Norrköping, Sweden
Sept 21, 2017

Dear Prof. Maria Kanakidou,

Hereby we submit a revised version of the manuscript ACP-2017-338, Reanalysis of and attribution to near-surface ozone concentrations in Sweden during 1990-2013, by Camilla Andersson and co-authors.

We found most of the comments by the two anonymous referees to be of much use. Due to that fact we spent quite a lot of time doing additional sensitivity simulations and analyses, as you will see described in the replies. We feel this has added strength to the research and the manuscript has gained additional quality.

We have attached the manuscript and supplements with track changes to allow you to get a full view on all changes made to the manuscript.

We hope you will find it a strong and interesting study well suited for publication.

Best regards,

Camilla Andersson and co-authors

Author reply to Anonymous Referee #1

We wish to thank the referee for insightful comments and questions, as well as technical improvements. The study has much benefited from the additional work performed as a consequence of the major points. Point by point replies follow below to all issues raised by the referee.

Major points:

1. The attribution results for the "bound" and "meteo" simulations. The text in this section needs to be clarified, and it would be highly beneficial to establish the sensitivity of the results to the methodology/underlying assumptions. In section 2.5 the method for attributing trends to meteorology and other factors is given as: "The respective contributions to the trend are formed by subtracting the MFG with the corresponding sensitivity simulation" . How exactly how the contributions are calculated i.e is the difference in O3 between the MFG and the sensitivity simulations first calculated to produce a _O3 and then trend for _O3 or the residual O3 then calculated? Is this trend in _ O3 assumed to be the contribution to the O3 trend as plotted in Figs 9 and 10? Please be explicit about this calculation in the text.

Reply from the Authors (RA, in italics): The contributions the trend is calculated by 1. Forming gridded metrics (annual/monthly), 2. Gridded trends, 3. Regional average trend, 4. Difference between MFG and the corresponding sensitivity simulation. We have clarified this in the method description Sect. 2.5:

"The respective contributions presented in Sect. 3.3 are formed through the following sequence: 1. Calculation of gridded metrics (focusing on monthly 1h maximum, monthly mean and annual 1h percentile levels); 2. Calculation of secular gridded trends over the monthly or annual metrics; 3. Calculation of regional (North, Central, South, cf. Fig. 3) mean of the secular trends, 4. Calculation of the difference between the regional mean trend in MFG and the corresponding sensitivity simulation."

A key question is how does this methodology and assumptions made influence the results? in particular:

a. In particular how sensitive are the results in all sensitivity experiments to the choice of the year 2011?

RA: This is indeed an important question.

It is too much for us to test all 24 years as base year. To restrict the amount of CPU-efforts, we have made a sensitivity test where we use 1990 as base year instead for bound, meteo and emis (where emis SE and emis Eur are combined, this contribution is included in Table 1b). 1990 was chosen as it differs from 2011 both for European emissions and climatologically. For example the NAO index was high in early winter 1990 and low in 2011, whereas the summer index was positive but close to 0 in 1990 and negative in 2011.

We choose to show the differences in contributions to the trend due to base year by scatterplots in Fig 5c and the Supplements (Fig. S5). They show that the contributions are robust for percentiles and most monthly means. Whereas the contributions of variations in meteorology and emissions to the secular trend in monthly max 1h mean differ for some months.

We have included a description on how we conduct this investigation in Sect. 2.5 and 2.6., and present the results in Sect. 3.1.:

"Third, the attribution may be sensitive to the chosen base year. Sensitivity simulations using 1990 as base year instead of 2011 are also conducted, to investigate the robustness of the results. To investigate all 24 years as base years would take too much computational efforts, we choose 1990 as it differs from 2011 both for European emissions and climatologically. If the contributions to the trend differ too much between the base years 1990 and 2011 then the results are not robust. If they are similar it is not a guarantee that the results are robust but it is an indication. The contributions with 1990 as base year are formed in the same way as for the 2011 sensitivity runs. The contributions due to change in top and lateral boundaries (bound) and variations in meteorology are included in the same manner, while we compare the total footprint of the change in emissions, i.e. the sum of FD emis and SE emis (emis) rather than the two parts."

"*Finally, investigating the impact of the selected base year in the sensitivity simulations, we compare the contributions (bound, emis and meteo) based on keeping the year 2011 constant in the sensitivity simulations to keeping the year 1990 constant (Fig. 5c). Most contributions to the trend in percentiles are robust (Fig. 5c), falling close to the 1:1 line. Only a few of the very weakest O_3 contribution trends fall outside the factor 2 lines (for the meteo contribution). The contributions to the secular trend in some of the monthly mean and the monthly maximum 1h mean near-surface O_3 differ more for the two base years than the percentiles (Supplements, Fig. S5). For monthly mean the trend due to changes in meteorology is stronger for some months (one month is weaker) when 2011 is used as base year compared to 1990. The other contributions fall within the factor 2 lines. For monthly maximum the deviation is larger, even differing in sign for the contribution due to variation in meteorology for some months, and a few contributions due to emission change also fall outside the factor 2 lines."*

b. For the "bound" simulations, is the year for constant boundary conditions not important, given the assumption of a constant background of O3 from 2000 onwards? The reader also needs to understand further about the O3 boundary conditions: the source of these boundary conditions and why they are assumed constant after 2000. Would there be a much larger contribution to the O3 trend if the boundary conditions varied after 2000?

RA: There is observational evidence that near surface ozone in Europe has increased up to ca. 2000, with less clear trends after that date (e.g. Cooper et al., 2014). We have therefore selected a conservative approach and let all the ozone concentrations at the boundaries be fixed from 2000 onwards .This is now clarified in the text.

In the present model set-up we also do not have the possibilities to assign different trends at different geographical locations or different trends for different ozone metrics (e.g. maximum concentrations or average concentrations).

If we would have assumed continued ozone increase after 2000 (in opposition to observations) our results would likely shift towards a larger increase (or smaller decrease) of near surface ozone during the period 1990-2013. It is not possible, at this stage, to discuss exactly how that would impact the different ozone percentiles.

c. The authors state that the impact of meteorology is difficult to interpret and note meteorology variations cause a positive trend on O3. This does seems rather unintuitivewhy is there a positive trend due to meteorology, is this because of the methodology, rather than a robust finding? Varying meteorology is usually noted as the cause of difficulties in O3 trend detection (e.g Colette et al. 2016; Lefohn et al. 2017). Hence there may possibly be a larger O3 trend in a simulation with constant compared to varying meteorology, leaving a residual positive trend? Alternatively, there could be a trend in a given meteorological variable over this period that would cause a trend in O3. It would be highly beneficial to investigate the above points to establish why this trend is positive, and as noted in 1) the sensitivity of this result to the constant meteorological year selected should be assessed for any robust statement to be made.

RA: It is indeed a fact that meteorology introduces interannual variations that can cause a trend not to be significant. The trend can also be sensitive to extreme years in the beginning/end of the time series. We have made a few sensitivity tests: i/ using 1990 as base year (see 1b above). ii/ investigating the overall trend over shorter time period periods (1990-2012, 1990-2011, 1991-2013, 1992-2013).

The below table (replica of Table 3 for different periods, investigation of point ii) shows that the trend is not sensitive to removal of 1 or two years in the beginning. We choose not to include this sensitivity analysis since the manuscript is long already.

	1990-2013			1991-2013			1992-2013		
	North	Central	South	North	Central	South	North	Central	South
Mean [μg m^{-3} $year^{-1}$]	+0.18*	0.13	+0.18*	0.14	0.09	0.16	0.07	0.04	0.12
SOMO35 [ppb(v) d $year^{-1}$]	14.32	-3.05	-4.65	10.91	-5.84	-4.2	4.68	-12.15	-10.81
Maximum 8h mean [μg m^{-3} $year^{-1}$]	-0.11	-0.68**	-1.16**	-0.06	-0.61*	-0.99**	-0.23	-0.82***	-1.30***
Maximum 1h mean [μg m^{-3} $year^{-1}$]	-0.14	-0.82**	-1.36***	-0.08	-0.71**	-1.17**	-0.27	-0.94***	-1.50***
AOT40c [ppm(v) h $year^{-1}$]	-0.01	-0.07*	-0.09	-0.02	-0.08*	-0.09	-0.04*	-0.11**	-0.13*
AOT40f [ppm(v) h $year^{-1}$]	0.03	-0.09	-0.12*	0.01	-0.10*	-0.12	-0.02	-0.14**	-0.16*
#hours >80 μg m^{-3} [# $year^{-1}$]	+26.32*	1.7	6.61	20.95	-2.18	5.56	12.3	-8.51	-0.29
#days >70 μg m^{-3} [# $year^{-1}$]	1.31	0.73	1.12	1.03	0.39	1	0.54	-0.03	0.69
#days >120 μg m^{-3} [# $year^{-1}$]	0.01	-0.12*	-0.32**	0.01	-0.11*	-0.28*	0	-0.15**	-0.36**
	1990-2012			1990-2011					
	North	Central	South	North	Central	South			
Mean [μg m^{-3} $year^{-1}$]	0.16	0.13	0.15	+0.21*	+0.18*	0.17			
SOMO35 [ppb(v) d $year^{-1}$]	13.87	-1.99	-7.46	+18.92*	3.56	-4.66			
Maximum 8h mean [μg m^{-3} $year^{-1}$]	-0.23	-0.63*	-1.10**	-0.06	-0.53*	-1.05*			
Maximum 1h mean [μg m^{-3} year-	-0.23	-0.76**	-1.32**	-0.11	-0.64*	-1.26**			

^{1}]									
AOT40c [ppm(v) h year^{-1}]	-0.01	-0.06	-0.09	0	-0.05	-0.09			
AOT40f [ppm(v) h year^{-1}]	0.03	-0.08	-0.13*	0.05	-0.05	-0.12			
#hours >80 μg m^{-3} [# year^{-1}]	+26.03*	1.95	2.6	+32.94*	8.07	5.73			
#days >70 μg m^{-3} [# year^{-1}]	1.14	0.82	0.92	1.53	1.28	1.12			
#days >120 μg m^{-3} [# year^{-1}]	0	-0.11*	-0.32**	0.01	-0.1	-0.31*			

However, we do not agree that it is un-intuitive that the meteorological variations could cause a positive ozone trend in Sweden. Warmer climate could cause more biogenic isoprene emissions in Sweden and elsewhere and more high pressures and transport events of high ozone from the European continent could also contribute. This is in agreement with what was found in the retrospective simulations of Andersson et al. (2007), a positive but not significant trend of ca 0.5% decade due to meteorological variability over the periods 1958-2001 and 1979-2001. It is outside the scope of this study to investigate the reason behind this.

Changes in manuscript: see 1a.

2. The regional-average trends. How sensitive are these trend results to when the averaging is performed in the calculation, especially in the case of calculating trends in O3 percentile ranges? In this study, regional averaging is done after calculating percentiles at each grid box and then a regional trend is calculated, but trends could be calculated for each grid box first and then averages calculated subsequently or the data could be pooled.

RA: We have also calculated the trend per grid box, after calculating the percentiles per grid box (see also Fig. 7 and supplements) and then conducted the spatial averaging to compare this to Table 3. The results are identical, see below.

The trends are the same under these two methods as a result of us using first order linear regression. We do not think it is correct to average the p-value over a region, still we include this as well in the below table (same indication as in Table 3). The two methods result in somewhat different significances, which most often are stronger than those of the trend in the regional average (except for annual mean and number of hours above 80 ppbv).

To calculate pooled trend statistics is outside the scope of this study. We do not include these results as the manuscript is long already.

	Spatial mean of grid box trend			Trend in spatial mean (original Table 3)		
	North	Central	South	North	Central	South
Mean [μg m^{-3} year^{-1}]	+0.18	+0.13	+0.18	**+0.18***	+0.13	**+0.18***
SOMO35 [ppb(v) d year^{-1}]	+14	**-3.1*****	**-4.7*****	+14	-3.1	-4.7

Maximum 8h mean [μg m^{-3} $year^{-1}$]	**-0.11*** **	**-0.68*** **	**-1.2*** **	-0.11	**-0.68****	**-1.2****
Maximum 1h mean [μg m^{-3} $year^{-1}$]	**-0.14*** **	**-0.82*** **	**-1.4*** **	-0.14	**-0.82****	**-1.4*****
AOT40c [ppm(v) h $year^{-1}$]	**-0.01*** **	**-0.07*** **	**-0.09*** **	-0.01	**-0.07***	-0.09
AOT40f [ppm(v) h $year^{-1}$]	**0.03***	**-0.09*** **	**-0.12*** **	+0.03	-0.09	**-0.12***
#hours >80 μg m^{-3} [# $year^{-1}$]	+26	+1.7	+6.6	**+26***	+1.7	+6.6
#days >70 μg m^{-3} [# $year^{-1}$]	+1.3	+0.73	+1.1	+1.3	+0.73	+1.1
#days >120 μg m^{-3} [# $year^{-1}$]	**+0.01****	**-0.12*** **	**-0.32*** **	+0.01	**-0.12***	**-0.32****

3. Spring and summer peaks. A change in when peak O3 occurs throughout the 24-year period is commented on in several places in the text. However, as noted in Fig S6 there is large inter- interannual variability, such that it is hard to make any robust conclusions about shifts in maxima and dominance of spring vs. summer-time peaks over the course of the trend period. Is there any further evidence to illustrate this point?

RA: as the manuscript is long already we remove the discussion on the shift in peak rather than adding more analyses.

4. The data assimilation process. In Figure 1 it seem that the match model simulation feeds into data assimilation, whilst I thought it would be a combination that produces a new simulation. Data assimilation is a process that combines a "background field" or "first guess" with observations to produces new physically consistent model fields. This is usually an iterative process that occurs as the model runs forward. However, the figure makes this appears as a post –processing correction, although section 2.4 discusses a "first guess". If the process is iterative, the figure does not capture this flow and should be revised for clarity. If this is a post-processing effort the text should be revised to state this.

RA: The variational data analysis was conducted as a post processing, i.e. in retrospect. Thus the figure is correct.

We have clarified in the text by refraining from calling it data assimilation and instead calling it (retrospective) variational data analysis. We choose to keep the terminology "first guess" of the model background field to the variational data analysis since we think the text is clear enough by the introduction of these changes.

5. Figure 5 discuses trends in O3 percentile levels. Which percentile levels?

RA: We do not expect the reader to understand which circle belongs to which percentile (although this can be achieved by comparison to e.g. Supplement Table S2 or Fig. 10), however the levels included were lacking. We have included the percentile levels in the figure legend and manuscript text.

Minor or technical points:

Page 1, Line 13: As above. The second sentence is confusing. I assume the observations are assimilated into the CTM before performing the model simulations.

AR: The data assimilation was conducted as a post process. We have clarified this by stating "using retrospective variational data analysis".

Page 1, line 20: Please clarify why including all observations leads to artificial trends and why using only time consistent measurements avoid this? Or else remove the text on artificial trends from the abstract for simplicity. See also below. This text appears in number of places in the manuscript without clarification.

RA: we have removed the statement about "artificial trends" from the abstract. See more on the issue below.

Page 1, line 20: add "Distribution of the" before "surface O3"

AR: Done.

Page 1, line 26: change "processes" to something like "factors".

AR: Done.

Page 2, line 11: change the IPCC (2013) reference to reference the specific IPCC chapter.

AR: Done.

Page2, line 17, is the clause "the right weather conditions" needed? If so why? Also noted below.

RA: Yes. This is to emphasize that the weather does affect ozone formation, e.g. cloudy and/or cold weather affects the ozone formation negatively whereas warm and clear weather means favorable conditions. We have changed the language to "under favorable weather conditions"

Page 4, line 8; Page 5 line 13, page 6 line 12, page 7 line 30, page 10 line 30- in all these places it is highlighted that artificial trends can be introduced, this seems an important and challenging point, can this text be expanded upon so the reader understand why an artificial trend could appear. The text on page 7 is expanded but

still not clear. The text on page 10 seems useful earlier. Some of the repetition could be removed.

RA: We have now included an explanation at the first mentioning of artificial trend in the introduction: "Artificial trends can for example arise from the introduction of new observation sites, which reduce the model bias in the area surrounding the measurement site during the time it is included but not before." Thus, if the model bias is prone to be constant over-estimation (or under-estimation) then the introduction of more (or less) observations by time will cause artificial trends in the data set. We have also removed some of the repetition but as it is a vital point in this study we keep most of them.

Page 8, line 25, As comment 1. above. Please explain the basis of the scaling used and depicted in fig 2a. i.e. what observed changes? Why has this been assumed to be constant from year 2007 onwards? The basis of the seasonal cycle (Fig S1) should also be given.

RA: The temporal trend of the boundary concentrations used in the present study is, admittedly, a crude estimate but should grossly represent the evolution of atmospheric species relevant to tropospheric ozone. Trends in NOy concentrations follow emission changes in the USA, CH4 concentrations closely follow global average background concentrations. O3 concentrations are assumed to increase by 1% per annum until 2000 and held constant after that date. The reason for this is to use a pragmatic and conservative approach considering all involved uncertainties (as introduced in an earlier published study; Engardt et al, 2017).

Fig. S1 illustrates the absolute levels and annual variation of O3 boundary concentrations during the year 2011. The values and their seasonal variation are from MATCH standard configuration as outlined in Andersson et al. (2007).

Andersson, C., Langner, J., and Bergström, R.: Interannual variation and trends in air pollution over Europe due to climate variability during 1958 2001 simulated with a regional CTM coupled to the ERA40 reanalysis, Tellus B 59, 77-98, 2007.
Engardt, M., Simpson, D., Schwikowski, M. and Granat, L.: Deposition of sulphur and nitrogen in Europe 1900-2050. Model calculations and comparison to historical observations, Tellus B, 69, 1328945, 2017.

Page 9, line 10: it would be useful for the reader to provide some insights into the impacts of using a higher resolution emissions data at 1km by 1km over Sweden as compared to EMEP 50 km by 50km when the data are subsequently interpolated to 44km resolution.

RA: The gain is mainly in the interpolation of the 50km resolution emissions to 44km, on a different projection, where we lose spatial information. SMED and EMEP are otherwise very similar for Sweden. This is already stated in the text: "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 reason for us doing this is also technical. The high-resolution emission inventory is used in later steps in the MATCH Sweden system. This is not explained in this manuscript, but a later, where this issue will be investigated. We wished to use as similar input data as possible also in this study. We ask the referee to wait for the next paper to get a more in-depth analysis.

Page 9, line 20, The text on the Swedish contribution of emissions to the domain would benefit from a map of the domain, otherwise is this text needed as the methods are already long?

RA: Yes. Actually the manuscript will benefit from a map of both domains: the total European domain and the domain of the "2dvar analysis". We have added this to the manuscript (Fig. 1b).

