

## ***Interactive comment on “Long term changes in the upper stratospheric ozone at Syowa, Antarctica” by K. Miyagawa et al.***

**K. Miyagawa et al.**

koji.miyagawa@gmail.com

Received and published: 16 April 2013

Dear Referee #3,

We thank the referee for valuable comments on our work. Our responses to the specific comments are given below. We marked the reviewer's and the author's comments by RC: and AC:.

RC: This paper presents an analysis of the long-term Umkehr record from Syowa. The authors show qualitative agreement with SBUV overpass data, and that the Umkehr record follows the EESC curve for an age of air of 5.5 years, or somewhat longer. It is not clear to me why they claim that “Ozone recovery during the austral spring over Syowa station appears to be slower than predicted”. There is a great deal of statistical

C1383

analysis attempting to identify dynamical parameters affecting ozone, but it is not very convincing, and the authors' conclusion that “dynamical and other chemical changes in the atmosphere are delaying the recovery over this station” does not really seem to be supported by any clear evidence.

AC: The conclusion is based on the matches of the ozone residuals (CUSUM method) with the two sets of EESC curves shown in Fig.7b. Unfortunately there are no long-term EESC measurements for the South Pole stratosphere, and therefore, we rely on the modeled estimates of ozone depleting substances (ODS) to evaluate changes in the Syowa Umkehr ozone time-series. The cumulative residuals of ozone in the upper stratosphere do not seem to follow the Polar 2 curve after 2001, which is the turning point for the EESC concentrations estimated for the Polar region. The Polar 2 curve is simulated by the Goddard automailer program, and is based on the current state of knowledge (WMO Ozone assessment, 2011) for the transport of Halogens from tropics to high latitude region and the release of chlorine and bromine radicals from halogens, which is determined by their chemical life-time and fractional release factors (FRF). If stratospheric ozone measured at Syowa station does not follow the recovery rates predicted by the Polar 2 curve (Fig.7), it could mean that the modeled EESC did not account for the actual transport of ODS to the South Pole or the stratospheric lifetime of halogens was not estimated correctly. The atmospheric circulation in climate models is not well characterized, and thus models have hard time to predict ozone recovery at high latitudes. For example, the “leaky pipe” theory (Neu et al., 1999; Ray et al. 2010) suggests that the stratospheric air re-circulates from subtropics back to the tropics, then mixes with the younger air coming from the troposphere, and thus extends the long-tail of the life-time distribution of long lived halogens in the stratosphere. Recently published papers (Laube et al., 2012; Rigby et al, 2013) re-assessed the stratospheric life time of many halogens regulated by the Montreal Protocol and estimated somewhat longer life-time for major ODSs, such as CFC-11, CFC-12 and CH<sub>3</sub>CCl<sub>3</sub>. The non-linear relation between the age of air and fractional release time complicates the assessment and prediction of the EESC concentrations. The authors of both papers

C1384

found the age of air to be longer as compared to results published by Newman et al (2006) and Ozone assessment (2011), which in turn would affect the EESC curves. Since Syowa data do not agree with the Polar 2 curve, it could mean that the age of air in the upper stratosphere over Syowa could be longer than 5.5 years. In addition, Laube et al (2012) and Rigby et al (2013) emphasize that the FRF and age of air correlations are done under the assumption of the steady-state of the stratospheric transport. Since transport can change in the future according to climate change scenarios (acceleration of the Brewer-Dobson circulation, more wave breaking in stratosphere, re-circulation, etc), thus the concentrations of the ODS in the stratosphere could change over time that is not well predicted by EESC curves. Therefore, the fact that increase in Syowa stratospheric ozone data is not following the predicted rate of chlorine reduction (provided by the EESC Polar 2 or Polar 3 curves) cannot be used with high certainty to state that other than the ODS reduction processes are therefore affecting ozone levels (i.e. increase in green-house gases).. We agree with the reviewer that we can not quantify the change in circulation with Syowa ozone data, but we can state that Syowa ozone data are not in agreement with currently proposed polar EESC curves, and further studies/measurements are needed to improve these predictions.

