

## ***Interactive comment on “Ozonesonde climatology between 1995 and 2009: description, evaluation and applications” by S. Tilmes et al.***

### **Anonymous Referee #1**

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Review of "Ozonesonde climatology between 1995 and 2009: description, evaluation and applications" by Tilmes et al., 2011

### GENERAL COMMENTS

This paper presents and analyzes a new vertical ozone climatology derived from ozonesonde flights made at a number of global stations.

This was an extremely tedious paper to review. The paper appears to have been written hastily with little or no proof reading and the numerous errors considerably detract from the research presented. The science may be good but the writing style is extremely sloppy to the point where it simply becomes annoying. If I weren't reviewing this paper I would not have managed to read it to the end. Actually, I didn't make it to the end

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(see below). I know that the authors' first language is not English, but the poor quality of the writing seriously detracts from the research being presented. I am very annoyed by the fact that there are a number of authors on this paper whose first language is English and I suspect that they made little effort to improve the quality of this paper. Were they really comfortable with the paper being submitted in this state? I have made a number of suggestions below for how ambiguities, obscurity and opacity of the text can be clarified, although after a while I tired of this having realized that this should not be the job of a reviewer. As it stands the paper does not meet the quality standards for ACP by a very long way and I would strongly recommend that the authors improve the quality of the writing in this paper if they hope to have it published in this journal.

Large parts of the paper are dedicated to listing in great detail the characteristics of an ozone climatology which simply repeats conclusions that have been reported on in numerous earlier papers. This paper needs to be significantly streamlined and simplified to only be discussing those results that are new.

#### SPECIFIC COMMENTS

Page 28748, line 2: The first line of the abstract does not make it clear that this climatology is constructed from ozonesondes only. It refers to 'ozone soundings' but this is ambiguous. Ozone soundings does not exclude measurements from satellite e.g. the Microwave Limb Sounder (MLS).'

Page 28748, line 5: You refer here to the compilation of ozone profiles from 1980-1994 but the title of the paper suggests that you are only considering a climatology from 1995 to 2009. Why are the data from 1980 to 1994 also needed?

Page 28748, line 10: By 'compare the variability of ozone distributions within each region' do you mean compare the inter-station differences in each region or do you mean compare the ozone variability between regions?

Page 28749, line 3: I am surprised that you pick out 'fossil fuel combustion, industrial

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processes, and biomass burning' as the primary anthropogenic activities that affect ozone. I would have had CFCs and halons at the top of my list, followed by N<sub>2</sub>O, and then GHGs in general. Or did you mean only tropospheric ozone? If you meant only tropospheric ozone you should say so.

Page 28749, line 9: All of the factors that influence ozone that you have discussed so far are chemical. So it comes as a bit of a surprise when you say 'therefore strongly controlled by long-term changes in chemistry and transport'. You haven't said anything about effects of changes in transport on ozone and so there is no support for your conclusion that ozone is also affected by transport.

Page 28749, lines 11-16: This sentence confuses a number of different causes of tropospheric ozone variability. What exactly is impacted by gravity and Rossby wave signatures? The weather systems, tropospheric ozone, or the sensitivity of ozone in different regions to stratosphere-tropospheric exchange? This sentence needs to be clarified.

Page 28749, line 18: What do you mean by 'in remote areas'? If you define a remote area as an area which is beyond the influence of cities, then what you have stated is, by definition, false.

Page 28749, lines 18-19: What you have said here is factually true i.e. 'Most of these ozone sources are expected to increase with a warmer climate' but the implicit attribution made in this statement is false. Yes, the tropospheric ozone source from strat-trop exchange is expected to increase with a warmer climate but not because the climate is warmer - it increases because stratospheric ozone increases.

Page 28749, line 21: Is it the changes in the global ozone background that are poorly understood or is it the contribution of CH<sub>4</sub> to those changes that is poorly understood?

