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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Reanalysis of and attribution to near-surface ozone concentrations in Sweden during 1990–2013" *by* Camilla Andersson et al.

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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 (as a supplement to this reply) 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.

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Discussion paper

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): 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 O3 precursors in Sweden resulting in weak horizontal gradients of near-surface O3 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.

Page-by-page comments:

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 O3 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 O3 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 O3 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 (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 O3 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 night-

time 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 O3 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

Jenkin, ME, Trends in ozone concentration distributions in the UK since 1990: Local, regional and global influences, Atmos. Environ., 42, 5434-5445, 2008

Fiore, AM., Levy II, H. & Jaffe, D., A. North American isoprene influence on intercontinental ozone pollution, Atmos. Chem. Physics, 11, 1697-1710, 2011

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

Please also note the supplement to this comment:
https://www.atmos-chem-phys-discuss.net/acp-2017-338/acp-2017-338-AC2-supplement.zip

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