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Atmospheric Chemistry and Physics Discussions

# *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.

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**General comments**

*The most valuable aspects of this study are self-consistency in the analysis approaches used and the fixed time period, although it is clear that a 10 yr period is insufficient for robust results. The analysis excluding individual years provides valuable new insight into the sensitivity of the trends, and is probably the most novel and interesting part of the study. However, it also demonstrates that meteorological events in a single*



*year (e.g., 2003) can be sufficient to change the sign of the trend, and hence that the uncertainty on the mean decadal trends are greatly underestimated. It is important to bring this out in the discussion, and suggest ways in which this might be addressed. Does extending the analysis to 15 or 20 years at sites where data is available make substantial changes to the trend?*

In our paper we show that meteorological events in a single month (i.e. August 2003 heatwave) can be sufficient to alter the sign of the trend in a ten year time period and have suggested that these meteorological effects would be masked if longer-term time series were used. Zvyagintsev and Tarasova (2011) similarly identified that changes in relative humidity and in temperature at eight German sites decreased the ozone trends by 0.5 - 1.0 ppb per 10 years in the time periods analysed (11 to 16 years), with a significant proportion of O3 trends (up to several tens of percent) attributed to trends of these meteorological variables. Weatherhead et al. (1998) calculate that $\sim$ 30-35 years of monthly mean data is required to detect a trend of 5% per decade ($\sim$0.25 ppbv/yr) at a 95% confidence level for a species such as tropospheric O3 . Statistical approaches to account for the effects of meteorological variables on tropospheric O3 trends has been discussed extensively in Thompson et al. (2001).

More recently, the approach of using a generalised additive model (Stephan Henne, personal communication) or a generalized linear mixed model Chan et al. (2009) to remove part of O3 variability that can be explained by meteorology provides a clearer indication of O3 trends resulting from emission reduction measures. This discussion as the referee has pointed out is essential and has been added to the paper.

*The 2-sigma error term quoted here does not provide a true reflection of the uncertainty in the trend. This is clearly illustrated by the variability in values when a single year is omitted (shown in Table 4). The mean trend from these ten studies is 0.16 +/- 0.11 ppb/yr (1 sigma error), and this appears a much more robust assessment of the expected uncertainty. While I appreciate the statistical methods used here,*

*it is not clear that the assumptions inherent in these approaches are defensible. If the authors intended to retain this approach, stronger justification is required, along with an assessment of errors not considered (site distribution, short 10-yr time-scale, inter-annual variation) and additional discussion to aid the reader in interpreting them.*
The referee is correct in that the $2\sigma$ reflects the statistical uncertainty on the trend analysis and does not account for the other factors. The ensemble approach to giving a regional view of the ozone trend given the number of sites does drive down the variability element for trend detection. A more rounded discussion of the factors that affect the ability to detect a trend and their contribution to uncertainty has been added to the text:

There are a number of factors that affect the ability to detect a trend (Weatherhead et al. (1998), Weatherhead et al. (2002)) that include: a) Measurement uncertainty b) Long-term stability of the measurements c) Autocorrelation d) Natural variability e) Extreme events e.g. heatwaves In general, the number of years required to detect a trend is dependent on the magnitude of the variance and the autocorrelation coefficient of the noise (Weatherhead et al. (1998)).

In this work an ensemble approach has been developed using 158 sites, this may drive the variance down for trend detection but will not be able to offset the influence of meteorological variability. Further, the ensemble it taken from sites that pass the data availability and quality thresholds for this work and has spatial limitations (Figure 2).

Additional text to the end of section 2:
The statistical uncertainties quoted in this paper must also be convolved with systematic uncertainties typical for the UV absorption measurement of ozone. Generally, O3 data sets of this variety are believed to be accurate to within $\pm$3%.



*Comparison with the model is greatly underexploited. The model trends are compared side by side with the measurements, but little attempt is made to explain or interpret them. Did the chemical boundary conditions change from year to year? If so, then what would the trends have been if they had been kept constant? The model study should allow the cause of the trends to be clearly diagnosed. If emissions were held constant, how much larger would the trends have been? Further analysis of this type is required to answer the question expressed in the title of the paper. There is much valuable additional detail available from this part of the study that is not included in the discussion here!*

The authors would like to emphasise that the focus of this paper is the analysis of observational data and its links to modelling studies. Modelling sensitivity of the type suggested here has been done before Jonson et al (2006), Vautard et al. (2006). In this work, the model considers only the evolution of meteorological conditions and emissions. The boundary conditions do not account for the year to year variability nor for global trends. However, as shown by Vautard et al. (2006), an imposed trend in boundary conditions does not improve the skills of CHIMERE to reproduce the observed trends. For these reasons and because there was no global climatology consistent and continuous over the 1990-2008 time period available at the time we did this simulation, we do not consider the long-term boundary condition variations. The referee is correct in suggesting that for the title question there is always room for more analysis of this type.

