

Interactive comment on “Have primary emission reduction measures reduced ozone across Europe? An analysis of European rural background ozone trends 1996–2005” by R. C. Wilson et al.

R. C. Wilson et al.

p.s.monks@leicester.ac.uk

Received and published: 11 November 2011

General comments

There is a lot in this paper. Too much actually. I recommend that the statistical analysis of the observations and the modelling be separated into two papers.

The concept of the study was to bring in a self-consistent manner the observations and models together and explore whether an ensemble approach of challenging models

C11780

with this data was useful. We would not like split either aspect. We find the comment a compliment in that we have tried to make a thorough and comprehensive analysis.

Although the methods are not very well described, it appears that the authors have done extensive smoothing of the data, which will reduce the noise and give statistically significant trends where none were present in the original data. I would argue that taking out the seasonal cycle is an appropriate tool, but smoothing out noise and other variations is not appropriate.

The referee is incorrect about this there has been no smoothing of the data. To this end a clarification of statistical methods has been made. Only deseasonalising of the data in order to remove the seasonal component from the trend has been conducted. Altered text p. 18440 l.24:

A variety of tools have been developed to assess ozone trends using the statistical programming language R (R Development Core Team, 2009). As a first approach, decadal and seasonally disaggregated O₃ trends have been characterised using the non-parametric loess regression (Cleveland, 1979) to fit more complex trends of daily or monthly O₃ concentrations with at least 75 % of hourly data coverage required.

To identify changes in mean, background and peak O₃, the openair package (Carslaw and Ropkins, 2011) was first used to deseasonalise O₃ time series using an stl-based method i.e. decomposing the time series into seasonal, trend and irregular components using loess regression - a technique extensively used on environmental datasets (Cleveland et al., 1990). Subsequently, with seasonality removed from the time series, annual linear trends (ppbv/yr) were then quantified using the non-parametric Mann-Kendall approach to trend detection which is commonly implemented in environmental sciences (Hirsch et al., 1982). In the openair package, the *MannKendall* function combines the non-parametric Mann-Kendall analysis for

C11781

trend (Hirsch et al., 1982) with Sen-Theil slope estimates (further details are available in Carslaw and Ropkins (2011)). Annual linear trends of O3 monthly means, 5th and 95th percentiles were calculated with at least 75 % of hourly data coverage required during each month of the time-series. Bootstrap and block-bootstrap techniques were used to quantify uncertainties and account for autocorrelation in the air quality data. Annual linear trends were also quantified in units of %/yr from log-transformed (Parrish et al., 2002; Parrish, 2006) data. Annual and seasonal trends are reported including 95 % confidence intervals and with a statistical significance of $p < 0.1$

In many respects, calculating seasonal O3 trends is a more useful and relevant metric, rather than an annual trend, even if the data have been deseasonalised.

We agree with the reviewer that seasonal trends are a more useful metric than the annual trend. However, seasonal trends calculated in this study were generally of reduced statistical significance (Table A5-A7, supplement).

In addition, the authors have chosen to use a P-value of 0.1, which will also exacerbate the number of significant trends observed. Keep in mind that at least 10% of the significant trends are in fact due to random deviations.

We are aware of that and have used a significance level of $p < 0.1$, however it is consistently and clearly stated throughout this study. In the supplement, the individual significance levels of $p < 0.1$, $p < 0.05$, $p < 0.01$ and $p < 0.001$ for annual trends at all sites has been provided (Tables A2-A4, supplement) should they be required.

The finding of No significant trend is still an interesting policy result. If in fact the emission reductions have not led to reductions in O3 then this still needs a better reconciliation with model forecasts that suggest O3 should have declined.

Yes we agree, see comment above.

C11782

In addition, I am not convinced that calculating a single trend for all Europe is meaningful when there are so many regional variations. The model comparison with this single European value is not useful.

In an attempt to characterise and quantify regional European trends we have used this new ensemble approach of a "European-averaged" trend. This has been in order to challenge simulated O3 trends with a better approach than just an individual site by site comparison. A single European-averaged value is not important, however a comparison of the distribution of the trends in the observed and simulated data is more useful (see Figure 13 - which has now been relabelled to show O3 mean, 5th and 95th percentiles (Left - right) and observed and simulated (top, bottom, respectively)).

Specific comments

18436, lines 8-10: Rewording needed. On a ten year time-scale presented in this study, O3 trends related to anthropogenic NOx and VOC reductions are being masked as a result of a number of factors including meteorological variability, changes in background ozone and shifts in source patterns.

18437, line 18: There are a number of important references that should be mentioned including the HTAP reports and the US NAS/NRC report (2009) on background air quality.

altered to include references in I.20-24:

Much of the focus in hemispheric background trends has been on the detection, transport and attribution of ozone trends in marine air entering the western seaboard of the USA/Canada and Europe (Carslaw, 2005; Chan and Vet, 2010; Derwent et al.,

C11783

2007a; Jaffe and Ray, 2007; Oltmans et al., 2006; Parrish et al., 2009; Simmonds et al., 2004; HTAP (2010); Cooper et al. (2010); Law et al (2010)).

