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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.

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We thank for the valuable comments of reviewer 1. Following our replies:

1) Page 911, line 21: The authors used a level of 650 hPa for JFJ in the backward trajectory calculations. They should also specify the altitude of the topography for the respective grid point in their trajectory calculations. I guess much higher than 650 hPa. This means that 650 hPa is not at surface but well above surface and hence the PBL influence is significantly reduced in their calculations. The authors should commend on this important issue which similarly applies for Kislovodsk.

The selected pressure levels correspond to the measured average pressure levels at the measurement cites. JFJ is situated at 3580 m. a.s.l. topographically, hence the corresponding closest pressure level is 650hPa. KHMS is at 2070 m. a.s.l., so the pressure level of 750hPa roughly corresponds to the topographic altitude of the site. This implies that the selected altitude for JFJ is not within the PBL in the trajectory model. This altitude was selected in order to take into account that several studies showed, that JFJ often resides in the free troposphere. This point was clarified in the revised version of the manuscript (see chapter 2.2).

2) Page 915, lines 16-18: "The presence of two maxima ...; of ozone concentration."; What do the authors mean with "existence of the two typical regimes of ozone concentration" ? For JFJ especially, this two maxima distribution can be attributed to different processes e.g. tropospheric ozone production versus stratospheric ozone contribution from STE or NW winds versus SE winds due to the well-known JFJ wind channeling effect (see Schuepbach et al., JGR, 2001).

We agree, that different regimes should reflect different factors or mechanisms. We rephrased this sentence to clarify this concept (see last paragraph of 3.1).

3) Page 916, lines 2-5: The authors claim that mount venting at JFJ occurs from March to August. Based on studies using aerosol and specific humidity data I have the impression that the mount venting at JFJ due to thermal convection is mainly activated from May to September maximizing in July-August. March and April are not really months that thermal convection is active at JFJ. The authors should commend on that.

We agree, that the ozone maxima observed from March to August overlaps in time with maximal mountain venting. We changed the text to clarify this point.

4) Section 3.3.1, Page 920: "The FT trends with P<400 hPa at KHMS are more negative than PBL trends". How does that fit with justification 1) "quite substantial impact of the PBL on the ozone levels at KHMS (especially in summer with developed convection over the continent, hiding the effects of ozone increase at the higher levels in the

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troposphere") ?

In summer time, the FT trends are not more negative than the PBL ones. They are about the same. In general as lined out in the paper, we think that deep in the continent, most airmasses arriving KHMS will be influenced by boundary layer at some point. In summer, high pressure systems lead at the same time to large scale downward flow from higher altitude to the station but also the enhanced convection increasing the PBL influence. The strong downward trends suggest a strong influence of the PBL even in those "Free troposphere" cases.

5) Section 3.3.2: The authors report on clear positive trends at JFJ for the earlier period 1991-2001. Why the year 1990 is excluded from this earlier period? Is this simply because the ozone levels in 1990 are a bit higher than in the years 1991, 1992 and 1993 (according to Figure 2a)? Mind please that there is an earlier study investigating homogeneities and trends of the ozone record at JFJ from 1988 to 1996 where a shift discontinuity was found in 1991 (see Zanis et al., Atmospheric Environment, 1999). Furthermore there are earlier studies on the seasonality and trends for JFJ which are not cited. See for example: Bronnimann et al., Atmospheric Environment, 2000.

It is true, that ozone was measured at JFJ prior to 1991. Among the reasons of the selection of 1991 as the beginning of the dataset for trend analysis were the results of Zanis et al. (1999) who provided evidence for some data quality problems in the JFJ ozone measurements. On the other hand both sites have a big gap in the measurements in 1990. As reviewer mentioned the trend analysis is very sensitive to the selection of the initial year. Use of the year with half missing values as the beginning of the time series could introduce even more uncertainty to the obtained trend estimates. We added the corresponding references on the early seasonality analysis and trends in the paper.

