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Reanalysis of and attribution to near-surface ozone concentrations in Sweden during 1990-2013

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Abstract

We have constructed two data sets of hourly resolution reanalyzed near-surface ozone (O_3) concentrations for the period 1990-2013 for Sweden. Long-term simulations from a chemistry-transport model (CTM) covering Europe were combined with hourly ozone concentration observations at Swedish and Norwegian background measurement sites using data assimilation. The reanalysis data sets show improved performance than the original CTM when compared to independent observations.

In one of the reanalyzes we included all available hourly near-surface O_3 observations, whilst in the other we carefully selected time-consistent observations in order to avoid introducing artificial trends. Based on the second reanalysis we investigated statistical aspects of the near-surface O_3 concentration, focusing on the linear trend over the 24 year period. We show that high near-surface O_3 concentrations are decreasing and low O_3 concentrations are increasing, which is mirrored by observed improvement of many health and vegetation indices (apart from those with a low threshold).

Using the chemistry-transport model we also conducted sensitivity simulations to quantify the causes of the observed change, focusing on three processes: change in hemispheric background, meteorology and anthropogenic emissions (Swedish and other European). The rising low concentrations of near-surface O_3 in Sweden are caused by a combination of all

three processes, whilst the decrease in the highest O_3 concentrations is caused by O_3 precursor emissions reductions.

While studying the relative impact of anthropogenic emissions changes, we identified systematic differences in the modelled trend compared to observations that must be caused by incorrect trends in the utilised emissions inventory or by too high sensitivity of our model to emissions changes.

1 Introduction

Elevated concentrations of near-surface ozone (O_3) are a major policy concern, given their ability to damage both vegetation (e.g. Royal Society, 2008) and human health (e.g. WHO, 2006). It is also an important greenhouse gas (e.g. IPCC, 2013). Elevated O_3 concentrations are formed in the troposphere by the oxidation of volatile organic compounds (VOCs) and carbon monoxide, driven by solar radiation in a polluted air mixture that includes nitrogen oxides (NO_x). Close to combustion sources, the background O_3 concentration is reduced through reactions with directly emitted nitric oxide (NO; see for example Finlayson-Pitts and Pitts, 2000). However, further away from the source and with sufficient availability of VOCs and the right weather conditions, these NO_x emissions can lead to rises in the O_3 concentration. O_3 can be transported to regions far away from the area where it was formed and even across continents (e.g. Akimoto, 2003; Derwent et al. 2015). Oxidized nitrogen can also be transported to remote regions as reservoir species, such as peroxy-acetyl nitrates (PANs). These can be a significant source of NO_x and alongside naturally emitted biogenic VOCs cause O_3 formation in otherwise non-polluted areas (e.g. Jacob et al., 1993).

European and North American anthropogenic emissions of NO_x increased over most of the 20^{th} century, but decreased strongly since the 1980s due to emission control (e.g. Lamarque et al., 2010; Granier et al., 2011). Asian emissions have continued to rise under the same period (Ohara et al., 2007). Jonson et al. (2006) showed that the trend in O_3 concentration in Europe cannot be fully explained by changes in European precursor emissions. By inter-continental transport the increasing precursor emissions in Asia could contribute to increasing background levels with at least a strong impact in North America (Vestraeten et al., 2015), whilst the trend in European background O_3 seasonal variation could also be affected by the decreases in North American precursor emissions (Derwent et al., 2015). Climate also changes over time, causing both changes to the O_3 forming potential, biogenic emissions of

O_3 precursors and deposition processes (Andersson and Engardt, 2010). Variability in climate, such as the North Atlantic Oscillation (NAO), contributes to the variation in O_3 concentration in the upper troposphere through variations both in the stratospheric contribution and in the transport patterns (Gaudel et al., 2015). Although the stratospheric contribution to the O_3 concentration at the surface is generally small (3-5 ppb(v)) in Europe (Lelieveld and Dentener, 2000), it can be a relevant contribution to near-surface O_3 in certain areas and time periods (Zanis et al., 2014) and could affect the observed trend in near-surface O_3 (e.g. Fusco and Logan, 2003). Despite the large number of studies of tropospheric O_3, a number of challenges still remain, such as explaining the near-surface concentration trends (Monks et al., 2015).

Observations in the northern mid-latitudes, either at the surface (Oltmans et al, 2006) or from ozone-sondes and commercial aircraft (Logan et al., 2012), present the picture of increasing tropospheric O_3 concentrations during the second half of the 20th century (Parrish et al., 2012; Cooper et al., 2014). The strong increase in near-surface O_3 concentration until the late 1990s at three widely separated North Atlantic sites, including Mace Head, seems to have peaked or remained stationary (Simmonds et al 2004; Oltmans et al., 2006; Derwent et al., 2007). At Pico Mountain Observatory in the Azores, a decreasing O_3 concentration trend was observed during 2001-2011 which was believed to be mainly caused by decreasing precursor emissions in North America (Kumar et al., 2013). Air masses with European origin observed at Mace Head show a decrease in summertime peak O_3 concentrations and increase in wintertime, which is believed to be connected to European NO_x policy (Derwent et al., 2013). O_3 concentrations observed at European alpine sites and in ozone-sonde data (MOZAIC) above European cities have decreased since 1998 with the strongest decrease in summer (Logan et al., 2012).

Several modelling efforts have been conducted to describe the past near-surface O_3 concentration development (e.g. Fusco and Logan, 2003; Schultz et al., 2007; Pozolli et al., 2011, Xing et al. 2015). Parrish et al. (2014) present past trends in tropospheric O_3 concentrations modelled with three chemistry-climate models and conclude that while there is considerable qualitative agreement between the measurements and the models, there are also substantial and consistent quantitative disagreements. These include that the models capture only 50 % of the change observed during the last 5-6 decades and little of the observed seasonal differences, and that the rate of the trends are badly captured. There are ways

forward to improve the description of the trends: 1) understanding the processes and improving the model description of the physics and chemistry for processes of greatest importance in these models, 2) improving the input data quality and 3) incorporating observations in the model by data fusion methods to accurately represent the past statistics in a reanalysis. The first two are important for conducting scenario calculations, whilst the last is an option for producing mappings.

If correctly conducted, data fusion will improve the modelled estimates. If temporal and spatial consistency is not considered, it may however introduce artificial trends. Data assimilation, a subset to data fusion (Zhang et al., 2012), is the process by which observations of a system are incorporated into the model state of a numerical model, in this case into the chemistry transport model (CTM) (Kalnay, 2003; Denby and Spangl, 2010). Advanced data assimilation schemes like the 4 dimensional variational (4dvar; e.g. Courtier et al., 1994; Inness et al., 2013) technique utilize information provided by satellites and propagate this in space and time from a limited number to a wide range of chemical components to provide fields that are physically and chemically consistent with the observations. Inness et al. (2013) performed a reanalysis of global chemical composition, including O_3 concentration, for 2003-2010 using advanced data assimilation of satellite observations within the framework of the monitoring atmospheric composition and climate (MACC) project. They demonstrated improved O_3 and CO concentration profiles for the free troposphere, but biases remained for the lower troposphere. Another global reanalysis using data assimilation of satellite data for 2005-2012, showed improved performance for chemical species (Miyazaki et al., 2015) but for the O_3 concentration at the surface errors remain associated with low retrieval sensitivity in the lower troposphere and gaps in spatial representation between the model and observations. In order to improve surface characteristics, in situ observations of O_3 need to be included in data assimilation. Another reanalysis of near-surface O_3 concentration in Europe was conducted for the period 2003-2012 within the MACC project (Katragkou et al., 2015). The reanalysis was based on the MACC global model, which consists of the European Centre for Medium-Range Weather Forecasts' Integrated Forecast System (IFS) coupled to the MOZART-3 CTM. In this reanalysis 4dvar data assimilation was used to incorporate in situ measurement from the databases EMEP and Airbase. The data assimilation reduced the bias in near-surface O_3 concentration in most of Europe, and it reproduced the summertime maximum in most parts of Europe, but not the early spring peak in northern Europe. When restricting the observations to in situ measurements in Europe, the beginning of the time

period of the reanalysis can be extended further back in time utilizing simpler data assimilation techniques than 4dvar. Variational analysis in 2 dimensions (2dvar) and the analytical counterpart optical interpolation can be used as a CPU-efficient diagnostic tools to improve modelled near-surface O_3 retrospectively (e.g. Alpfjord and Andersson, 2015; Robichaud and Ménard, 2014).

The MATCH (Multi-scale Atmospheric Transport and CHemistry) Sweden system (Alpfjord and Andersson, 2015) includes an operational CTM and methods for data assimilation of atmospheric concentrations in air and precipitation. The system is used for annual assessments of the near-surface O_3, SO_2, NH_3 and NO_2 background concentrations and deposition of nitrogen, sulfur and base cations in Sweden. In this study, the MATCH Sweden system is used to conduct a reanalysis of the hourly near-surface O_3 concentration for Sweden and Norway during the 24-year period 1990-2013 using 2dvar. We use time-consistent input data to avoid the introduction of artificial trends in the results. In an attempt to understand the trends, we perform model sensitivity analyses and apply the CTM without data assimilation. This approach brings new knowledge to explain the trends in O_3 concentrations found in Sweden.

The aims of this study are:

- To create a state-of-the art long-term, temporally and spatially consistent, reanalysis of hourly near-surface O_3 concentrations covering the geographical areas of Sweden and Norway (see Sect. 2)
- To evaluate the performance of the O_3 reanalysis of the MATCH Sweden system, used in the annual assessment of air quality in Sweden (see Sect. 3.1)
- To investigate trends and extreme values in near-surface O_3 in Sweden (see Sect. 3.2) and its implications on health and vegetation (see Sect. 3.4)
- To understand the causes of the change over time, focusing on contributions of emission change, lateral and upper boundary and meteorological variability. (see Sect. 3.3)

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2 Method

In this study we utilize data assimilation in order to combine the respective best qualities of both a CTM and long-term measurements to map near-surface O_3 concentrations during a long historical time period (1990-2013). We focus our study on Sweden, but also include Norway in the data assimilation.

For the data assimilation we use the MATCH Sweden system, which is briefly explained in Sect. 2.1. Here variational analysis in two dimensions is applied, and further details are given in Sect. 2.4. Concentration fields provided by the CTM at each grid point are considered as the "first guess" (background field/prior information) of our "best estimate" of the state of the atmosphere before the introduction of observations (Kalnay, 2003). The method used for the production of the "first guess" is explained in Sect. 2.2. The selection of measurements that are included in the data assimilation is important, both to avoid artificial trends in the reanalysis data and in order to select observation sites with corresponding spatial and temporal representations as in the model. We explain our method for the selection of measurements in Sect. 2.3.