Page 10, line 4: Do these measurement sites have a station classification in EMEP such as "rural", if so please add, so the reader can see that these represent the regional background.

RA. The sites are classified as regional background sites. We have added a more thorough description of the origin and scale the sites represent.

Page 16, line 20 and Figure 6: Please explain how the regional average trends were calculated. Is the regional average calculated first then the trend?

RA: Here we averaged over the region before computing the trend. We have clarified this in the figure legend and in the main text.

Page 17, line 3: Fig7a shows only two colours so the highest values in the northerly mountains are hard to discern. Could the scale be improved?

RA: Of course. We have changed the scale and improved the resolution of the figures.

Page 17, line 19: Although there are some similarities in spatial patterns the colour scales suggest comparable magnitudes in the south and westernmost part of the region shown for the annual maximum 1 hr mean, whilst the south is the area with highest values for the annual mean metric.

*RA: we have clarified the sentence: "…*have similar spatial variation as their respective period means (Fig. 7), i.e. the maximum of annual means peaks in the north and the maximum of the annual maximum 1h mean peaks in the south*"*

Page 17, line 30 should "in the North" be added after March monthly mean?

RA: we have added "in the Central and North".

Page 18, line 13, as noted above there is large interannual variability that it is hard to say anything about shifts in maxima and dominance of spring vs. summer-time peaks.

RA: we have removed this since the manuscript is long already (major point 3).

Page 19, line 20, please explain what weather states favourable for O3 formation means and why relevant for NOx-VOC regimes for O3 production?

*RA: We apologize for an unfortunate typo; the text should have been "**… not** favorable for O3 formation". We have now reformulated to emphasize when this effect is the strongest: "when there is little photolysis (especially in the winter and during night-time)"*

Page 20, line 9, there is also greater photolysis of NO2 to form O3 which is why many locations in the northern hemisphere have a peak in Boreal summer. So greater production as well as destruction.

RA: The discussion deals with the fact that the trend in the boundaries contribution to the trend in near-surface concentrations over Sweden is lower during summer than during rest of the year despite the trend in boundary forcing being the same throughout the year in this numerical experiment. Our conclusion is that this is related to processes responsible for decreasing the life time of near surface ozone. We listed two such processes "dry deposition to vegetation and photolysis of ozone". We agree that O3 production is also greater in summer, but it is not relevant to include it in that paragraph.

Page 22, line 18: SOMO35 is not primarily used an indicator of long-term health effects. It is used as a metric compatible with short-term exposure and a threshold for adverse effects to occur- see HRAPIE report by WHO (2013).

RA: Yes. This was a mistake. As the paragraph was too political (see referee #2) and not completely correct as you point out we choose to remove the whole lot. We define SOMO35 including references that we think are well suited earlier in the section.

WHO 2013b. Health risks of air pollution in Europe – HRAPIE project: New emerging risks to health from air pollution – results from the survey of experts. http://www.euro.who.int/__data/assets/pdf_file/0006/238956/Health-risks-of-airpollution-in-Europe-HRAPIE-project,-Recommendations-for-concentrationresponsefunctions-for-costbenefit-analysis-of-particulate-matter,-ozone-and-nitrogendioxide.pdf

Page 23, line 26: data assimilation only reduces the impacts of boundary conditions at the surface.

RA: we now clarify that we mean "reduces the impact at the surface caused by errors in the lateral and upper model boundaries".

Page 24, line 11: although a number of studies do find that emissions changes are larger than climate change the time period of the climate change and the metric being analysed is important. Whilst this may be the case for annual or summertime means for higher frequency metrics such as percentiles used here this has not been well established.

RA: Thank you for pointing this out. We have clarified that the statement is meant for annual and summertime means. Further we include a new paragraph in the discussion:

"Our study shows that the impact of meteorological variability on the trend changes strongly from lower percentile levels to the very highest (in the South), with a shift from a positive to a negative contribution (cf. Fig. 10). Thus, conclusions drawn on the importance of meteorological variability in comparison to other factors such as changes in emissions will vary strongly depending on the metric that is studied. We have also studied the impact of base year in the sensitivity study (1990 vs 2011; cf. Fig. 5c and Supplement Fig. S5). The attribution to the trend is robust for all percentiles, including the annual maximum, whereas the monthly maximum is not robust for emissions and meteorological variation. So far studies of the future development of near-surface O_3 have focused on long-term means such as summer mean (e.g. Langner et al, 2012a,b; Watson et al., 2016), whereas the the direction of cause of high-frequency metrics, such as the higher percentiles we show here, have not been established and should be investigated further."

And also as a new bullet in the conclusions.

Page 25, line 8, SOMO35 in the north part of Sweden increase, hence this conclusion should be modified.

RA: Yes. We have clarified that this is true for central and south. In the north the changes are not significant so we refrain from conclusions on that.

Figure 1: see comment 4 above.

RA: The figure is correct.

Figure 5: See comment 5 above. This figure describes trends in percentile levels but please include the percentile levels that are plotted.

RA: the percentile levels are included in the main text as well as in the legend. See also reply above.

Figure 8-It is hard to see any trend in this figure because of the scale that covers all the percentile ranges. Could separate panels for low –median and median to high percentiles help with clarity? Or perhaps a figure like Fig 5 in Simpson et al. 2014 which depicts the table results clearly. An alternative would be to swap Figure 8 to the supplement and Table S3 to the main text as the table shows the trend results more clearly than the Figure.

RA: We have redrawn the figure to increase the legibility. We prefer to retain the figure with its three panels as it gives a comprehensive graphical overview of the trends of the different percentiles in the different parts of Sweden. The exact numbers of the trends and their significance, provided in the Supplement, are not the main message, but is available in other places.

Figure 9- the legend is hard to read.

RA: ok, we have updated the resolution of the figure.

Figure S6- this figure displays two y-axes but only one set of points are plotted?

RA: The left hand axis shows the running day of the year and the right hand axis the date this means. We remove the figure, see above.

Author reply to Anonymous Referee #2

We would like to thank the anonymous referee #2 for insightful comments and questions. We appreciate the raising of the two significant reservations, by addressing these we feel the manuscript has become more focused but also more nuanced. The technical issues has also helped us to improve the manuscript greatly. We have attached the manuscript and supplements with track changes included to allow for an overview of all changes included. Please note that this includes all changes to the manuscript, also including those based on anonymous referee #1. Below follow our replies point by point to the issues raised by the referee.

I have two significant reservations:

1.As noted on page 11, the length scale used for the 2dvar is 1000 km. The authors note that this is large, but claim that this is justified by the sparse network and the weak gradients in Sweden. However, Fig. 7 makes clear that the gradients can be rather large, especially in Southern Sweden. I would like to see more discussion of this problem, ideally with results from some sensitivity runs to help demonstrate if this really is a serious issue or not.

Reply from the Authors (RA, in italics): Unfortunately we cannot perform any new sensitivity runs testing the length scale due to technical reasons. We will have to leave this for future work.

We agree that the length scale may be too large in the southern part of the country. The impact of this may be too weak gradients in the south in the reanalysis. We have added a discussion on this in the in Sect. 4, whilst also shifting parts of the text from the methods to the discussion to focus the discussion in one place:

"*The length scale of the variational data analysis is set to 1000 km, implying a large horizontal influence of the observation increments. This is related to the sparse network of regional background observations but also 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 large length scale is also a filtering of local influences in the observations, consequently suppressing sharp gradients in the analysis. However, the horizontal variation in near-surface ozone is larger in the south than in the north, and the large length scale chosen in the data analysis may cause too weak horizontal gradients in the reanalysis data set, especially in the south. An improvement to this would be to describe the geographical variation of near-surface ozone in the background error field, rather than representing this by a constant value as done in this study.*"

2. Much of the discussion around annual mean O3 values results from the problem of nocturnal ozone depletion, which is said to be more important in southern Sweden. As nocturnal O3 itself is quite irrelevant for most health and vegetation metrics, why wasn't the analysis focused on some ozone-indicator that actually reflects these problems? This could be daytime ozone, M7, M12, or the daily 8-h values mentioned in Table 3.

RA: We have included also an evaluation of the daily maximum 1h mean ozone in 2013 (Table 2). We choose the daily maximum rather than other metrics, since the annual/monthly maximum is one of the main focuses of the manuscript. The daily maximum is highly likely to occur during daytime, meaning it should not be impacted by the nocturnal ozone depletion.

The evaluation shows overall improved scores compared to the hourly/annual mean. However, similar to the annual mean evaluation, the spatial correlation is 0.1 units worse in the cross validation as compared to the MFG simulation.

*Our conclusion is therefore that it is **not only** the nightly inversions that cause problems in the spatial variation, but this also occurs during daytime (for the highest values). This can be caused for instance by the distance to oceans and emission sources.*

We have included the evaluation (in Sect 3.1) and a discussion on this topic, also reducing the focus on the nocturnal ozone depletion (night-time inversions) in the manuscript:

"The evaluation of the daily maximum generally shows better correlation but slightly larger bias than the evaluation of the hourly mean. The spatial correlation is also worse in the cross validation compared to the MFG, but the spatial error is improved."

We removed the discussion on the impacts of the local topography in Sect. 3.1 (evaluation) and focus this to the discussion in Sect. 4.

<u>Page-by-page comments:</u>

Page 1, Abstract

l16 - ... 'performance over' rather than 'performance than'

RA: Done

Page 2

l9-11 - use more recent refs.

RA: The introduction is opened by general and non-controversial statements. We feel the cited overview literature is relevant although they may be old according to some standards.

l14 - define NOx as NO + NO2.

RA: Done. We also moved the definition of NO and NO2 to the Introduction where NOx is defined.

l21-22 - the paper of Fiore et al (2011) provides a much more recent example of this PAN effect.

RA: a reference to Fiore et al 2011 is added.

l31 - here the HTAP results presented in Fiore et al (2009) would also be relevant.

RA: a reference to Fiore et al. 2009 is added.

Page 3

l16 - use more recent refs

RA: We agree the references are outdated. To simplify we changed the sentence and references to: "The strong increase in near-surface O_3 concentration until the late 1990s at Mace Head, has levelled out to relatively stationary annual values throughout the 2000s (Derwent et al., 2013; Cooper et al., 2014)."

Page 4

l13 - move technique before ()

RA: Done

l30 - explain or provide references for 'databases EMEP and Airbase'

RA: We found an error in this sentence. EMEP is now referred to later in the manuscript, whereas Airbase is not included anymore. The text on this MACC reanalysis was updated and we shifted its location in the introduction:

"*Another reanalysis of near-surface O_3 concentration in Europe, also within the MACC project, was conducted for the period 2003-2012 (Katragkou et al., 2015). In this reanalysis 4dvar data assimilation was also used to incorporate retrievals from satellites. 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.*"

Page 7

l22-23 - states that MATCH only calculates chemistry for the lowest 5 km of the model domain. Is this a sufficient depth, when looking at the impact of tropospheric boundary conditions? I was puzzled as to why the boundary conditions are given as

mass units. Usually the volumetric mixing ratio is used as this is the more conserved quantity, and independent of pressure and temperature. Why this choice?

RA: MATCH has been constructed to describe near-surface concentrations and surface depositions. MATCH does not include stratospheric chemistry. In its standard European configuration we therefore typically limit the vertical domain to the lowest 5-6 km of the troposphere. Through numerous comparisons with observations and other similar models (see references in the main text) we have great confidence in the model's ability to reproduce near-surface O3 in Europe. We believe the present set-up is adequate for assessments of the impact of general trends in hemispheric background concentrations although trends in stratospheric chemistry or physically driven changes in stratospheric-tropospheric exchanges will likely not be captured. This comment is now also introduced to the main text.

In the original version we chose to present mass concentrations in order to use consistent units throughout the manuscript, but agree it is better to show the conserved quantity that is actually used by the model. We have now updated Figure S1 to display boundary concentrations in ppb(v) (MATCH uses g/kg).

Page 8

l18 - states that the model uses 22km grid-spacing, but on page 9 the emissions are interpolated to a 44km grid. Which is correct?

RA: This statement of resolution refers to that of the original HIRLAM meteorology. The resolution of MATCH (which we also interpolate the meteorology to) is given previously in section 2.2, as we wish to explain the meteorology separately to the MATCH setup. We added "and emissions" to the sentence in section 2.2.

The reason for running MATCH on a coarser resolution than that of our Swedish emissions and the original meteorology is the need to limit CPU resources consumed by the project.

l26 - maybe add 'see also Andersson et al, 2007' here as a ref also, since it isn't obvious from the text where the time-development comes from. Further confusion arises on page 23, when it is stated that the trends are taken from Engardt et al., 2017. Please clarify which statement is correct?

RA: Sorry for the confusion. We have amended the text and now refer to Engardt et al. (2017) already in section 2.2.1.

The secular trends are from Engardt et al. (2017), while the reference boundary concentrations -valid for the year 2000- are taken from Andersson et al. (2007). As explained in Andersson et al. (2007) do we include seasonal and geographical variations of the boundary concentrations in the reference case.

Page 9

l25-26 - states that no trend is assumed in the intra-annual variation in emissions, but such a trend is likely to exist. There have been quite large changes in the sources and fuel-mix over this period. Will this matter? I think you should also mention that the spatial distribution of emissions is also held constant, which is possibly a bigger source of uncertainty. (Which year was used for the spatial distribution?)

RA: Both the spatial distribution and the seasonal variation is updated by the year year, e.g. for SMED see http://extra.lansstyrelsen.se/rus/SiteCollectionDocuments/Statistik%20och%20data/Nationell%20emissionsdatabas/Metod_och_kvalitetsbeskrivning_Geografisk_f%C3%B6rdelning_sub2016.pdf (in Swedish). *We have removed the incorrect statement from the manuscript.*

Page 10. Are these sites all part of EMEP? If not, are the data-quality criteria equivalent to those of EMEP sites?

RA: We have added a detailed description of all utilized sites in the main text:

"The Swedish observations were delivered by the Swedish data host (at that time, July 1, 2017, Swedish Environmental Institute, IVL). The Norwegian observations were extracted from EBAS (http://ebas.nilu.no; extracted on July 6, 2017). All sites except Norr Malma and Rödeby are classified as regional background measurement sites by EMEP (Internet URL: http://www.nilu.no/projects/ccc/emepdata.html; Hjellbrekke and Solberg, 2015). Norr Malma is located ca 70 km north-east of Stockholm and is considered a regional background measurement site by Stockholm Air and Noice (http://slb.nu), who are responsible for the site. Rödeby is located 10 km north of the small town Karlskrona, and is considered a rural location (personal communication with Titus Kyrklund, Swedish EPA)."

Page 12

What is 'full-domain'? Is all of Europe covered? Does 'Eur' include Russia?

RA: We have included a map showing the full domain in Fig. 1 (panel b). We refer to this in the bullet. We have also re-defined and explained the labels of the two bound simulations: "SE emis" is Swedish only emissions (see below)," FD emis" is full domain minus Swedish emissions.

Page 12 and onwards
The 2-letter code 'Se' is a little confusing, neither English nor an accepted abbreviation. The UN code is 'SE', so why not use that directly?

RA: We have changed the code to SE emis from Se emis.

Page 13

l1 - mention that numerics can also cause non-linearity in CTMs.

RA: yes. We added this to the end of the sentence: ", or as a numeric effect in the model."

Page 15

l26 - this says Fig. 5 gives 'annual' O3, but Fig.5 mentions just 'percentiles'. Which percentiles? I couldn't figure out what this Figure was showing.

RA: The figure shows whether the trends averaged over the three regions for different percentile levels are the same between the SUM, MFG and LONGTERM data sets (values for the latter are also given in Supplementary Table S3). The issue is not to specifically understand which circle that belongs to which percentile, however the information on the levels used was lacking.

To clarify, we have changed the description in the description in Sect 3.1 to "In Fig. 5 we compare regionally averaged linear trends in annual percentiles (levels: 0, 2, 5, 10, 25, 50, 75, 90, 95, 98 and 100) of hourly near-surface O_3 over the period 1990-2013…" . We have also updated the figure legend.

The figure was also updated with a panel c due to a comment by Referee #1.

Page 17

l14 - why say 'possibly caused by'? You have the data, so you can say exactly what caused this.

RA: We have checked the data. Both daytime and nighttime means are lower in the south than in the northerly mountains although the difference is larger for the nighttime mean. Thus we conclude that it is mainly an impact of the higher altitude in the northerly mountains that cause higher mean values in the north, while in the south the lower values are partly caused by the lower nighttime values and the higher max-values are caused by high ozone events originating mainly from continental Europe.

We have edited the text accordingly: "The lower period mean of the near-surface O_3 in the south than in the north is mainly caused by the higher altitude, of the latter, mountainous, region whereas the opposite gradient for the annual maximum 1h mean is caused by the distance to continental Europe where the high ozone events originate from."

Page 18

l21-22. This explanation of Fig.8 would have been better presented before it is first referenced

RA: We have moved the explanation to the first paragraph in Sect. 3.2.

Page 19

l14-15. This Figure reminds strongly of that presented by Jenkin (2008), so it would be good to reference that paper.

RA: The paper is now included as a reference.

Page 20

l22-26. The big change in sign for 'meteo' between the 98th and 100th percentile deserves some comment.

RA: yes. We have emphasized this. We have also included a discussion on this in Sect. 4 (as it was also commented on by Referee #1) and a new bullet in the conclusions.

Page 22

l23-26. This sounds like a political statement of the authors views. I agree that NOx control is essential for many reasons, but cite scientific papers to support your statement.

RA: we have removed the paragraph as we agree it was too political and the manuscript is long anyway.

Page 25

l21-22. This statement is unclear. Which earlier studies?

RA: We have added two references as examples.

Figures

Generally, the figure quality is quite poor and should be improved. (Some of the figures look like screen-dumps of excel plots, and the various Sweden maps (e.g. S10-S11) have awful color schemes.)

RA: We will make sure to make the final figures at a better quality to allow zooming in. In the updated manuscript we have made an effort to include high-quality figures.

We use the color schemes of Fig. S7-S10, as they allow many different levels to be shown. A person prone to colorblindness may have difficulty in interpreting some of the colors, having to turn to the text for interpretation. We have tried to explain the figures as thoroughly as possible with that in mind without making the text too heavy.

Fig. 2. The C5H8 emissions are so close to zero here that the plot doesn't show anything except that the emissions are very small. These could either be presented on a

separate plot, or just described in the text. Are C5H8 emissions really so small by the way? I have seen larger estimates for Europe.

RA: The C5H8 emissions are now lifted out to a separate panel (Fig 2c) to highlight the inter-annual variations and trend. The average C5H8 emissions in the model domain in the present study is 3.1 Tg year per year. Earlier model comparisons has shown that isoprene emissions in MATCH are lower than in other, similar, models. In Langner et al. (2012b) MATCH had average isoprene emissions of 1.6 Tg per year while the EMEP model and SILAM had 3.4 and 4.1 Tg per year, respectively. That study covered a similar domain but used meteorology from a climate model. One important conclusion from that study, was that isoprene emissions are highly uncertain and variable across models. The present C5H8 emissions are within the variability of the Langner et al. (2012b) study.

