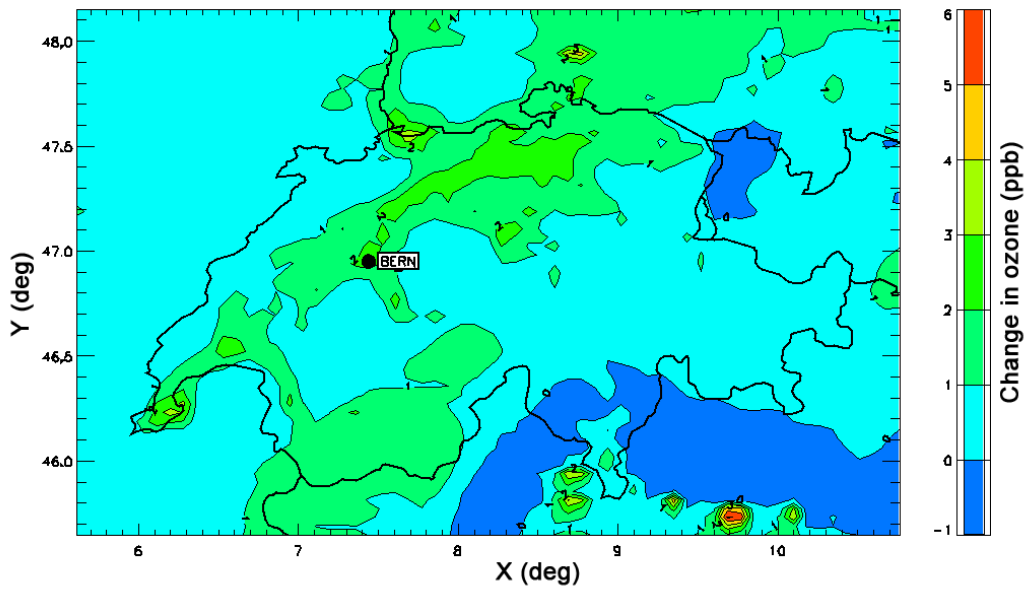
4 Figure 6: Relative changes in annual average PM_{2.5} concentrations over the European
5 domain for two scenarios: BL 2020 - 2005 (upper panel) and MTRF 2020 - 2005 (lower
6 panel).

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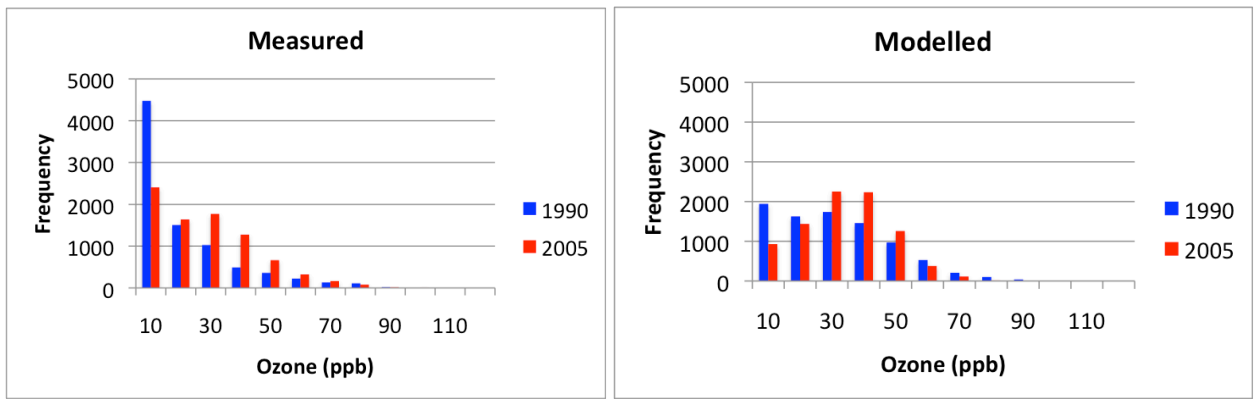
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3 Figure 7: Changes (ppb) in annual average ozone mixing ratios over the European
 4 (upper panel) and Swiss (lower panel) domains, 2005 – 1990.