Page 28750, line 1: I think that the way in which this is worded is misleading. The stratospheric cooling speeds the rate of the O+O<sub>2</sub>+M reaction and slows the rate of the

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O<sub>3</sub>+O reaction which results in more ozone (see e.g. Jonsson, A. I., J. de Grandpré, V. I. Fomichev, J. C. McConnell, and S. R. Beagley (2004), Doubled CO<sub>2</sub>-induced cooling in the middle atmosphere: Photochemical analysis of the ozone radiative feedback, J. Geophys. Res.(109), D24103, doi:24110.21029/22004JD005093). It just biases the Chapman equilibrium. Is it right to call O<sub>3</sub>+O chemical ozone destruction?

Page 28750, line 2: Why only the Arctic? Why not also over Antarctica?

Page 28750, lines 9-10: 'A precise description of the geographical and vertical distribution of ozone' cannot improve a chemistry-climate model. Even if you knew perfectly the 4D ozone field, it would not improve CCMs at all.

Page 28750, line 14: There is no such thing as an Umkehr instrument.

Page 28750, line 18: 'Little information' is very much a subjective judgment. I am quite sure that the PIs of the SBUV instruments would disagree with you that their instruments provide 'little information on the vertical distribution of ozone'. Why not just be more precise and say something about the typical vertical resolution of nadir sounding instruments and let the reader make the call as to whether this is 'little information' or not.

Page 28750, line 20: Do the nadir sounders not have large uncertainties in the troposphere and the UTLS region?

Page 28750, lines 21-23: This sentence leaves me totally confused. Used for what? And why can't ozonesonde measurements outside of the tropics also be used (for whatever the aircraft data are being used for)?

Page 28750, line 23: 'Surface measurements are of excellent quality'. This is a very broad generalisation. Is *every* surface ozone measurement of excellent quality? No improvements in surface ozone measurements anywhere are required?

Page 28750, line 24: Influenced by local changes in what?

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Page 28751, line 3: But ozonesondes do not sample the entire globe.

Page 28751, line 4: They also exhibit excellent vertical resolution above 10 hPa if they get that high and so I would just say 'measure at high vertical resolution throughout the flight which typically extends from the surface to 10 hPa'.

Page 28751, line 9: Do you mean ozone stations in general here or specifically ozonesonde stations? You have been talking exclusively about ozonesonde stations in this section and so I suspect that you mean ozonesonde stations but I can't be sure.

Page 28751, line 17: And you mean specifically ozonesonde soundings right?

Page 28752, line 2: You mention the temporal resolution for the validation data from the WDCGG database but say nothing about the temporal resolution of the other validation databases. Why?

Page 28752, lines 8-9: This sentence is not clear at all. Are you saying that Section 5 discusses the time evolution of ozone or that Section 5 discusses the time evolution of the sampling of ozone within your database? They are two very different things.

Table 1: This would make far more sense if you listed the number of ozonesonde flights per year rather than the total in the period. Otherwise the values are just not comparable. Some sites appear to have made more flights than others but that's just because they ran over a longer period. I think that the caption should also point out that values are omitted when the station makes less than 12 profiles per season for at least 5 continuous years.

Figure 1 caption: You say that the subtropical stations are shown in black and also that 'Those stations that are not included in selected regions are shown in black'. I don't know how to interpret that. Does that mean that all sub-tropical stations are not in a selected region?

Page 28753, line 10:  $\pm 2-3\%$  is ambiguous and is not the correct way to report precision values. Do you mean 2-3% or do you mean  $2.5 \pm 0.5\%$ ? The same comment

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applies elsewhere in this paragraph.

Page 28753, line 14: A negative bias of -3% is the same as a positive bias of 3%. I just want to make sure that that's what you mean i.e. the Brewer-Mast sondes measure consistently high by 3% compared to a reference measurement. Likewise for the Japanese sondes.

Page 28753, line 15: But isn't accuracy and bias the same thing? I believe that the correct terminology, as used by the metrology community, is that random error (precision) and bias (accuracy) combine to form the net measurement uncertainty. If you don't use that convention, please be sure to define exactly what you mean by precision, bias and accuracy. Also, are these quoted accuracy values for the stratosphere, the troposphere or both? I would suggest that these values would be far better presented in a table that lists, for all 3 different types of sondes, the precision (random error), the accuracy (the bias), and the resultant measurement uncertainty, for both the troposphere and stratosphere.