Langner, J., Engardt, M., Baklanov, A., Christensen, J.H., Gauss, M., Geels, C., Hedegaard, G.B., Nuterman, R., Simpson, D., Soares, J., Sofiev, M., Wind, P. and Zakey, A. 2012b. A multi-model study of impacts of climate change on surface ozone in Europe. Atmos. Chem. Phys. 12, 10423-10440. doi:10.5194/acp-12-10423-2012

Fig. 3. Given the frequent discussion of the topographic location of these sites, I think a Table with altitude would also help.

RA: As we have removed a lot of the discussion on local topographical effects we do not see the need to include the altitude of the sites are needed anymore.

Fig. 4. I found the color choice unusual. Usually one uses red to indicate a warning, e.g. that data-quality is poor. Here red is used to indicate good data-quality,

RA: The colors are chosen to i/ avoid troubles for the colorblind, ii/ show well on the map in Fig. 3. The colors are coordinated in Fig. S2-S3.

Fig. 5. As noted above, I don't know what 'ozone percentiles' means if one doesn't specify which percentile. The blue and green colors here can also be hard to distinguish.

RA: Our aim with the figure is not for the reader to understand specifically which circle belongs to which percentile, but the engaged reader derive this by combining the panels with the tabulated trends in percentiles of the LONGTERM reanalysis in Supplementary Table S3 or extract the values from Fig. 10. We have clarified the percentile levels included in the legend.

Fig. 7. Poor quality.

RA: we have made updated the figures at a higher resolution and will make sure the final figures are zoomable.

Fig. 8. Increase the font-size for the percentile labels - they are really hard to see.

RA: we have now amended the plots including the use of larger and clearer symbols and bigger font size to increase legibility.

Fig. 9. Improve quality. I really liked the content of this Figure, and also Fig. 10, but they both look like screen dumps.

RA: The figures are updated at a higher resolution and we will make sure the final figures are zoomable.

References

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Fiore, A., Dentener, F., Wild, O., et al., A., Multi-model estimates of intercontinental source-receptor relationships for ozone pollution, J. Geophys. Res., 114, 2009

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 retrospective variational data analysis. The reanalysis data sets show improved performance over 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. Based on the second reanalysis we investigated statistical aspects of the distribution 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 reflected in observed improvement of many health and vegetation indices (apart from those with a low threshold).

Using the CTM we also conducted sensitivity simulations to quantify the causes of the observed change, focusing on three factors: change in hemispheric background concentrations, meteorology and anthropogenic emissions. The rising low concentrations of near-surface O_3 in Sweden are caused by a combination of all three factors, whilst the

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decrease in the highest O_3 concentrations is caused by European O_3 precursor emissions reductions.

While studying the 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. Stocker et al., 2013). Elevated O_3 concentrations are formed in the troposphere by the oxidation of volatile organic compounds (VOCs) and carbon monoxide (CO), driven by solar radiation in a polluted air mixture that includes nitrogen oxides (NO_x, sum of nitric oxide and nitrogen dioxide: $NO+NO_2$). Close to combustion sources, the background O_3 concentration is reduced through reactions with directly emitted NO (see for example Finlayson-Pitts and Pitts, 2000). However, further away from the source and with sufficient availability of VOCs and under favorable 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; Fiore et al., 2011).

European and North American anthropogenic emissions of NO_x increased over most of the 20th 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 (Fiore et al., 2009; Derwent et al., 2015).

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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 Mace Head, has levelled out to relatively stationary annual values throughout the 2000s (Derwent et al., 2013; Cooper et al, 2014). 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. The models capture only 50 % of the change observed during the last 5-6 decades and little of the observed seasonal differences, and the rate of the trends are poorly captured. There are ways forward to improve the

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description of the trends in models: 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. Artificial trends can for example arise from the introduction of new observation sites, which reduce the model bias in the area surrounding the measurement site during the time it is included but not before. Data assimilation, a subset to data fusion (Zhang et al., 2012), is the process by which observations of the real world 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 technique (4dvar; e.g. Courtier et al., 1994; Inness et al., 2013) 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 reanalysis of near-surface O_3 concentration in Europe, also within the MACC project, was conducted for the period 2003-2012 (Katragkou et al., 2015). In this reanalysis 4dvar data assimilation was also used to incorporate retrievals from satellites. 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. A third global reanalysis using data assimilation of satellite data for 2005-2012, showed improved performance for many 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 the data assimilation.

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Borttaget: 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 variational data analysis techniques. Variational data analysis in 2 dimensions (2dvar) and the analytical counterpart optimal 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 variational data analysis of atmospheric concentrations in air and precipitation. 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 variational data analysis. 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 concentrations and meteorological variability. (see Sect. 3.3)

2 Method

In this study we utilize variational data analysis in order to combine the best qualities of a CTM and long-term measurements to map near-surface O_3 concentrations during a long

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historical time period (1990-2013). We focus our study on Sweden, but also include Norway in the variational data analysis.

For the variational data analysis 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 variational data analysis 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 variational data analysis. 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, ammonia (NH_3) and sulfur dioxide (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 variational data analysis (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.

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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 retrospective variational data analysis 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 and emissions 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, which is the the model's standard configuration for pan-European simulations. The selected set-up has been demonstrated (e.g. Andersson et al., 2007; Langner et al., 2012a; Markakis et al., 2016) to be adequate for describing near-surface O_3 across Europe although trends in stratospheric chemistry or physically driven changes in stratospheric-tropospheric exchanges will likely not be captured.

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

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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. 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. 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).

Borttaget: 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).

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 HIRLAM was run as forecasts from 6-hourly analyses, composed of three-dimensional variational upper air analyses and optimal interpolation surface analyses (Dahlgren et al., 2016). 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.

Borttaget: (http://www.euro4m.eu)

Borttaget: in 3 dimensions

Although the present study focuses on Sweden it is necessary to realistically describe the fluxes of O_3 and its precursors from continental Europe and further afield. Hemispheric background concentrations of all species for the modelled year 2000 are similar to the ones used by Andersson et al. (2007). As in Andersson et al. (2007), boundary values representative for the average concentrations at 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 furthermore scaled to mimic observed changes in the hemispheric background during the period 1990 through 2013 following the work of Engardt et al. (2017), cf. Fig. 2a. Note that the hemispheric background ozone concentrations are assumed constant from 2000 onwards following recent assessments

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of the evolution of near surface ozone in Europe (e.g. Cooper et al, 2014). CO and NMVOC boundaries are held constant throughout the simulation. 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).

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 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.

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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 (Hjellbrekke and Solberg, 2015; 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. Emissions of biogenic isoprene are calculated online in MATCH following the E-94 isoprene emission methodology proposed by Simpson et al. (1995).

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Both the total domain and Swedish national 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 (cf. Fig 2c). The national 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

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

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). The amount and spatial distribution of the emissions is updated each year.

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Borttaget: We assume that there is no trend in the temporal intra-annual spatial variation of the emissions.¶

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. The Swedish observations were delivered by the Swedish data host (at that time, July 1, 2017, Swedish Environmental Institute, IVL). The Norwegian observations were extracted from EBAS (http://ebas.nilu.no; extracted on July 6, 2017). All sites except Norr Malma and Rödeby are classified as regional background measurement sites by EMEP (Internet URL: http://www.nilu.no/projects/ccc/emepdata.html; Hjellbrekke and Solberg, 2015). Norr Malma is located ca 70 km north-east of Stockholm and is considered a regional background measurement site by Stockholm Air and Noice (http://slb.nu), who are responsible for the site. Rödeby is located 10 km north of the small town Karlskrona, and is considered a rural location (personal communication with Titus Kyrklund, Swedish EPA). The sites included are all instrumentation sites, where O_3 is measured continuously and reported with hourly temporal resolution. The retrospective variational data analysis 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):

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- The first set 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 (personal communication with Sverre Solberg, NILU). The inclusion of these two sites could introduce an abrupt change 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 variational data analyses. 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. We return to whether these reanalyzes differ in Sect. 3.1.

2.4 Variational data analysis

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.

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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.

The variational data analysis 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 different 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 variational data analysis 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 variational data analysis*. 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)
   - Full domain (see Fig. 1b) non-Swedish anthropogenic emissions (FD emis)

B. Change in lateral and upper boundaries (bound)

C. Change in meteorology, including online modelled biogenic isoprene emissions (meteo)

Borttaget: , 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.

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Four sensitivity simulations are conducted; in which each of the four listed factors are kept constant at ~~their~~ level ~~in~~ 2011. ~~The respective contributions presented in Sect. 3.3 are formed~~ through the following sequence: 1. Calculation of gridded metrics (focusing on monthly 1h maximum, monthly mean and annual 1h percentile levels); 2. Calculation of secular gridded trends over the monthly or annual metrics; 3. Calculation of regional (North, Central, South, cf. Fig. 3) mean of the secular trends, 4. Calculation of the difference between the regional mean trend in MFG and 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 ~~three~~ 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 simulations are non-additive. This could occur when changes to mixtures of complex chemistry, weather situations and emissions take place, or as a numeric effect in the model. For this reason we compare the sum of the trend in the estimated contributions to the MFG trend. Second, our methodology quantifies 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 MFG trend to make sure the base case simulation does not deviate too strongly from the reanalysis results. If the deviation is large, i.e. the modelled trend is far from the observed, this means that the MFG simulation 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-additive or non-representative) for the trend in a specific metric then our method cannot be used to explain that specific trend. Third, the attribution may be sensitive to the chosen base year. Sensitivity simulations using 1990 as base year instead of 2011 are also conducted, to investigate the robustness of the results. To investigate all 24 years as base years would take too much computational efforts, we choose 1990 as it differs from 2011 both for European emissions and climatologically[2]. If the contributions to

[2] The NAO index was high in early winter 1990 and low in 2011, whereas the summer index was positive but close to 0 in 1990 and negative in 2011.

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Borttaget: The respective contributions to the trend are formed by subtracting the MFG with the corresponding sensitivity simulation.

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the trend differ too much between the base years 1990 and 2011 then the results are not robust. If they are similar it is not a guarantee that the results are robust but it is an indication. The contributions with 1990 as base year are formed in the same way as for the 2011 sensitivity runs. The contributions due to change in top and lateral boundaries (bound) and variations in meteorology are included in the same manner, while we compare the total footprint of the change in emissions, i.e. the sum of FD emis and SE emis (emis) rather than the two parts.

2.6 Evaluation

We evaluate two aspects of the reanalysis. The first is an independent evaluation for a single year with focus on the variational data analysis method. The second is an evaluation of the simulated near-surface O_3 concentration trend over the 24-year period and our ability to explain the causes of the trend.

For an independent evaluation of the variational data analysis method we conduct a cross validation at the included Swedish measurement sites. In this method we exclude one measurement site at a time from the variational data analysis, and evaluate the results at the excluded location. This means we conduct one 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.

For the evaluation of the long-term trend we focus on the three 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 and 3/ whether the contributions to the secular trend are sensitive to base year. We focus this investigation on 11 different percentiles of hourly mean O_3 concentrations, for an estimate of the scores at

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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.

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).

Borttaget: The comparisons are presented as scatterplots in Fig. 5 and compared to the 1:1 line, factor 2 line and equal sign quadrants.

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 and daily maximum of hourly mean 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 evaluation of the daily maximum generally shows better correlation but slightly larger bias than the evaluation of the hourly mean. The spatial correlation is also worse in the cross validation compared to the MFG, but the spatial error is improved. Overall, the independent cross validation shows that the 2dvar method improves the performance of the modelled hourly mean and daily maximum 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 Fig. 5 we compare regionally averaged linear trends in annual percentiles (levels: 0, 2, 5, 10, 25, 50, 75, 90, 95, 98 and 100) of hourly near-surface O_3 over the period 1990-2013 for the MFG simulation, the LONGTERM reanalysis, the sum of contributions and the contributions to the trend for different base years. Investigating the additivity of the four contributions (bound, meteo, SE emis and FD 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. Finally, investigating the impact of the selected base year in the sensitivity simulations, we compare the contributions (bound, emis and meteo) based on keeping the year 2011 constant in the sensitivity simulations to keeping the year 1990 constant (Fig. 5c). Most contributions to the trend in percentiles are robust (Fig. 5c), falling close to the 1:1 line. Only a few of the very weakest O_3 contribution trends fall outside the factor 2 lines (for the meteo contribution). The contributions to the secular trend in some

Borttaget: 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).

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of the monthly mean and the monthly maximum 1h mean near-surface O_3 differ more for the two base years than the percentiles (Supplements, Fig. S5). For monthly mean the trend due to changes in meteorology is stronger for some months (one month is weaker) when 2011 is used as base year compared to 1990. The other contributions fall within the factor 2 lines. For monthly maximum the deviation is larger, even differing in sign for the contribution due to variation in meteorology for some months, and a few contributions due to emission change also fall outside the factor 2 lines.

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, but for the contribution of meteorology to the monthly maximum is not robust under the choice of base year for all months. 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). The seasonal variation in the linear trend of the spatially averaged monthly values is also included in the figure. Spatially resolved statistics for annual mean and annual maximum of 1h mean near-surface O_3 are provided in Fig. 7. The temporal evolution of 11 percentile levels from the 0th (annual minimum 1h mean) to the 100th (annual maximum 1h mean) are shown in Fig. 8, and the corresponding trends with indication of significance levels are recaptured in the Supplements (Table S2).

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,

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Borttaget: Time series of annual percentiles averaged over the three regions are shown in Fig. 8.

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 October-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 mainly caused by the higher altitude of the latter, mountainous, region whereas the opposite gradient for the annual maximum 1h mean is caused by the distance to continental Europe where the high ozone events originate from. The difference in spatial pattern between the southern, 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 their respective 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.

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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 in the Central and North.

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.

We proceed by investigating the trend in annual percentiles (Fig. 8) of hourly near-surface O_3 concentration, averaged[3] over the three Swedish regions (cf. Fig. 3). 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 in northern Europe were 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 significant and stronger than in the North, 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

[3] 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.

Borttaget: 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).

Borttaget: 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).

Borttaget: The temporal evolution of 11 percentile levels from the 0th (annual minimum 1h mean) to the 100th (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).

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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 UK by Jenkin (2008) for 1990 until the early 2000s and in the US by Simon et al. (2015) for 1998-2013, both studying urban and regional background measurements across the respective countries. Jenkin (2008) interpret it as caused by three major influences: i/ increasing hemispheric background, ii/ decreasing severity in high ozone events arising from the European continent and iii/ decreasing local-scale removal of ozone due the control of NOx emissions. Simon et al (2015) interpret the US evolution 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 when there is little photolysis (especially in the winter and during night-time). 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.

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3.3 Attribution of the change in near-surface ozone

In this section we quantify the contributions of physical factors 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 (i.e. full domain, non-Swedish) anthropogenic emission change. In Figs. 9 and 10 the contributions to the trend in seasonal variations and percentiles are delineated for the North and South regions.

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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 contribute to 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 lower impact during summer. This variation is likely a result of an O_3 destruction process that is

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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 variational data analysis. 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. Note the shift from a generally strong positive contribution to a strong negative contribution from the 98th percentile to the 100th percentile in the South. 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, "FD 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

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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. SOMO35 (the Sum of Ozone Means[4] Over 35 ppb(v)) is used as a metric describing human exposure. The cut-off value of 35 ppb(v) is often used in risk assessments as a statistically significant increase in mortality has been observed at daily ozone concentrations >25-35 ppb(v) (Bell et al., 2006; Amann et al., 2008; Orru et al., 2013). In Table 3 we present the linear trends in our reanalysis data set for these metrics, and

[4] 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.

have collected geographically resolved statistics, such as the period mean, maximum and linear trend in the Supplements (Figs. S6-S10).

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) 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 S2). 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.

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 data 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 variational data analysis 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 length scale of the variational data analysis is set to 1000 km, implying a large horizontal influence of the observation increments. This is related to the sparse network of regional background observations but also 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 large length scale is also a filtering of local influences in the observations, consequently suppressing sharp gradients in the analysis. However, the horizontal variation in near-surface

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ozone is larger in the south than in the north, and the large length scale chosen in the data analysis may cause too weak horizontal gradients in the reanalysis data set, especially in the south. An improvement to this would be to describe the geographical variation of near-surface ozone in the background error field, rather than representing this by a constant value as done in this study. 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 variational data analysis scheme will spread such features to parts of the model results where they are not valid. Some of the southerly sites in the variational data analysis are known to experience night-time inversions with associated depletion of near-surface O_3 and the reanalysis will thus be affected by this. Introducing a geographically varying length scale and background error in the variational data analysis and an improvement in the spatial resolution of the model would improve the spatial representation of the analysis, the latter 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 top boundary conditions are taken from the work by Engardt et al. (2017) and are based on observed trends at regional background location in Europe. The upper boundaries are especially poorly constrained in our study, and as a consequence so is the stratospheric contribution to the inter-annual variation and trend. The variational data analysis reduces the impact at the surface caused by 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 FD emis). The resulting figure is included in the Supplements (Fig. S11). 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

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in the Central and South regions. This calls for emission inventories to be improved in order to assure 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 annual or summer-time mean 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, 2012b; 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.

Our study shows that the impact of meteorological variability on the trend changes strongly from lower percentile levels to the very highest (in the South), with a shift from a positive to a negative contribution (cf. Fig. 10). Thus, conclusions drawn on the importance of meteorological variability in comparison to other factors such as changes in emissions will vary strongly depending on the metric that is studied. We have also studied the impact of base year in the sensitivity study (1990 vs 2011; cf. Fig. 5c and Supplement Fig. S5). The attribution to the trend is robust for all percentiles, including the annual maximum, whereas the monthly maximum is not robust for emissions and meteorological variation. So far studies of the future development of near-surface O_3 have focused on long-term means such as summer mean (e.g. Langner et al, 2012a,b; Watson et al., 2016), whereas the the direction of cause of high-frequency metrics, such as the higher percentiles we show here, have not been established and should be investigated further.

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

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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 provided upon request from the corresponding author.

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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 central and south 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 weaker impact on future

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near-surface O_3 concentrations than shown by earlier studies (e.g. Langner et al., 2012a,b; Watson et al., 2016).

- The results show that the impact of meteorological variability changes strongly from lower percentiles levels to the very highest in the South. In studies of future development the maximum ozone, and the causes for change in this, should be investigated further.