Neu, J. L., and R. A. Plumb, The age of air in a “leaky pipe” model of stratospheric transport, *J. Geophys. Res.*, 104, 19,243 – 19,225, 1999. Ray, E. A., et al. (2010), Evidence for changes in stratospheric transport and mixing over the past three decades based on multiple data sets and tropical leaky pipe analysis, *J. Geophys. Res.*, 115, D21304, doi:10.1029/2010JD014206. Laube, J. C., Keil, A., Bönisch, H., Engel, A., Röckmann, T., Volk, C. M., and Sturges, W. T.: Observation-based assessment of stratospheric fractional release, lifetimes, and Ozone Depletion Potentials of ten important source gases, *Atmos. Chem. Phys. Discuss.*, 12, 28525-28557, doi:10.5194/acpd-12-28525-2012, 2012. Rigby, M., Prinn, R. G., O’Doherty, S., Montzka, S. A., McCulloch, A., Harth, C. M., Mühle, J., Salameh, P. K., Weiss, R. F., Young, D., Simmonds, P. G., Hall, B. D., Dutton, G. S., Nance, D., Mondeel, D. J., Elkins, J. W., Krummel, P. B., Steele, L. P., and Fraser, P. J.: Re-evaluation of the lifetimes of the major CFCs and CH<sub>3</sub>CCl<sub>3</sub>

C1385

using atmospheric trends, *Atmos. Chem. Phys.*, 13, 2691-2702, doi:10.5194/acp-13-2691-2013, 2013

RC: The presentation in general is unclear, and seems unfocussed. There are too many figures, as some of them appear to merely illustrative and others are presented with almost no discussion. For example, Figure 2 presents no information that is not already in the text; the information in Figure 3a is also in Figure 4a; and Figure 11 apparently repeats (part of) Figure 7. Figure 9 doesn’t present any information that is pertinent to the authors’ conclusions. AC: Thank you for pointing out the repetition. We have removed some figures and added more explanation to the text.

RC: I think the authors need to focus on the story they wish to tell, and drop some of the distracting side discussion. In some cases I think they need simply to omit discussion of some of the factors that they (commendably) examined; we don’t need to know about every dynamical factor they looked at; just those that showed a statistically significant correlation with ozone. The paper should be rewritten, and made shorter. There is a little too much discussion of what other people have done, and not enough detailed discussion of what the authors did.

AC: Thank you for pointing the lack of the discussion. We ran additional analysis to evaluate contributions from all explanatory parameters that explain ozone variability at Syowa in spring and summer. A new summary table and text are added to help with the discussion of results.

Detailed comments:

RC: Page 385, lines 19-21: I believe this anticorrelation has more to do with the rate of the O+O<sub>3</sub> loss reaction.

AC: We have corrected this sentence. “Conversely, the temperature decline of the upper stratosphere leads to the increase in ozone (O<sub>3</sub>), via a Chapman mechanism and the ClO<sub>x</sub> cycle first described by Molina and Rowland (1974)”

C1386

RC: Page 387, lines 3-6: Apparently all 1508 profiles were usable?

AC: Approximately 10% were removed by a data quality evaluation, leaving 1360 profiles (Aug–Dec: 828, January–April: 532) in the analysis. The text has been updated.

RC: Page 388: This is titled “Uncertainty for ozone profile retrieval”, but doesn’t actually discuss those uncertainties at all (not quantitatively), except to assure us that they have been “successfully corrected”. Also, what are the special uncertainties of making Umkehr measurements at high latitude, where the sun takes a long time to change SZA and consequently makes a considerable change in azimuth as well? The authors note that few stations poleward of 60 even try.