Page 28753, line 17: What does it mean to 'employ' an ozonesonde? I also don't know how one 'corrects' a station. I'm sorry, but I just have no idea what this sentence is trying to say.

Page 28753, line 22: You refer here to 'factors outside the range of 0.8 and 1.2'. What are these 'factors'? This is the first time they have been mentioned and no description of them is given anywhere. You only say that profiles are ignored if these 'factors' are outside of the range 0.8 to 1.2. If the reader has no clue as to what these factors are, how can this statement make any sense?

Page 28754, line 1: But how do you interpolate from an altitude grid in meters to a pressure grid in hPa? Somewhere you must be making a coordinate conversion? And surely if you are doing the interpolation in pressure coordinates then you are doing it in  $\ln(P)$ ?

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Page 28754, lines 1-4: If you did what you said you did i.e. averaged all available ozone observations between 1980 and 1994, and between 1995 and 2009 at all pressure levels, there is no way that this produces seasonal ozone profiles.

Page 28754, lines 6-7: Can you put a number on 'good agreement' otherwise this is a meaningless, subjective statement. Surely you can quote one number that is indicative of the differences between the two climatologies?

Page 28754, line 11: But that deviation could simply be the result of interannual variability i.e. we don't expect seasonal means to be the same from year to year. Or, again, I don't understand what you are saying.

Page 28754, lines 12-14: But how will knowing the number of profiles that were used in each 15 year average help the modelers using your climatology? If they knew the exact dates and times of the profiles, and where they were located, they could sample the models to match those. But I don't think that that is what you are suggesting. Surely it would make more sense if the uncertainties on your derived climatologies included any uncertainty resulting from temporal and spatial sampling bias in each region and season?

Page 28754, line 16: You say 'sufficient ozone soundings per season' but sufficient in what regard? What is your criterion for sufficiency?

Page 28754, line 20: You say 'obtain a sufficient sample size for tests of significance' but tests of significance with regard to what? An averaged ozone profile cannot be flagged as being significant or not significant. It can be flagged as being statistically significantly different from the true profile, or statistically significantly different from the mean profile from another region. What is the benchmark against which you are testing for 'significance'?

Page 28755, lines 8-9: You say 'Further, gravity wave activity is most prevalent over the Pacific and Indian Oceans' but it is not clear at all how this connects to the discussion

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of the variability of tropospheric ozone in the tropics. Nowhere have you discussed (with citations of the relevant literature) exactly how gravity wave activity affects the distribution of ozone in the troposphere.

Page 28755, lines 9-12: I am confused here. Surely the fact that the vertical distribution of ozone and its year-to-year variability over a 10 year period shows significant differences between stations in the tropics is a compelling reason to regionally separate the tropical stations, not a reason to combine them? You would be more justified in combining stations within a region if they showed ozone behavior that is homogeneous in space and time. So the reason that you provide for not regionally separating the tropical stations is, I believe, the very reason why you should. In fact a few lines later you say 'For detailed model evaluation of tropospheric ozone in the tropical region a comparison of single stations is likely to be more meaningful' i.e. the regional heterogeneity of the ozone distribution is a reason NOT to combine all stations in the tropics. Now maybe you don't have enough tropical stations to do this regional separation, but that's a different issue.

Page 28755, line 21: There is one question I had in mind which was not addressed by your brief description here of the Hellinger distance. If two distributions are identical in shape, but do not overlap at all because their medians are very different, is the Hellinger distance 0 or 1? I think that providing some such pedagogical examples would greatly help the reader is gaining an intuitive understanding of what the Hellinger distance represents.

Page 28756, line 4: It's not clear to me what you mean when you say that the ozone distribution depends on the altitude interval. Do you mean that it depends on the altitude region? Ah, maybe you mean the shape of the PDF and not the shape of the ozone profile itself? Can you please clarify this.

Figure 2: It is not necessary to label every panel in the Figure with 'West Europe' since the whole figure is for Western Europe and for Western Europe only. And why are you

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not showing the ozone PDF around the thermal tropopause i.e. from -1 km to +1 km?