6) There are a number of statements which are too speculative e.g. We agree, that some of the following statements are qualitative (speculative)

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i. Page 921: "On average the most negative trend is observed (among the most contributing clusters) in the cluster 4, covering the Southern and Central Europe and originating over Central Atlantic. This may be connected with ozone decrease downwind of the area (Central and Southern Europe) with strong emissions regulations of ozone precursors. Similar negative trends are found in cluster 5 (-0.89 ppb/year), which originates over Central Europe, confirming the idea that negative trends at KHMS may be connected not only with local emissions decrease (see below) but also can be impacted by the air advection from Europe."

As it was mentioned above the strongest advection in the clusters possibly occurs via the free troposphere (if the speculation presented above should be valid), which is in agreement with the trend magnitude in the subset of P<400 hPa. As it has been mention in the answer to the comment 4) both mentioned clusters cover a continental area, where in summer high pressure system and developed convection persist.

ii. Page 922: "In summer cluster 3 is the most frequent one and ozone trends in this cluster are attributable to the decrease of ozone precursor's emissions of the local scale. Summer trends in the clusters 5 and cluster 4 might be impacted by Southern and Western European emissions decrease due to legislation". If the trends in summer cluster 3 shows only the effect of local scale emissions then it would be sensible to assume positive ozone trend in winter due to reduced titration.

"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 in to same extent. We tried to include these arguments in the revised version of the paper.

iii. Page 923: "Taking into consideration that ozone source areas for KHMS are situated

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much lower than for JFJ, the shape of the trends seasonality (slight negative trend in winter and slight positive trend in summer might be the signs of the increased ozone production in the PBL over the Atlantic due to ship emission increase."

We agree, that the increase of shipping emissions is a qualitative argument and we are not able to quantify its effect on ozone KHMS, which is indeed expected to be rather small. We removed this statement from the paper.

iv. Page 925: "The trends remain mostly positive and statistically significant is winter, unlike the other seasons. The only FT subset where trend remains positive and statistically significant in summer corresponds to the cases with " P<400 hPa", i.e. for the air which travels quite high but not necessarily had contact with the stratosphere. The cases which fulfill this criterion may correspond to the long-range transport of precursors and ozone from Asia."

This point has been removed from the paper.

7) Page 924, lines 13-15: The authors state that "Ozone increase in winter is consistent with the ozone response to NO emissions reduction as expected from air pollutants abatement regulation (less titration of ozone in winter and less production in summer)." In spring we have also strong positive trends (0.83 ppbv/year) which are only slightly lower than in winter (0.92 ppbv/year) and possibly not statistically different from winter. Is the justification of ozone titration for winter holds also for spring trends?

We rather believe, that NO titration plays a minor role in spring because sunlight intensity is much higher, but indeed it depends on specific meteorological conditions (which is probably true for summer as well, e.g. some effects of NO-titration by fast frontal transport probably also occur in summer)

8) Page 925, lines 4-8: The authors state "Therefore we can conclude that for the period 1991-2001 the trends at JFJ may mainly be caused by two factors, namely in situ emissions regulations, causing ozone decrease in summer and increase in winter

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in PBL overlapping with systematic increase due to the growth of the stratospheric contribution (mostly seen in spring)." However, still we have (Table 7) a strong positive trend for PBL cases in summer (0.76 ppbv/year) which is of similar magnitude with FT/ST ozone trends (0.72 ppbv/year). How that fits with their conclusion?

The trends have different importance/contribution connected with relative number of cases of air arrival from PBL or FT. Moreover, the used trajectories cannot describe mixing, which is a limitation of the method (see 2.2) .An extended approach making additional use of the residence times of the parcels may be useful for further studies.

9) Figure 6: The clusters 1, 3 and 5 seem to be very similar for the case of JFJ while clusters 1 and 6 are also similar for the Kislovodsk. What does it make them different?

We agree that this similarity of clusters is surprising. However, the graphs give the clusters centers, implying that even if the clusters look similar the clusters by themselves consist of the trajectories covering different spatial areas and traveling at different altitudes.

10) Figure 9: I think in Figures 9a and 9c it should be P<400 hPa. Thank you for your comment, this was a mistake which we changed in the revised version of the manuscript.

Most of the minor comments are incorporated in the text.

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