AC: We added the following sentences. The systematic errors in Umkehr measurements are evaluated via simultaneous intercomparisons of each instrument (Dobson # 119 and Dobson #122) with the reference instrument (Dobson #116). The Dobson #119 instruments at Syowa is comparable with Dobson #122 instrument, with about 5% uncertainty estimated in the retrieved profile for the period when these instruments were used. After SZA correction, N values still have discernible uncertainty errors. Some of these errors are dependent on total ozone (TO), which was suggested by the results of the intercomparison campaign. After the total ozone correction, the uncertainty error for N values is reduced by about 5-10%. Through the reevaluation of Syowa’s record as described in detail by Miyagawa et al. (2009a), most discontinuities in station’s Umkehr time series have been successfully corrected.

The uncertainties in the Umkehr profile retrieval are summarized by Petropavlovskikh et al. [2005 and 2009]. During spring it takes sun 2-3 hours to move between 80 and 90-degrees SZA, and ~ 2.5 hours during summer, which is comparable to the time at middle latitudes. Ozone profile retrievals are done under the assumption that ozone does not change during the time taken to complete an Umkehr measurement, which is true for most of the chemical processes in stratosphere, with the exception of the upper stratosphere where some ozone photochemical reaction rates are under an hour. The

C1387

azimuth angle of the sun changes by 25-45 degrees during an Umkehr measurement. However, since the Umkehr technique relies on the light scattered downward from the zenith direction, the profile is derived overhead of the station and is not affected as much as the direct sun measurement where the line of pointing to the sun might cross different ozone fields as the sun moves and azimuth angle changes. The accuracy of the ozone profile derived from Umkehr measurements is limited to ~ 5 % in the upper and middle stratosphere. Moreover, there is a known bias between ozone derived from Umkehr and other ozone methods. We have removed this bias by de-seasonalizing the time series; thus it does not affect the trend estimates, which represent relative change in the measurement over time. The errors in the retrieval are minimized by using seasonal averages, while the 95 % confidence level defines uncertainties of the trend estimates. The reason that not many Dobson stations use the Umkehr method is that it is time consuming, expensive and limited by available personnel in Antarctica during spring. There was an attempt to retrieve Umkehr profiles at the Arrival Heights station (77S), which was successful, but was not continued because of the above-mentioned considerations. As latitude increases, the period during the year during which the zenith angle changes the required 90 to 70 (or reverse) in a half day decreases. At latitudes greater than 80, no days are possible in the year

RC: Page 390, Equation (1): I think the terms are monthly averages. This should be stated.

AC: These data are, as the reviewer suggests, summarized and presented as monthly averages.

RC: Page 391, lines 20-21: This is surprising. The heat flux is presumably a dynamical proxy for meridional (ozone) transport. Since atmospheric motions are primarily zonal, wouldn’t monthly zonal averages of the heat flux make more sense than coincident measurements?

AC: Since Umkehr measurements are not done every day, it makes sense to use daily

C1388

coincidences between the two data sets to avoid sampling errors. Also, especially in spring, the atmospheric motions depart significantly from zonal symmetry, as indicated by, e.g., the variations in equivalent latitude shown herein.

RC: Page 392, lines 3-4: That sounds like a good idea, but it doesn't appear to have been done. From my reading of the text, it seems that all the proxies were used, even those that made the fit worse (Table 2).

AC: We performed further studies to evaluate the contribution of each proxy to the ozone variability. Results were evaluated and only proxies that reduce ozone variance are now kept in statistical model. This is now described in the text.

RC: Table 2 is quite puzzling, and needs more explanation. In most of the summer cases, the minimum RMSD is achieved without all of the dynamical proxies, so that adding more proxies makes the fit worse! This should be a red flag that there is no information being added. Each proxy added adds one additional degree of freedom to the fit, so the fit should get better, even if the additional proxy represents random noise. The fact that it improves only very slightly in many cases with the addition of more proxies suggests that these carry no new information. I am quite concerned as well about the time lags, which of course represent seven additional degrees of freedom to the fit. Most of the lags found are so long that it is difficult to imagine a physical mechanism responsible for them, as the authors admit. We are not told how much they improve the fit, and so one suspects that the improvement is actually coming from the additional degree of freedom added in each case.