18438, Line 15: *where should be were* addressed.

18439, lines 3=5: *Don't understand this* altered text:

The mapping of these emission trends to models and observations remains a challenge (Jonson et al., 2006), particularly the ability to pull out relatively small trends from inter-annual variability (Koumoutsaris et al., 2008; Voulgarakis et al., 2010). Derwent et al. (2010) carried out sensitivity studies with a photochemical trajectory model to show how European changes in VOC and NO_x levels would have been the cause of reductions in episodic peak ozone levels.

18441, lines 1-11: *The authors mentioned three different methods (Lowess, Mann-Kendall, Sen-Theill) but they do not describe in detail how they use the three methods. This is a critical omission.*

The statistical methods section has been clarified, see response to "general comments".

It is not clear how they have done the spatial averaging presented in Table 1. Are the trends calculated individually for each site within each country then averaged, or are the O₃ values averaged first?

C11784

altered text from p. 18438 from l. 8 to clarify:

.....have been observed. More recent annual ozone trends at background European sites summarised by country in Table 1. Many previous studies investigate O₃ trends at a single site in a country. Where available, the annual trend (ppbv/yr or %/yr) of multiple sites has been averaged from literature values with statistical significance ($p < 0.1$) to ascertain an average trend by country. Values reported from this study in Table 1 are the average of annual trends (ppbv/yr and %/yr) calculated for individual sites within each country with statistically significant trends ($p < 0.1$). The period of trend analysis varies in each case study, rendering it difficult to compare all sites in a uniform manner. Despite the increasing ozone trends in the Northern Hemisphere, the upward trends appear to be continuing at a reduced rate since the 1980s (Guicherit and Roemer, 2000).

18445, line 7: *The huge Siberian fires were probably also important in 2003.*

altered text p 18445 l 4-9:

The relatively high O₃ monthly means in 1998/1999 have been linked to large-scale global biomass burning, in conjunction with an El Niño event between 1997–1998 (Simmonds et al., 2004). Similarly, Jaffe et al (2004) identified that Siberian biomass burning in the Summer of 2003 contributed to increases in background O₃ of 5 - 9 ppbv at 10 sites in the northern hemisphere. Additionally, the European heatwave during the summer of 2003 is a contributory factor leading to high O₃ levels that year (Solberg et al., 2008; Lee et al., 2006; Ordóñez et al., 2005).

The modeling section really needs to be in a separate paper. There are a large amount of details about the model that are not included and the comparison with observations

C11785

are handled in just a few summary figures.

The details of the model set up have been clarified in response to reviewer 1's comments. Also the authors would like to reiterate that the emphasis of the study was to provide a single self-consistent methodology for assessing trends in observational O3 data at multiple sites across a region (Europe). The focus is not the simulation studies themselves but enough detail is given to have confidence in the model runs.

Table 1: Need to explain how multiple sites used to calculate a single trend in each country.

Addressed above.

Figure 6: Seems to duplicate information in Table 4.

Whilst Figure 6 does duplicate information in Table 4, we feel it provides a good visual indicator for the impact of individual years on the trend 1996-2005.

Figure 12: Using this single metric (all Euro trend) to compare with the model is a massive simplification. The model discussion and evaluation need to be expanded greatly in a separate paper.

This has been addressed above in response to earlier comments.

References

Carlaw, D. C. and Ropkins, K.: openair an R package for air quality data analysis, Environmental Modelling and Software, 2011.

Committee on the Significance of International Transport of Air Pollutants; National Research Council: Global sources of local pollution: An assessment of longrange transport of key air pollutants to and from the United States, The National Academies Press,

C11786

Washington, D. C., 2009.

Cooper, O., Derwent, D., Collins, B., Doherty, R., Stevenson, D., Stohl, A., and Hess, P.: Conceptual overview of hemispheric or intercontinental transport of ozone and particulate matter, Chapter 1, pp. 1–24, United Nations, New York and Geneva, 2010.

Jaffe, D., Bertschi, I., Jaegl'e, L., Novelli, P., Reid, J. S., Tanimoto, H., Vingarzan, R., and Westphal, D. L.: Long-range transport of Siberian biomass burning emissions and impact on surface ozone in western North America, Geophysical Research Letters, 311, L16 106, doi:10.1029/2004GL020093, 2004.

Law, K., Parrish, D., Arnold, S., Chan, E., Chen, G., Cooper, O., Derwent, D., Edwards, D., Jaffe, D., Koch, D., Laj, P., Martin, R., Methven, J., Monks, P., Penkett, S., Prospero, J., Quinn, P., Remer, L., Staehelin, J., Scheffe, R., Takami, A., Tanimot, H., Thouret, V., Turquety, S., Zdanowicz, C., and Ziemke, J.: Observational evidence and capabilities related to intercontinental transport of ozone and particualte matter, Chapter 2, pp. 25–75, United Nations, New York and Geneva, 2010.

C11787