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Figure and table legends

Figure legends

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

Figure 2. (a) Temporal trend of factors used for scaling boundary concentration of relevant species (based on Engardt et al., 2017). (b) Temporal trend of total domain (circles; left vertical scale) and Swedish (triangles; right vertical scale) annual anthropogenic O_3 precursor emissions utilized by MATCH from 1990 to 2013. Emissions of nitrogen oxides (NOx), non-methane volatile organic compounds (NMVOC) and carbon monoxide (CO) are indicated by different colors (cf. legend); emissions of sulfur oxides (SO_x) and ammonia (NH_3) are excluded from the panel.

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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. (a,b) Temporal trends in annual percentiles of hourly mean near-surface ozone (levels: 0, 2, 5, 10, 25, 50, 75, 90, 95, 98 and 100, cf. also Table S2) 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 reanalysis LONGTERM vs the MATCH model simulation MFG (b). Filled circles indicate significant trends ($p \leq 0.05$) in the MFG simulation, whereas non-significant MFG trends ($p > 0.05$) are indicated by an empty circle. (c) Sensitivity in contributions to the secular trends in regionally (North: squares, Central: circles, South: diamonds) averaged annual percentiles (levels: 0, 2, 5, 10, 25, 50, 75, 90, 95, 98 and 100) of hourly near-surface O_3 over the period 1990-2013 due to choice of base year (1990 vs 2011). Modelled contributions to the near-surface ozone trend due to

change in top and lateral boundaries of relevant species (dark yellow, bound), change in full domain emissions (blue, SE emis + FD emis), and variation in meteorology (fair yellow, meteo). 1:1 line in black, factor 2 lines in dark grey.

Figure 6. Seasonal cycle of monthly mean (a) and monthly maximum (b) of 1h mean near-surface ozone concentrations averaged over the period 1990-2013 (circles; left vertical scale) and region (North, Central and South Sweden, cf. Fig. 3) and the linear trend over the same period of the respective spatially averaged monthly values (triangles; right vertical scale). The different regions are identified by the 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 S2, 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 (brown 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, FD emis), emissions, trend in top and lateral boundaries of relevant species (dark yellow, bound) and variation in

meteorology (fair yellow, meteo). The sum of the modelled contributions is indicated by the dashed brown 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 diamond; LONGTERM reanalysis) and modelled MFG near-surface ozone trend (brown 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, FD emis), emissions, trend in top and lateral boundaries of relevant species (dark yellow, bound) and variation in meteorology (fair yellow, meteo). The sum of the modelled contributions is indicated by the dashed brown line.

Table legends

Table 1a. Model calculations and scenarios, all covering the years 1990-2013, including the "first guess" to the retrospective variational data analysis and base case to the sensitivity simulations (MFG), two reanalysis data sets (LONGTERM and ALL), sensitivity scenarios (MFD, 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 and daily maximum of 1h mean 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/days 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 evaluation 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).

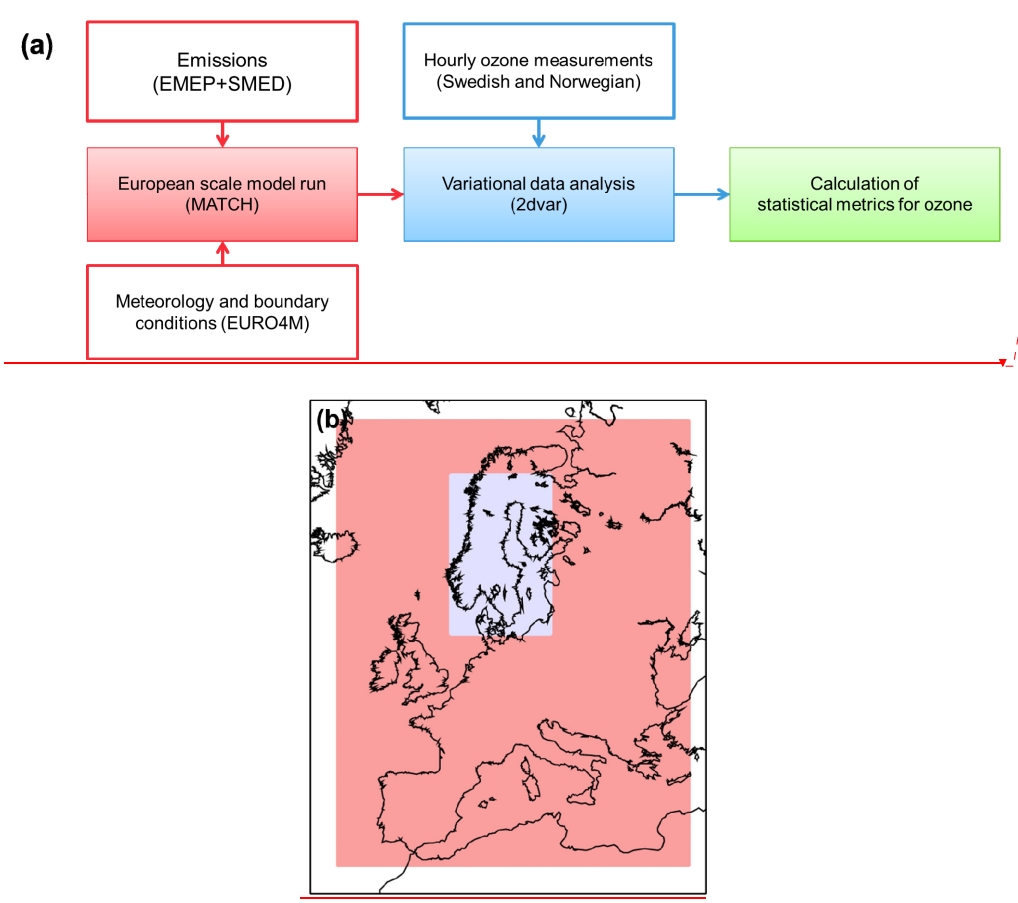
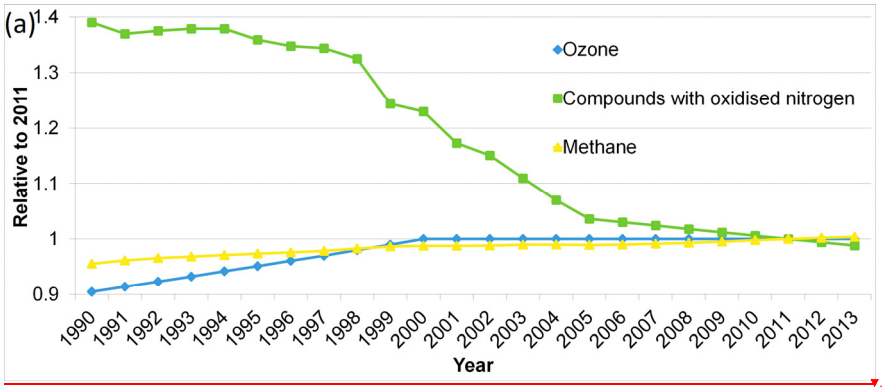
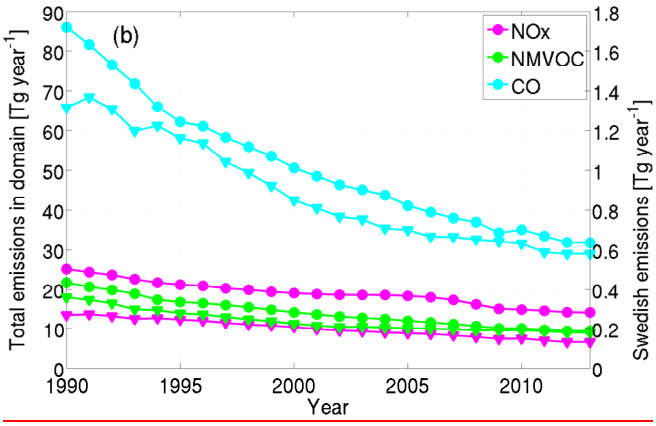


Figure 1. a) A flow-chart of the relevant part of the MATCH Sweden system for this reanalysis study. b) The total domain of the European scale model run (pink + light blue) and the domain of the variational data analysis (light blue).





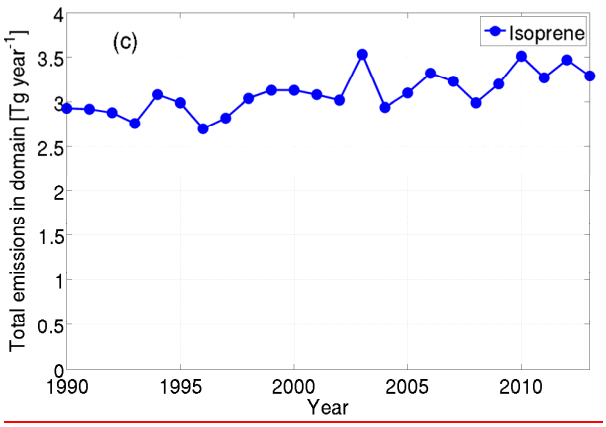


Figure 2. (a) Temporal trend of factors used for scaling boundary concentration of relevant species (based on Engardt et al., 2017). (b) Temporal trend of total domain (circles; left vertical scale) and Swedish (triangles; right vertical scale) annual anthropogenic O_3 precursor emissions utilized by MATCH from 1990 to 2013. Emissions of nitrogen oxides (NOx), non-

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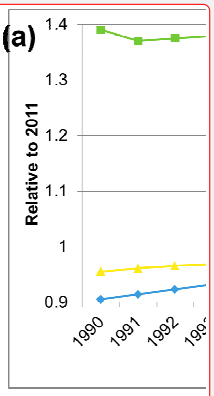

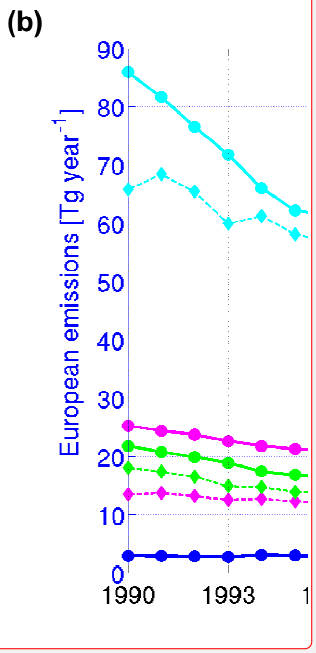

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Borttaget: Note that the hemispheric background ozone concentrations are assumed constant from 2000 onwards. CO and NMVOC boundaries are held constant throughout the simulation.

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methane volatile organic compounds (NMVOC) and carbon monoxide (CO) are indicated by different colors (cf. legend); emissions of sulfur oxides (SO_x) and ammonia (NH_3) are excluded from the panel. (c) Temporal trend of total domain biogenic isoprene emissions.

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Borttaget: and biogenic isoprene (C5H8)

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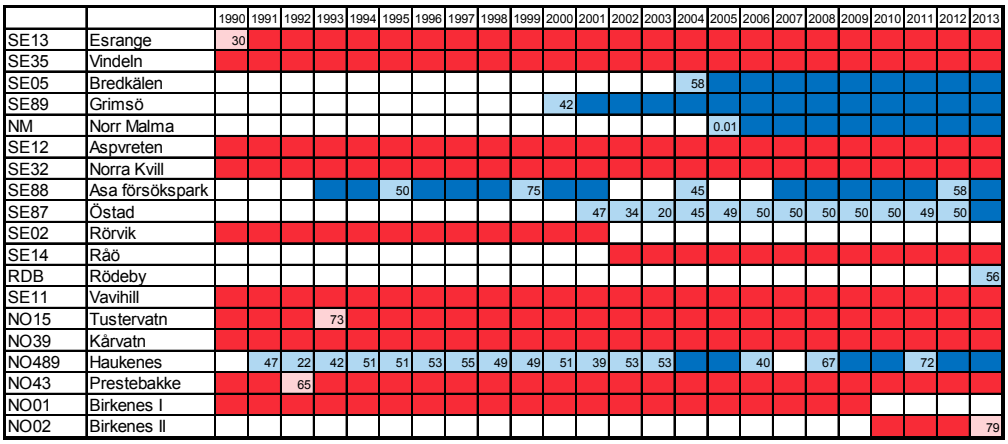
Figure 3. Instrumentation sites for hourly near-surface ozone concentration observations in Sweden and Norway, which are used in the variational analysis. Red circles: sites with full data coverage. Blue circles: sites with restricted data coverage. The subdivision of Sweden into three regions (North,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



		1990	1991	1992	1993	1994	1995	1996	1997	1998	1999	2000	2001	2002	2003	2004	2005	2006	2007	2008	2009	2010	2011	2012	2013
SE13	Esrange	30																							
SE35	Vindeln																								
SE05	Bredkälen															58									
SE89	Grimsö											42													
NM	Norr Malma																0.01								
SE12	Aspvreten																								
SE32	Norra Kvill																								
SE88	Asa försökspark						50				75					45								58	
SE87	Östad												47	34	20	45	49	50	50	50	50	50	49	50	
SE02	Rörvik																								
SE14	Råö																								
RDB	Rödeby																								56
SE11	Vavihill																								
NO15	Tustervatn				73																				
NO39	Kårvatn																								
NO489	Haukenes		47	22	42	51	51	53	55	49	49	51	39	53	53			40		67			72		
NO43	Prestebakke			65																					
NO01	Birkenes I																								
NO02	Birkenes II																								79

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. White squares: no observations are available for that year and site. The LONGTERM reanalysis includes the red measurement sites, the ALL reanalysis includes both red and blue.

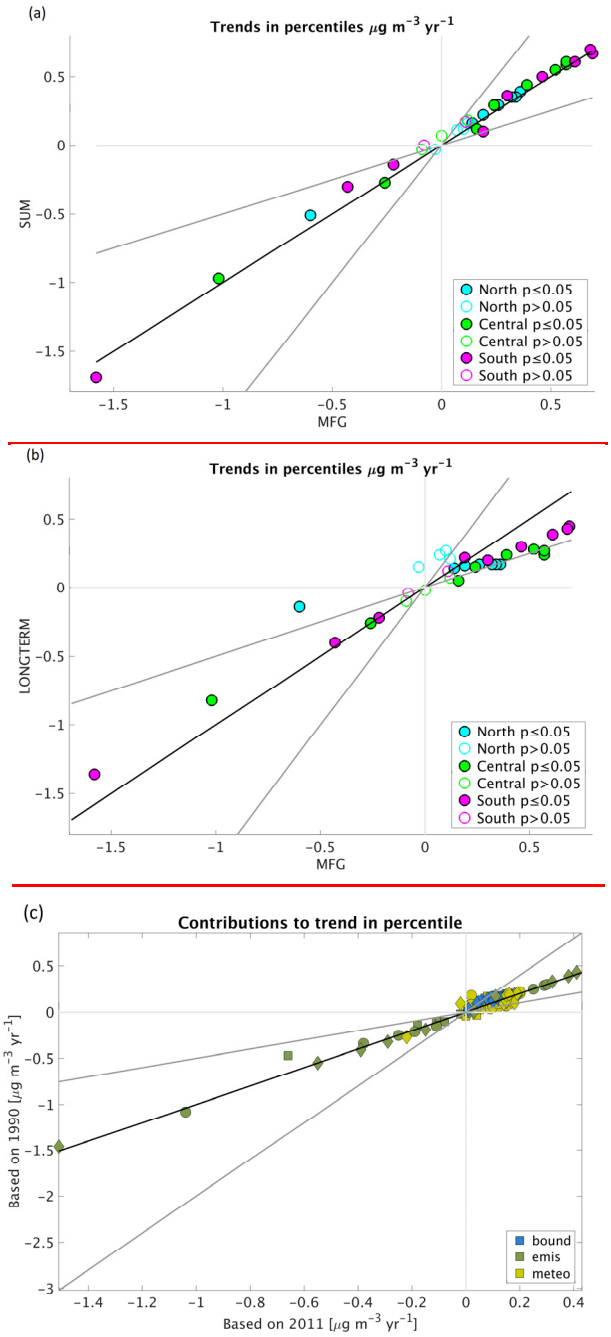


Figure 5. (a,b) Temporal trends in annual percentiles of hourly mean near-surface ozone (levels: 0, 2, 5, 10, 25, 50, 75, 90, 95, 98 and 100, cf. also Table S2) averaged for the three regions North (blue), Central (green) and South (magenta) for the sum of the contributions to

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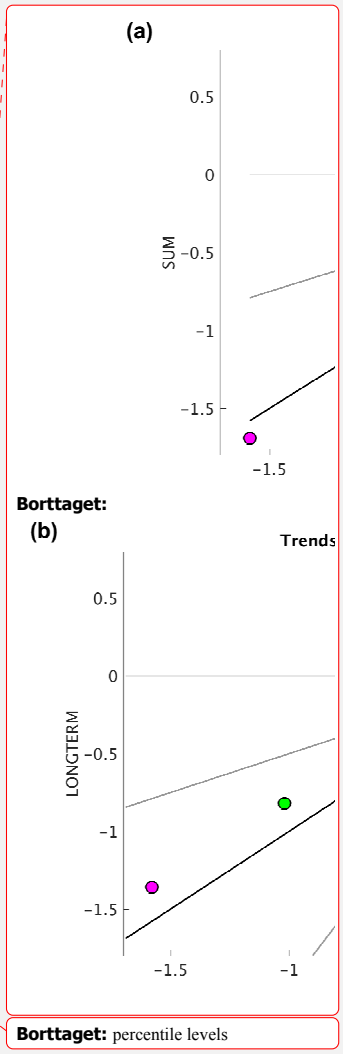

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the trend (SUM) vs the MATCH model simulation MFG (a) and the reanalysis LONGTERM vs the MATCH model simulation MFG (b). Filled circles indicate significant trends ($p \leq 0.05$) in the MFG simulation, whereas non-significant MFG trends ($p > 0.05$) are indicated by an empty circle. (c) Sensitivity in contributions to the secular trends in regionally (North: squares, Central: circles, South: diamonds) averaged annual percentiles (levels: 0, 2, 5, 10, 25, 50, 75, 90, 95, 98 and 100) of hourly near-surface O_3 over the period 1990-2013 due to choice of base year (1990 vs 2011). Modelled contributions to the near-surface ozone trend due to change in top and lateral boundaries of relevant species (dark yellow, bound), change in full domain emissions (blue, SE emis + FD emis), and variation in meteorology (fair yellow, meteo). 1:1 line in black, factor 2 lines in dark grey.