AC: We performed more analysis and now the statistical model has been changed to use only proxies that improve the fit. See the new version of Table 2. We have also added discussion to the text to explain how the proxies were chosen (also see above response to the question about Heat Flux contribution to ozone variability at Syowa station). As the reviewer correctly points out, the contribution of the ENSO proxy was not significant as compared to other listed proxies. We find the contribution of Heat

C1389

Flux to ozone variability comparable to the EqLat850K in the springtime season and only in the lower and middle stratosphere, According to our analysis of ozone variability in the summer season, we find that Solar, QBO, AOD and EqLat explain 2-35 % of the variability, whereas EqLat at 520 K is preferred to fit time series in layers 1 and 2+3, whereas for layers 5, 6, 7 and 8, EqLat at 850 K is a better proxy. It is also interesting to note that ozone variability in layer 2+3 and 4 during summer time is defined by Solar, AOD and QBO, EqLat 850 K and SAM proxies. Layer 7 and 8 appears to have additional improvement in the explained variances when Heat Flux proxy is used. On the other hand the improvement is similar when SAM is used in place of Heat Flux. Therefore, it is inconclusive results and thus we cannot say what exactly attributed to the improvement of the fit. We also have to notice that Heat Flux does not have large variability in summer time. During spring, stratospheric ozone (layers 2+3, 4, 5, and 6, ) is explained by major contributions from Solar, QBO, AOD, EqLat 850K and Heat Flux , whereas ozone in layers 1, 7, 8 and 8+ requires all the above, but Heat Flux proxy. The best fit with the statistical model was obtained by applying time-lag for SOR, QBO, and AOD proxies (See Table2b and 2c) and no lag to other proxies.

Here are some findings conclusions from working on Syowa dataset analysis. 1) We find it very important to match dates between proxies and observed ozone (the same date proxy works better than monthly averaged proxy that includes all days). It is very important for analysis of the station data where variability is very high, such as in spring time at Syowa. 2) The reduction in variance can be marginal, which makes the method of finding the best lag estimate less robust. 3) Ideally, for each proxy there should be a physical mechanism that connects the variability in proxy to variability in ozone. For example, we do not know the mechanism that would correlate ENSO to the variability in the upper stratospheric ozone. However, we could find reference in the literature that relates effects of ENSO with the middle, lower stratosphere in tropics (Randel et al, 2009, Calva et al, 2010). Since tropics are connected to the Polar regions through Brewer-Dobson circulation, we can expect that ozone in the Polar stratosphere can be correlated to the ENSO variability. Since ENSO is correlated to ozone in the

C1390

lower/middle stratosphere, to the extent that ozone in the middle/lower stratosphere is correlated to that in the upper stratosphere, then ENSO should be correlated to ozone in the upper stratosphere. Therefore, part 2) makes part 3) extremely important for selecting explanatory parameters for the best model fit... 4) The EqLat proxy is useful, since it helps to separate the chemical and dynamical source in ozone variability. However, it does not explain what physical mechanism influences the dynamical variability of the atmosphere in the vortex and over Syowa. 5) It also means that EqLat proxy can be possibly related to SAM, ENSO or any other dynamical proxy, and while we do not know the exact mechanism yet, we would like to point out the high correlation of EqLat to ozone variability in the Polar regions.