*Sections 3.2, 4 and 5 describe the results of the study, but are short on detailed analysis or interpretation. How do we expect the seasonal behaviour to change if changes are due to background ozone or to local emissions? Do we see these features in the current analysis? How does this compare with what other studies have found? What further measurements or analysis do we need in order to reduce the uncertainty or to permit better attribution of the cause of the observed changes.*

In order to address the issue raised by the referee Figure 1 has been added and some text. It is worth noting that the introduction does deal in some detail with a discussion of the literature context and the factors that are important for the observed European rural background ozone trends answering some of the referee's questions.

Figure 1 gives an idealised view of the seasonal cycles for ozone from a marine and (W) European regional-background viewpoint. The basis for the diagram is from some work where the ozone data is classified as a function of the NOx emissions integrated along back trajectories from the time of measurement, allowing the influence of pollution on observed ozone to be assessed Trotrep (2003).

Figure 1 is a schematic diagram of an idealized marine background and European ozone seasonal cycle. In winter, there is an ozone decrement over continental Europe below the nominal marine background driven by the suppression of ozone levels by NO. In general, reductions in urban NOx may lead to an increase in wintertime urban and regional ozone. The origin and occurrence of springtime ozone maximum has been discussed in Monks(2000). In summer, the period of highest photochemical activity there is a substantial ozone increment over marine background controllable by pre-curser reductions. Seasonal trends can give insight into the relative contribution of photochemical ozone production/peak episodes (Summer/95$^{th}$ percentile), NOx titration (Winter) and the hemispheric background (5$^{th}$ percentile).

For addition to conclusions text:
It is clear that the identified year-by-year variations, caused by meteorological variability could be masking the impact of emission reductions on long-term ozone trends. In order to better assess the influence of emission changes on the observed ozone, alternative statistical approaches are required to remove the influence of the



meteorological variability such as using general additive or generalised linear models e.g. (Chan et al. (2009), Chan et (2010)).

*The last sentence of the abstract is not clearly demonstrated in the paper. It is clear that the observed ozone trend does not match the perceived reduction in precursor emissions, but this does not indicate that the emission reduction had no substantial effect, only that it cannot be seen in the period and locations available. This is presumably because the effects are relatively small compared with the masking effects of background ozone changes, shifts in source patterns, and meteorological variability. This sentence needs to be rephrased to state that the effects of emission reductions are not obvious (or observable?) at most locations, but that their effects have not been quantified.*

We agree with the referee that 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.

**specific comments**

*p.18437, l.15: "Within ozone trend work [there has been substantial focus to date on quantifying hemispheric backgroundÂİ trends in ozone (The Royal Society, 2008)]" - please rephrase (or remove).*

There has been substantial focus to date on quantifying hemispheric backgroundÂİ trends in ozone (The Royal Society, 2008).

*p.18437, l.24-27: Note that this focus on marine inflow is somewhat simplistic, both*

*because of the effects of recirculation of continental air over the oceans associated with synoptic-scale systems, and the sensitivity to air mass history. Some controversy remains here.*

Yes, the referee is correct and the controversy is alluded to in the references in this paragraph which details some of the discussion. In order to more explicitly address this concern the text has been changed to:

Measurements of marine inflow can provide the opportunity to sample ozone in an environment without the confounding influence of continental emissions while at the same time having acted as an integrator of upwind continental-scale emissions. It is worth noting that marine in-flow measurements could be influenced by the effects of recirculation of downwind continental air over the oceans associated with synoptic-scale systems. There remains some discussion as to the attribution of the observed trends (Oltmans et al., 2006, 2008; Parrish et al.,2009; Cooper et al., 2010).