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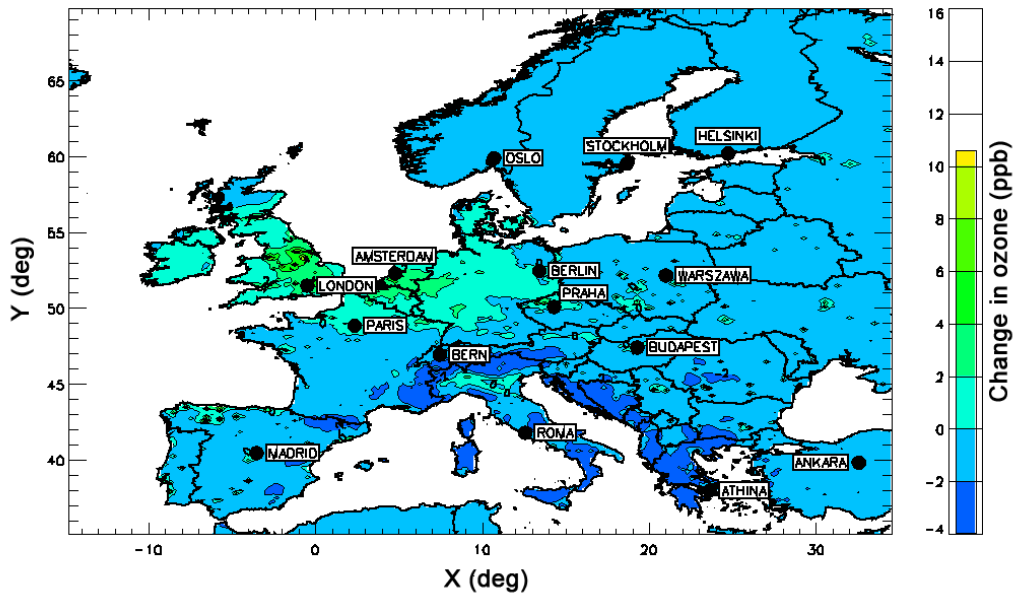
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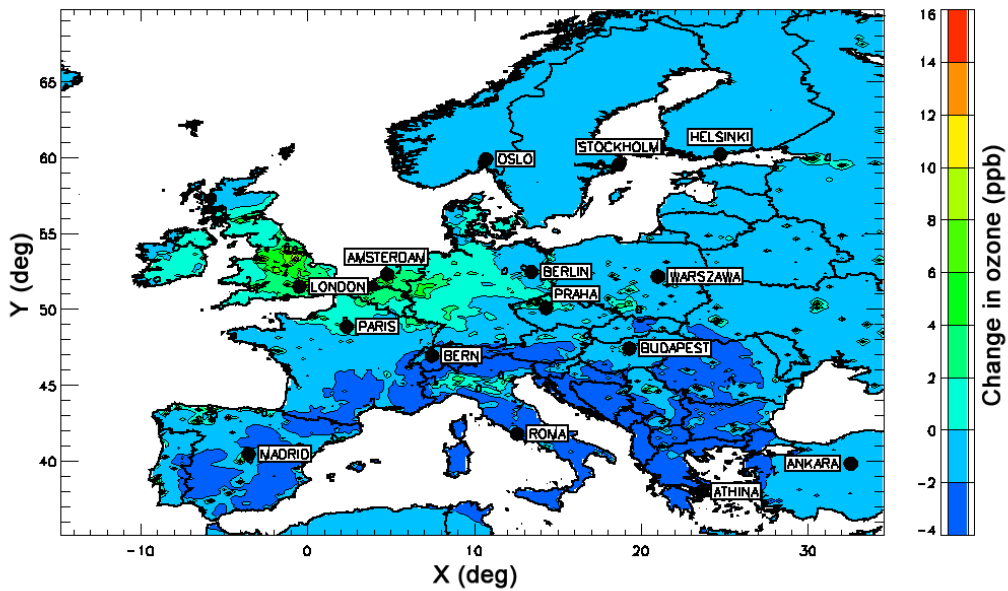


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 2 Figure 8: Changes in frequency distributions of measured (left) and modelled (right)
 3 ozone between 1990 and 2005 in Zurich.
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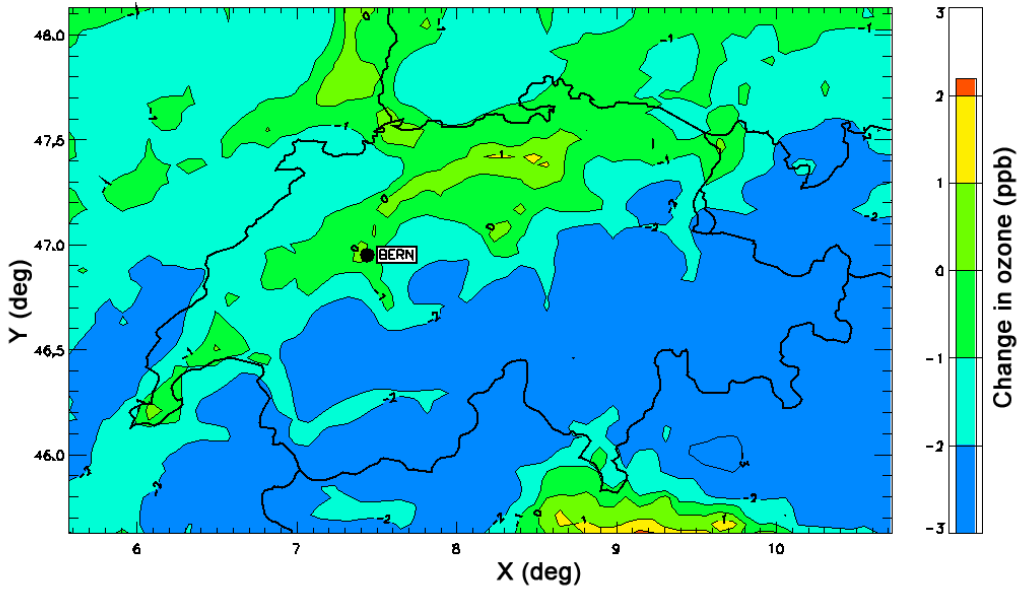


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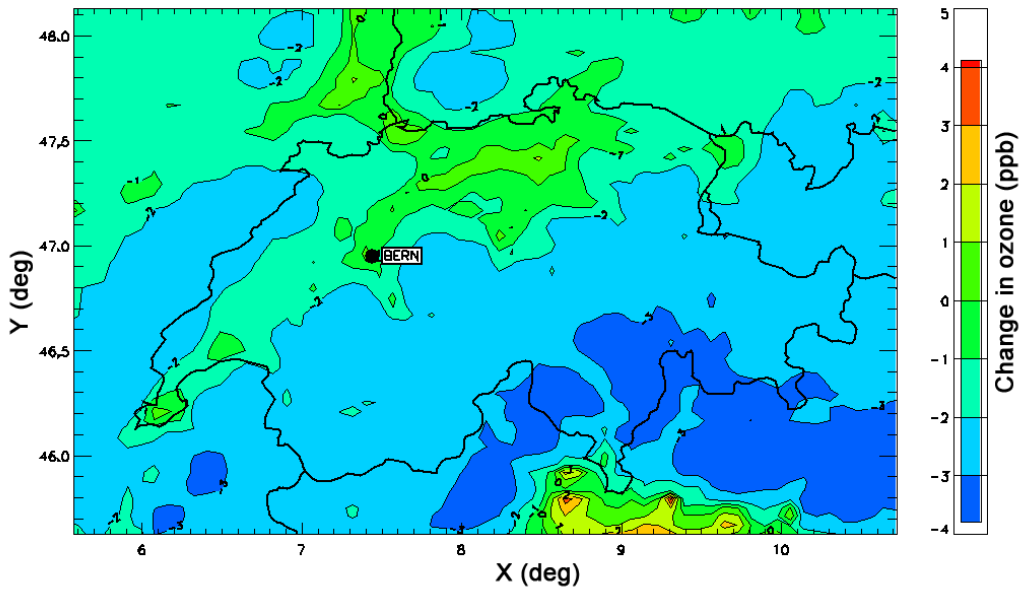
4 Figure 9a: Changes in annual average ozone mixing ratios (ppb) over the European
5 domain, BL 2020- 2005 (upper panel), MTFR 2020- 2005 (lower panel).