One aim of this study is to investigate trends in near-surface O_3 in Sweden. To understand the long-term changes in concentration we try to quantify the causes of change, through model sensitivity analyses, and applying the MATCH model without data assimilation. We investigate the respective contributions to the trends of change in European emissions by separating the impact on O_3 trends of changes in local emissions in Sweden, in hemispheric background concentrations (including changes to the top and lateral boundaries) and in meteorology (including changes to biogenic emissions, transport, O_3 forming capacity, O_3 deposition etc.). The method for this quantification is described in Sect. 2.5. The methods we use for evaluation are given in Sect. 2.6.

2.1 The MATCH Sweden system

The MATCH Sweden system is an operational system used for annual assessments of near-surface regional background concentrations in air of O_3, NO_2, NH_3 and SO_2 as well as deposition of sulfur, nitrogen and base cations over Sweden (Alpfjord and Andersson, 2015). The system includes an operational CTM (MATCH; Multi-scale Atmospheric Transport and Chemistry; Robertson et al., 1999) and methods for data assimilation (using 2dvar) of

atmospheric concentrations in air and precipitation. The yearly results from the mapping can be found at www.smhi.se/klimatdata/miljo/atmosfarskemi.

The flow-chart in Fig. 1 describes the parts of the MATCH Sweden system that are used in this reanalysis of near-surface O_3 concentrations. Explanations are provided in Sect. 2.2 to 2.4. For a description of the whole MATCH Sweden system, see e.g. Alpfjord and Andersson (2015).

2.2 First guess – model assessment

The starting point (cf. Fig. 1) for the two-dimensional variational data assimilation of near-surface O_3 is hourly fields of modelled O_3, produced by MATCH. The MATCH model includes ozone- and particle-forming photo-chemistry with ~60 species (Langner et al., 1998; Andersson et al., 2007, 2015). Part of the gas-phase chemical scheme was updated based on Simpson et al. (2012), except for some reaction rates (following the recommendations by the International Union of Pure and Applied Chemistry, IUPAC), and the isoprene chemistry mechanism that was based on an adapted version of the Carter one-product mechanism (Carter, 1996; Langner et al., 1998). A selection of compounds with different ozone forming potentials is used to represent all hydrocarbons emitted into the atmosphere. The photolysis rates depend on the photolytically active radiation, which is dependent on latitude, time of day, cloud cover etc. In this study MATCH interpolates the input meteorology to a domain covering Europe and surrounding areas with 44 km grid point spacing. MATCH uses all meteorological model layers for vertical wind calculations, but restricts the calculations of chemistry and transport to the lower troposphere using the vertical levels of the meteorological model from the surface up to ca 5 km height.

MATCH is an offline model, thus, driven by meteorological data generated externally and as such it is often a challenge to undertake long (multi-decadal) simulations due to non-homogenous input data. Dynamical meteorological models, which provide the three-dimensional meteorology for the offline CTMs, are constantly updated to higher resolutions and more advanced physical schemes. Emission inventories are typically constructed for certain target years and different methods may have been used to compile total emissions and/or the geographical distribution of the emissions. Careless combination of different emission data or meteorology from varying model configurations can introduce artificial

secular trends in the modelling of atmospheric pollutants. Emissions of biogenic isoprene are calculated online in MATCH following the E-94 isoprene emission methodology proposed by Simpson et al. (1995). Further details of MATCH in the present model version and its ability to simulate near-surface O_3 can be found in separate publications, for example Markakis et al. (2016), Lacressoniere et al. (2016) and Watson et al. (2015; 2016). In this study, we specifically aimed for internally coherent input data, although it led to compromises in e.g. the temporal coverage of the meteorology and the resolution of the gridded pan-European emissions. In the following sections we briefly describe the utilized input data.

2.2.1 Meteorology and boundary concentrations

In the present study we force MATCH with three-dimensional meteorology from the numerical weather forecast model HIRLAM. Within the EURO4M-project (http://www.euro4m.eu) HIRLAM was run as forecasts from 6-hourly analyses, composed of variational upper air analyses in 3 dimensions and optimal interpolation surface analyses. Lateral and lower (sea surface temperature and sea ice) boundaries were taken from ERA-Interim (Dee et al., 2011). Full three-dimensional model states needed to run MATCH are available from 1979 through February 2014. Under EURO4M, HIRLAM was running on a domain covering Europe and Northern Africa with 22 km grid point spacing and 60 vertical layers from the surface to 10 hPa.

Although the present study focuses on Sweden it is necessary to realistically describe the fluxes of O_3 from continental Europe and further afield. Hemispheric concentrations of all species are similar to the ones used by Andersson et al. (2007) for the modelled year 2000. As in Andersson et al. (2007), boundary values representative for the lateral and top boundaries of relevant species are interpolated spatially with a monthly temporal resolution. Boundary concentrations of O_3, oxidized nitrogen and methane are scaled to mimic observed changes in the hemispheric background during the period 1990 through 2013 (cf. Fig. 2a). The same factor is used for all months of the respective year, although most species also undergo a seasonal cycle in the boundary concentrations used by MATCH (see supplement Fig. S1).

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2.2.2 Emissions

The version of MATCH utilized in this study needs anthropogenic emissions of sulfur (SO_2 and sulfate), nitrogen oxides (NO and NO_2), carbon monoxide (CO), non-methane volatile organic compounds (NMVOCs), and ammonia (NH_3). The model uses annually accumulated values for each species, which are distributed with different temporal or vertical profiles based on species and sectors.

For countries outside Sweden (as well as international shipping) we utilize the gridded (50 km × 50 km) annual data available at EMEP's web-page (http://www.emep.int; downloaded 23 June, 2015). All emission data were split into congruent 5 km × 5 km cells where we replaced the coarse-resolution data over Sweden with the original emission data from SMED (Svensk miljöemissionsdata; http://www.smed.se; 1 km × 1 km converted to 5 km × 5 km cells in EMEP's geometry). National totals from SMED are very similar to the national totals available in the EMEP database, but our methodology enables higher resolution emission data over Sweden. The gridded 5 km × 5 km emission data were interpolated to MATCH's 44 km resolution domain during the simulations.

Both the total domain and Swedish anthropogenic O_3 precursor emissions decrease strongly over the period 1990-2013 (cf. Fig. 2b). The total domain anthropogenic precursor emissions decrease on average[1] by 1.8 % yr^{-1}, 2.4 % yr^{-1}, 2.6 % yr^{-1} during 1990-2013 for NO_x, NMVOC and CO respectively, whereas biogenic isoprene emissions (calculated online by MATCH) increase by 0.8 % yr^{-1} according to our simulations. The Swedish emissions decrease by similar amounts (2.4 % yr^{-1}, 2.1 % yr^{-1} and 2.9 % yr^{-1}). The Swedish contribution to the total domain emissions is 1.0 % for NO_x and 1.7 % for NMVOC and CO on the average, with a slight decrease in the relative Swedish contribution over the period for NO_x (0.01% yr^{-1}), and a slight increase for NMVOC and CO (0.01 % yr^{-1} and 0.003 % yr^{-1} respectively). We assume that there is no trend in the temporal intra-annual variation of the emissions.

[1] The trend is calculated by linear regression over the period 1990-2013 and related to the 1990 emission level.

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Atmospheric Chemistry and Physics Discussions

2.3 Measurements

Figures 3 and 4 summarize the observations of hourly O_3 concentrations used in the variational analysis and the corresponding hourly data coverage per year in the period 1990-2013. These measurements represent the regional background in Sweden and Norway. The sites included are all instrumentation sites, where O_3 is measured continuously and reported with hourly temporal resolution. The data assimilation is conducted on hourly resolution, which means that measurements with a coarser time resolution, such as diffusive samplers, are not included in the variational technique. Two measurement data sets were compiled (see Table 1):

- The first includes data from all available instrumentation sites in Sweden, and a selection in Norway based on data availability, quality and location. These are all the red and blue sites in Figs. 3 and 4 also including years where the data capture is lower than 80 %. The reanalysis based on these measurement data is called ALL.
- The second data set includes data from instrumentation sites for which the data coverage exceeds 80 % for at least 23 out of the 24 years. These are the red sites in Figs. 3 and 4. The reanalysis based on these measurement data is called LONGTERM. Råö is seen as the replacement for the site Rörvik, and therefore these sites form a pair, which is included in this data set. Birkenes I was replaced by Birkenes II in 2009, and the two sites were run in parallel for a few years. We choose to include Birkenes II from 2010 and onwards. The reason for the change of site location is that Birkenes I was influenced by local effects, such as night-time inversions (personal communication with Sverre Solberg, NILU). The inclusion of these two sites could introduce an artifical trend in the reanalysis, but since it is outside the main focus area (Sweden) and mainly during night we choose to include the site in the LONGTERM reanalysis.

The two measurement data sets are input to two otherwise similar data assimilations. The ALL-reanalysis is our best estimate of gridded near-surface O_3 over Sweden for a given time. The LONGTERM-reanalysis is used for trend and statistical analyses. This is because changes in the number of sites and data coverage in the ALL data set can introduce artificial trends due to model biases being corrected by observations included in the later part of the period but not in the first. We return to whether these reanalyzes differ in Sect. 3.1.

Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2017-338
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Discussion started: 20 June 2017

Atmospheric Chemistry and Physics Discussions

2.4 Data assimilation

The spatial analysis problem can be formulated as how to best distribute observational information at a discreet number of locations to a spatially consistent field. We have adopted the 2dvar approach, which includes a modelled background field (from a CTM simulation, "first guess") combined with available in situ observations (Robertson and Kahnert, 2007), as indicated in Fig. 1. With this method the error estimates of both the background field and the observations play a central role. The observational errors are assumed independent and uncorrelated, while the background errors have spatial correlations that form a background error matrix. The solution is found by the best combination of the background field and observations given their respective error estimates. This can be described as a variational problem, defined by a cost function,

$$J(x)=0.5\,[x-x^b]^T\,\mathbf{B}^{-1}\,[x-x^b]+0.5\,[y-\mathbf{H}(x)]^T\,\mathbf{O}^{-1}\,[y-\mathbf{H}(x)]$$

where x is the state to be found (the reanalysis), x^b the background state (our "first guess"), y the vector of observations, **H** is the observation operator, and **B** and **O** are the error covariance matrices of the background field and the observations, respectively. In order to find the optimal solution the cost function is stepwise minimized by a variational method, starting with $x=x^b$, and ending with the state x, which represents the optimal balance between the two terms. During the process the co-variances in the B matrix acts to extrapolate the observational information in space.