*p.18438, l.3-6: some uncertainty values needed on the stated trends.*

A review of Northern Hemisphere ground-based "background" ozone trends (including several European sites) has shown a slowly increasing average ozone concentration in the Northern Hemisphere of 0.5 - 2 % yr (variable uncertainties Vingarzan (2004) and references therein). Long-term European background ozone trends of 1.5 $\pm$ 0.9 ppbv/yr at Hohenpeissenberg (1971 - 1983) (Logan, 1985), 1.48 $\pm$ 0.51 ppbv/yr at Zugspitze (1978 - 1995) Oltmans et al. (1998), 0.31 $\pm$ 0.12 ppbv/yr at Mace Head (1987 - 2007) (Derwent et al., 2007b) have been observed, with more recent annual ozone trends at background European sites summarised by country in Table 1.

*p.18440, l.16: given the distribution of sites shown in Fig 2, which are heavily weighted towards Central Europe, some comment on their suitability for assessing European ozone is required. Western, southern and eastern Europe are relatively poorly*



*covered. This point is not mentioned until the conclusions - it needs addressing much earlier in the text.*

The following will be altered from page 18440 l.14:

158 sites categorised as rural/background (not directly impacted by nearby precursor emissions), with O3 data available between 1996−2006, were selected for analysis from the overall data set (see Fig. 2 and Table 2). The majority of these sites have a continuous time series for the 1996−2005 inclusive period. The selection criteria was a maximum of one year with less than 75 % of data and more than 75 % of the data for all other years. These sites have been assessed in terms of their representativeness and recategorised accordingly (Henne et al., 2010) (see Table 2), allowing a characterisation of continental-scale O3 trends. The advantage of this approach at the continental-scale, is the use of a large number of sites which are less influenced by measurement/sampling bias of any single site. The majority of sites are located within central Europe, due to a lack of long-term monitoring sites within France, Spain and the Mediterranean region within the monitoring networks used to compile the GEOmon harmonised dataset. Although the data used within this study may be biased towards Central European locations, the individual sites selected are representative of rural and/or background O3 sites (with the exception of BE0211A, BE0345A, DE0754A, GB0033R, NL0223A, NL0232A that are recategorised as suburban) and are categorised in full within Table 2.

*p.18441, l.14: the meaning of this introductory sentence is not clear - do you mean that different methods are suitable for different purposes? Please rephrase and/or explain this.*

The following will be altered from page 18441 l.14:

A number of statistical methods have been used to quantifying trends in ozone and other environmental datasets, the simplest and most commonly used is a linear regression of which methods are reviewed in Hess et al 2001. Whilst a loess regression and autoregressive integrated moving average (Tripathi et al. 2010, Oltmans et al. 2008) sometimes combined with a cubic polynomial fit (Oltmans et al. 2008) have been used to highlight and characterise seasonality and longer term variations in ozone time-series. Trend tests include t-test and Mann-Kendall (Hirsch et al (1982), Sicard et al. (2009), (2010)). Loess regression is a non-parametric method that can be used to derive a good fit and characterisation of complex ozone time-series that have a seasonal component, however the trend is not readily quantifiable. Consequently, a Mann-Kendall trend test is used to quantify linear trends over the ten year time period in this study.

*p.18442, l.23-25: two explanations for changes in the 5th percentile are outlined here, but no attempt is made to separate them in the analysis. In the remainder of the paper, this low tail in the distribution is assumed to represent the background, but it is possible that NOx reductions play a part at many sites, particularly in the winter. Why is this discounted? Further discussion of this would be highly relevant in Section 3.2.*

We agree with the referee's comments that there should be different seasonal O3 signatures, however the seasonally disaggregated trends given in Tables A5-A7 in the supplement drop in statistical significance. Within the limits of the data used in this study, it is not possible to comment any further.

*p.18444, l.11: section 3.1.1 should be relabelled 3.2, and section 3.2 should be 3.3.*

yes, this has now been changed.



*p.18444, l.25: climatological changes are usually defined over 30 yr periods to avoid significant influence from interannual variability, so it should come as no surprise that 10 years is too short. The excluded-years approach is good, and should be combined with the full 10-year analysis to provide a more robust assessment of the uncertainty in the trend.*

The issue of length of time series has been dealt with earlier in the replies

As suggested by the referee the years excluded methodology has been incorporated as a measure of the uncertainty i.e.

O3 mean trend = 0.16 (+0.31, -0.04)
O3 5th trend = 0.13 (+0.26, 0)
O3 95th trend = 0.16 (+0.36, -0.14)

*p.18445, l.18-20: Given current constraints on the global economy, this continuation is not inevitable. This would be a good place to emphasize the importance of continuing long-term measurements, as without them there will not be sufficient data to be confident about trends.*

It is necessary that the monitoring of trace gas measurements across Europe is continued in order to provide the lengthy time-series required for future trace gas trend analysis on a regional scale, without the masking by meteorological and background ozone variabilities.