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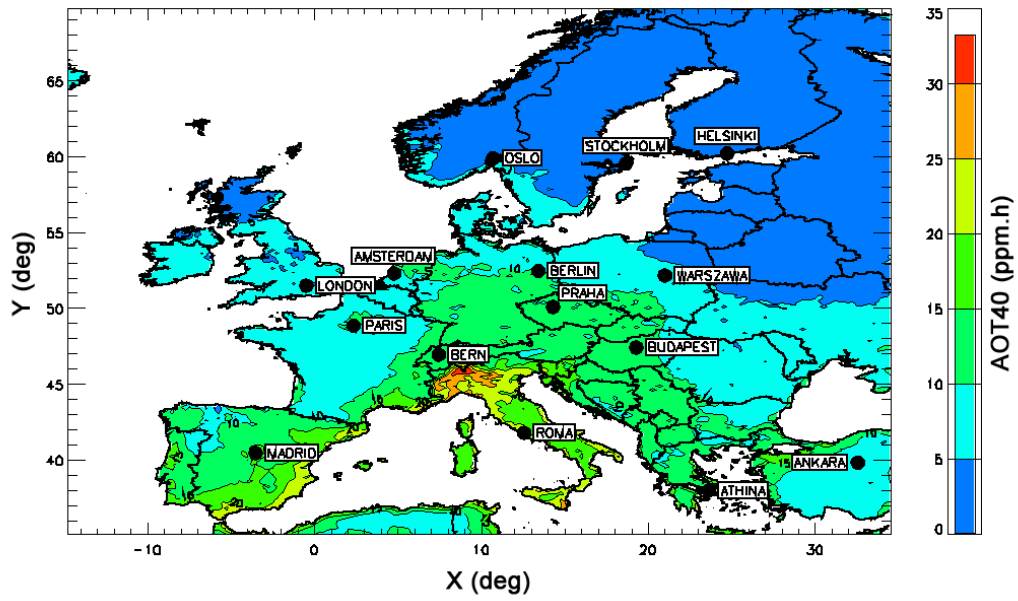
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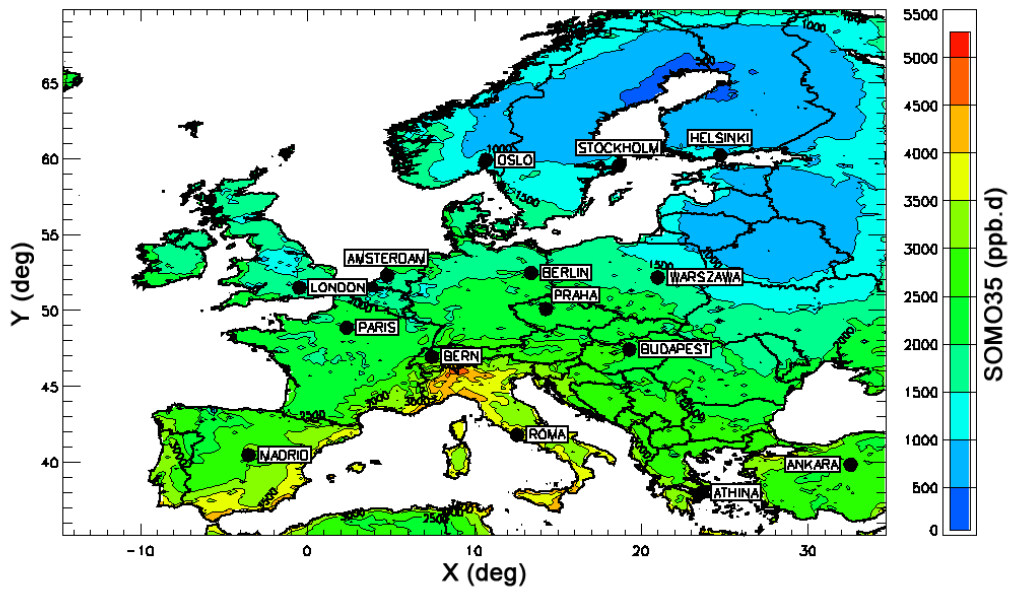
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 3 Figure 9b: Changes in annual average ozone mixing ratios (ppb) over the Swiss domain,
 4 BL 2020- 2005 (upper panel), MTFR 2020- 2005 (lower panel).