We restrict our study to reanalyze near-surface O_3 on the regional background scale, which means we only include regional background measurement sites. We also restrict our study to 2dvar, rather than using higher dimensional variational analysis. The background covariance matrix is modelled in a simplified fashion with a constant background error, 20 times larger than the observation error, and Gaussian spatial correlations with a length scale of 1000 km. This implies a strong weight towards the observations and assuming a rather large horizontal influence of the observations, which is related to the rather sparse network of regional background observations and the relatively small emissions of O_3 precursors in Sweden resulting in weak horizontal gradients of near-surface O_3 on the regional background scale.

The data assimilation was conducted on a 22 km resolution grid with hourly temporal resolution, combining the modelled "first guess" for near-surface O_3 (the MATCH base case

scenario, MFG in Table 1) and regional background measurements. Two 24-year reanalyzes were formed, using the two sets of hourly measurement described in Sect. 2.3 (ALL and LONGTERM in Table 1). If an included measurement site was lacking an observation for a specific hour, the site was excluded from the data assimilation for that specific hour.

The resulting spatially resolved hourly O_3 data are used to form annual and seasonal statistical metrics for O_3, such as the mean value and the maximum 1-hour mean value, and annual policy and impact related metrics (cf. Fig. 1). We analyze these annual and seasonal data for the 1990-2013 mean, trend and extreme values in Sect. 3.2 (annual/seasonal mean and maximum) and Sect. 3.4 (health and vegetation impact metrics).

2.5 Understanding the trends

We include also a quantification of the causes to the trend in near-surface O_3 concentration. For this investigation we conduct model simulations with MATCH, *excluding data assimilation*. We investigate the respective contributions to the modelled total trend due to

A. Change in emissions, which is separated between
    - Swedish anthropogenic emissions (Se emis)
    - European (full domain) non-Swedish anthropogenic emissions (Eur emis)
B. Change in lateral and upper boundaries (bound)
C. Change in meteorology, including online modelled biogenic isoprene emissions (meteo)

Four sensitivity simulations are conducted; in which each of the four listed processes are kept constant at the level in 2011. The respective contributions to the trend are formed by subtracting the MFG with the corresponding sensitivity simulation. All model simulations and scenarios are described in Table 1a. The method of forming the contributions from these simulations is shown in Table 1b.

There are two critical points in the investigation of the causes of the trend: First, this quantification methodology assumes linearity, whereas the sum of contributions (SUM) is not necessarily equal to the trend in the MFG simulation. If they are not equal, this means that the simulation is non-additive. This could occur when changes to mixtures of complex chemistry,

weather situations and emissions take place. For this reason we compare the sum of the trend in the estimated contributions to the MFG trend. Second, we quantify the contributions to the trend in the MFG simulation, which may differ from the reanalyzed trend. Thus we will compare the reanalyzed trend and the base case trend to make sure the base case simulation does not deviate too strongly from the reanalysis results. If the deviation is too large, i.e. the modelled trend is far from the observed, this means that it is non-representative. Such discrepancies could arise from over-sensitivity in MATCH to one process and insensitivity to another, compared to the real world, or imperfections/artificial trends in the input data such as erroneously estimated emissions or erroneous assumptions on the trend in hemispheric background concentrations. If either is true (non-representative or non-additive) for the trend in a specific metric, such as the trend in the January mean, then our method cannot be used to explain that specific trend.

2.6 Evaluation

We evaluate two aspects of the reanalysis. The first is an independent evaluation for a single year with focus on the data assimilation method. The second is an evaluation of the simulated near-surface O_3 concentration trend over the period and our ability to explain the causes of the trend.

For an independent evaluation of data assimilation method we conduct a cross validation at the included Swedish measurement sites. With this method we exclude one measurement site at a time from the data assimilation, and use the analysis results from the excluded location in the evaluation of all sites. This means we conduct one such 2dvar simulation for each considered measurement site. Due to the large amount of computation involved we evaluate one year only by this method. We choose the year 2013, which is when the data coverage is the largest. This means that we have the opportunity also to investigate whether we see any difference in performance between the reanalysis with the larger number of measurement sites (ALL) and the long-term reanalysis (LONGTERM). The evaluation metrics used here are mean value (mean), standard deviation (σ), model mean bias normalized by the observed mean (%bias), Pearson correlation coefficient (r) and the root mean square error (RMSE), see Supplement Sect. S1.

Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2017-338
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Discussion started: 20 June 2017

For the evaluation of the long-term trend we focus on the two critical points raised in the previous section: 1/ the additivity of the trend in the contributions as compared to the trend in O_3 concentration from the MFG simulation, and 2/ whether the MFG trend is representative of the O_3 concentration trend in the LONGTERM reanalysis results. We focus this investigation on 11 different percentiles of hourly mean O_3 concentrations, for an estimate of the scores at different concentration levels. We focus specifically on averages over the three Swedish regions North, Central and South (cf. Fig. 3), to investigate whether there is any variation in performance in Sweden. The comparisons are presented as scatterplots in Fig. 5 and compared to the 1:1 line, factor 2 line and equal sign quadrants.

Additional evaluation and comparisons of the temporal variation over the whole period is included in the Supplements for the two reanalyzes LONGTERM and ALL, the MATCH simulation MFG and observed annual mean (see Supplement Sect. S2 and Figs. S2-S4 and Table S1).

3 Results

3.1 The performance of the model simulations and reanalyzes

Before turning to the evaluation results, we investigate whether the two ozone reanalyzes differ. We do this by comparing time series of annual O_3 metrics for the two data sets. The investigation is presented in the Supplements and shows deviations in the latter years as the number of sites in the ALL data set increases beyond the sites included in the LONGTERM data set (see Supplement Sect. S3 and Figs. S2-S4). The deviation in annual mean near-surface O_3 concentration is larger than for annual maximum 1 hour mean given that many of the newer sites are sensitive to night-time inversions. Due to the visible deviation in results, we use the LONGTERM for the trend and statistical analyzes in the paper, whereas both are used for the evaluation of the 2dvar-method in this section. Both are included in the method evaluation because the evaluation scores may be dependent on the density and specific locations of the measurement sites. The ALL data set is to be used as a best estimate of geographically resolved near-surface O_3 concentrations for Sweden for a subset period within the full period 1990-2013.

In Table 2 we show the evaluation statistics from the validation of hourly near-surface O_3 in 2013. The near-surface O_3 concentrations from the MFG simulation compare well with

observations, and the 2dvar-technique leads to improvements. The spatially averaged correlation coefficient of hourly near-surface O_3 concentrations (se Supplement Sect S1 increases from 0.67 when comparing the MFG O_3 concentrations to observations, to 0.76 when comparing the ALL reanalysis independently to observations through a cross validation (Table 2). The %bias decreases from 1.4 to -0.3 and the RMSE is also improved in the independent evaluation of the ALL reanalysis. Similar improvements are also obtained when using fewer measurements (LONGTERM, Table 2), showing that the method is stable with the number of measurement sites. The cross validation spatial error (RMSE) is however larger than that obtained when evaluating the MFG simulation against independent observations, where the cross validation results indicates that the 2dvar reduces the quality of the annual mean spatial variation in 2013. The lowest annual means in 2013 (Supplement Table S2) are found in the sites Rödeby, Aspvreten, Östad, Norr Malma and Asa, where the annual means are below 30 ppb(v). The highest annual means are found in Esrange, Norra Kvill, Råö and Vavihill. This is likely caused by strong night-time inversions in the sites with lower annual means. These night-time inversions depend to a large extent on local topography, and are not uncommon in inland sites positioned at a low altitude in the local landscape compared to the average of the surrounding area. This variation occurs at a higher resolution than is captured by the MFG simulation (44km resolution). Simultaneously the correction of the model by the data assimilation based on the differences between the model and the measurements, results in readjustments of the model results for the surrounding area and specifically for other sites not affected by night-time inversions. This is illustrated in the Supplements (Fig. S5). Overall, the independent cross validation shows that the 2dvar method improves the performance of the modelled hourly mean O_3 compared to the MFG simulation. This is true not only in the measurement sites, but also elsewhere, with exception for the spatial variation in annual mean.

In Fig. 5 we compare trends in annual near-surface O_3 percentiles over the period 1990-2013 for the MFG simulation, the LONGTERM reanalysis and the sum of contributions. Investigating the additivity of the four contributions (bound, meteo, Se emis and Eur emis), we compare the O_3 concentration trends in the MFG simulation to the trend in the sum of the contributions (SUM, Fig. 5a). Almost all values fall close to the 1:1 line. Only a few of the very weakest O_3 trends fall outside the factor 2 lines. Thus, the contribution experiment can be used to explain the MFG O_3 trend. Comparing the LONGTERM and MFG trends in near-surface O_3 (Fig. 5b), the values are within a factor of 2 for most percentiles and regions. There

is a general tendency for the positive MFG trends to be stronger than the reanalyzed trend (LONGTERM). The largest deviations in the O_3 trends are in the North and the relationship between these two is not as linear as in the other two regions. Most of these trends are however not significant. This demonstrates the added value of the measurement model fusion, where errors in the modelled trend are corrected by the analysis. The deviations are small enough to conclude that in most cases the MFG is representative, showing that the MATCH model can be used to understand the trends in the LONGTERM data set.

In conclusion we have shown that the MFG performs well for hourly near-surface O_3 concentration and the 2dvar analysis improves the performance to almost perfect correspondence to the measurements in the measurement locations, and improved performance elsewhere (cf. the cross-validation), with the exception of the spatial variation. There is an added value of a reanalysis when investigating the trend of near-surface O_3 concentrations. The MATCH model can be used to investigate the causes to the reanalyzed O_3 trend. In the North the trends in the reanalyzed and the MFG O_3 concentration deviates by more than a factor of 2 for some percentiles. We will focus on this deviation more in the final discussion (Sect. 4).

3.2 Reanalyzed near-surface ozone in Sweden 1990-2013

The mean 1990-2013 seasonal variations in monthly mean and monthly maximum of 1h mean near-surface O_3 are presented in Fig. 6, averaged over the three regions: North, Central and South (as defined in Fig. 3). Spatially resolved statistics for annual mean and annual maximum of 1h mean near-surface O_3 are provided in Fig. 7. Time series of annual percentiles averaged over the three regions are shown in Fig. 8.

3.2.1 1990-2013 period statistics

The near-surface O_3 in Sweden exhibits a seasonal variation, which peaks during spring (Fig. 6). In the North the seasonal maximum concentration occurs in April, whereas it occurs later, in May, in the regions further south. The earlier peak in the North, as compared to the South, was also shown by Klingberg et al. (2009) for in situ observations. In the North, the seasonal peak in monthly mean O_3 concentration is higher than the corresponding seasonal peaks in the other two regions, and this is a feature throughout the whole winter half-year: the monthly

mean O_3 concentrations are higher in the North than the more southerly regions during Oct-April. During the summer, the monthly means are higher in the South than in the other two regions. This leads to a 24-year period mean value (Fig. 7) that is highest in the northerly mountains and lowest in central Sweden. This pattern is also supported by Klingberg et al. (2009) based purely on observations, but including a larger number of observation sites through the inclusion of passive diffusion samplers.