*p.18445, l.22: At the start of this section it would be helpful to explain why it is important to look at seasonal trends.*
Yes, please see previous comment above on seasonal ozone trends.

*p.18446, l.13-16: Not all air at these remote sites comes from the ocean, and at the 95th percentile it is likely that local or regional sources make a significant contribution.*

As per the previous comment in the same vain, we accept that not all air at these remote sites comes from the ocean, the absence of summer maxima in the 95$^{th}$ percentile seasonal cycle at Lough Navar and Macehead (Figure A2, supplement) suggests that there is no significant observable contribution by local/regional sources and they reflect the influence of marine air. The same sort of feature has been seen in Canada (Chan and Vet, 2010). We have changed the emphasis to reflect the referees concern:

In the case of O3 5$^{th}$ percentiles, sites in peripheral Europe display a spring time maximum, whilst those in more central locations exhibit either a secondary summer maximum or a broad spring-summer maximum. GB0006R (Lough Navar) and IE31 (Mace Head) display spring maxima in O3 95$^{th}$ percentiles, consistent with the influence of marine air received at these sites, which is not strongly affected by locally emitted human induced O3 precursors. The same sort of feature in baseline ozone has been observed in continental Canada (Chan and Vet, 2010).

*p.18446, l.22-29: The list of sites is not useful here unless there is some underlying thread that links them together. The discussion in this section is principally descriptive, and could be greatly simplified and/or replaced with more interpretation of what the observed seasonal difference in trends might indicate.*
The paragraph has been simplified in line with the referees comments and site names removed. A discussion on the seasonal influences has been added to the overall text.

*p.18449, l.5: Additional interpretation is needed here. It is clear from Fig 9 that sites with a positive NOx emission trend have a higher ozone trend than those with a neg-*



*ative NOx trend. This suggests that local emissions do play a role, and it allows this impact to be removed (by looking at the mean ozone trend where the NOx emission trend is zero).*

We are not sure that the correlation or increase in variation is significant enough in Figure 9 to draw the conclusion the referee makes. We would more interpret this figure as not showing any strong correlation between NOx emission inventory trend and observed ozone trend.

*p.18449, l.9: also transport of ozone, not just its precursors*

Altered text:

Consideration must also be given to meteorological conditions, chemical reactions (e.g. NOx titration) and the trans-boundary transport of precursors and ozone to the measurement sites.

*p.18449, l.15: Please explain "fixed climatologies". What is the temporal and spatial variation in these boundary conditions? Most importantly, did they vary from year to year? This has important implications for the analysis, given that a positive trend is also seen in the model results.*

Altered text:

The boundary conditions used are monthly varying climatologies calculated by averaging multi-year simulation (1999 to 2005) of the LMDZ-INCA climate-chemistry. Therefore the boundary conditions reflect an annual cycle of background pollutants.

*Table 2 adds little to the paper and should be moved to the Supplement.*
moved.

*Fig 13: Which trends are observed and which are from the model? What do the different panels show (presumably mean, 5th, 95th percentiles, but in what order)? Please add appropriate labels to the figure.*
These graphs have now been relabelled.

*Typos: Short title: reduction > reductions*
*p.18443, l.11 (end of line): trends > trend*
*p.18444, l.12: effected > affected*
*p.18448, l.6-7: meaning unclear.*
We are unsure what is unclear to the referee. Ground level ozone production is driven by the availability of NOx and VOC. Therefore, a key part of the puzzle is to understand the trends of these precursors as expressed in emission inventories.

We have changed the sentence to read ...

In order to explore one of the factors driving the observed ozone trends, the trends in the corresponding emission inventories for the anthropogenic ozone precursors have been examined

*p.18449, l.17: models > model*
*p.18452, l.10: remove one "each"*
*p.18452, l.22: includes > included*

**References**

Chan, E.: Regional groundlevel ozone trends in the context of meteorological influ-



ences across Canada and the eastern United States from 1997 to 2006, Journal of Geophysical Research, 114, D05301, 2009.

Chan, E. and Vet, R. J.: Baseline levels and trends of ground level ozone in Canada and the United States, Atmospheric Chemistry and Physics, 10, 8629–8647, 2010.