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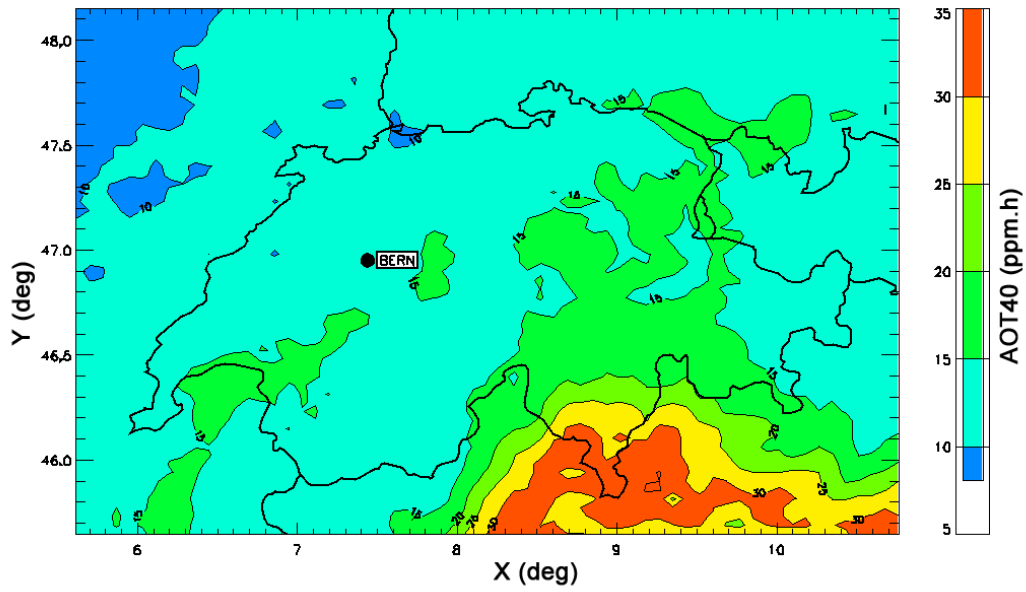


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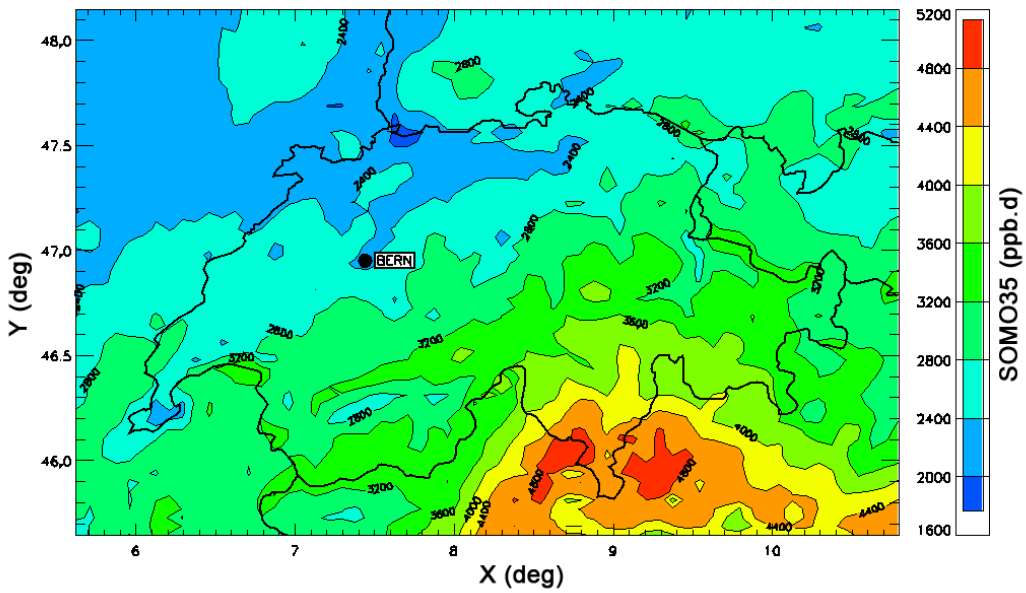
4 Figure 10: Modelled AOT40 (ppm h) (upper panel) and SOMO35 (ppb d) (lower panel)
5 over the European domain for the reference year (2005).

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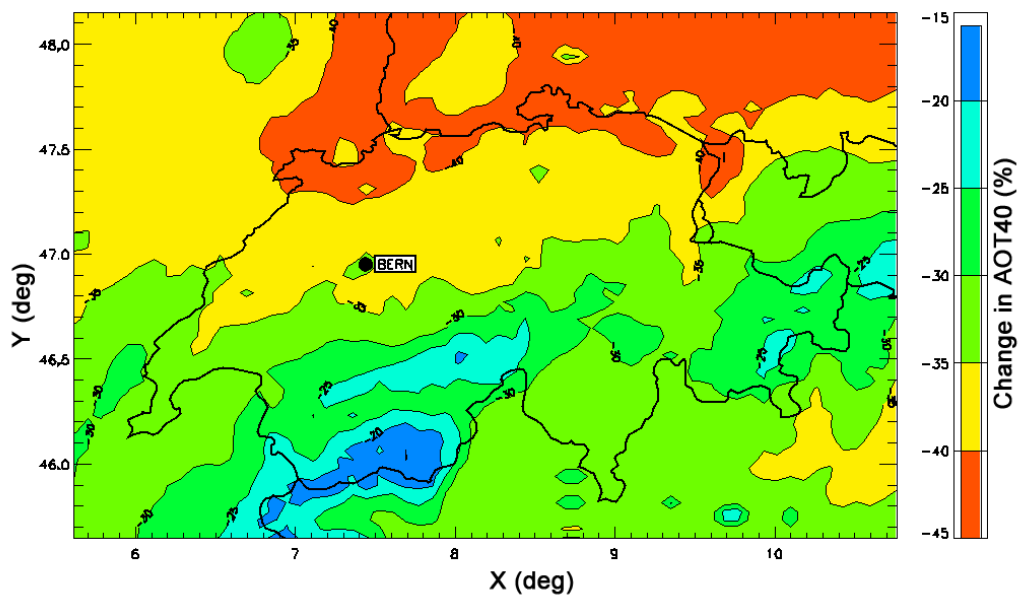