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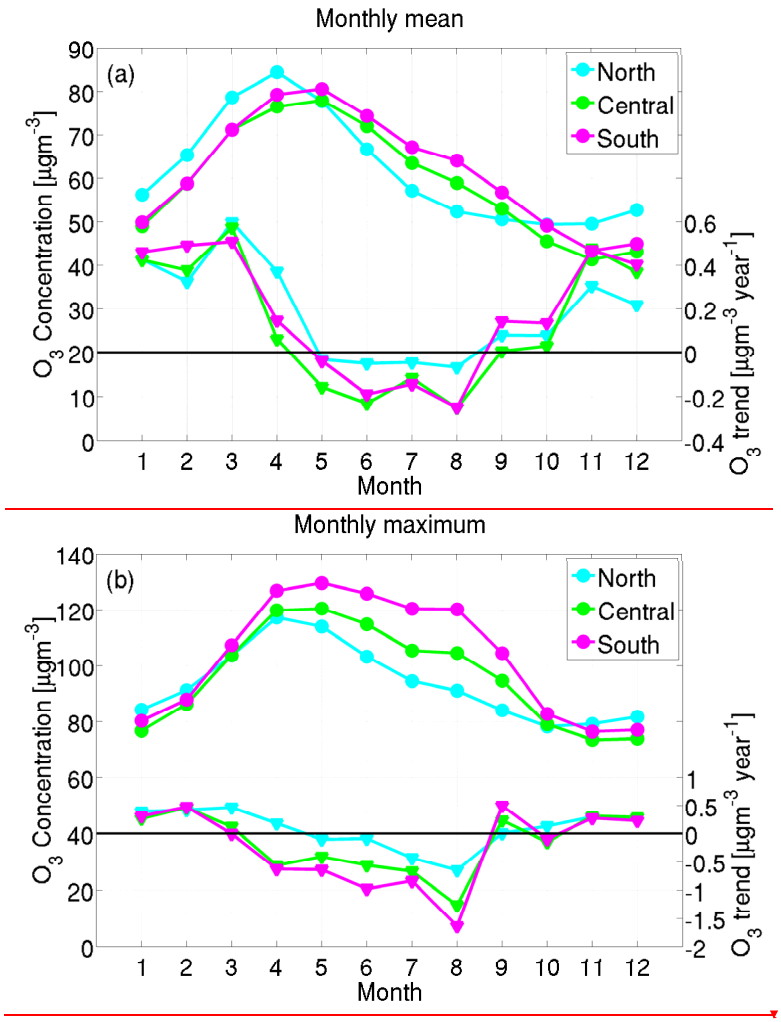
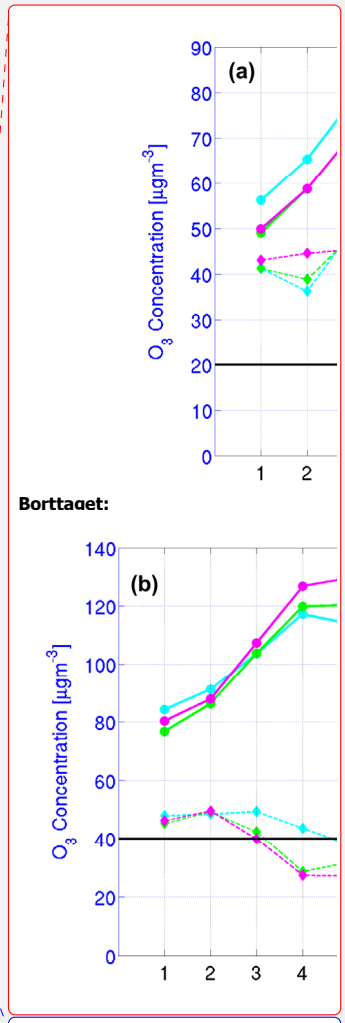


Figure 6. Seasonal cycle of monthly mean (a) and monthly maximum (b) of 1h mean near-surface ozone concentrations averaged over the period 1990-2013 (circles; left vertical scale) and region (North, Central and South Sweden, cf. Fig. 3) and the linear trend over the same period of the respective spatially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



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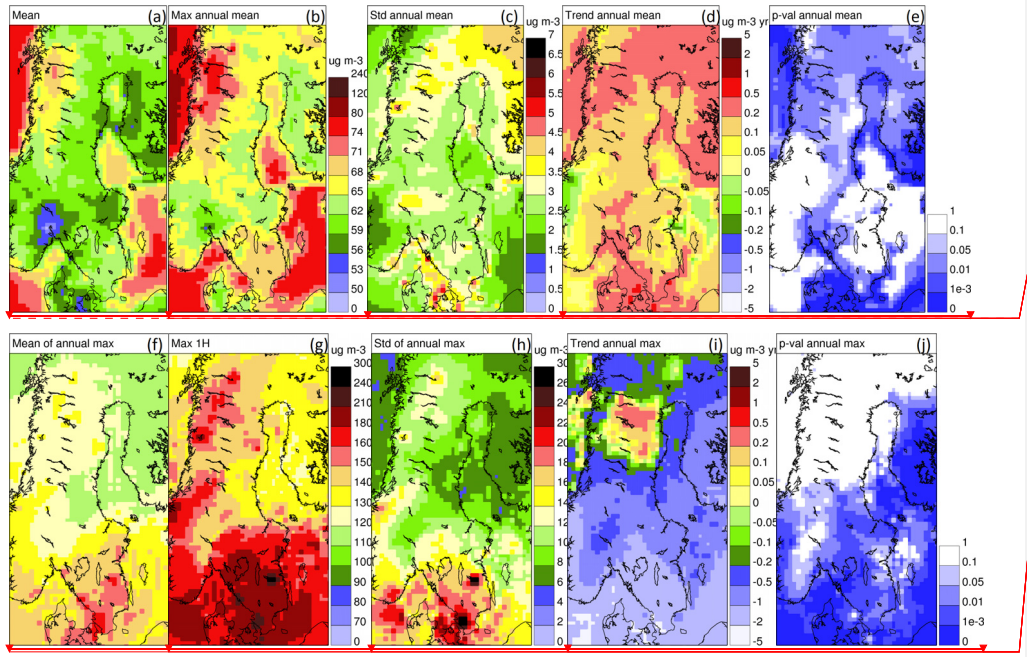


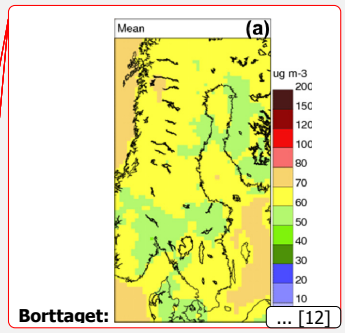
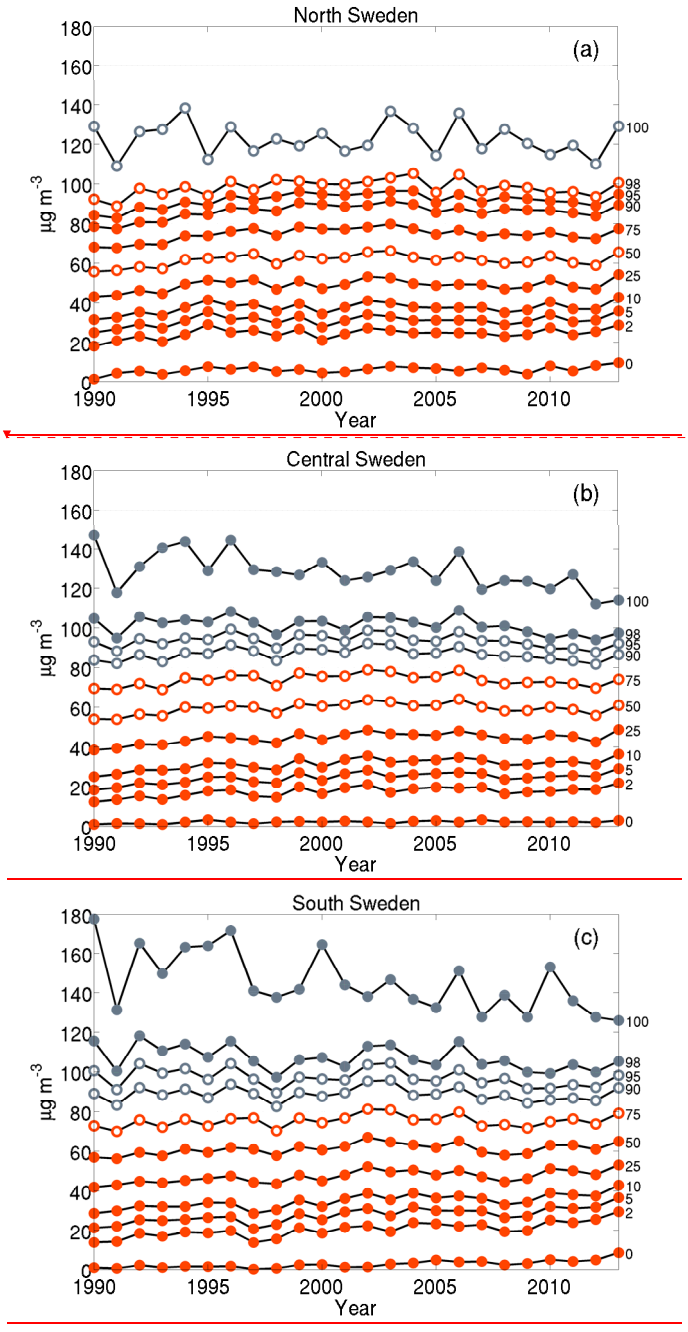


Figure 7. Statistical properties of the annual mean (top row; (a)-(e)) and annual maximum 1h mean (bottom row; ((f)-(j)) near-surface ozone concentration. In the columns from left to right: 1990-2013 mean ((a),(f)), 1990-2013 maximum ((b),(g)), 1990-2013 standard deviation ((c),(h)), linear trend over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



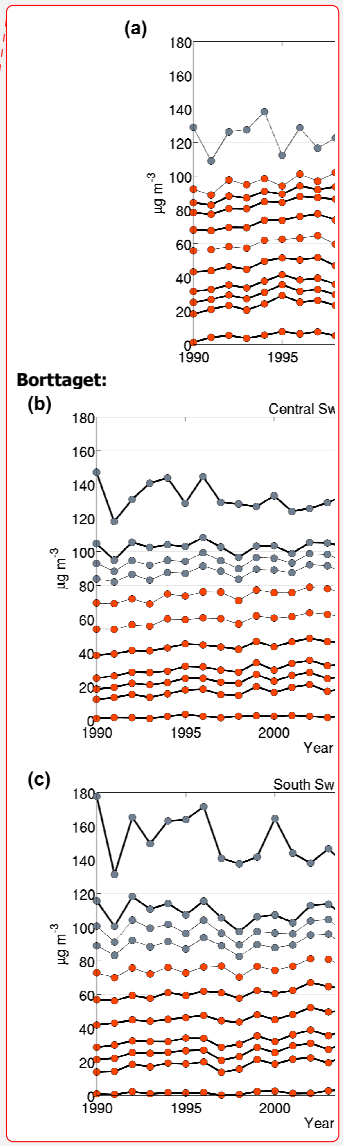


Figure 8. Temporal variation of annual percentiles of near-surface ozone concentrations averaged over the three regions North (a), Central (b) and South (c) of Sweden (cf. Fig. 3). The line marked 0 is the zero-percentiles (lowest hourly mean near-surface ozone

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 S2, 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.

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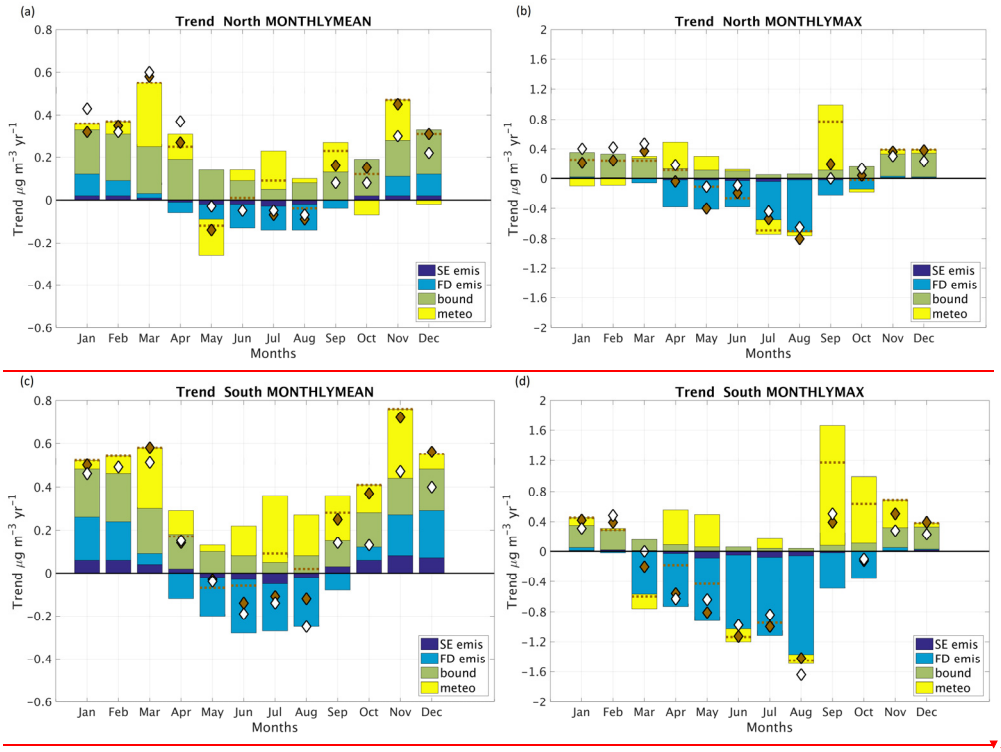
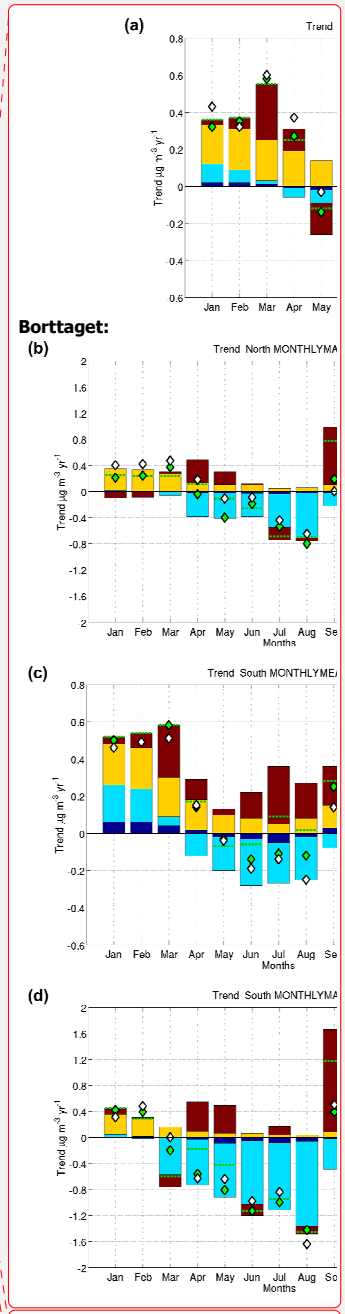


Figure 9. Linear trend over 1990-2013 in monthly mean ((a),(c)) and monthly maximum 1 hour mean ((b),(d)) near-surface ozone concentration for the North ((a),(b)) and the South ((c),(d)) Swedish regions (cf. Fig. 3). Reanalyzed (white diamond; LONGTERM reanalysis) and modelled "first guess" (MFG) near-surface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



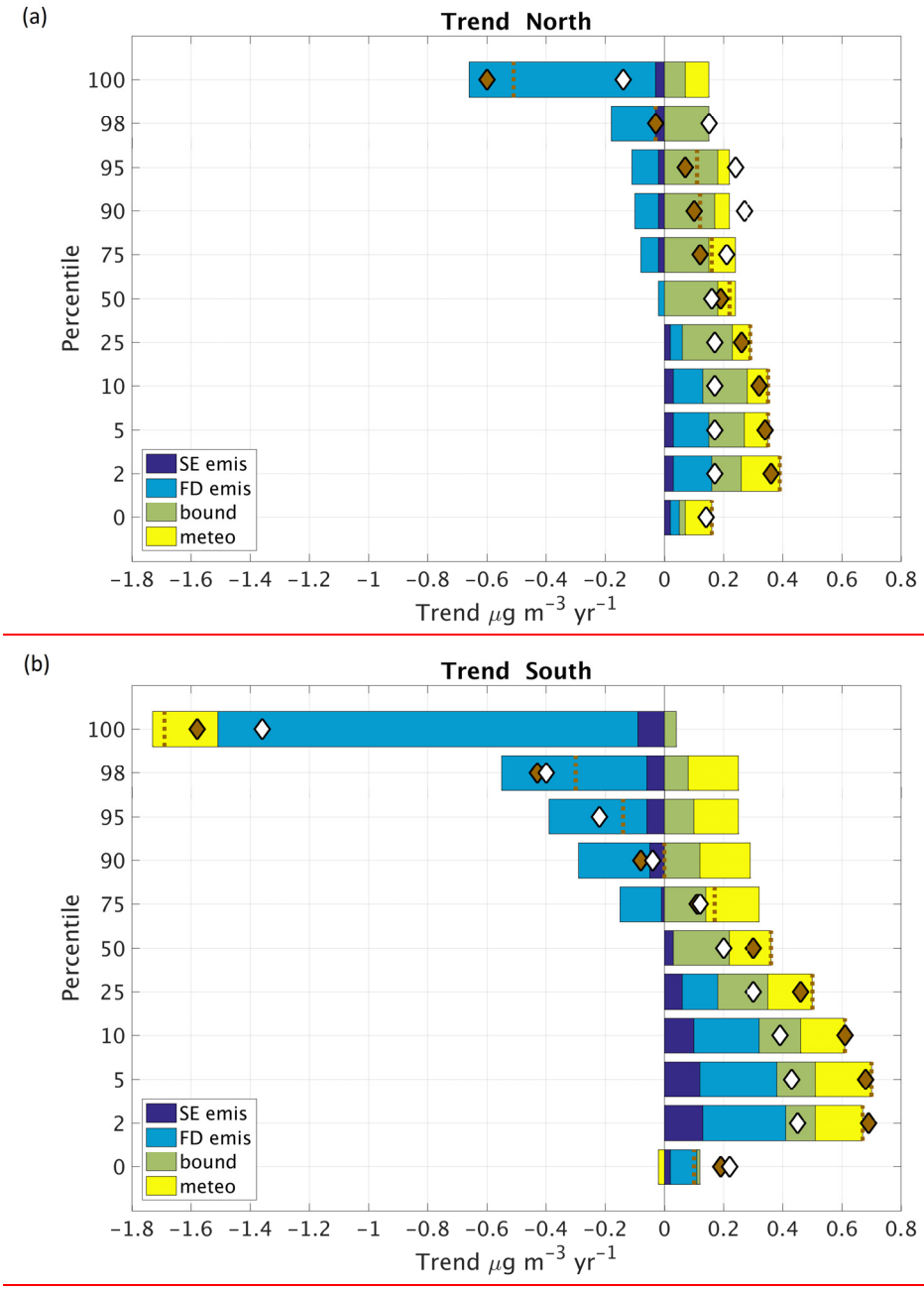


Figure 10. Linear trends over 1990-2013 in annual percentiles of hourly mean near-surface ozone concentrations for the North (a) and the South (b) Sweden regions. Reanalyzed (white diamond; LONGTERM reanalysis) and modelled MFG near-surface ozone trend (brown

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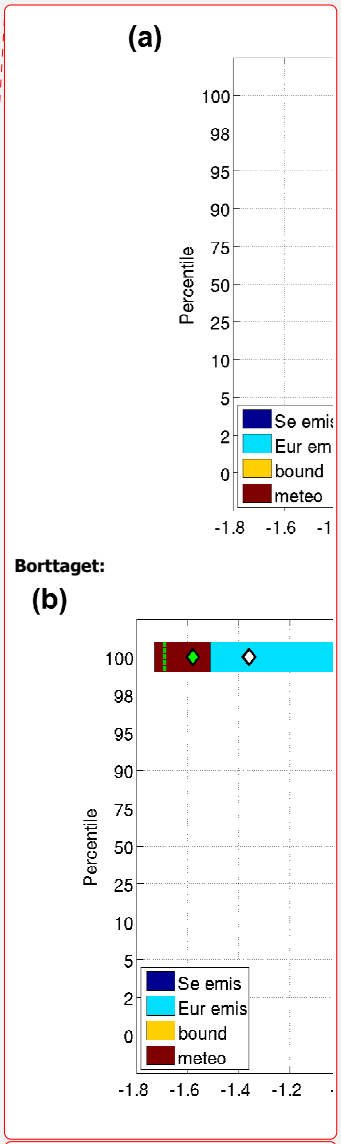

Borttaget: green

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, FD emis), emissions, trend in top and lateral boundaries of relevant species (dark yellow, bound) and variation in meteorology (fair yellow, meteo). The sum of the modelled contributions is indicated by the dashed brown line.