For the period mean seasonal variation in monthly maximum 1h mean near-surface O_3 (Fig. 6b) there is a similar seasonal peak in April-May, but there is also a secondary peak during summer (in August). The further south the higher is the monthly maximum 1h mean near-surface O_3 during March-October. This applies to both the primary and the secondary seasonal peaks in monthly maximum. The 24-year period mean of the annual maximum of 1h mean near-surface O_3 (Fig. 7) is lower in central Sweden than in the South and the North, and it is highest in the South.

The lower period mean of the near-surface O_3 in the South than in the North is possibly caused by night-time inversions at some of the southerly sites, and also therefore the reason for the opposite gradient for the annual maximum 1h mean as compared to the annual mean. The difference in spatial pattern between the south, central and northern parts of Sweden is why we choose the three regions defined in Fig. 3. The period maximum of the annual means and period maximum 1h mean near-surface O_3 concentrations have similar spatial variation as the period means (Fig. 7). The overall 24-year maximum 1h mean near-surface O_3 reaches above 240 μg m^{-3} in isolated parts of the South, and is generally above 180 μg m^{-3} in the south and 130 μg m^{-3} in the central and northern part of Sweden.

3.2.2 Trend over the period

Seasonal variations are also present in the trend of both monthly mean and monthly maximum 1h mean near-surface O_3 concentrations (Fig. 6). Monthly means increase strongly during winter and spring (approx. Nov-April), and decrease moderately (North) or strongly (Central and South) during summer (May-Aug). The trends in monthly maximum 1h mean follow a similar pattern. Generally, the rate of change is stronger or at the same level in the Central and South as compared to the North. The strongest decrease is in the August maximum 1h mean in the South and Central, and the strongest increase is in the March monthly mean. The day of the year when the annual maximum 1h mean near-surface O_3 occurs shifts to earlier in the

year in the later part of the period, although there is large inter-annual variation, which is stronger in the South than in the North (Supplements Fig. S6).

The annual mean near-surface O_3 (Fig. 7d,e) increases almost everywhere in Sweden over the time period. The trend is however only significant in restricted parts of Central and South regions, due to considerable inter-annual variation in the areas with the highest trend. The annual maximum 1h mean near-surface O_3 (Fig. 7i,j) is significantly decreasing in South and Central regions, whereas the change in the North is a mixture of increase and decrease, and it is without significance in most areas. The decrease in the South and Central annual maximum 1h mean is a result of the strong decrease in the summer-time O_3 maximum; in the beginning of the 24-year period, the southern summer-time maximum is more often the annual peak rather than the spring-time maximum, whereas the summer-time maximum is more often secondary to the annual maximum in the end of the period. The annual maximum 1h mean is shifted to earlier in the year (Supplements Fig. S6). In a study of four rural European sites and one in western United States, Parrish et al. (2013) showed that not only are springtime O_3 concentrations larger in recent years than in earlier decades, but also that the seasonal maximum now also occurs earlier, as in our results for Sweden. This change in seasonal cycle is also supported by the work by Cooper et al. (2014). The change in the annual maximum 1h mean near-surface O_3 from summer-time peak to spring-time peak means that more than one process can be the cause of the change (increasing spring-time and decreasing summer-time).

We proceed by investigating the trend in annual percentiles of hourly near-surface O_3 concentration, averaged[2] over the three Swedish regions (cf. Fig. 3). The temporal evolution of 11 percentile levels from the 0^{th} (annual minimum 1h mean) to the 100^{th} (annual maximum 1h mean) are shown in Fig. 8, and the corresponding trends with indication of significance levels are recaptured in the Supplements (Table S3). In all three regions the low and medium percentiles are increasing, while the highest percentiles are decreasing from 1990 until 2013. This was also shown by Simpson et al. (2014) based on observations for northern Europe and based on observations for Europe, US and East Asia by Lefohn et al. (2017). Further, using hourly O_3 observations, Karlsson et al. (2017) showed that reduced concentrations were

[2] The percentile is calculated per grid square for all hours in each year, then regional mean annual percentiles are calculated and finally the trend is calculated based on these averaged percentiles.

restricted to the highest O_3 concentrations during summer daytime, while the increase in low and mid-range concentrations occurred during wintertime at both day and night.

In Central and South regions the decrease in the highest near-surface O_3 percentiles are stronger than in the North, and significant and this decrease is evident throughout the maximum 10% percentile range (although the change is not significant for the 90th and 95th percentile levels; cf. Fig 3). This change is mainly caused by decreased high values during the summer-time. In the North, only the annual maximum 1h mean is decreasing and the inter-annual variability is stronger than the rate of change, indicated by the lack of significance for this percentile. The medium and low percentile increase in the North is moderate, but significant, for most percentiles up to the 95th, with very similar rates of change. In the Central and South the change in the low percentiles is highly significant and stronger than in the North. This is an indication that the increase in low near-surface O_3 concentrations cannot only be explained by increasing background. As a result of the decrease of high and increases of low percentiles, there has been a narrowing of the range of the near-surface O_3 concentrations over the period. This was also observed in the US by Simon et al. (2015) for 1998-2013, studying urban and regional background measurements across the US. They interpret this as a response to the substantial decrease in O_3 precursor emissions in the US over the time period. Decreased primary NO emissions results in decreased O_3 titration close to combustion sources, but also reduces local O_3 further away from the emissions sources under weather states favorable for O_3 formation. In the next section we investigate the impact of Swedish and European emission decrease over the period, and relate this to the impact of change in the chemical composition of the hemispheric background and meteorological variations.

3.3 Attribution of the change in near-surface ozone

In this section we quantify the contributions of physical processes to the modelled trend of near-surface O_3 concentration in Sweden during the period 1990-2013. We investigate the impact of the trend in lateral and upper boundaries, meteorological variations and Swedish and European (non-Swedish, full domain) anthropogenic emission change. In Figs. 9 and 10 the contributions to the trend in seasonal variations and percentiles are quantified for the North and South regions.

We start our attribution by analyzing the impact of changing hemispheric background levels of relevant chemical species ("bound" bars in Figs. 9 and 10). These show an increase in monthly mean and maximum 1h mean throughout the year and for all percentiles, mainly as a result of our assumption of an increasing O_3 concentration trend in the lateral and upper boundaries during the 1990s and constant boundary conditions for O_3 during the rest of the period. There is a seasonal variation in the trend of the boundary contribution, with a minimum during summer. This variation is likely a result of an O_3 destruction process that is stronger during summer than winter, such as dry deposition to vegetation and photolysis of ozone. The seasonal variation in the contribution to the trend from the boundary impacts both monthly mean and maximum 1h mean. Our representation of the trend in the concentration of species at the model domain boundary is climatological. The climatological upper boundary means that the inter-annual variations in near-surface O_3 are likely underestimated in remote locations. The impact on inter-annual variations may be largest at high altitudes or far away from the major anthropogenic sources. Hess and Zbinden (2013) showed the importance of the stratospheric contribution to the inter-annual variation at Mace Head and Jungfraujoch; it is possibly also important in the north of Sweden, especially in the mountainous areas. Such variation is not captured by the boundary settings, but it is indirectly included in the reanalyzes data sets through the variation in the measurements included in the data assimilation. As a consequence, the MFG and "bound" simulations underestimate the inter-annual variability as compared to observations and the reanalysis (cf. Table 2), and this could also affect the "bound" trend.

The impact of meteorological low-frequency variations ("meteo") during the 24 years is also an important factor, but more difficult to interpret. The meteorological variation acts to cause a positive trend in near-surface O_3 concentration for most monthly means and maxima, as well as for most percentiles. The meteorological influence on the trend is as large as the impact of the change in boundary, for most percentile levels in the South, while it is weaker for most percentile levels in the North.

During the period 1990-2013 both European (full domain, non-Swedish) and Swedish emissions have decreased strongly. There is a strong seasonality in the impact of the decreasing European emissions, and the contribution to the trend of the Swedish emissions follows the same pattern but with smaller magnitude (cf. Fig. 9, "Eur emis" and "Se emis" respectively). During summer the decreasing emissions have acted to lower both the monthly

mean and maximum 1h mean. During winter the trend in monthly maximum 1h mean is unaffected by the change in emissions, indicating that the highest near-surface O_3 concentrations during winter are due to other sources than local O_3 production. Emission decreases have acted to cause increases in monthly mean near-surface O_3 concentrations in the winter, due to reduced O_3 destruction by primary NO emission. Trends in percentiles (Fig. 10), show that the emission decrease has caused decreases to percentiles higher than the 50th level, and increases below. The impact is stronger in the South than in the North, which is expected due to the South being closer to the European continent. The contribution of the trend in emissions is often stronger than the changing boundary, e.g. in the South for most percentiles and for monthly maximum 1h mean during the summer half-year in both regions. Thus, the observed increase in low and medium near-surface O_3 levels is caused by a mixture of both changes to the hemispheric background levels and emission reductions of O_3 precursors, while the decrease in the high percentile levels is mainly caused by emission decrease.

3.4 Implications for health and vegetation impacts

For the protection of vegetation, the target value by EU (EU directive 2008/50/EC) states that the 5-year mean AOT40 (near-surface O_3 concentration above 40 ppb(v) accumulated over May-July; AOT40c) must not exceed 9 ppm(v) h, and as a long-term goal AOT40c must not exceed 3 ppm(v) h during a calendar year. For protection of human health the target value by EU (EU directive 2008/50/EC) states that the daily maximum running 8 hour mean near-surface O_3 concentration must not exceed 120 μg m^{-3} more than 25 days per year as a 3-year mean, and as a long-term goal the daily maximum of 8h mean near-surface O_3 concentration must not exceed 120 μg m^{-3} at all. Sweden has formulated 16 environmental quality objectives, including clean air, alongside specifications to help reach these objectives. The following specifications are currently valid for near-surface O_3 concentration in Sweden (NV, 2015): the hourly mean must not exceed 80 μg m^{-3}, the daily maximum 8h mean must not exceed 70 μg m^{-3} and AOT40f (O_3 concentration above 40 ppb(v) accumulated over April-September) must not exceed 5 ppm(v) h. In Table 3 we present the linear trends in our reanalysis data set for these metrics, and have collected geographically resolved statistics, such as the period mean, maximum and linear trend in the Supplements (Figs. S7-S11).