Page 28756, line 22: You refer here to the region within 3 km of the thermal tropopause, but as far as I can tell, you are nowhere considering the region within 1 km of the thermal tropopause.

Page 28756, line 25: You refer here to the 'Hellinger distance between different observations' but it is not possible to calculate the Hellinger distance between observations. You can only calculate the Hellinger distance between distributions.

Page 28757, line 6: But MOZAIC doesn't actually collect air samples right? If that is true, then referring to 'aircraft samples from the MOZAIC program' is misleading.

Figure 4: I am confused by the legends in the leftmost panels of this figure. The legend states that soundings at 800 hPa are shown with open squares, soundings at 900 hPa are shown with open triangles, and soundings at 1000 hPa are shown with open diamonds, and yet none of these symbols appear in the top row panel (only filled diamonds) and while there are some open diamonds in the bottom row panel there are also filled diamonds and so I have no idea how to reconcile the information provided in the figure legend with what I actually see in the panels. In the text it says 'Altitude information of surface stations is included in Fig. 4 (right panels, different sizes of diamonds, going from small to large with increasing altitude)'. Sorry, but at least in my version of Figure 4 I see no variation at all in the size of the symbols. The caption for Figure 4 does not say what the ' $r=0.85$ ' and ' $r_0=0.99$ ' refer to.

Page 28757, line 28: There was no upper bound on the altitude range for the 3rd altitude interval?

Page 28758, line 13: I don't know what you mean by 'different regional coverage of those data'. First of all, different compared to what? And where is this shown? Figure 4 shows correlations for NH Polar West and SH Midlat and SH Polar. Figure 5 shows the correlations for Western Europe, North America, SE America and Japan. I don't

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see the plot for the eastern NH polar region anywhere. I must be missing something. Wherever these differences are shown, explaining their anomalous behavior with 'different regional coverage of those data' is no explanation at all. You need to be more specific.

Page 28758, line 16: What do you mean by 'the different data sets in the SH Polar region are also highly correlated'. Do you mean that the ozonesonde data from Antarctic stations are highly correlated? If so, this is not shown anywhere. It seems to be a throw away statement. What should the reader interpret from this? Are datasets from other regions not highly correlated?

Page 28758, line 18: Is this temporal or spatial variability that you are referring to? If it is temporal variability then I don't understand the point since no matter how high the temporal variability, if that variability is correlated over a large region, then the spatial sampling of the region should not lead to discrepancies between the ozonesonde and validation data.

Page 28758, line 24: 'a remarkable agreement' is a value judgment. Why not just say 'For Western Europe, ozonesonde observations and surface measurements agree to within  $\pm 4$  ppbv with a correlation coefficient of 0.97, for all pressure levels and seasons.'

Page 28759, line 13: Can't this statement of 'a reasonable agreement' be replaced by some quantitative metric of the degree of agreement?

Page 28759, line 16: I don't know what you mean by 'The shape of the ozone distributions'. Do you mean the shape of the ozone PDF, the shape of the ozone profile, the seasonal evolution of the ozone or something else? And whatever it is that you mean, can you please point to a figure where this is shown.

Page 28759, line 22: What is the basis for the statement that a correlation coefficient of 0.86 is 'rather good'. What is the benchmark against which you make this judgment?

Page 28759, line 26-29: These sentences confuse a whole lot of things. The temporal

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variability in surface ozone over China ('large daily variability of ozone') is used to explain the spatial variability in ozone between Japanese stations ('large variability among the stations'). That just makes no sense to me.

Figure 6 Caption: 'Time evolution of .. from'. And why is the Northern Hemisphere Tropics region (incorrectly labeled in the actual figure) now arbitrarily included in the analysis?

Figure 7 Caption: I don't know what you mean by 'For the same region, this level is not shown for the tropics'. For the same region as what? I'm sorry, I just can't make sense of this.

Is the idea that Figure 6 shows the time evolution of ozone for the troposphere while Figure 7 shows the evolution for the stratosphere? If so, why not just say so. As it stands, it is not clear what differentiates Figures 6 and 7.