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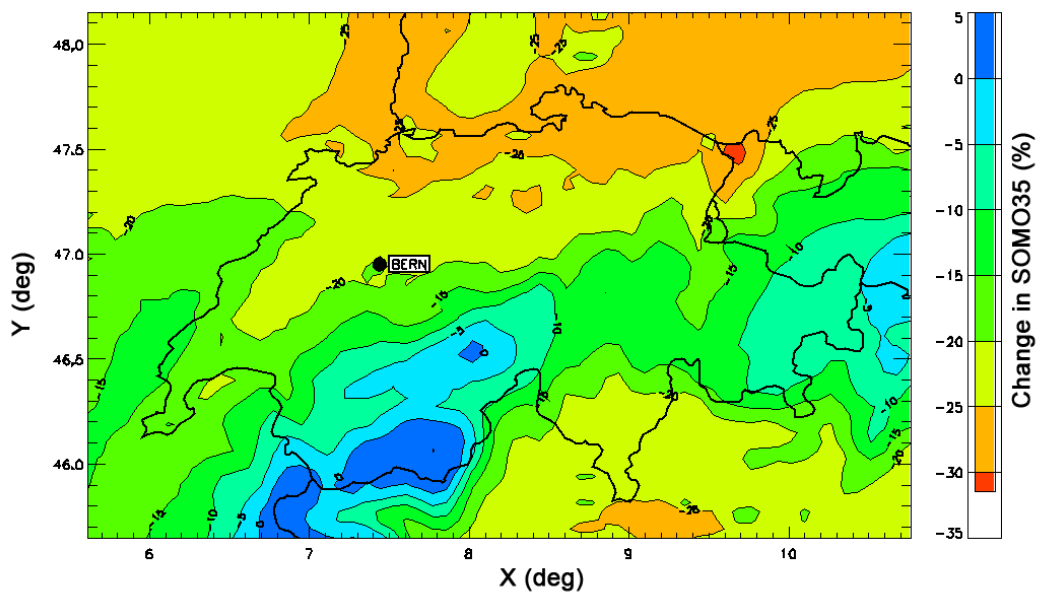
4 Figure 11: Modelled AOT40 (ppm h) (upper panel) and SOMO35 (ppb d) (lower panel)
5 over the Swiss domain for the reference year (2005).

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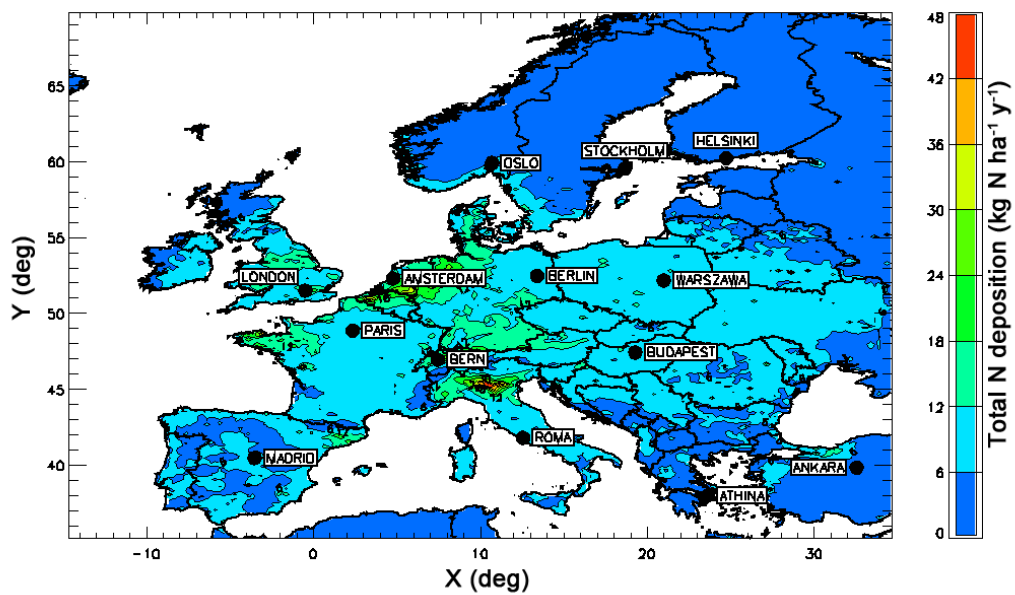
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3 Figure 12: Relative changes in AOT40 (upper panel) and in SOMO35 (lower panel) over
 4 the Swiss domain, 2005 – 1990.