Hess, A., Iyer, H., and Malm, W.: Linear trend analysis: a comparison of methods, At- mospheric Environment, 35, 5211 – 5222, doi:10.1016/S1352-2310(01)00342-9, 2001.

Hirsch, R. M., Slack, J., and Smith, R. A.: Techniques of trend analysis for monthly water-quality data, Water Resources Research, 18, 107–121, 1982.

Jonson, J. E., Simpson, D., Fagerli, H., and Solberg, S.: Can we explain the trends in European ozone levels?, Atmospheric Chemistry and Physics, 6, 51–66, 2006.

Monks, P., Richard, A., Dentener, F., Jonson, J., Lindskog, A., Roemer, M., Schuepbach, E., Friedli, T., and Solberg, S.: Tropospheric ozone and precursors, trends budgets and policy: TROTREP synthesis and integration report, 2003.

Monks, P. S.: A review of the observations and origins of the spring ozone maximum, Atmospheric Environment, 34, 3545 – 3561, 2000.

Oltmans, S. J., Galbally, I. E., Brunke, E.-G., Meyer, C. P., Lathrop, J. A., Johnson, B. J., Shadwick, D. S., Cuevas, E., Schmidlin, F. J., Tarasick, D. W., Claude, H., Kerr, J. B., Uchino, O., and Mohnen, V.: Trends of ozone in the troposphere, Geophysical Research Letters, 25, 139–142, doi:10.1029/97GL03505, 1998.

Oltmans, S. J., Lefohn, A. S., Harris, J. M., and Shadwick, D. S.: Background ozone levels of air entering the west coast of the US and assessment of longer-term changes, Atmospheric Environment, 42, 6020 – 6038, 2008.

Sicard, P., Coddeville, P., and Galloo, J.: Near-surface ozone levels and trends at rural stations in France over the 1995-2003 period, Environmental Monitoring and Assess-

ment, 156, 141 – 157, 2009.

Sicard, P., Mangin, A., Hebel, P., and Malla, P.: Detection and estimation trends linked to air quality and mortality on French Riviera over the 1990-2005 period, Science of the Total Environment, 408, 1943 – 1950, 2010.

Thompson, M. L., Reynolds, J., Cox, L. H., Guttorp, P., and Sampson, P. D.: A review of statistical methods for the meteorological adjustment of tropospheric ozone, Atmospheric Environment, 35, 617 – 630, doi:10.1016/S1352-2310(00)00261-2, 2001. Tripathi, O. P., Jennings, S. G., O'Dowd, C. D., Coleman, L., Leinert, S., O'Leary, B., Moran, E., O'Doherty, S. J., and Spain, T. G.: Statistical analysis of eight surface ozone measurement series for various sites in Ireland, Journal of Geophysical Research, 115, D19 302, 2010.

Vautard, R., Szopa, S., Beekmann, M., Menut, L., Hauglustaine, D. A., Rouil, L., and Roemer, M.: Are decadal anthropogenic emission reductions in Europe consistent with surface ozone observations?, Geophysical Research Letters, 33, L13 810, 2006.

Weatherhead, E. C., Reinsel, G. C., Tiao, G. C., Meng, X., Choi, D., Cheang, W., Keller, T., DeLuisi, J., Wuebbles, D. J., Kerr, J. B., Miller, A. J., Oltmans, S. J., and Frederick, J. E.: Factors affecting the detection of trends: Statistical considera- tions and applications to environmental data, Journal of Geophysical Research, 103, 17 149–17 161, 1998.

Weatherhead, E. C., Stevermer, A. J., and Schwartz, B. E.: Detecting environmental changes and trends, Physics and Chemistry of the Earth, 27, 399 – 403, 2002. Zvyagintsev, A. and Tarasova, O.: Trends of surface ozone concentrations in germany and their connections with changes in meteorological variables, Russian Meteorology and Hydrology, 36, 258–264, 2011.

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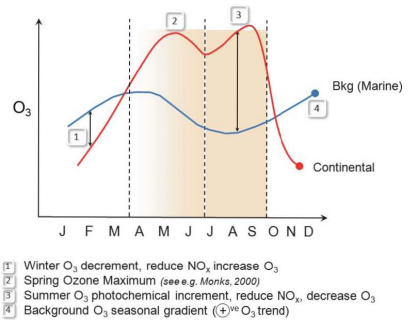


**Fig. 1.**