The narrowing of the O_3 concentration range, especially through increasing lower percentiles, can impact human and vegetation exposure to O_3. The effect metrics based on accumulation of values above a threshold (AOT40c; AOT40f; SOMO35, Sum of Ozone Means[3] Over 35 ppb(v)) and the number of days with daily maximum of 8h mean near-surface O_3 concentration exceeding 120 μg m^{-3} have been decreasing over the period in the South and Central regions, as have the highest values in the year. This is in accordance with the decrease in the highest percentiles in these regions (cf. Supplements Table S3). Conversely, the metrics with lower threshold values increase, such as the number of hours exceeding 80 μg m^{-3} and the number of days with daily maximum 8h mean near-surface O_3 concentration exceeding 70 μg m^{-3}. This increase is significant in the North, whilst it is not significant in the South and Central. This agrees with the change in medium and low percentiles. A continued increase in low values would cause a continued increase in these metrics, and would eventually reverse the decreasing trend to an increase. This is valid specifically for those metrics with accumulation of values or higher thresholds, such as SOMO35 and AOT40c.

The highest near-surface O_3 concentrations, associated with short-term (acute) health impacts, show a clear and significant decrease in the South (where the highest values occur), leading to an improvement in health impacts. For long-term health effects, there is no established threshold below which there are no adverse effects, even if SOMO35 often is used. The increase in low values (and e.g. the annual mean) has negative impacts on health, although SOMO35 is decreasing in the South and Central Sweden. This increase is also of concern given that policy choices will cause further reductions in local NO emissions – which are highly correlated to where people reside – thus increasing the sensitivity of O_3 to the background and hemispheric background level. Despite this, the solution is not to reverse policies that reduce local NO production, given that this would negatively impact both the highest values and the hemispheric background. A solution must therefore be sought via international policy regulations.

[3] For SOMO35 the Mean is defined as the daily maximum of running 8h mean near-surface O_3 concentrations and the accumulation is over a year unless otherwise is stated.

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Atmospheric Chemistry and Physics Discussions

4 Discussion

This work improves upon previous studies by investigating the trends in near-surface O_3 concentration via a combination of both observed and modelled knowledge. The respective advantages of modeling (geographical and temporal coverage) and observations (the most reliable O_3 concentration estimate at a discreet point) can be exploited through data assimilation to reach a greater understanding of the atmospheric state, and the model can further be used as a tool to explain what is described.

Our results should, however, also be viewed in the context of their limitations. The model simulations have a relatively coarse horizontal resolution, meaning that processes that are more local in origin are not captured by the model – these include the role of local topography or coastal climate for the night-time boundary layer stability (Klingberg et al., 2011), or local emission sources. As a result, the data assimilation scheme will spread such features to parts of the model where they are not valid. Some of the southerly sites in the data assimilation are known to experience night-time inversions and the reanalysis will thus be affected by this. We choose however not to exclude these data from the data assimilation, on the basis that this is restricted to occasional events during night-time. An improvement in the spatial resolution of the model would improve the spatial representation of the analysis, since the difference between observation and model has the potential to decrease at these observation sites.

As with all modeling studies, the model cannot perform better than the quality of the forcing input data. Knowledge of emissions in the beginning of the 24-year period is less comprehensive than at the end, which could introduce artificial trends to the MFG. The trends in lateral and boundary conditions are taken from the work by Engardt et al. (2017) and are based on observed trends at the lateral boundaries of Europe. The upper boundaries are especially poorly represented, and as a consequence so is the stratospheric contribution to the inter-annual variation and trend. The data assimilation reduces the impact of errors in the lateral and upper model boundaries. However, the reanalysis may still be affected in regions with sparse measurement coverage. This can affect the attribution to the trend. In this study the MFG simulation captures the observed (reanalyzed) trend reasonably well, but there is a discrepancy between the reanalysis and MFG trend for most percentile levels in North Sweden. To investigate this in more detail, we have compared the error in trend by percentile (the difference between the trends in MFG and LONGTERM) to the trend caused by the four contributions (bound, meteo, Se emis and Eur emis). The resulting figure is included in the

Supplements (Fig. S12). There is a 1:1 relation between the impact of the trend in the European emissions and the deviation between the MFG and the LONGTERM trends. This could be caused by overestimation of the European emissions trend. A similar tendency is seen for the Swedish emission contribution in the Central and South regions. This calls for emission inventories to be improved in order to make sure the trend in ozone precursor emissions is correct. Another reason for this could be too strong model sensitivity to the European emission trend in the North. If this was true, it would have implications for sensitivity studies that consider the future development of near-surface O_3. In studies relating the impacts of future climate change to future anthropogenic precursor emission change, a robust conclusion for most models is that the impact on near-surface O_3 concentration of future precursor emissions is much stronger than the impact of climate change (e.g. Engardt et al., 2009, Langner et al, 2012; Watson et al., 2016). If models are too sensitive to trends in emissions in remote areas, compared to other processes, such a conclusion might change. Parrish et al. (2014) also compared observed and modelled trends and found that the three chemistry-climate models studied failed to reproduce the observed trends – the modelled O_3 concentration trend was approximately parallel to the estimated trend in anthropogenic precursor emissions of NO_x, whilst observed O_3 concentration changes increased more rapidly than these emission estimates. This implies that there is a lack of knowledge relating to controls of concentrations of tropospheric O_3. Whether it is the trend in ozone precursor emissions or the model sensitivity to emissions that need improving is left for future studies.

Finally, we conducted a trend analysis of the reanalyzed near-surface O_3 using linear regression. We have chosen to present the trend in the LONGTERM data set in all analyzes, regardless of whether it is statistically significant or not. We stress that a trend contains valid information even where it is not statistically significant – and it will become significant if the change and variability remains the same over time. We also recognize that there are other methods of investigating the statistical behavior of the data set, and therefore welcome further use of the data, which may be accessed upon request from the corresponding author.

5 Conclusions

- We have constructed two hourly reanalyzes of near-surface O_3 for Sweden for the period 1990-2013: one time-consistent reanalysis and one using all available hourly

measurements. Both data sets are available upon request from the corresponding author.

- We have evaluated the performance of the reanalyzed near-surface O_3 and mainly found improved performance compared to the MATCH model.
- Our results show:
  - High near-surface O_3 concentrations in Sweden are decreasing and low O_3 concentrations are increasing.
  - Health and vegetation impacts due to high near-surface O_3 concentrations (quantified by policy related threshold metrics) have decreased in Sweden as a result of the decrease in the highest ozone values.
  - Decreasing emissions in Europe have led to decreasing summer-time near-surface O_3 concentrations, as well as a decrease of the highest concentrations.
  - The rising low concentrations of near-surface O_3 in Sweden are caused by a combination of rising hemispheric background O_3 concentrations, meteorological variations and O_3 response to European O_3 precursor emission regulation.
  - There is a discrepancy between modelled and observed (reanalyzed) O_3 trends in northern Sweden. This could be caused by erroneous trends in the historical anthropogenic ozone precursor emissions used here or that our model is too sensitive to changes in emissions. If the latter is true, it implies that the evolution of future precursor emissions may have a smaller impact on future near-surface O_3 concentrations than shown by earlier studies.

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Discussion started: 20 June 2017

Atmospheric Chemistry and Physics Discussions

Figure and table legends

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Figure 1. A flow-chart of the relevant part of the MATCH Sweden system for this reanalysis study.

Figure 2. (a) Secular trend of factors used for scaling boundary concentration of relevant species. Note that the hemispheric background ozone concentrations are assumed constant from 2000 onwards. CO and NMVOC boundaries are held constant throughout the simulation. (b) Temporal trend of total domain (solid lines; left vertical scale) and Swedish (dashed lines; right vertical scale) annual O_3 precursor emissions utilized by MATCH from 1990 to 2013. Emissions of nitrogen oxides (NOx), non-methane volatile organic compounds (NMVOC), carbon monoxide (CO) and biogenic isoprene (C5H8) are indicated by different colors (cf. legend); emissions of sulfur oxides (SO_x) and ammonia (NH_3) are excluded from the panel.

Figure 3. Instrumentation sites for hourly near-surface ozone concentration observations in Sweden and Norway, which are used in the variational analysis. Red circles: sites with full data coverage. Blue circles: sites with restricted data coverage. The subdivision of Sweden into three regions (North, Central and South) follows county borders, as indicated by the fat black lines.

Figure 4. Data availability at instrumentation sites for hourly near-surface ozone concentration observations in Sweden and Norway. Red squares: years with at least 80 % annual data for sites with full data coverage (see also Fig. 3). Light red: sites with <80 % annual data (data capture indicated in square) for sites with full coverage. Blue and light blue squares: as for the red squares, but for sites with restricted data coverage.

Figure 5. Trends in near-surface ozone percentile levels averaged for the three regions North (blue), Central (green) and South (magenta) for the sum of the contributions to the trend (SUM) vs the MATCH model simulation MFG (a) and the MATCH model simulation MFG vs the reanalysis LONGTERM (b). Filled circles indicate significant trends ($p \leq 0.05$) in the MFG simulation, whereas non-significant MFG trends ($p > 0.5$) are indicated by an empty circle. 1:1 line in black, factor 2 lines in dark grey and equal sign quadrants are separated by light grey lines.

Figure 6. Seasonal cycle of monthly mean (a) and monthly maximum of 1h mean (b) near-surface ozone concentrations averaged over the period 1990-2013 (solid lines; left vertical scale) and the linear trend over the same period of the respective monthly values (dashed lines; right vertical scale) in the three regions North, Central and South Sweden (cf. Fig. 3). The corresponding regions are referred to by different colors, see legend. Results from the LONGTERM reanalysis.

Figure 7. Statistical properties of the annual mean (top row; (a)-(e)) and annual maximum 1h mean (bottom row; ((f)-(j)) near-surface ozone concentration. In the columns from left to right: 1990-2013 mean ((a),(f)), 1990-2013 maximum ((b),(g)), 1990-2013 standard deviation ((c),(h)), linear trend over the period 1990-2013 ((d),(i)) and significance in the linear trend over the period ((e),(j)). Results from the LONGTERM reanalysis.

Figure 8. Temporal variation of annual percentiles of near-surface ozone concentrations averaged over the three regions North (a), Central (b) and South (c) of Sweden (cf. Fig. 3). The line marked 0 is the zero-percentiles (lowest hourly mean near-surface ozone concentration of the year), 100 is 100-percentile (highest hourly mean near-surface ozone concentration of the year), 50 is the 50-percentile (i.e. annual median of the hourly mean near-surface ozone concentration). The sign of the corresponding linear trend (cf. Supplements Table S3, including a statistical analysis of the trend) of each percentile is indicated by colour: a negative linear trend over 1990-2013 is indicated by grey symbols; a positive trend by orange symbols. Statistically significant trends ($p \leq 0.05$) are indicated by thick lines. Results from the LONGTERM reanalysis.