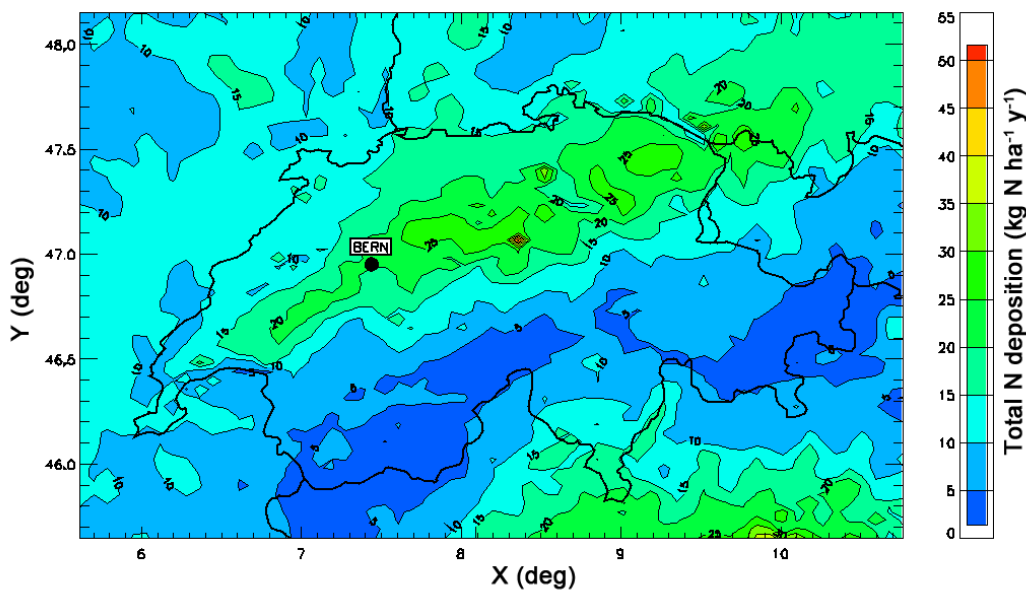
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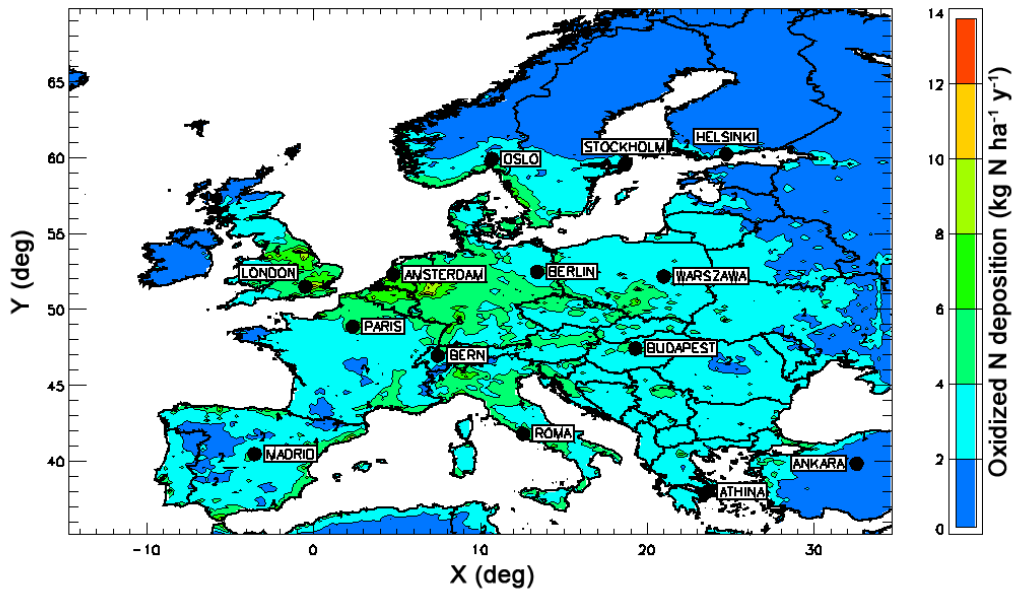


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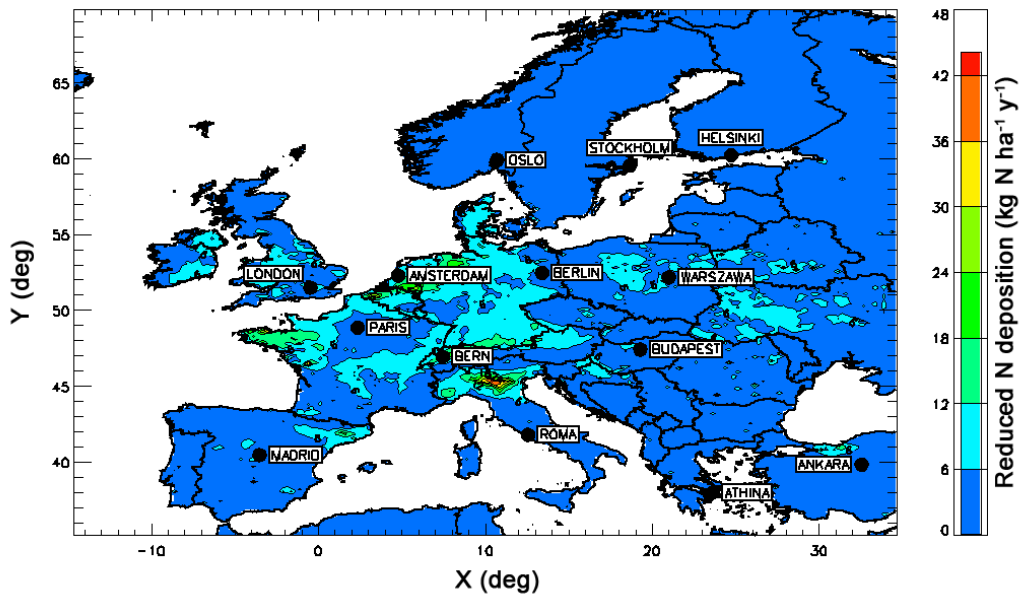
3 Figure 13: Total N deposition ($\text{kg N ha}^{-1} \text{y}^{-1}$) over the European (upper panel) and the
 4 Swiss (lower panel) domains (2006).

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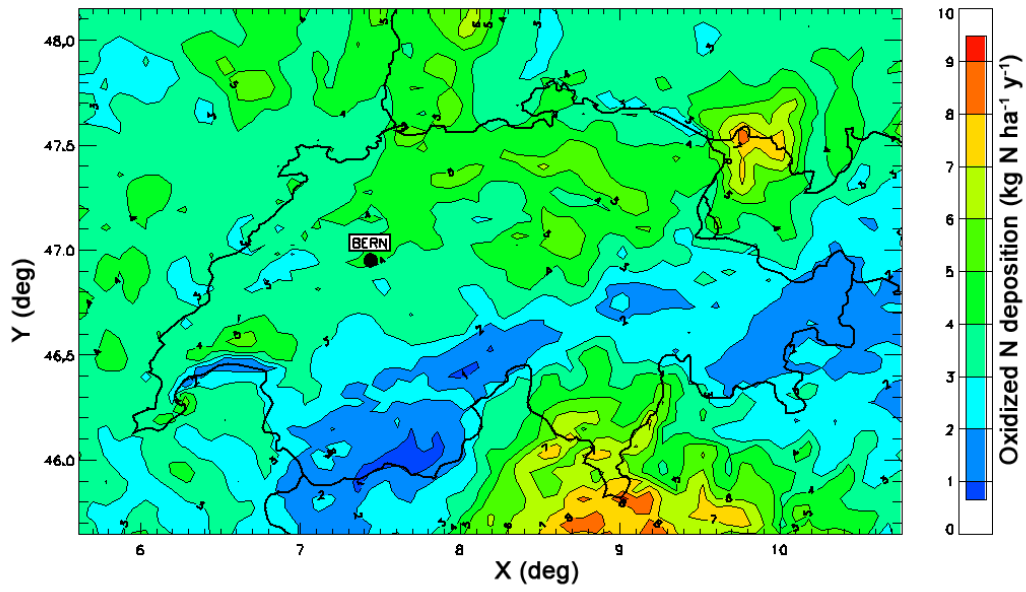