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

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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.
MFD	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.

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Table 1b. Formation of contributions to the linear trend over the period 1990-2013 from the sensitivity simulations (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.
FD emis	Contribution to the trend caused by the change in full domain anthropogenic, non-Swedish, emissions, calculated as the model scenario difference: (MFG-MFD)-(MFG-MSE).
emis	Contribution to the trend caused by the change in full domain anthropogenic emissions, calculated as the model scenario difference: MFG-MFD. Used only for the base year sensitivity investigation.
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+FD emis+Bound+Meteo.

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Table 2. Evaluation of modelled hourly and daily maximum of 1h mean 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/days[6] 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 evaluation 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 evaluation statistics					
Hourly mean		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	
Daily maximum		mean (ppb(v))	std dev (ppb(v))	%bias (%)	r	RMSE (ppb(v))	#days
ALL	obs	39.4	8.7				365
	MFG	37.7	7.6	-4.3	0.79	5.8	
	cross	38.3	7.9	-2.6	0.83	5.2	
	2dvar	39.5	8.5	0.3	0.97	1.4	
LONGTERM	obs	40.0	8.7				365
	MFG	37.8	8.0	-5.6	0.79	6.0	
	cross	38.7	7.9	-3.3	0.81	5.6	
	2dvar	40.4	8.9	1.0	1.00	0.8	
		spatial statistics of annual means					

[6] A daily data coverage of 75% (>18 hours) is required to include the observed daily maximum as a valid observation.

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Annual mean		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	
Annual mean of daily maximum		mean (ppb(v))	std dev (ppb(v))	%bias (%)	r	RMSE (ppb(v))	#stns
ALL	obs	39.4	1.3				12
	MFG	37.7	1.2	-4.4	0.43	2.5	
	cross	38.3	1.6	-2.7	0.31	2.3	
	2dvar	39.5	1.6	0.3	0.90	1.0	
LONGTERM	obs	40.0	1.6				6
	MFG	37.8	1.4	-5.7	X	2.9	
	cross	38.7	2.0	-3.4	X	2.7	
	2dvar	40.4	1.6	1.0	X	0.4	

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1 Table 3. Linear trend during 1990-2013 of policy related metrics in the 3 Swedish regions
2 North, Central and South (cf. Fig. 3). Stars (*, **, and ***) indicate that the trend is
3 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****

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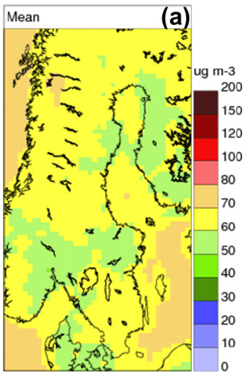

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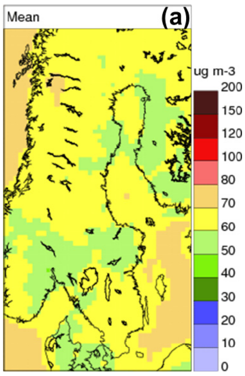

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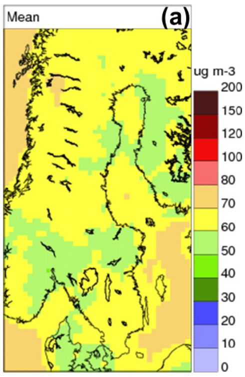

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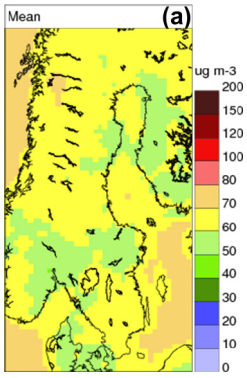

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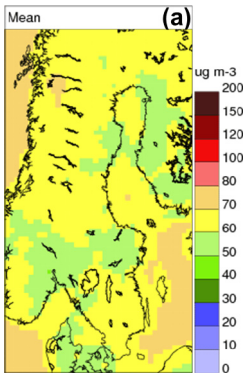

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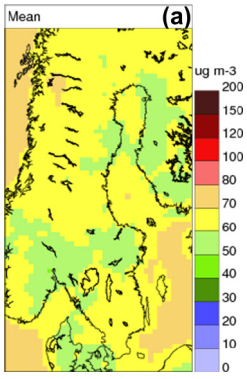

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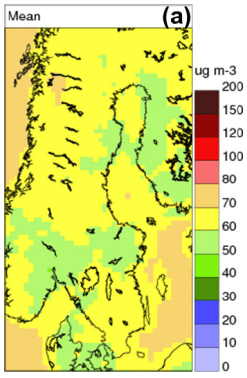

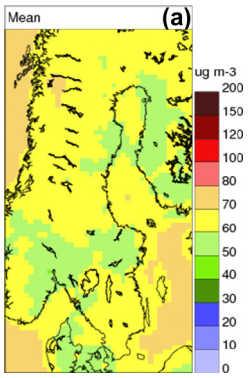

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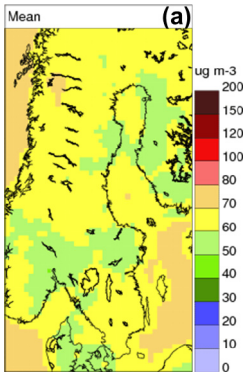

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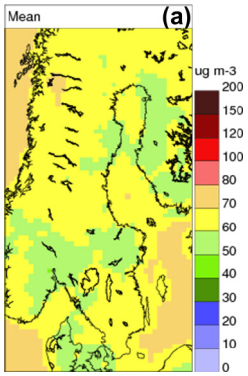

S1. Evaluation metrics

The evaluation is made using the metrics defined through the following equations,

$$mean = \overline{x_a} = \sum_{i=1}^{N} \frac{x_a^i}{N} \quad (1)$$

$$\sigma = \sigma_a = \sqrt{\frac{1}{N-1}\sum_{i=1}^{N}\left|x_a^i - \overline{x_a}\right|^2} \quad (2)$$

$$\%bias = 100 * \frac{\bar{x}_{mod} - \bar{x}_{obs}}{\bar{x}_{obs}} \quad (3)$$

$$r = r(x_{obs}, x_{mod}) = r(x_{mod}, x_{obs}) = \frac{1}{N}\sum_{i=1}^{N}\left(\frac{x_{obs}^i - \overline{x_{obs}}}{\sigma_{obs}}\right)\left(\frac{x_{mod}^i - \overline{x_{mod}}}{\sigma_{mod}}\right) \quad (4)$$

$$RMSE = \sqrt{\frac{1}{N}\sum_{i=1}^{N}(x_{mod}^i - x_{obs}^i)^2} \quad (5)$$

Where x_a^i is the observed (x_{obs}) or modelled (x_{mod}) values, σ is the standard deviation of obsevations (σ_{obs}) and model values (σ_{mod}), %bias is the model mean bias normalized by the observed mean, r is the Pearson correlation coefficient between observed and modelled values, and the RMSE is the root mean square error of the modelled values compared to the observed.

Table 2 in the main paper includes spatial mean of hourly statistics and spatial statistics of annual means:

- The spatial mean of hourly statistics is calculated as follows: The above metrics (eqns. (1)-(5)) are calculated for hourly near-surface O_3 concentrations at each of the measurement sites. These statistics are then averaged spatially over the measurement sites (as in eqn. (1)). This evaluates the temporal performance.
- The spatial statistics of hourly means are calculated as follows: The annual mean (2013) is calculated from the observed respective modelled hourly near-surface O_3 at each of the observation sites. These observed and modelled annual mean pairs are then used in the calculation of the above metrics (eqns. (1)-(5)). This evaluates the spatial performance.

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S2. Evaluation of annual means

The 2dvar analysis significantly improves the correlation coefficient and RMSE at the observation sites of modelled annual mean near-surface O_3 as compared to the MFG simulation (Table S1): The average correlation coefficient, 0.46, in the MFG, is improved to 0.87 in the LONGTERM reanalysis, and reaches 0.99 in the ALL reanalysis. The RMSE is also improved in the ALL and LONGTERM reanalyzes. This is expected, since the ALL reanalysis is dependent on all the observations included in the evaluation, LONGTERM is dependent on part of the observations, whereas the MFG simulation is independent of the observations. It is striking that the mean bias is very low for all simulations, including the observation independent MFG. The MFG simulation underestimates the inter-annual variation, whereas the variations in the reanalyzes are similar to the variations in the measurements. The spatial statistics of the 2dvar analysis are similar to or better than the MFG simulation. The correlation coefficient of the multi-year means is poor for the MFG simulation and the spatial variation is underestimated, but both are strongly improved in the LONGTERM and ALL reanalyzes.

S3. Time series comparison of ALL and LONGTERM

To understand how the number of measurement sites included in the two assimilated data sets affects the time series for a larger spatial area, we compare the trends in annual mean and annual max (Fig. S2-S3) obtained with the two simulations. The annual values are averaged for three regions (North, Central and South, as illustrated in the main paper Fig. 3). The time series of ALL and LONGTERM diverge, especially in the later part of the period, which is due to an introduction of more measurement sites in the later part of the ALL simulation. Several of these sites experience strong night-time temperature inversions, which in turn result in very low night-time O_3 concentrations. For this reason the annual max does not diverge as much as the annual means. Thus the estimated trend differs for the two simulations, with the largest difference for annual mean in southern and central Sweden. To eliminate such impacts on the trend statistics, we will therefore focus on the LONGTERM simulation in the assessments of these metrics. In Fig. S3 we also include a comparison of the annual mean time series for observations and the MFG, LONGTERM and ALL simulations at each of the measurement sites. The trend figures illustrate the evaluation scores: good performance by MFG at many sites and improvements due to the variational analysis, with best performance

compared to the observations by the ALL simulation. Further, it is clear that the time series of LONGTERM and ALL diverge at the measurement sites with fewer years of data, further strengthening our conclusion about using the LONGTERM data set for trend and extreme estimation.

S4. Figures and tables

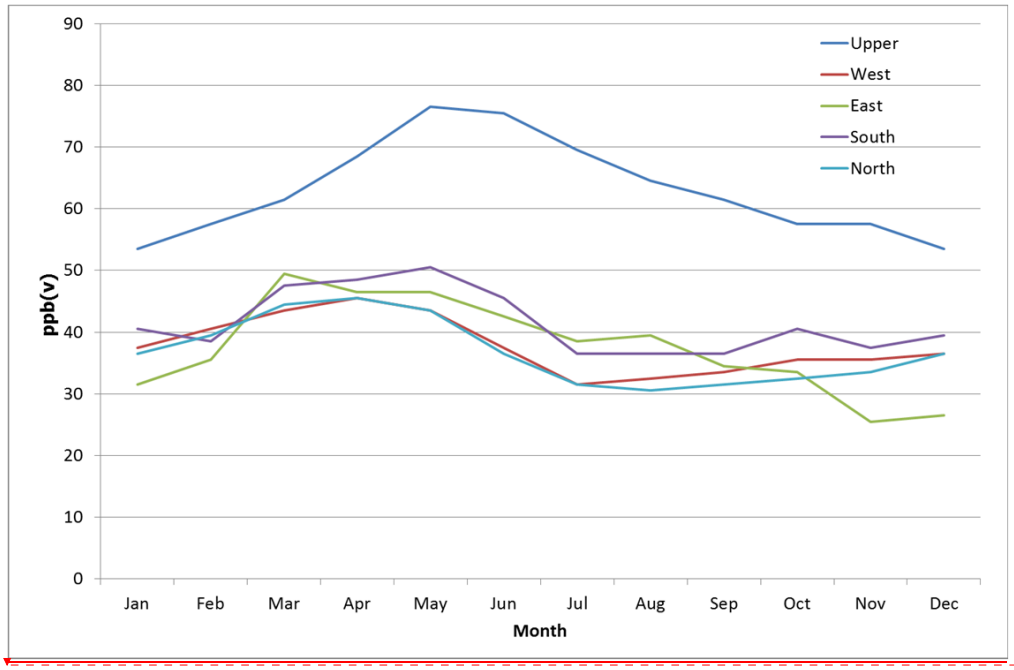

Figure S1. Seasonal variation in lateral and upper boundary conditions for ozone in the year 2011. The upper boundary is at approximately 5 km height. Unit: ppb(v)

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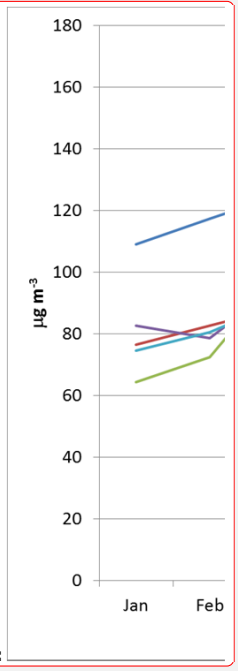

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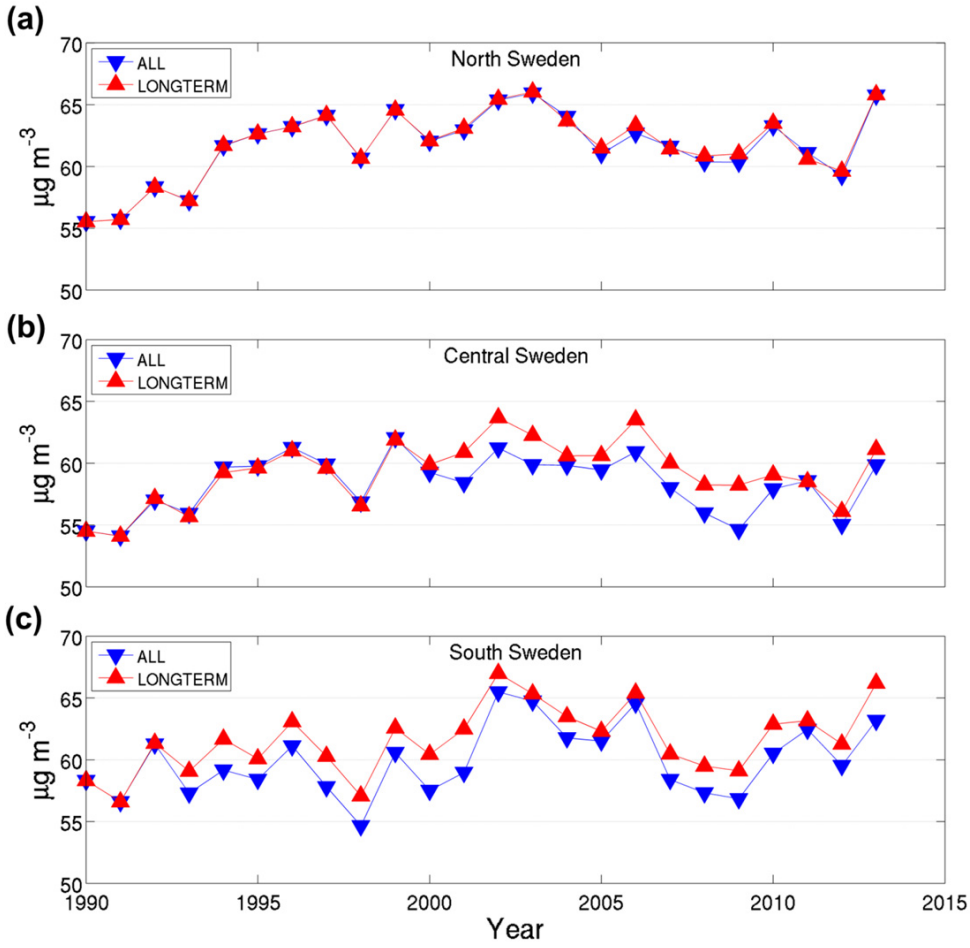


Figure S2. Time series of annual mean near-surface ozone concentrations averaged over three regions (North (a), Central (b) and South (c) Sweden, cf. main paper Fig. 3), for the two reanalyzes ALL (blue) and LONGTERM (red).

