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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Surface ozone at the Caucasian site Kislovodsk High Mountain Station and the Swiss Alpine site Jungfraujoch: data analysis and trends (1990–2006)" *by* O. A. Tarasova et al.

O. A. Tarasova et al.

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We are grateful for the valuable comments of reviewer 2. Following our replies:

The discussion on trends is rather lengthy, it suffers from an excess of detail, and it is hard to digest. Eight tables on trends are presented showing about 300 linear trend results.

We accept the critics, that the discussion was too lengthy including too many details. We tried to focus on the most important results by clarifying some of the statements on trends which also allowed us to shorten the text to some extent. Some details

were removed but some are still included when they were needed to support some key statements.

The main criticism on this article refers to the trend section for the following reasons: 1) it fails to address properly the main features of the trend results: 1a) why are there (large and significant) opposite trends at the two sites for the first interval (1991-2001)?

We agree that the strong downward trends at KHMS and the strong upward trends at JFJ which occurred in the first period of the measurements are very surprising. We were also surprised that the used classes based on classifications of the backward trajectories did not separate more strongly the trends within the subsets (almost all classes showed upward or downward trends in the respective stations), which indeed can question the used approach. We therefore addressed the limitation of the approach (see chapter 2.2. and 3.3., first paragraph). We therefore need to work sometimes with rather indirect scientific evidence and we found, that the main reasons of the opposite signs of the trends at two sites are their geographical position together with ozone precursor emission changes, which relates to summer trends at KHMS. We are aware, that "ozone precursor changes" are usually leading to ozone increases in winter because of the effect of NO-titration, which is obviously not found at KHMS. It is well documented that VOC-emissions in the former USSR dramatically declined in the first part of the 1990s, but we could not find any data related to NO-emission changes during this period. Since NO-emissions originate from fossil fuel combustion we believe, that those emissions did not decrease at least not to the same extent. We tried to include these arguments in the revised version of the paper. JFJ is more impacted by the free tropospheric air over Atlantic, but the scientific reasons of the ozone increase over the Atlantic are obviously not fully explained at the present time, though changes in flux of ozone from the stratosphere into the troposphere are discussed as one possibly reason, which is mentioned in the manuscript (note that ozone concentrations in the lowermost stratosphere in the early 1990s rather strongly increased which is no longer the case in the more recent years). KHMS is situated much further in

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continent, hence it is impacted to higher degree by the continental air and possibly by more complicated dynamical processes (the Main Caucasian Crest to the south, Middle East and Mediterranean ozone maxima), which, however, could not be resolved by the used backward trajectory analysis, making the statement based on the presented results speculative (note that TES satellite data shows that for example in summer JFJ is situated in the zone of the descending air motion while KHMS is in the zone of the ascending motion (Liu et al., Tropospheric ozone over the Middle East, JGR, 2009).

1b) why do both sites show a discontinuity in their trends from the first to the second interval?

We first aimed to document, that this discontinuity in trends (which is known for ozone at the West coast of Europe as well as JFJ) were also found in KHMS. Whereas the change in summer ozone at KHMS can be reasonably well understood taking into account anthropogenic ozone precursor emission changes, the sign of winter ozone changes at KHMS as well as the reasons of the "background increase" over the Atlantic are not fully understood at the present time. Nevertheless, ozone in the lowermost stratosphere did not increase as in the early 1990s which might be viewed as one of the important reasons.

1c) the PBL and FT trends are rather similar. Why is that? Are the two cases not separated enough, is there an underlying common cause?

We agree that the classes used in the analysis did not yield to a significant separation. This is probably explicable by limitations of the used methodology. In our trajectory approach we tried to find a compromise between more uniform subsets impacted by particular source area and general subset representativeness. The filters which were applied to the ozone data series are overlapping. A case of the ST contact does not exclude further PBL contact. These cases were not separated further to keep enough statistical significance of the subset. Compare also the arguments presented under 1a (see above).

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2) it uses technique to cluster trajectories (1-8) and to separate FT and PBL parcels. A tacit objective of applying these techniques is to create more homogeneous subsets with less variability and consequently smaller margins of errors in the trend estimates. The article does not discuss variability of the ozone subsets.

Because of limitation in time resources and because, we found that the message of the paper is rather complex (and therefore difficult to communicate) we did not follow the valuable approach suggested by the reviewer.

The results as presented in table 9 (cluster 3 and 5) and table 10 suggest that there is a large portion of variability left in the subsets, which might have a large effect on the trend results, in particular the results of the smaller subsets.

If we have understood your comment we tried to discuss this item under point 1c.

3) in explaining the trend results different mechanisms are presented: changes in the stratospheric influx, emission reductions in Europe, ship emissions, etc. The trend section is a large compilation of possible explanations which are not discussed consistently and which are not presented as a whole. European emission reduction as an explanation for a trend in one of the 300 trend cases should also be considered as an explanation for a number of other cases. How does it impact the other trend cases?

We agree that the discussion of the individual subsets was somewhat arbitrary and not optimally structured in the ACPD paper. We tried to clarify the main results in the resubmitted manuscript.

4) The authors have chosen to analyse trends by means of a simple linear regression. Results based on linear regressions are known to be sensitive to the values of start and endpoints. How robust are the results against shifting the time frame of the regression with one or two years, e.g.: shifting the time interval from 1991-2001 to 1993-2003 or 1991-2003?

We are aware of the fact, that linear regression analysis can be strongly affected by

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the years at the beginning and at the end of the time series. In order to minimize this effect we divided the series in two periods and used for both periods each ten years of measurements and the periods were not separated by looking at the temporal evolution of the measurements. To keep the estimates statistical significance both selected subsets (1991-2001 and 1997-2006) include at least 10 years of data. Year 1990 was excluded from the consideration due to the ozone measurements problems reported for JFJ before that year (Zanis et al., 1999) and due to big gaps in the data at the both sites in 1990.

Meteorological fluctuations within cluster subsets might also offer explanations for trends. The literature provides examples of methods to address meteorological variability in ozone trends.

We are aware of such works. We focused in the current paper on the influence of the air masses origin looking at ozone formation, while the role of meteorology (local effects) was not the main objective of the work. The first author has a publication by herself on the topic (Tarasova O.A. and A.Yu. Karpetchko, Accounting for local meteorological effects in the ozone time-series of Lovozero (Kola Peninsula), Atmos. Chem. Phys., 3, 2003, p. 941-949.)

The other valuable suggestions were taken into consideration.

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 905, 2009.