Page 28760, line 19: Why the special emphasis on the tropics?

Page 28760, line 22: Surely the biggest dynamical influence on ozone is the Brewer-Dobson circulation and yet not mention of that is made here.

Page 28760, lines 22-24: This seems to be simply repeating the material that appeared in the first paragraph of the Introduction.

Page 28761, lines 6-7: As stated, this is not true since the largest ozone mixing ratios are found in the stratosphere. I think that what you meant to say is 'In general, ozone mixing ratios in the troposphere show the largest values, and the largest variability between the years, in spring (not shown) and summer (see Figure...)'.

Page 28761, line 9: Why is it that the tropospheric ozone only shows spatial inhomogeneity longitudinally? I would have thought that meridional separation between clean and polluted sites would also induce meridional inhomogeneity.

Page 28671, line 11: How can you have interannual variability within a season? I have

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not managed to decipher what you are trying to say here.

Page 28761, lines 13-15: As I read sentences such as this, I wonder that surely all of this has been observed before in many previously published papers. How much of this is new? Much of what appears in this paper seems to repeat previously published knowledge (and without citing those earlier papers). This paper can be, and must be, significantly shortened and simplified by citing the many papers that have already presented these conclusions.

Page 28761, lines 20-21: 'Significant differences between different regions are also observed in the stratosphere.' This is not a new result. This has been known for a century or more. Almost everything that you have said in this paragraph is already well known.

Page 28761, line 25: And yet from the title of your paper there is no indication that you will be considering ozone prior to 1995.

Page 28671, lines 27-28: But Oltmans et al. (2006) only analyzed tropospheric ozone. How then can you say that in general ozone trends have been shown to be small for the last 15 years? This may be true for stratospheric ozone but that statement is certainly not supported by the paper that you cite.

Page 28762, lines 13-15: You say that 'Strongly decreasing ozone mixing ratios between 1990–1999 and 2000–2009 occur in the lower stratosphere of the SH Polar region in austral spring of more than 50%'. When I look at the green line in the 'SH polar' panel of Figure 8, which, according to the legend is for spring, I see no values less than about 13 or 14%. So clearly the text and the figure disagree. The line for 'fall' may go to -50%, but who can tell since the figure cuts off at around -25%. This is a mess.

Page 28762, lines 15-17: You state that 'Minimum ozone values have strongly decreased in the early nineties as the result of increasing ozone depletion due to halogen

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chemistry' but your analysis can't possibly show this since you're only looking at the differences between 1990-1999 and 2000-2009. If this is based on other people's analysis, you need to cite the papers which show this.

Page 28762, line 24: I see no indication of referencing to tropopause altitudes in Figure 9.

Page 28763, lines 3-5: Surely yours is not the first study to note that there is seasonality in the height of the ozonopause in the tropics with a maximum in DJF and MAM?

Page 28763, lines 20-21: I don't know what you mean by 'The seasonality of tropopause-referenced altitudes'? Do you mean the seasonality of ozone in tropopause-referenced altitudes?

Page 28763, line 28: Again, surely this is not the first paper ever to observe that the seasonality in ozone in the lowermost stratosphere depends on the coordinate system considered?

Page 28764, line 11: Just to be clear here, are you talking about trends in the altitude of the thermal tropopause in the subtropics, or are you talking about trends in ozone references to the thermal tropopause? I believe the former but given the earlier opacity of the writing I cannot be sure.

Page 28764, line 14: Surely such speculative statements (and there are many in this paper) could be made more robust, either by extending the analysis to something more than a passive commentary on the ozone climatology, or to reading and citing existing literature. I know that there are many papers that have documented the hemispheric asymmetry of the effect of the Brewer-Dobson circulation on ozone.

Page 28764, lines 15-17: This is not the first time that you have made this statement in this paper, and it is certainly not the first time it has been observed. There is far too much repetition of statements in this paper and the paper would be significantly improved if this repetition was removed and the paper thus shortened.

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Page 28766, line 7: 'The year 2001 was simulated'. Didn't you just say that 3 or 4 lines above?