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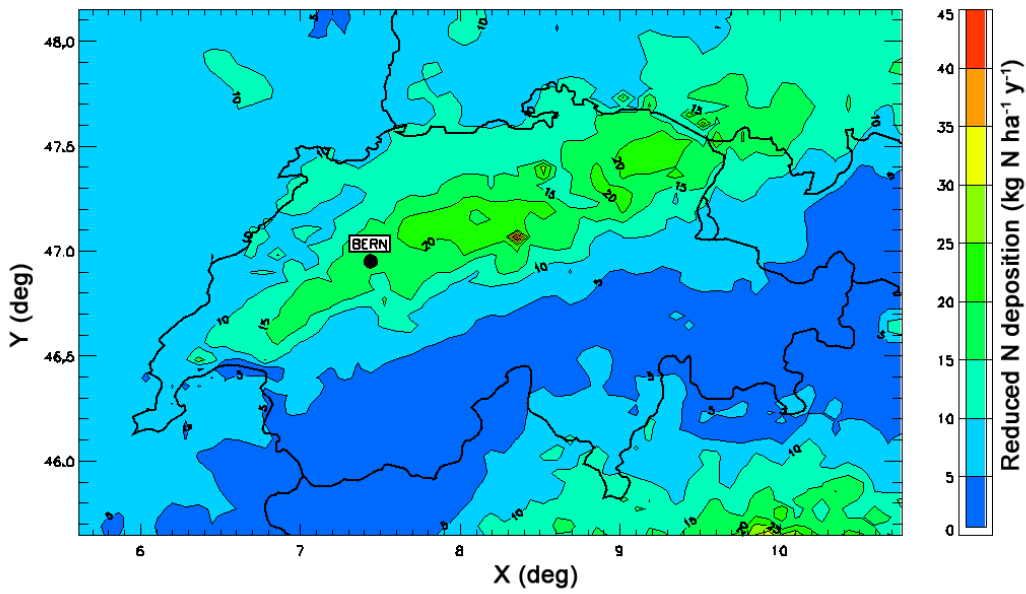
4 Figure 14: Deposition (kg N ha⁻¹ y⁻¹) of oxidized (upper panel) and reduced (lower panel)
5 nitrogen compounds over the European domain (2006).

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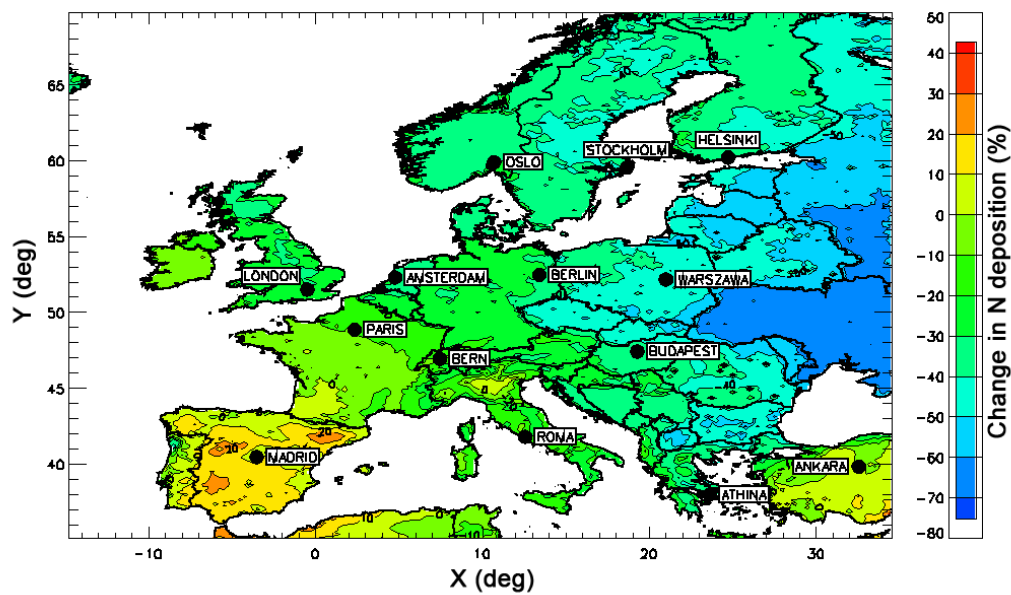
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4 Figure 15: Deposition (kg N ha⁻¹ y⁻¹) of oxidized (upper panel) and reduced (lower panel)
5 nitrogen compounds over the Swiss domain (2006).

