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Interactive comment on "Can we explain the trends in European ozone levels?" *by* J. E. Jonson et al.

Anonymous Referee #1

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The manuscript presents a well documented summary on the trends in both ozone precursors and tropospheric ozone over Europe since the 80s, based on results from the analysis of measurements and model calculations that are available in the literature. The manuscript adequately comments some of the possible factors (e.g. changes in the ozone precursor emissions, in the circulation within the troposphere, exchange through the tropopause, biomass burning, Ě) affecting those ozone trends. After a rather long introduction, the output of a European photochemical transport model is compared to the levels and trends in surface ozone observed at ensembles of European stations during 1990-2002. The use of sensitivity runs is a creative approach to put the modelled ozone changes in the context of both the changes in the ozone

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precursor emissions and the changes in the European background ozone. However, I think that the manuscript needs further clarification regarding the boundary conditions of the model and the presentation/discussion of the results. Here are my four major concerns:

1. Boundary conditions in the EMEP model (section 5.1): a. Although the Mace Head correction will be discussed in detail in another paper (Simpson et al., 2005, in prep.), the authors should provide more information on how they make the model fit the ozone measurements at Mace Head. Do they use the ozone monthly means at Mace Head? Do they consider all ozone data at Mace Head or only data from the unpolluted sector? Which are the implications of such a correction for the model results, especially for areas that are far from the Atlantic? b. The authors define the 10-year boundary conditions for ozone (AvgBC) as a "10-year climatology based upon the average of the annual data-sets over the period 1990-2000". What does this mean? Are those data sets the boundary conditions taken from both Logan (1999) and measurements at Mace Head? Is that climatology done by averaging the boundary conditions for 1990-2000 on a monthly basis? c. Boundary conditions for other species are mostly based on Simpson et al. (2003). As this is not a peer-reviewed paper but a report, the authors might include additional information.

2. As the authors point out, ozone trends are expected to be different in summer and winter so ozone trend studies should analyse the measurements on a seasonal basis. The authors perform two sensitivity runs to study the effects of changes in emissions and boundary conditions on summer and winter ozone. They also present results from model calculations and measurements of NO2 and O3 for different ensembles of stations during the whole period 1990-2002 (Figures 1 and 2). Both Figure 1 and Figure 2 are helpful because they show that on average the measurements are reasonably reproduced by the model. However, the authors discuss those figures in a rather qualitative way, and try to use them to explain the changes in NO2 and O3 during winter 1990-2002 and summer 1990-2002. I do not think that Figure 2 is appropriate to dis-

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cuss the changes in summer and winter O3 at the German and European sites. I suggest calculating the seasonal averages/medians of daily ozone maxima (for both measured and modelled data) and showing separate plots for the winter and summer values together with their corresponding trends. That way the discussion might be much more quantitative: it would be possible to compare the calculated ozone trends from both measurements and model calculations as well as to compare those trends with the results from the sensitivity runs, separately for summer and winter.

3. The authors show a plot for an ensemble of 54 European stations (Figure 2, top) and mention that there is a clear bias towards Northwest Europe in the location of those sites. The authors reckon that those sites might be more affected by changes in background ozone than by central European emissions, making it difficult to compare the time series of ozone in Figure 2 top with the changes in summer and winter ozone observed in Figures 3 and 4, respectively. However, the authors should consider that the ozone changes at the different sites might compensate each others. Depending on their location, those sites might present different photochemistry or even different emission changes, and might be influenced by different circulation patterns affecting the transport of ozone precursors (as an example see the different behaviour of Scandinavian sites in Solberg et al., 2005). Only if those stations exhibit a similar behaviour during the same season then it is justified to show the average results for the 54 stations like in Figure 2 top. If that is the case, it would also be helpful to show a map with the location of the 54 sites: the authors say that those sites are located in the Northwest of Europe but it is not clear to the reader where they are. Other possible options are grouping the stations in different classes according to their geographical location and characteristics, or showing results for individual stations which are representative either for background or for more polluted conditions.