Page 28766, line 26: Eyring et al. (2010) did not establish CCMVal2.

Page 28767, line 5: I have no idea what you mean by 'we evaluate tropopause-referenced altitudes'. The tropopause-referenced altitude is just the altitude above the tropopause. There is nothing to 'evaluate'. I have no idea what you are trying to say here.

OK, I have run out of stamina. I don't have the time or energy to finish this. I will leave it to the editor to decide what to do.

#### GRAMMAR AND TYPOGRAPHICAL ERRORS

Page 28748, line 16: Replace 'West Europe' with 'Western Europe'. Likewise for the caption for Figure 1, on line 26 of page 28754

Page 28748, line 19: The HTAP and CCMVal2 acronyms should be expanded here in the abstract.

Page 28748, line 26: Replace 'and different altitudes' with 'and at different altitudes'.

Page 28749, line 6: Replace 'VOC' with 'VOCs'.

Page 28749, line 7: Replace 'Trends and variability of' with 'Trends and variability in'.

Page 28750, line 14: Here you spell ozone sonde as two words but in the title of your paper as one word. At least be consistent. The same applies for the caption for Figure 1.

Page 28751, line 5: I don't understand the use of the word 'respectively' in this sentence. There is nothing else in the sentence that maps individually onto the troposphere and stratosphere and so I don't know what the 'respectively' refers to.

Page 28751, line 6: Replace 'with focus' with 'with a focus' and likewise elsewhere in

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the paper.

Page 28751, line 22: Replace 'in combining stations' with 'by combining stations'.

Page 28751, line 25: Replace 'measure for the' with 'measure of the'.

Page 28752, line 22: Replace 'Ultraviolet data Center' with 'Ultraviolet Data Center'.

Page 28752, line 23: Replace 'Earth system Research' with 'Earth System Research'.

Page 28753, line 3: I see you're switching back now to 'ozone sonde' instead of 'ozonesonde'.

Page 28753, lines 26-27: Replace 'employed Between' with 'employed. Between'.

Page 28753, line 28: I don't understand what it means to 'employ' a station.

Page 28754, line 8: Replace 'and present' with 'and the present'.

Page 28754, line 19: Replace 'stations of regions' with 'stations in regions'.

Page 28755, line 6: You say 'Walker circulations'. Is there more than one Walker circulation?

Page 28755, line 10: Replace 'interannual variations of a 10-yr period' with 'interannual variability over a 10 year period'.

Page 28755, line 11: Replace 'A grouping of the tropical region into sub-regions' with 'A separation of the tropical region into sub-regions'. You can't 'group' a region.

Page 28755, line 18: Replace 'in calculating' with 'by calculating'.

Page 28755, line 21: Replace 'distributions, not necessarily Gaussian' with 'distributions which need not necessarily be Gaussian'.

Page 28756, line 12: Replace 'distributions of all stations' with 'distributions at all stations'.

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Page 28756, line 22: You've already got the  $\pm$ , I don't think you also need the double squiggle.

Page 28756, line 25: Replace 'variation in median' with 'variation in the median'.

Page 28757, line 15: Replace 'from EMEP and WDCGG database' with 'from the EMEP and WDCGG databases'.

Page 28757, line 17: Replace 'A large number' with 'Data from a large number'.

Page 28757, line 23: Replace 'available in altitudes below 500 m' with 'available at altitudes below 500 m'.

Page 28757, line 26: Replace 'ozone sounding' with 'ozone soundings'.

Page 28758, line 5: Replace 'NH Polar, West Europe' with 'NH Polar and Western Europe'.

Figure 5 caption: Replace 'Surface stations are taken' with 'Data at surface stations are taken'.

Page 28758, line 9: Replace 'data are shown' with 'data is shown' since the subject of the sentence is 'the correlation'.

Page 28758, line 17: Replace 'high northern latitudes, only a limited amount of data' with 'northern high latitudes, only a limited number of data'.

Page 28761, line 28: Replace 'changes of ozone' with 'changes in ozone' and likewise elsewhere.

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Interactive comment on Atmos. Chem. Phys. Discuss., 11, 28747, 2011.

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