Figure 9. Linear trend over 1990-2013 in monthly mean ((a),(c)) and monthly maximum 1 hour mean ((b),(d)) near-surface ozone concentration for the North ((a),(b)) and the South ((c),(d)) Swedish regions (cf. Fig. 3). Reanalyzed (white diamond; LONGTERM reanalysis) and modelled "first guess" (MFG) near-surface ozone trend (green diamond), and modelled contributions to the near-surface ozone trend due to change in emissions: anthropogenic Swedish (dark blue, Se emis) and full domain, non-Swedish (fair blue, Eur emis), emissions, trend in top and lateral boundaries of relevant species (yellow, bound) and variation in meteorology (brown, meteo). The sum of the modelled contributions is indicated by the dashed green line.

Figure 10. Linear trends over 1990-2013 in annual percentiles of hourly mean near-surface ozone concentrations for the North (a) and the South (b) Sweden regions. Reanalyzed (white

Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2017-338
Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 20 June 2017

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Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2017-338
Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 20 June 2017

Atmospheric Chemistry and Physics Discussions

Table legends

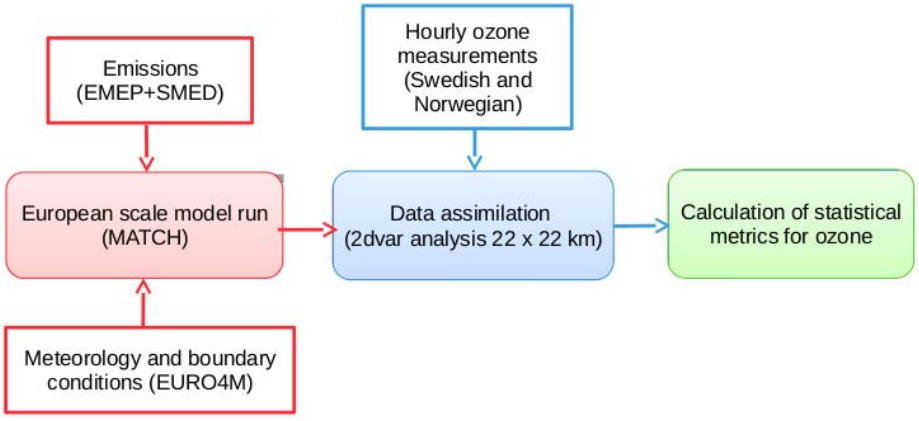
Table 1a. Model calculations and scenarios, all covering the years 1990-2013, including the "first guess" to the data assimilation and base case to the sensitivity simulations (MFG), two reanalysis data sets (LONGTERM and ALL), sensitivity scenarios (MEUR, MSE, MBC and MMET).

Table 1b. Formation of contributions to the linear trend over the period 1990-2013 from the sensitivity simulations (Se emis, Eur emis, Bound and Meteo, see Table 1a).

Table 2. Evaluation of modelled hourly near-surface ozone concentrations in 2013 at Swedish observation sites. Mean value (mean), standard deviation (σ), model mean bias normalized by the observed mean (%bias), Pearson correlation coefficients (r) for data including at least 10 pairs, the root mean square error (RMSE) and number of observed hours at the sites. The evaluation includes the reanalyzed data sets ALL and LONGTERM, where ALL is evaluated at the 12 Swedish sites included in that simulation, and LONGTERM is evaluated at the 6 Swedish sites included in that simulation (cf. Fig. 4). For each of these data set evaluations we include the observation *dependent* reanalysis (2dvar), the observation *independent* cross validation of the reanalysis (cross) and the MATCH base case simulation (MFG). The top half of the table shows the temporal performance (spatial mean of statistics, see Supplement Sect. S1). The bottom half of the table shows spatial performance (spatial statistics of annual means, see Supplement Sect. S1).

Table 3. Linear trend during 1990-2013 of policy related metrics in the 3 Swedish regions North, Central and South (cf. Fig. 3). Stars (*, **, and ***) indicate that the trend is significant ($p \leq 0.05$, $p \leq 0.01$, $p \leq 0.001$, respectively).

Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2017-338
Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 20 June 2017



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Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2017-338
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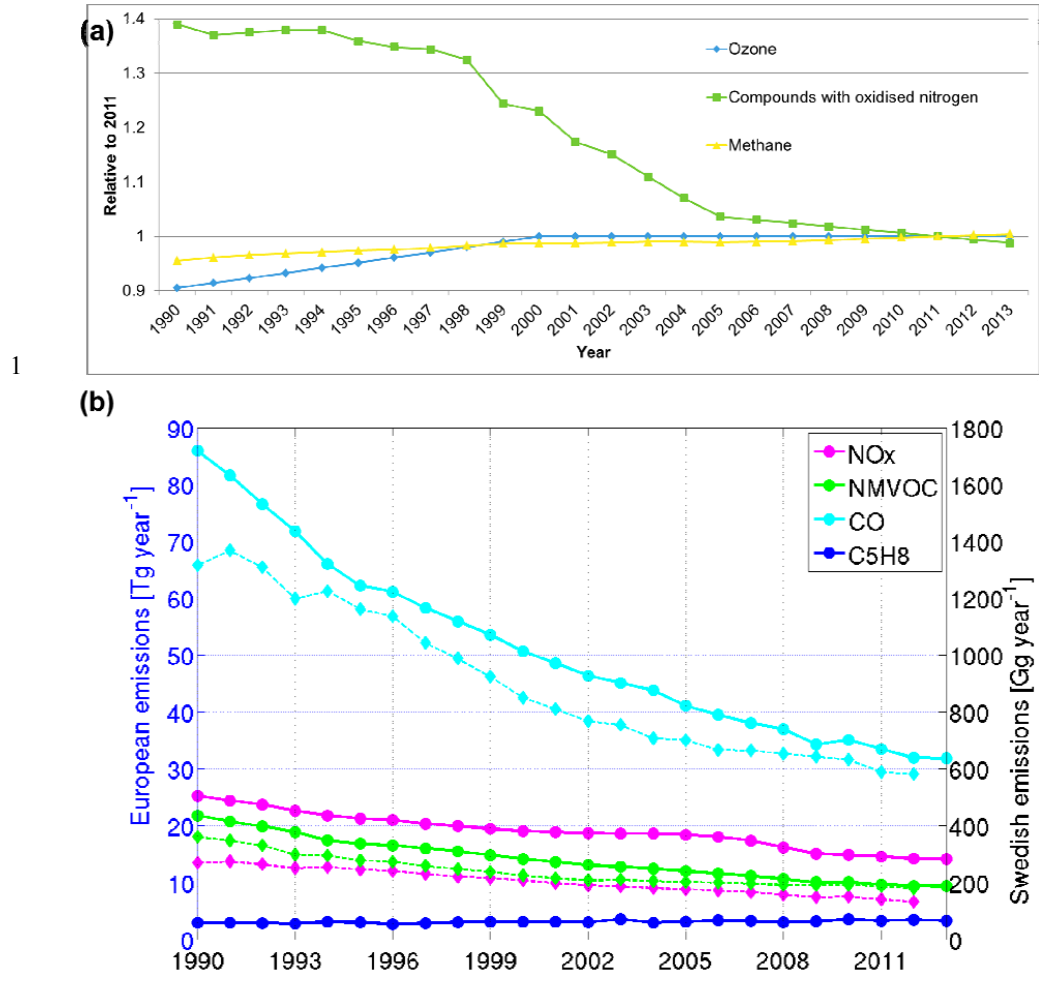
Discussion started: 20 June 2017



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Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2017-338
Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 20 June 2017

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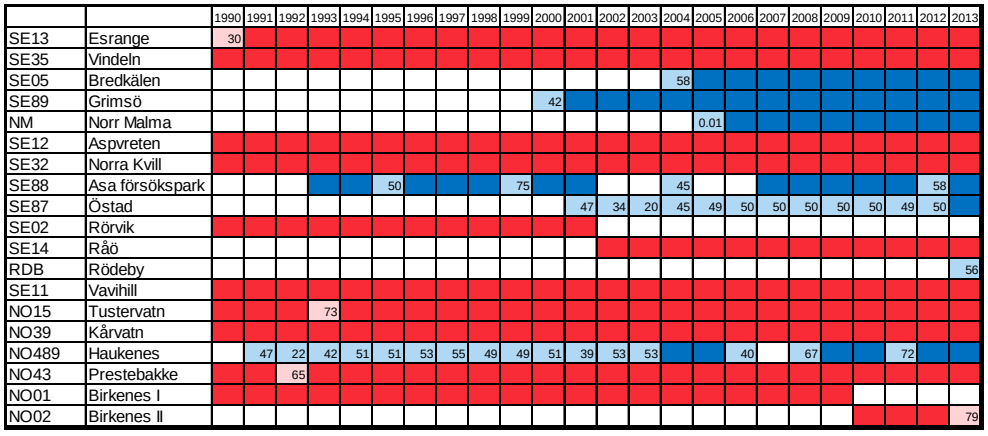
Discussion started: 20 June 2017

Atmospheric Chemistry and Physics Discussions



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Discussion started: 20 June 2017

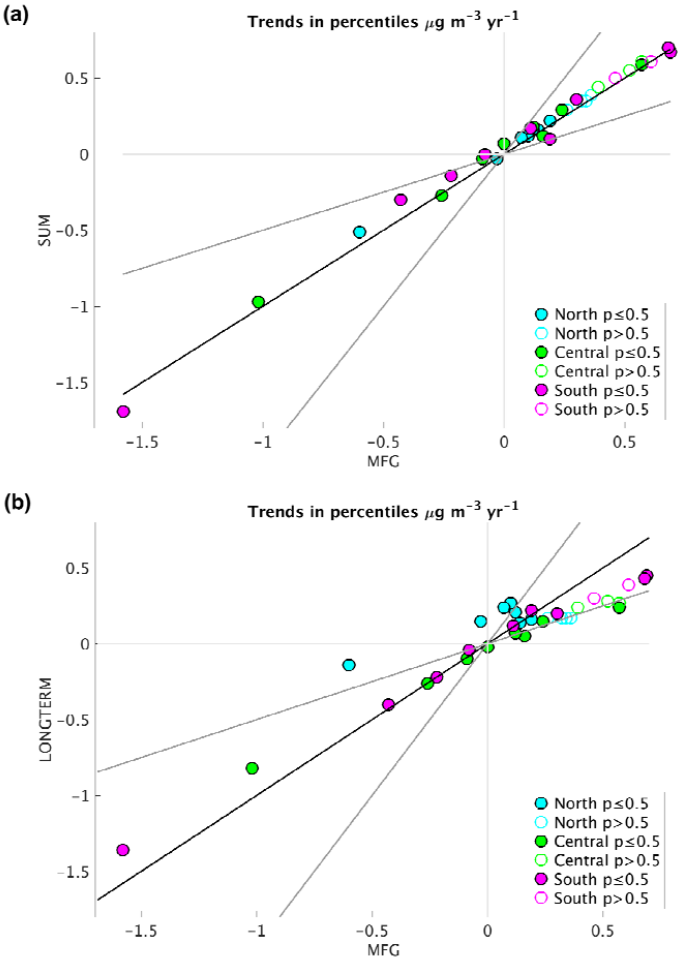

Figure 5. Trends in near-surface ozone percentile levels averaged for the three regions North (blue), Central (green) and South (magenta) for the sum of the contributions to the trend (SUM) vs the MATCH model simulation MFG (a) and the MATCH model simulation MFG vs the reanalysis LONGTERM (b). Filled circles indicate significant trends ($p \leq 0.05$) in the MFG simulation, whereas non-significant MFG trends ($p > 0.5$) are indicated by an empty circle. 1:1 line in black, factor 2 lines in dark grey and equal sign quadrants are separated by light grey lines.

Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2017-338
Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 20 June 2017

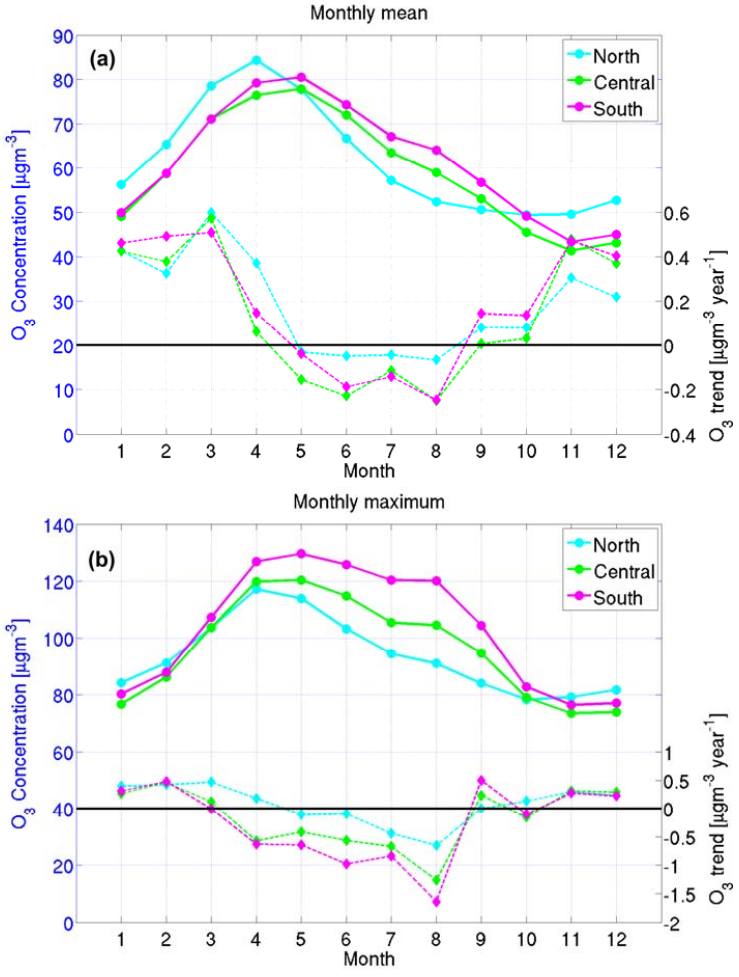

Figure 6. Seasonal cycle of monthly mean (a) and monthly maximum of 1h mean (b) near-surface ozone concentrations averaged over the period 1990-2013 (solid lines; left vertical scale) and the linear trend over the same period of the respective monthly values (dashed lines; right vertical scale) in the three regions North, Central and South Sweden (cf. Fig. 3). The corresponding regions are referred to by different colors, see legend. Results from the LONGTERM reanalysis.

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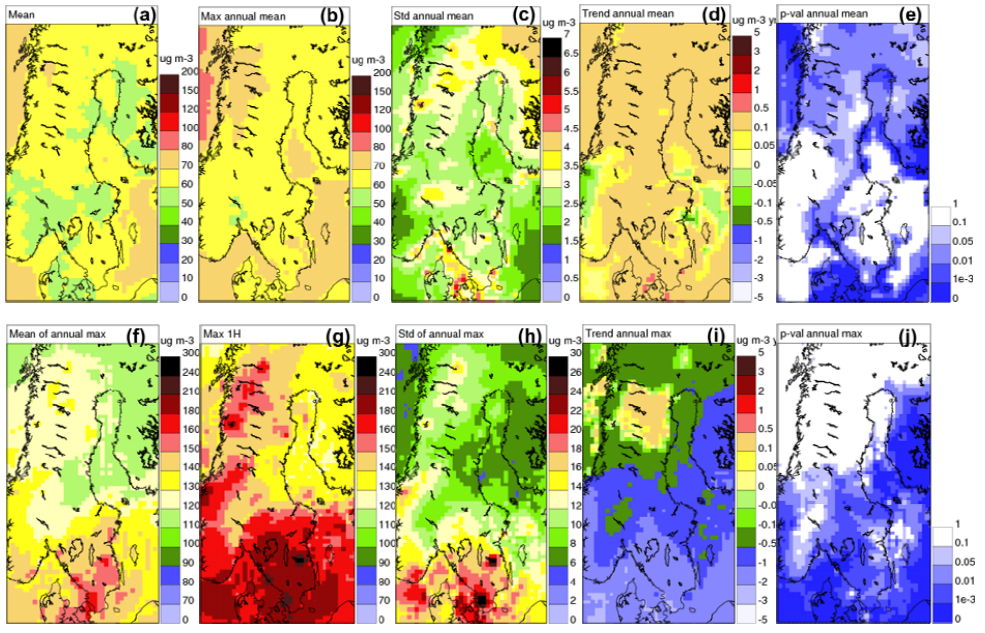

Figure 7. Statistical properties of the annual mean (top row; (a)-(e)) and annual maximum 1h mean (bottom row; ((f)-(j)) near-surface ozone concentration. In the columns from left to right: 1990-2013 mean ((a),(f)), 1990-2013 maximum ((b),(g)), 1990-2013 standard deviation ((c),(h)), linear trend over the period 1990-2013 ((d),(i)) and significance in the linear trend over the period ((e),(j)). Results from the LONGTERM reanalysis.

Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2017-338
Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 20 June 2017

Atmospheric Chemistry and Physics Discussions

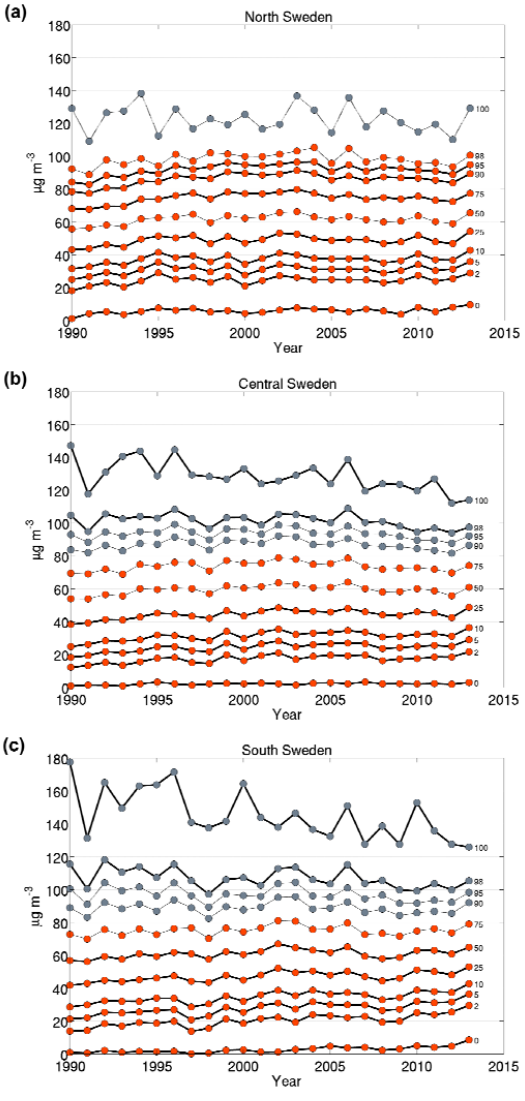

Figure 8. Temporal variation of annual percentiles of near-surface ozone concentrations averaged over the three regions North (a), Central (b) and South (c) of Sweden (cf. Fig. 3). The line marked 0 is the zero-percentiles (lowest hourly mean near-surface ozone concentration of the year), 100 is 100-percentile (highest hourly mean near-surface ozone concentration of the year), 50 is the 50-percentile (i.e. annual median of the hourly mean near-surface ozone concentration). The sign of the corresponding linear trend (cf. Supplements Table S3, including a statistical analysis of the trend) of each percentile is indicated by colour:

Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2017-338
Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 20 June 2017

Atmospheric Chemistry and Physics Discussions

a negative linear trend over 1990-2013 is indicated by grey symbols; a positive trend by orange symbols. Statistically significant trends ($p \leq 0.05$) are indicated by thick lines. Results from the LONGTERM reanalysis.

Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2017-338
Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 20 June 2017

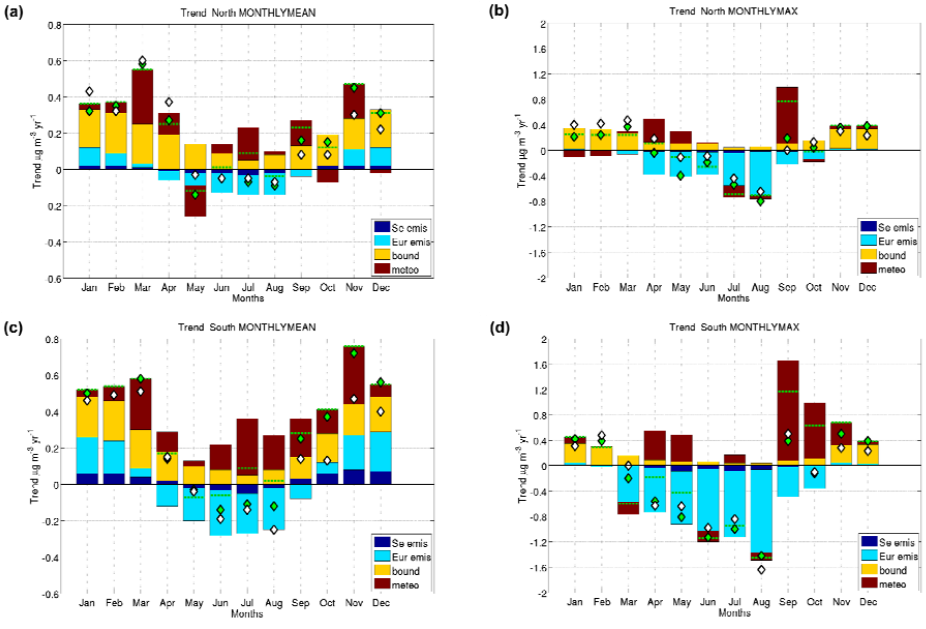

Figure 9. Linear trend over 1990-2013 in monthly mean ((a),(c)) and monthly maximum 1 hour mean ((b),(d)) near-surface ozone concentration for the North ((a),(b)) and the South ((c),(d)) Swedish regions (cf. Fig. 3). Reanalyzed (white diamond; LONGTERM reanalysis) and modelled "first guess" (MFG) near-surface ozone trend (green diamond), and modelled contributions to the near-surface ozone trend due to change in emissions: anthropogenic Swedish (dark blue, Se emis) and full domain, non-Swedish (fair blue, Eur emis), emissions, trend in top and lateral boundaries of relevant species (yellow, bound) and variation in meteorology (brown, meteo). The sum of the modelled contributions is indicated by the dashed green line.

Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2017-338
Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 20 June 2017

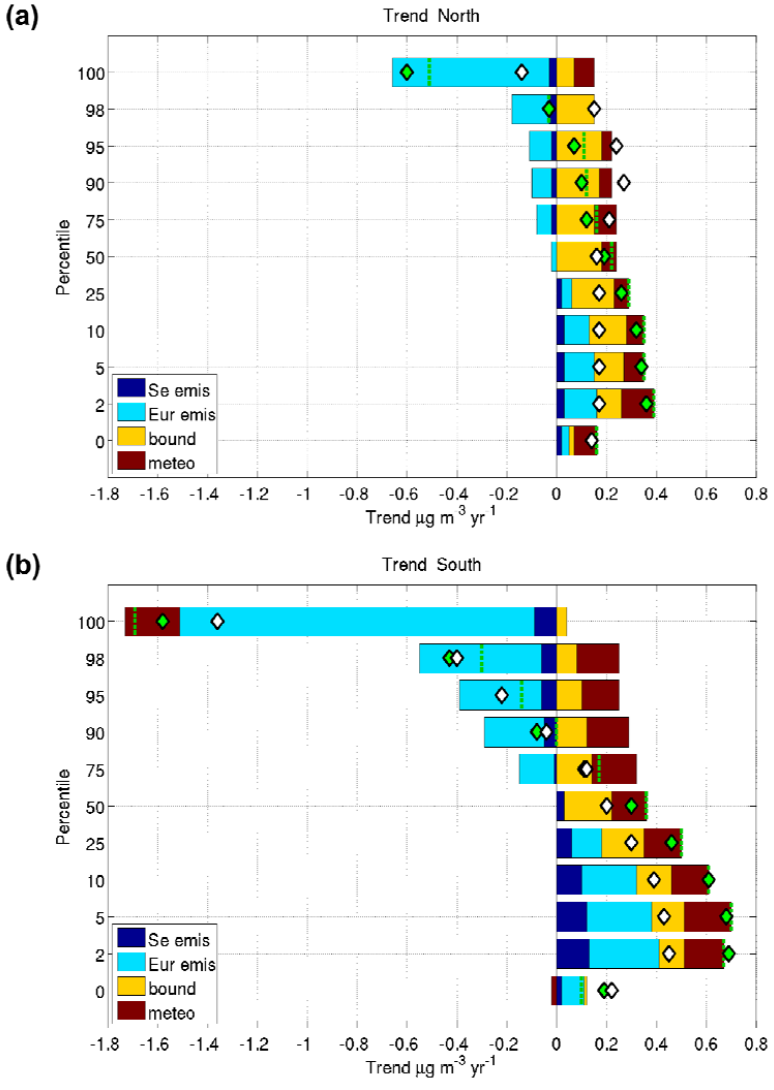

Figure 10. Linear trends over 1990-2013 in annual percentiles of hourly mean near-surface ozone concentrations for the North (a) and the South (b) Sweden regions. Reanalyzed (white diamond; LONGTERM reanalysis) and modelled MFG near-surface ozone trend (green diamond), and modelled contributions to the near-surface ozone concentration trend due to change in emissions: anthropogenic Swedish (dark blue, Se emis) and full domain, non-Swedish (fair blue, Eur emis), emissions, trend in top and lateral boundaries of relevant

Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2017-338
Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 20 June 2017

Atmospheric Chemistry and Physics Discussions

1 species (yellow, bound) and variation in meteorology (brown, meteo). The sum of the
2 modelled contributions is indicated by the dashed green line.
3

Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2017-338
Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 20 June 2017

Atmospheric Chemistry and Physics Discussions

Table 1a. Model calculations and scenarios, all covering the years 1990-2013, including the "first guess" to the data assimilation and base case to the sensitivity simulations (MFG), two reanalysis data sets (LONGTERM and ALL), sensitivity scenarios (MEUR, MSE, MBC and MMET).

Scenario/data set	Description
MFG	MATCH base case simulation and "first guess" used as input to the reanalyzes.
LONGTERM	Reanalysis data set of hourly near-surface ozone concentration covering Sweden and Norway based on 1) the MFG European MATCH simulation and 2) selected hourly near-surface ozone measurements in Sweden and Norway, based on temporal coverage of the measurement sites. Optimal for trend analyses. Analyzed and presented in Sect. 3.
ALL	Reanalysis data set of hourly near-surface ozone concentration covering Sweden and Norway based on 1) the MFG European MATCH simulation and 2) all available Swedish hourly ozone measurements and a selection of the Norwegian (as in LONGTERM). Not used for trend analyses in this study, but best estimate for the hourly near-surface ozone concentration in Sweden at any point in time.
MEUR	MATCH sensitivity simulation where the full domain anthropogenic emissions are kept constant from year to year, set to the level of 2011.
MSE	MATCH sensitivity simulation where the Swedish anthropogenic emissions are kept constant from year to year, set to the level of 2011.
MBC	MATCH sensitivity simulation where the top and lateral boundaries for all species are kept constant from year to year, set to the level of 2011.
MMET	MATCH sensitivity simulation where the meteorology is kept constant, using the meteorological year 2011.

Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2017-338
Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 20 June 2017

Atmospheric Chemistry and Physics Discussions

1 Table 1b. Formation of contributions to the linear trend over the period 1990-2013 from the
2 sensitivity simulations (Se emis, Eur emis, Bound and Meteo, see Table 1a).

Se emis	Contribution to the trend caused by the change in anthropogenic Swedish emissions, calculated as the model scenario difference: MFG-MSE.
Eur emis	Contribution to the trend caused by the change in anthropogenic European, non-Swedish, emissions, calculated as the model scenario difference: (MFG-MEUR)-(MFG-MSE).
Bound	Contribution to the trend caused by the change in lateral and upper boundaries, calculated as the model scenario difference: MFG-MBC.
Meteo	Contribution to the trend caused by the variation in meteorology, calculated as the model scenario difference: MFG-MMET.
SUM	Sum of the contributions to the trend, calculated as the sum of: Se emis+Eur emis+Bound+Meteo.

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Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2017-338
Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 20 June 2017

Atmospheric Chemistry and Physics Discussions

Table 2. Evaluation of modelled hourly near-surface ozone concentrations in 2013 at Swedish observation sites. Mean value (mean), standard deviation (σ), model mean bias normalized by the observed mean (%bias), Pearson correlation coefficients (r) for data including at least 10 pairs, the root mean square error (RMSE) and number of observed hours at the sites. The evaluation includes the reanalyzed data sets ALL and LONGTERM, where ALL is evaluated at the 12 Swedish sites included in that simulation, and LONGTERM is evaluated at the 6 Swedish sites included in that simulation (cf. Fig. 4). For each of these data set evaluations we include the observation *dependent* reanalysis (2dvar), the observation *independent* cross validation of the reanalysis (cross) and the MATCH base case simulation (MFG). The top half of the table shows the temporal performance (spatial mean of statistics, see Supplement Sect. S1). The bottom half of the table shows spatial performance (spatial statistics of annual means, see Supplement Sect. S1).

		spatial mean of hourly statistics					
		mean (ppb(v))	**std dev (ppb(v))**	**%bias (%)**	**r**	**RMSE (ppb(v))**	**#hours**
ALL	**obs**	30.9	11.0				8760
	MFG	31.1	9.4	1.4	0.67	8.8	
	cross	30.6	9.9	-0.3	0.76	8.0	
	2dvar	30.8	11.1	-0.6	0.94	3.5	
LONGTERM	**obs**	32.6	10.5				8760
	MFG	31.2	9.7	-3.3	0.67	8.7	
	cross	32.2	9.3	-0.1	0.72	8.5	
	2dvar	32.6	10.7	0.2	0.97	2.7	
		spatial statistics of annual means					
		mean (ppb(v))	**std dev (ppb(v))**	**%bias (%)**	**r**	**RMSE (ppb(v))**	**#stns**
ALL	**obs**	30.9	2.5				12
	MFG	31.1	1.2	0.6	0.21	3.0	
	cross	30.6	1.8	-1.0	0.11	3.5	
	2dvar	30.8	2.8	-0.5	0.98	0.7	
LONGTERM	**obs**	32.6	2.2				6
	MFG	31.2	1.0	-4.1	X	3.4	
	cross	32.2	1.6	-1.2	X	4.3	
	2dvar	32.6	2.2	0.2	X	0.2	

Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2017-338
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Discussion started: 20 June 2017

Atmospheric Chemistry and Physics Discussions

Table 3. Linear trend during 1990-2013 of policy related metrics in the 3 Swedish regions North, Central and South (cf. Fig. 3). Stars (*, **, and ***) indicate that the trend is significant ($p \leq 0.05$, $p \leq 0.01$, $p \leq 0.001$, respectively).

Metrics	**North**	**Central**	**South**
Mean [µg m^{-3} $year^{-1}$]	**+0.18***	+0.13	**+0.18***
SOMO35 [ppb(v) d $year^{-1}$]	+14	-3.1	-4.7
Maximum 8h mean [µg m^{-3} $year^{-1}$]	-0.11	**-0.68****	**-1.2****
Maximum 1h mean [µg m^{-3} $year^{-1}$]	-0.14	**-0.82****	**-1.4*****
AOT40c [ppm(v) h $year^{-1}$]	-0.01	**-0.07***	-0.09
AOT40f [ppm(v) h $year^{-1}$]	+0.03	-0.09	**-0.12***
#hours >80 µg m^{-3} [# $year^{-1}$]	**+26***	+1.7	+6.6
#days >70 µg m^{-3} [# $year^{-1}$]	+1.3	+0.73	+1.1
#days >120 µg m^{-3} [# $year^{-1}$]	+0.01	**-0.12***	**-0.32****