4. In my opinion the manuscript also needs to go through a large amount of copy-editing before it can be considered for publication. There are various typographic errors and some grammatical issues such as the subject-verb agreement (singular-plural) that

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should be revised in the manuscript.

Detailed comments: p. 5958, lines 19-21: “Such a trend in summer ozone is likely to be difficult to identify from the measurements because of large inter-annual variability”. This sentence can be removed from the abstract. It is known that there is a large inter-annual variability in the ozone levels for the different seasons. However, a variety of statistical approaches have become available in the literature in order to account for the effects of the changing meteorological conditions on the measured ozone levels. See next comment.

p. 5959, lines 9-10: “However, there are large inter-annual variations in ozone levels making it difficult to identify significant trends over the same period”. That is the reason why a variety of statistical approaches have been used for the meteorological adjustment of ozone before the estimation of ozone trends. Some examples for Europe might be cited by the authors (e.g. Kuebler et al., 2001; Brönnimann et al., 2002; TOR-2, 2003; Tarasova and Karpetchko, 2003; Ordóñez et al., 2005).

p. 5959, lines 10-12: “at many ozone sites sampling background and/or free tropospheric air, measured ozone has increased at all seasons, but in particular in winter and spring”. Add references if available.

p. 5960, line 14: “as much as 30% in recent years”. Specify the period.

p. 5962, lines 14-15: “Measurements from the early stages of industrialization indicate that ozone levels at that time may have been around 10 ppbv (Volz and Kley, 1988; Pavelin et al., 1999)”. Although air masses from Paris (influenced by SO2 and causing interferences with the measuring technique) were excluded from the analysis of Volz and Kley (1988), the very low ozone concentrations of 10 ppb observed at Montsouris might be associated with a very efficient ozone removal by dry deposition. Pre-industrial ozone levels might have been closer to 20 ppb as measured at Arosa during the 1950s (Staehelin et al, 1994). I suggest including both references.

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p. 5962, lines 23-24: The authors correctly quote Ordóñez et al. (2004) but include Ordóñez et al. (2005) in the reference list. Ordóñez et al. (2005) analyses O3 and Ox trends in Switzerland but not at different altitudes. See the right quotation (Ordóñez et al., 2004) at the end of the referee's comments.

p. 5963, lines 5-7: "Preliminary studies indicate a trend in recent years of about 1.7%/year in the upper free troposphere (Thouret et al., 2004)". I guess that the authors refer to the ozone trends calculated from vertical profiles measured over Paris and Frankfurt during the MOZAIC program. Has such a high trend (1.7%/year) been calculated for all seasons or only for winter or winter-spring? Please see Table 3 in Zbinden et al. (2005).

p. 5963, 16: Change "Mace Head Ireland" to "Mace Head, Ireland,".

p. 5963, 22: Change "Ozone trend analysis of the measurements are" to "Trend analysis of the ozone measurements is".

p. 5964, line 2: The word "annual" is repeated.

p. 5964, line 5: Replace "μg" to "μg/m3"

p. 5964, line 18: Remove "in ozone".

p. 5964, lines 20-21: "The increase in global free tropospheric ozone has contributed to the increase in surface ozone also in Europe". Add references. Are those references exactly the ones that are mentioned on lines 19-20 of the same page?

p. 5965, lines 5-17: Check typographic errors. Replace "was" to "were" (line 5), "with" to "by" (line 10), "where" to "were" (line 12) and "has" to "have" (line 17).

p. 5966, lines 2-5: "The corresponding results from Ě by Li et al. (2002)". This sentence is hard to understand.

p. 5966, lines 13-16: As Simpson et al. (2003) and Fagerli et al. (2004) are not peer-reviewed papers, a short description on the changes in the model with respect to ear-

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lier versions (Jonson et al., 2000a, 2001) might be included in section 5.1. Section 5.2: When comparing measured and modelled NO2 (figure 1), the authors should mention that the NO2 measurements at the EMEP network are performed by commercial instruments equipped with molybdenum converter. These instruments are cross-sensitive to some oxidised nitrogen compounds such as PAN and HNO3. As a consequence, the NO2 measurements present an upper limit for the real NO2 concentrations.