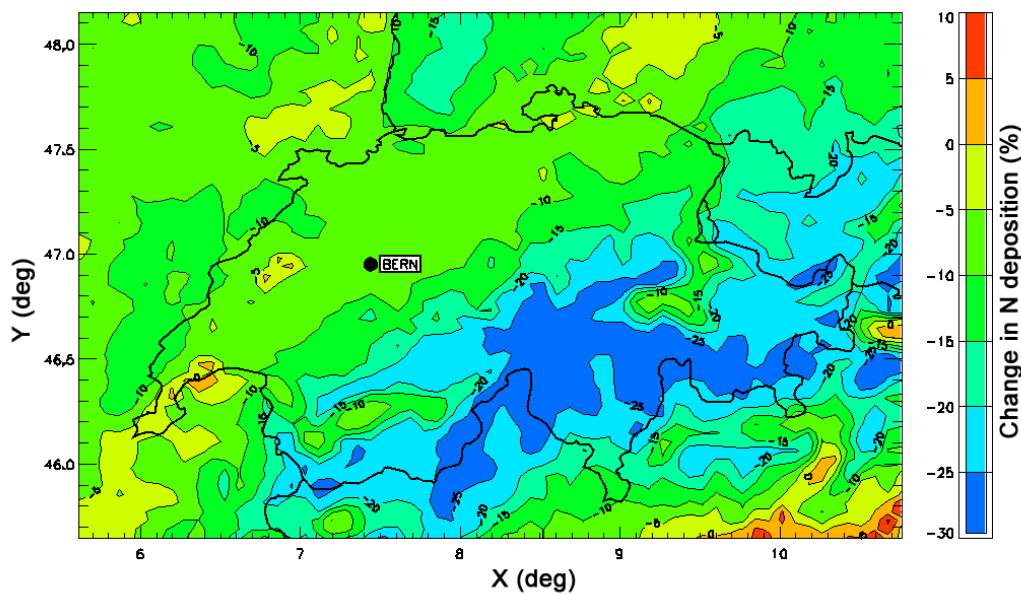
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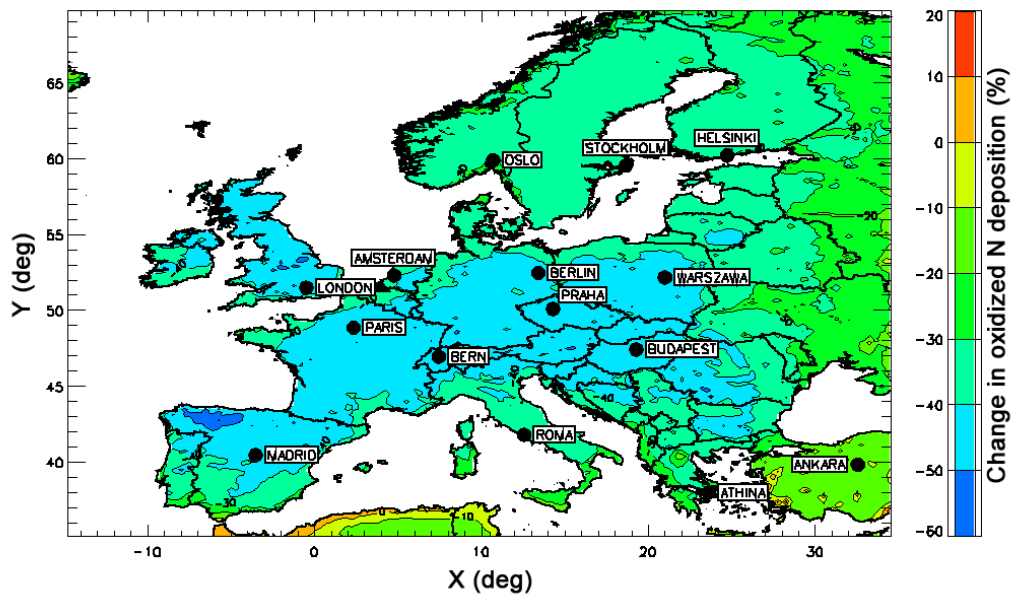
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3 Figure 16: Relative changes in nitrogen deposition over the European (upper panel) and
 4 the Swiss (lower panel) domains, 2005 – 1990.

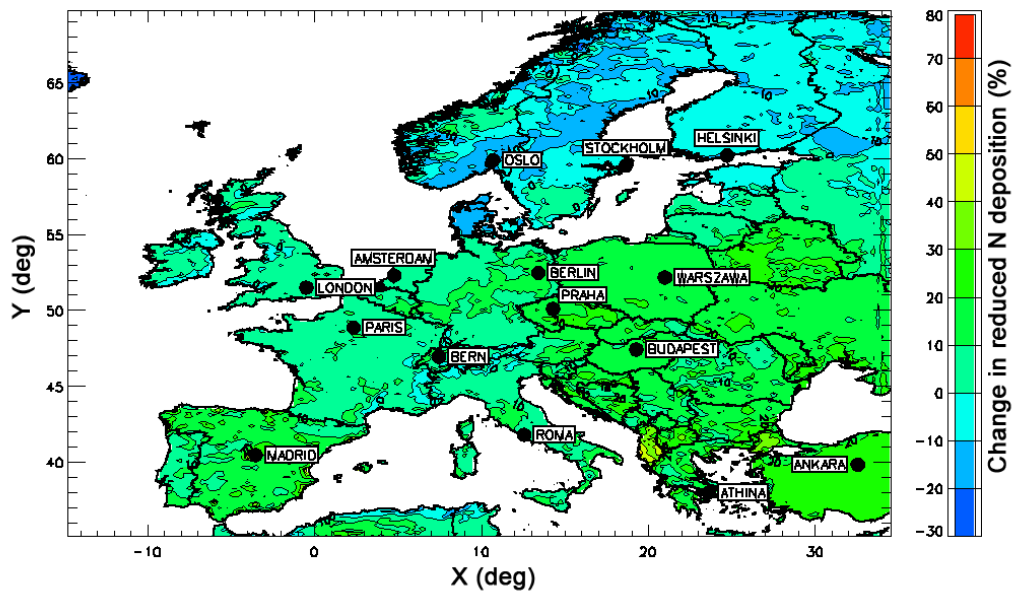
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3 Figure 17: Relative changes in deposition of oxidized (upper panel) and reduced (lower
 4 panel) nitrogen species over the European domain, BL 2020 – 2005.

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