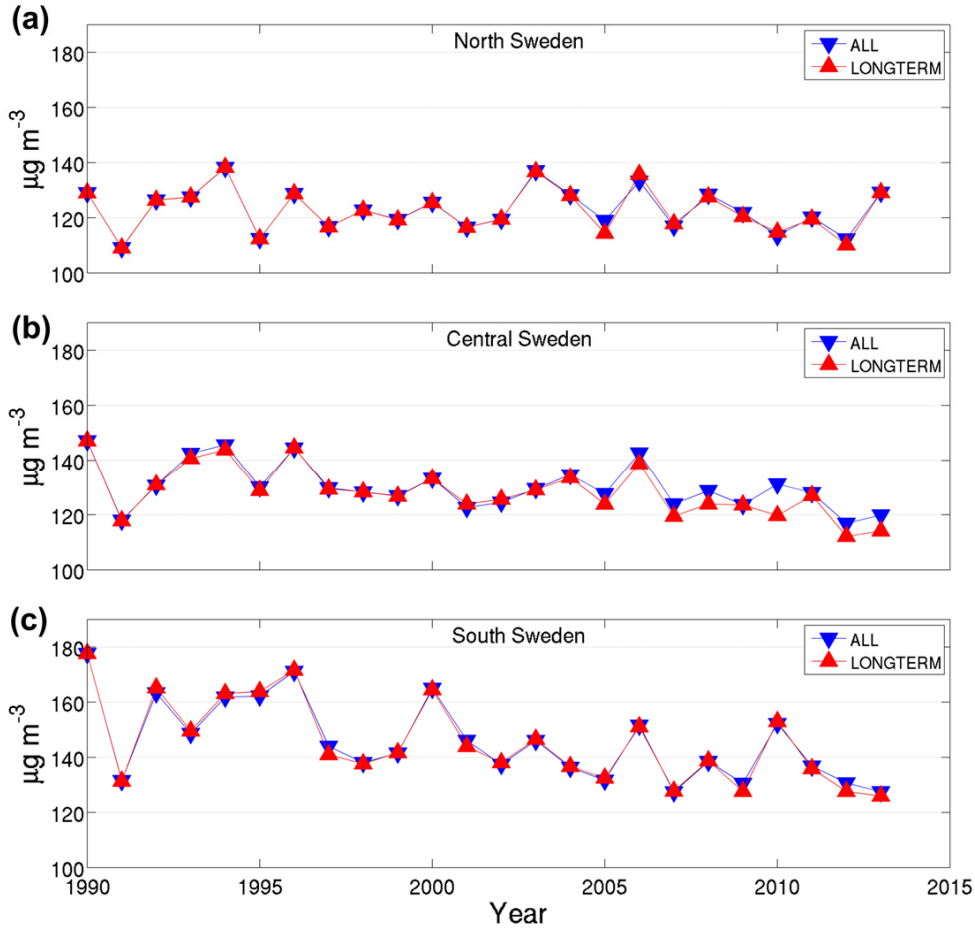


Figure S3. Time series of annual maximum of 1h mean near-surface ozone concentrations averaged over three regions (North (a), Central (b) and South (c) Sweden, cf. main paper Fig. 3), for the two reanalyzes ALL (blue) and LONGTERM (red).

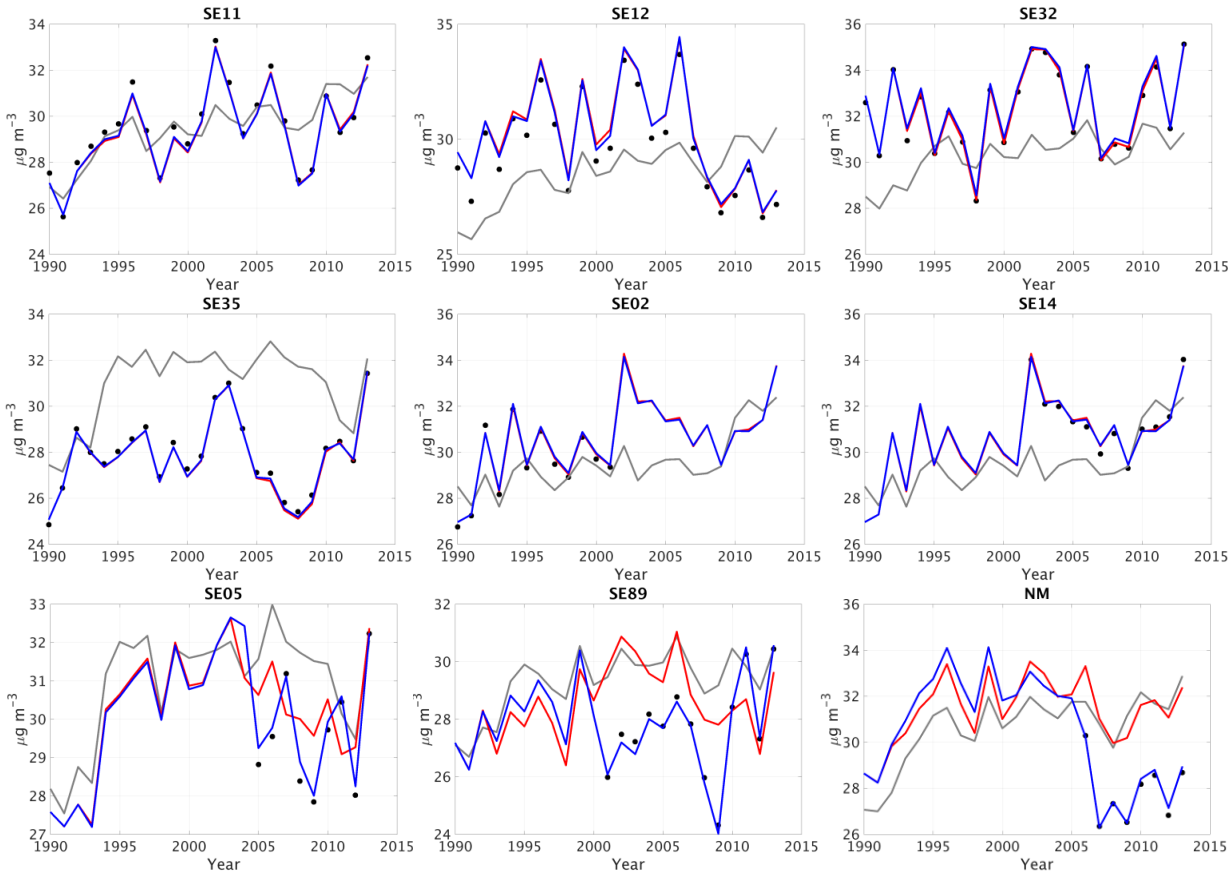


Figure S4. Time series of annual mean near-surface ozone at Swedish measurement sites with more than 1 year of measurement data. Observations (black circle), the "first guess" simulation MFG (grey line), the two reanalyzes LONGTERM (red) and ALL (blue).

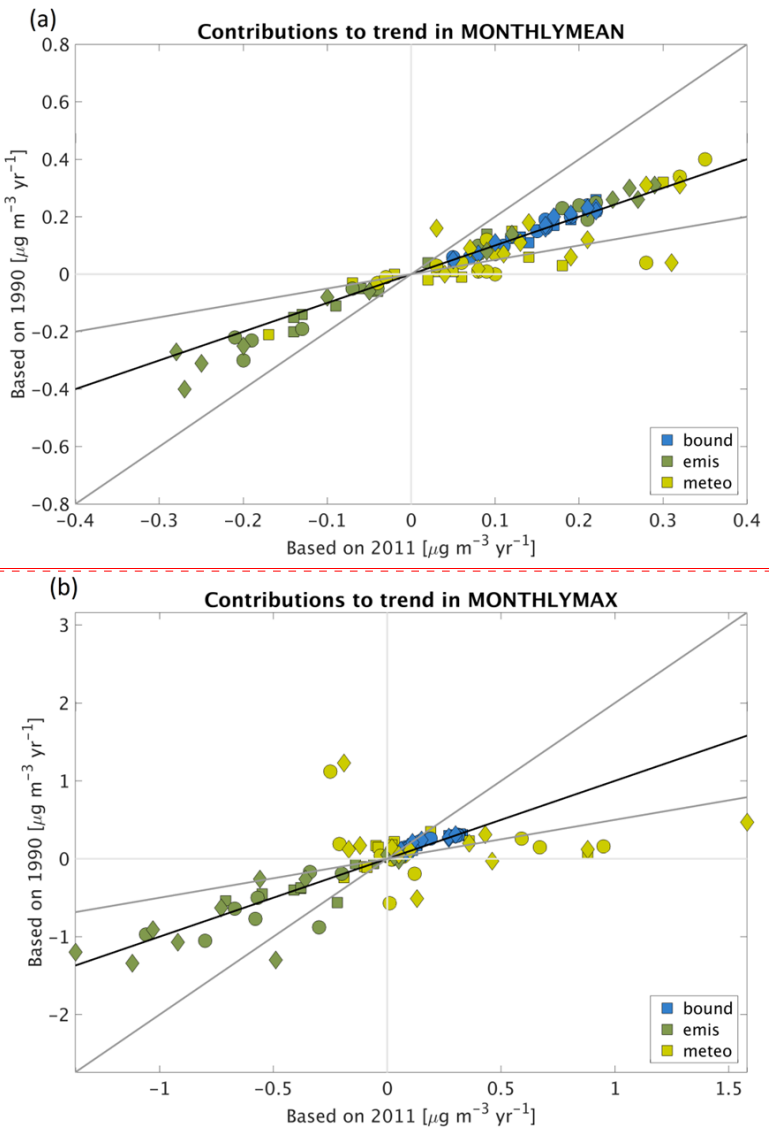
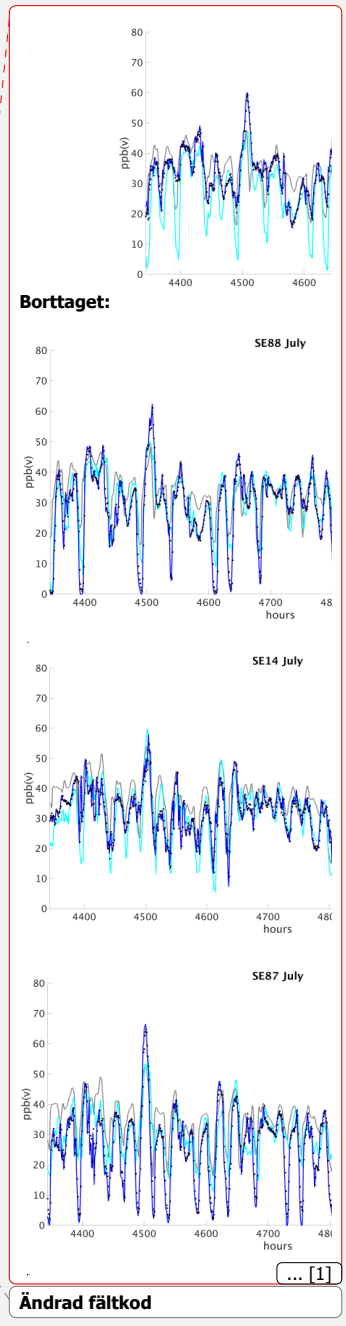


Figure S5. Sensitivity in contributions to the secular trends in regionally (North: squares, Central: circles, South: diamonds) averaged monthly mean (a) and maximum (b) hourly near-surface O_3 over the period 1990-2013 due to choice of base year (1990 vs 2011). Modelled contributions to the near-surface ozone trend due to change in top and lateral boundaries of relevant species (dark yellow, bound), change in full domain emissions (blue, SE emis + FD emis), and variation in meteorology (fair yellow, meteo). 1:1 line in black, factor 2 lines in dark grey.



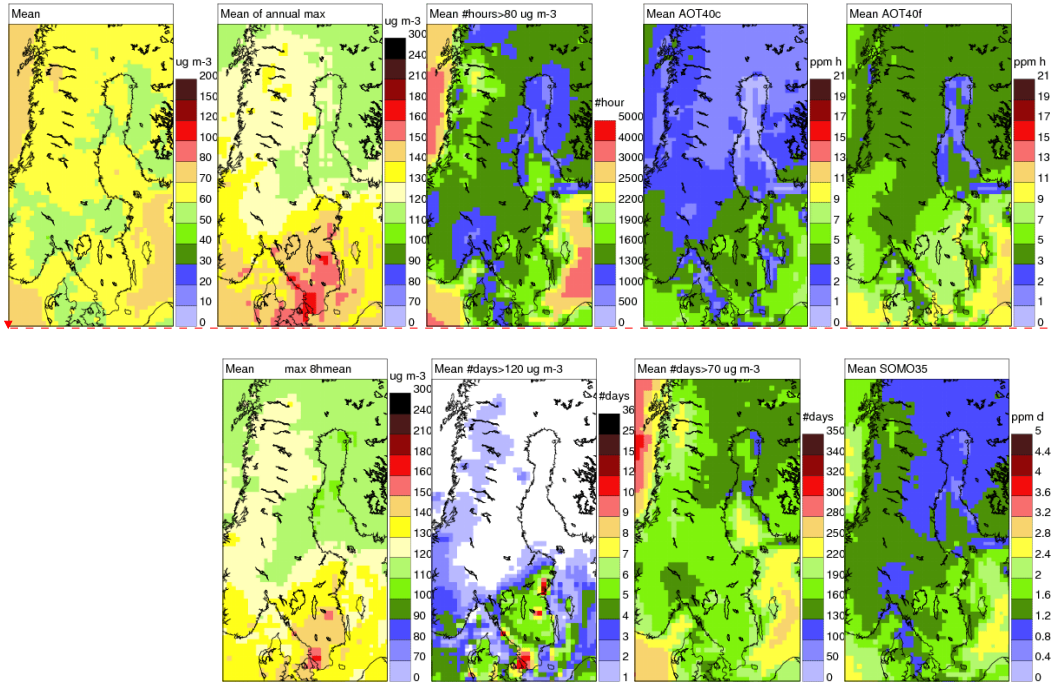


Figure S6. Period mean near-surface ozone during 1990-2013 from top left: Annual mean, annual max of 1hour mean (Max 1H), number of hours exceeding 80 µg m^{-3}, AOT40 in crop growing season (AOT40c; May-July), AOT40 in forest growing season (AOT40f; April-September), annual max of running 8hour mean, number of days with daily max of running 8hour mean exceeding 120 µg m^{-3} and 70 µg m^{-3}, and the health indicator SOMO35. Results from the LONGTERM reanalysis.

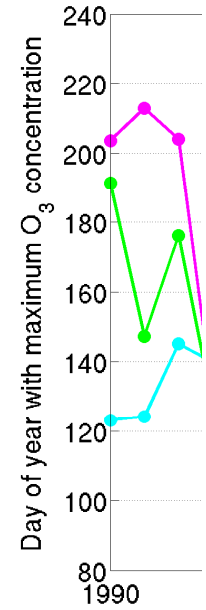


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Figure S6. Day of the year (1=1 Jan, 32=1 Feb. etc.) when the daily maximum 1h mean near-surface ozone reaches its annual maxima. The values displayed are spatial averages over the 3 Swedish regions: North, Central and South. Results from the LONGTERM reanalysis.¶
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Avsnittsbrytning (nästa sida)

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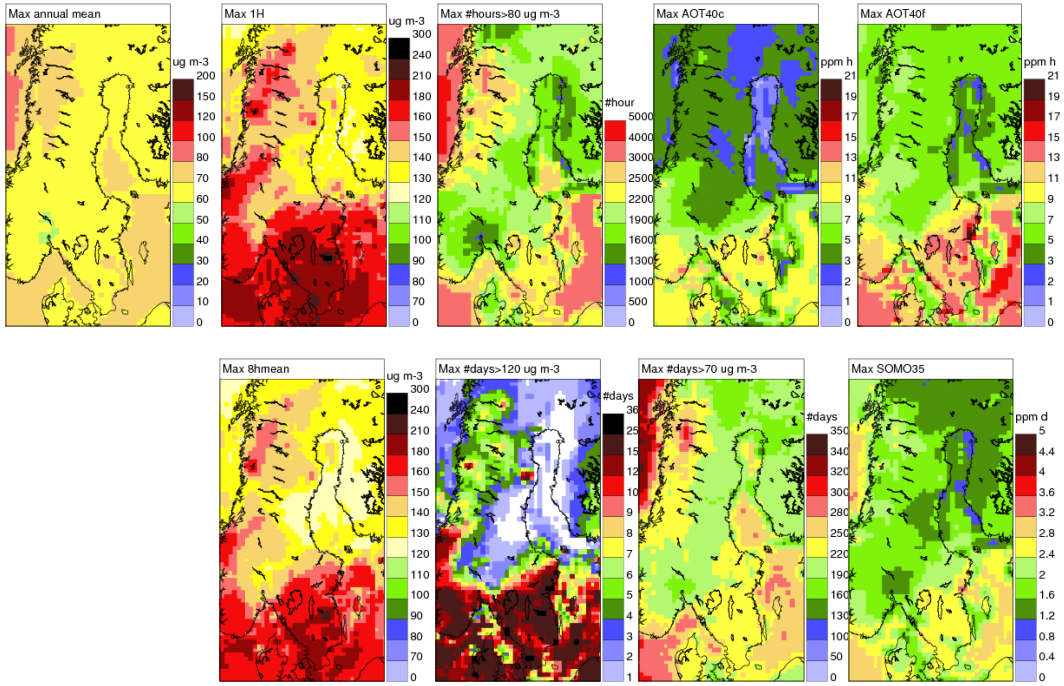

Figure S7. Period max value in near-surface ozone during 1990-2013 from top left: Annual mean, annual max of 1hour mean (Max 1H), number of hours exceeding 80 µg m^{-3}, AOT40 in crop growing season (AOT40c; May-July), AOT40 in forest growing season (AOT40f; April-September), annual max of running 8hour mean, number of days with daily max of running 8hour mean exceeding 120 µg m^{-3} and 70 µg m^{-3}, and the health indicator SOMO35. Results from the LONGTERM reanalysis.

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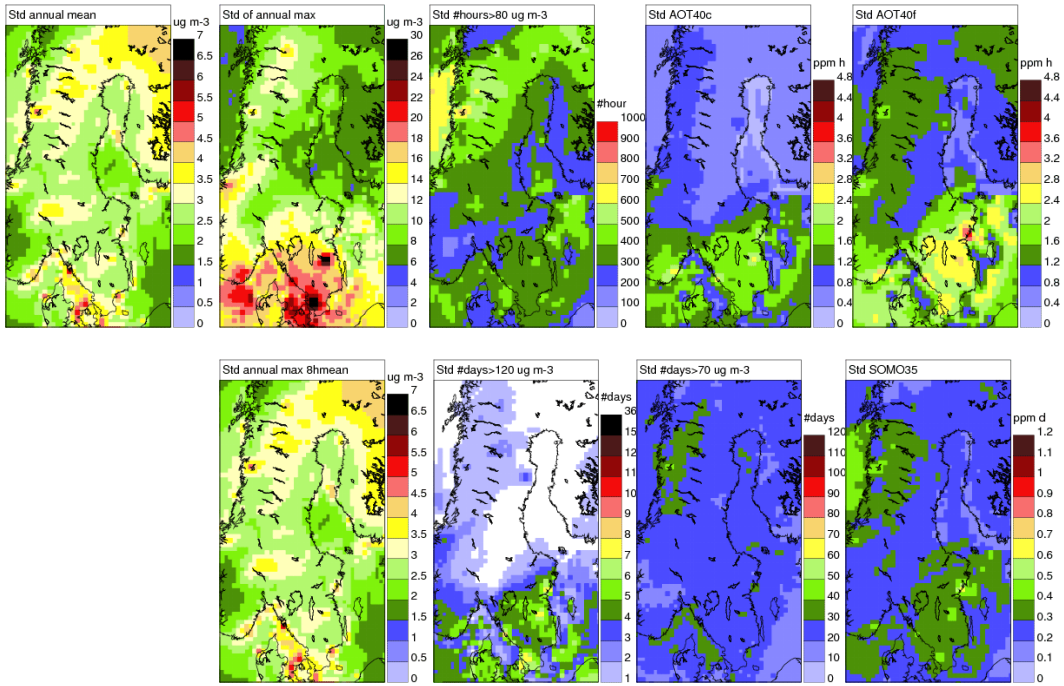

Figure S8. Period standard deviation in near-surface ozone during 1990-2013 from top left: Annual mean, annual max of 1hour mean (Max 1H), number of hours exceeding 80 µg m^{-3}, AOT40 in crop growing season (AOT40c; May-July), AOT40 in forest growing season (AOT40f; April-September), annual max of running 8hour mean, number of days with daily max of running 8hour mean exceeding 120 µg m^{-3} and 70 µg m^{-3}, and the health indicator SOMO35. Results from the LONGTERM reanalysis.