p. 5967, line 8: Replace “sued” to “used”.

p. 5967, line 16: Replace “13 sites” to “13 European sites”.

p. 5967, line 23: “This is probably because reductions in traffic emissions, with constant emissions throughout the year, are smaller than the total reductions in NOx emissions”. This conclusion is strange. It is difficult to understand what the authors mean by this sentence.

p. 5968, lines 7-9: “Model calculations with the EMEP photochemistry model have been made for 1990 and for the years 1995 to 2002”. However, Figures 1 and 2 show model results for 1990-2002. Do the authors mean that only emissions from 1990 and for 1995-2002 are used in the model runs? If so, why?

p. 5968, line 26: Replace “test” to “tests”

p. 5969, line 3: Replace “in favour of” to “rather than”

p. 5969, line 20: Replace “bottom left” to “top right” Sections 5.4.1 and 5.4.2: The authors mention that the ozone precursor emissions have increased over the Iberian Peninsula and Turkey. Add a reference: Vestreng et al. (2004), UNECE (2004), other, Ě?

p. 5970, line 3: The reaction is: NO + O3  NO2 + O2

p. 5970, line 7: Replace “quite strong” to “rather strong” Two different reports are referred as Roemer (2001). The authors should refer to them as Roemer (2001a) and

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Roemer (2001b) both in the text and in the reference list. Indicate if the values in Table 1 are taken from UNECE (2004) or from other sources. Write comma between EU25 and Germany. In which countries are the 13 stations used for Figure 1 top? As there are 13 stations, I guess the authors should write a comma between “Deuselbach” and “Langenbrügge”. Figures 3 and 4: The text should say “daily ozone maxima” instead of “daily maximum ozone”.

Conclusion: The authors have placed more emphasis on the literature than on their own analysis. It is up to them whether they want to shorten sections 1-4 and beginning of section 5 or not. A more detailed description of the setup of the boundary conditions is crucial to understand the implications of the Mace Head correction and other assumptions on the model results. A more quantitative comparison between model results and measurements (probably restricting the number of stations used in the analysis) is also needed, including results on ozone trends for both summer and winter. The authors might also discuss the results from the sensitivity tests in more detail (in section 5.4.5 or section 6) and possibly put them in the context of the trends calculated from the measurements: is it possible to asses whether the combined results from the two sensitivity runs are in agreement with the measurements? I think that this is a very interesting manuscript that, once these comments have been addressed, should be considered for publication.

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of daily surface ozone maxima in Switzerland in all seasons from 1992 to 2002 and discussion of summer 2003. Atmos. Chem. Phys., 5, 1187-1203, 2005. Solberg, S., Bergström, R., Langner, J., Laurila, T., and Lindskog, A.: Changes in Nordic surface ozone episodes due to European emission reductions in the 1990s, Atmos. Environ., 39, 179-192, 2005. Staehelin, J., Thudium, J., Buehler, R., Volz-Thomas, A., and Graber, W.: Trends in surface ozone concentrations at Arosa (Switzerland), Atmos. Envir., 28, 75-87, 1994. Tarasova, O. A. and Karpetchko, A. Yu.: Accounting for local meteorological effects in the ozone time-series of Lovozero (Kola Peninsula), Atmos. Chem. Phys., 3, 941-949, 2003. TOR-2: Tropospheric Ozone Research, EUROTRAC-2 Subproject Final Report, ISS GSF National Research Center for Environment and Health, Munich, Germany, 2003. Zbinden, R. M., Cammas, J. P., Thouret, V., Nedelec, P., Karcher, F. and Simon, P.: Mid-latitude Tropospheric Ozone Columns from the MOZAIC program: climatology and interannual variability. Atmos. Chem. and Phys. Discuss., Vol. 5, pp 5489-5540, 2005.

Interactive comment on Atmos. Chem. Phys. Discuss., 5, 5957, 2005.