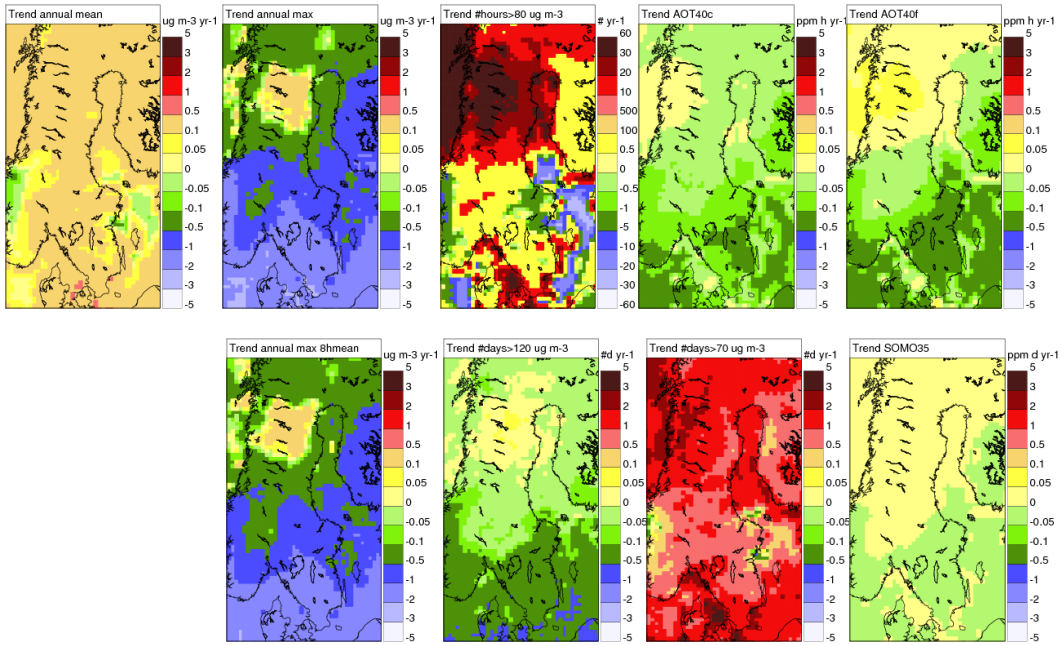

Figure S9. Period linear trend in near-surface ozone during 1990-2013 from top left: Annual mean, annual max of 1hour mean (Max 1H), number of hours exceeding 80 µg m^{-3}, AOT40 in crop growing season (AOT40c; May-July), AOT40 in forest growing season (AOT40f; April-September), annual max of running 8hour mean, number of days with daily max of running 8hour mean exceeding 120 µg m^{-3} and 70 µg m^{-3}, and the health indicator SOMO35. Results from the LONGTERM reanalysis.

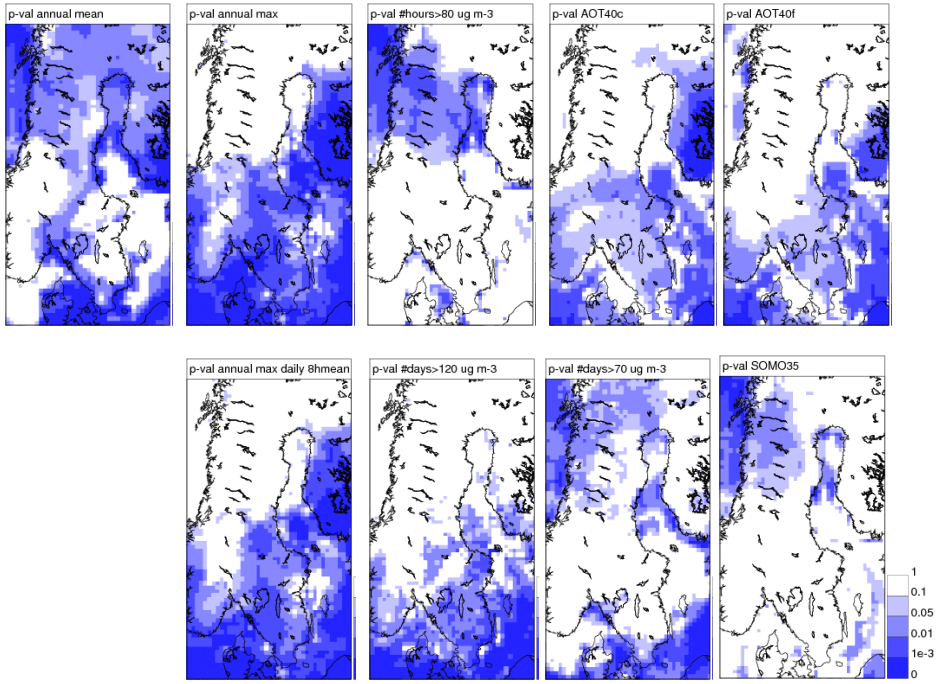

Figure S10. p-value in near-surface ozone linear trend over the period 1990-2013 from top left: Annual mean, annual max of 1hour mean (Max 1H), number of hours exceeding 80 µg m^{-3}, AOT40 in crop growing season (AOT40c; May-July), AOT40 in forest growing season (AOT40f; April-September), annual max of running 8hour mean, number of days with daily max of running 8hour mean exceeding 120 µg m^{-3} and 70 µg m^{-3}, and the health indicator SOMO35. p-values above 0.1 are non-significant (white). Results from the LONGTERM reanalysis.

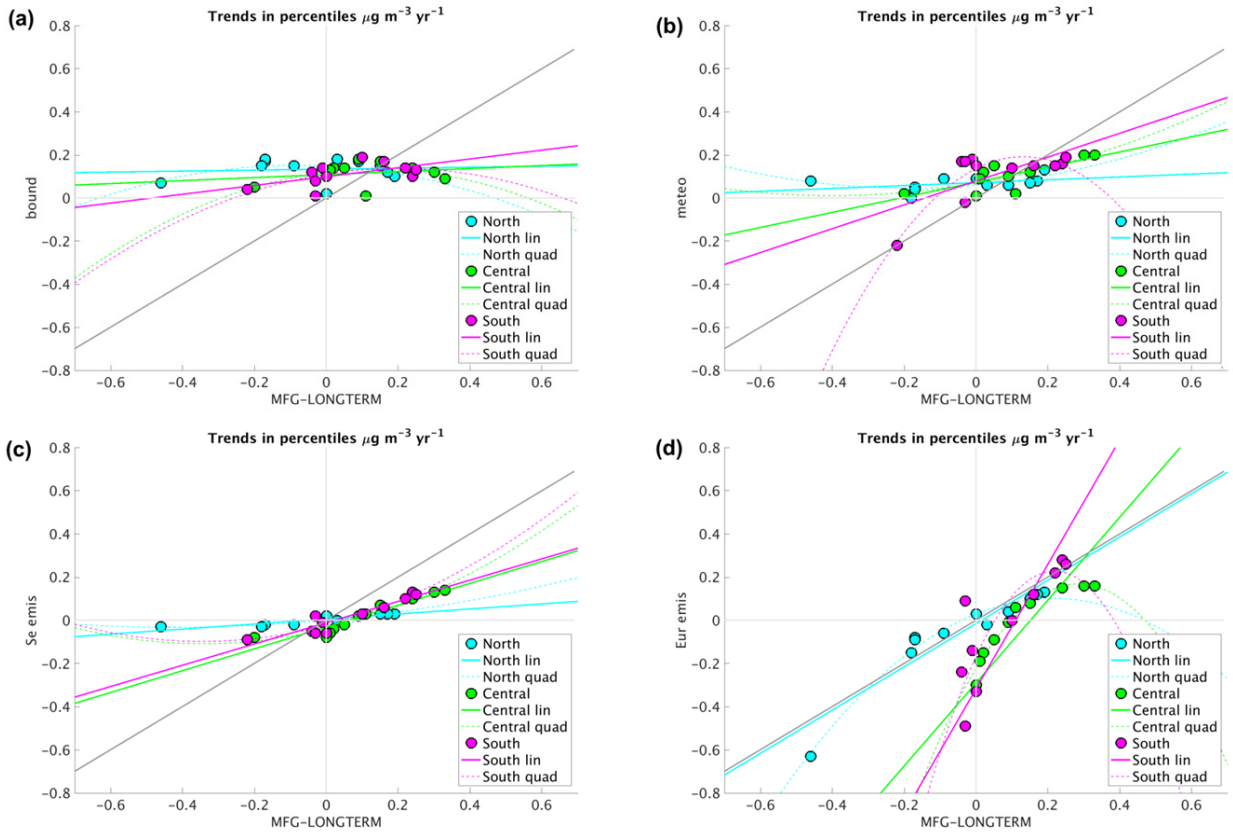

Figure S11. Trends in contributions (bound (a), meteo (b), Se emis (c), Eur emis(d)) versus the error in trend modelled by the CTM (difference between trends in the MFG reanalysis and the LONGTERM simulation) for the three regions (North (blue), Central (green) and South (magenta) Sweden, see Fig. 1 in the main paper). Circles represent different percentiles; solid line is the 1st degree and dashed line is the 2nd degree regression fit of all percentiles in the respective region.

Table S1. Evaluation of annual mean near-surface ozone concentration at Swedish measurement sites for the (observation independent) "first guess" (the MATCH base case simulation, MFG) and the two (observation dependent) reanalyzed data sets (ALL and LONGTERM) over the period 1990-2013. Mean value (mean), standard deviation (σ), mean bias normalized by the observed mean (%bias), Pearson correlation coefficients (r), root mean square error (RMSE) and mean number of years (#years) or measurement sites (#stns[1]). The top half of the table shows the mean over the 10 stations of the evaluation statistics at each measurement site (mean of yearly statistics). The bottom half of the table shows spatial evaluation statistics of the period (1990-2013) mean near-surface ozone concentration at the measurement sites (spatial statistics of multi-year means).

	mean of yearly statistics					
	mean (ppb(v))	**σ (ppb(v))**	**%bias (%)**	**r**	**RMSE (ppb(v))**	**#years**
Obs	29.8	1.7				17.3
MFG	30.0	1.1	0.5	0.46	2.65	17.3
LONGTERM	30.5	1.6	2.1	0.87	0.91	17.3
ALL	29.9	1.7	0.3	0.99	0.25	17.3
	spatial statistics of multi-year means					
	mean (ppb(v))	**σ (ppb(v))**	**%bias (%)**	**r**	**RMSE (ppb(v))**	**#stns**
Obs	29.8	1.4				10
MFG	30.0	0.9	0.5	-0.40	2.4	10
LONGTERM	30.5	1.3	2.1	0.78	1.3	10
ALL	29.9	1.5	0.3	0.99	0.2	10

[1] From the 13 Swedish measurement sites 12 were included in the evaluation, due to the requirement of a minimum of 6 years with more than 80% data coverage at observation sites. The station pair Rörvik and Råö, was considered as one site, thus the 10 sites in the spatial evaluation.

Table S2. Linear trend of percentiles in the 3 Swedish regions. Stars (*, **, and ***) indicate that the trend is significant (p≤0.05, p≤0.01, p≤0.001, respectively). Unit: μg m^{-3} $year^{-1}$.

Percentile	North	Central	South
100th	-0.14	-0.82**	-1.36***
98th	+0.15	-0.26*	-0.40*
95th	+0.24*	-0.10	-0.22
90th	+0.27*	-0.02	-0.04
75th	+0.21*	+0.07	+0.12
50th	+0.16	+0.15	+0.20*
25th	+0.17*	+0.24**	+0.30***
10th	+0.17*	+0.28***	+0.39***
5th	+0.17*	+0.27***	+0.43***
2nd	+0.17*	+0.24***	+0.45***
0	+0.14**	+0.05*	+0.22***

Flyttad (infogning) [1]

Borttaget: Table S2. Observed and modelled (MFG: MATCH modelled "first guess", cross: independent cross validation; ALL: observation dependent reanalysis) annual mean during 2013. Data sorted after the magnitude of the observational mean. Unit: ppb(v). Unit: ppb(v). ¶ ... [2]

Flyttad uppåt [1]: Unit: ppb(v).

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Sid. 8: [1] Borttaget **Andersson Camilla** **2017-09-20 17:55:00**

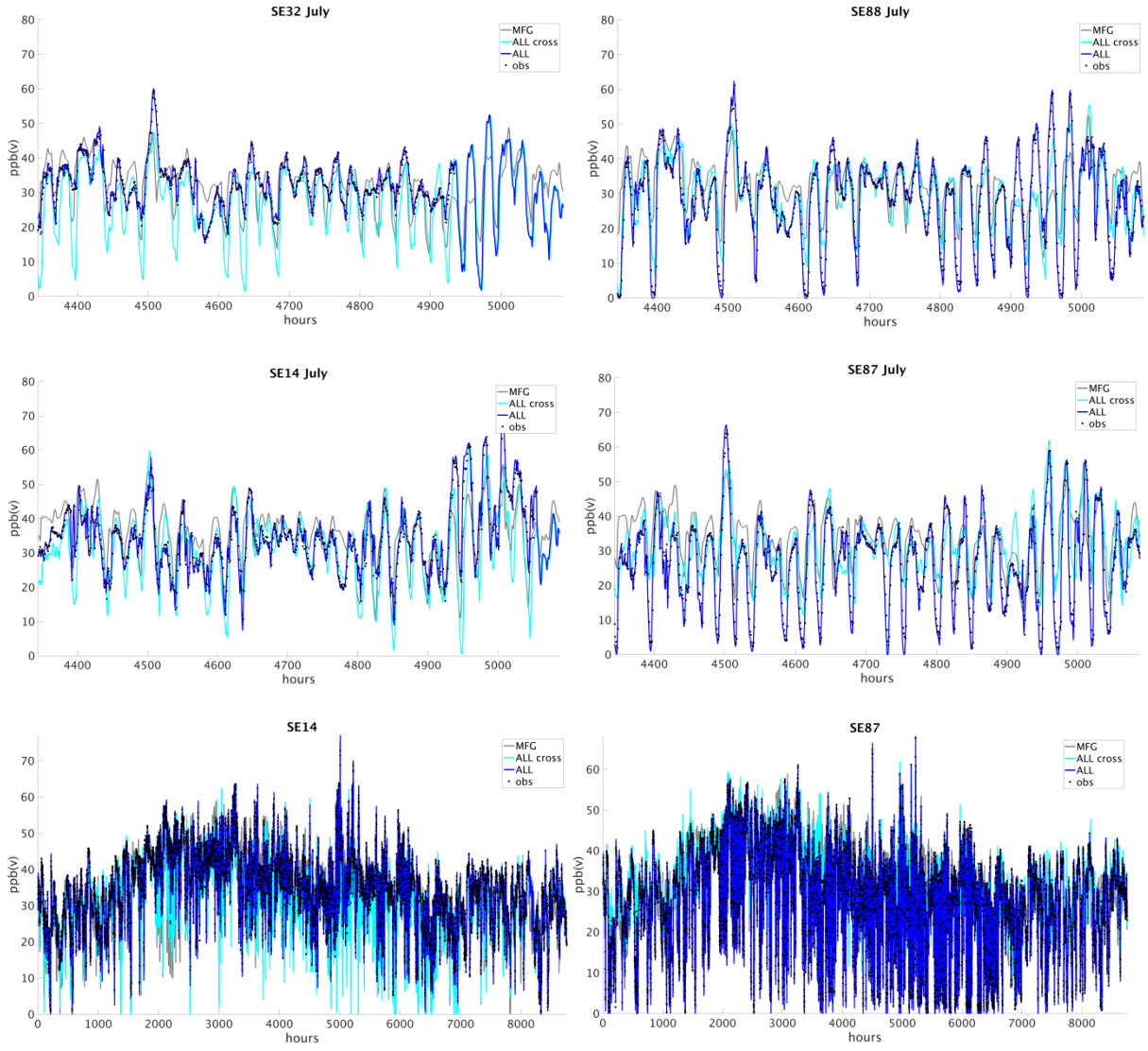

Figure S5. Hourly mean near-surface O_3 concentrations at selected sites including the MFG simulation (grey), ALL cross validation simulations (fair blue), the ALL reanalysis (dark blue) and observations (black circles). The bottom two panels show the full year 2013, the other zoom in on the month July 2013. Norra Kvill (SE32) and Råö (SE14) are less prone to be impacted by night-time inversions than Östad (SE87) and Asa (SE88).

Sid. 16: [2] Borttaget **Andersson Camilla** **2017-09-20 17:58:00**

Table S2. Observed and modelled (MFG: MATCH modelled "first guess", cross: independent cross validation; ALL: observation dependent reanalysis) annual mean during 2013. Data sorted after the magnitude of the observational mean. Unit: ppb(v). Unit: ppb(v).

	Obs	MFG	Cross	ALL	

RDB	27.0	27.1	24.4	24.6	Rödeby
SE12	27.2	30.5	31.4	27.4	Aspvreten
SE87	27.8	31.4	31.9	27.5	Östad
NM	28.7	32.8	33.1	28.9	Norr Malma
SE88	29.1	31.8	31.1	28.9	Asa försökspark
SE89	30.4	30.5	28.5	30.7	Grimsö
SE35	31.4	32.1	32.3	31.5	Vindeln
SE05	32.2	32.1	32.4	32.3	Bredkälen
SE11	32.5	32.2	32.0	32.7	Vavihill
SE14	34.0	32.4	31.2	33.8	Råö
SE32	35.1	31.3	29.1	35.6	Norra Kvill
SE13	35.1	28.8	29.5	35.2	Esrange
SE12	27.2	30.5	34.1	27.4	Aspvreten
SE35	31.4	32.1	32.3	31.4	Vindeln
SE11	32.5	32.2	34.1	32.6	Vavihill
SE14	34.0	32.4	33.1	33.8	Råö
SE32	35.1	31.3	30.0	35.4	Norra Kvill
SE13	35.1	28.8	29.5	35.2	Esrange