RC: The units for RMSD are not given, nor mean values, so it's hard to tell how good these fits are. I suggest adding correlation coefficients to the table as well. Are the numbers in (a) with or without the lags? How much difference do the lags make? The authors state (p. 391, line 27 to page 392, line 4) that "Sequential addition of proxies ... allows for evaluation of independency of the chosen proxies for trend analysis. Correlation analysis of variability in different proxies also helps to eliminate non-orthogonal terms from the model". This is very laudable, but I don't see that this evaluation has been performed. If so, it is not discussed. All I can find is "After removing inter-annual variability in ozone data associated with the above parameters...".

AC: The units for the RMSD are given in the Table 2. Table 2 now show the reduction in the explained variance of ozone data. We have checked the correlation coefficients between proxies and only use proxies that are not correlated. Please see the Table 2.

RC: Page 395, lines 18-20: "In summary, mean Umkehr ozone is lower than the homogenized SBUV ... and it shows good agreement with Umkehr. " This seems self contradictory.

AC: We have corrected this sentence: "In summary, it is shown that Umkehr ozone in the stratosphere is biased 6% low when compared to the homogenized SBUV dataset,

C1391

while in the troposphere Umkehr derived ozone is lower than the SBUV ozone by about 1%. At the same time, the correlation between Umkehr and MODv8.6 OP is significantly improved when the data are matched for temporal co-incidence within 24 hours (Table 3)".

RC: Page 396, lines 16-29: I don't follow this. Figure 4 seems rather to indicate that there are unresolved biases between the NOAA 17 and 18 platforms and previous ones. There are insignificant differences between the two curves in 4a. Also, in the figure caption, what is "ozone rate of overpass"?

AC: Figure 4 was removed from the manuscript. See further explanation in the next section.

RC: Page 397, lines 1-5: This may be true, but why would that make ZM data better? Overpass data should show the same drift. More importantly, Hassler et al. conclude that the use of equivalent latitude corrects for this drift, and the current authors have used that as one of their dynamical proxies, so this should not be an issue.

AC: We agree with the reviewer comment. We use Equivalent latitude proxy in the statistical model to address the dynamical variability of the vortex. We also considered the differences of the adjusted SBUV zonal product and the unadjusted product and confirmed the reviewer's comments for section 3.4: the differences between the overpass data and the adjusted zonal data are more due to the adjustment than the representativeness of the zone for the overpass. We have attached a plot showing the differences between the adjusted an unadjusted zonal products. It is similar to figure 4a in the document. It was recommended that the adjusted product be used to the 50N - 50S regions, and with extreme care otherwise. Unfortunately the zonal mean product does not work well for Syowa station latitude. We decided to remove Fig. 4 and the discussion from the paper. (see Fig A)

RC: Page 397, line 12: A correlation coefficient of 0.64 is really pretty awful, for measurements of the same thing! I would expect correlation coefficients of that magnitude

C1392

for some of the dynamical proxies.

AC: We have rewritten this sentence. The SBUV foot print does not exactly match the point location of the ground-based station and therefore represents the smoothed ozone field over 200 by 200 square km. Also, the low variability of ozone during the summer reduces correlation when signal to noise ratio becomes low. Therefore, correlation of 0.64 is restricted by the detection limit of both observing systems.

RC: Page 397, lines 24-25: This statement is made without substantiation, and Figure 4 suggests that it is not true.

AC: Figure 3b (new Figure 2c) shows that each satellite has different bias with Umkehr ozone at Syowa station at layer above 4 hPa. The MOD data offsets are similar to the averaged offsets of individual satellites (new Figure 2b), except for layers 7 and 8 where offset in the MOD data is reduced.

RC: Lines 27-28: Some of these correlations are terrible: a number of those in Table 3 are even negative! What does this mean? How can they be so bad?

AC: The sampling frequency is low during summer at Syowa because the sun does not descend to the elevations near 90-degrees SZA during mid-December to mid-January. It creates ozone profiles that are of poor quality. We thus removed December data from comparisons. Table 3 is updated.

RC: Page 398, lines 4-6: I don't see any ENSO correlation in the data presented. I believe this is wishful thinking.

AC: We have rewritten this section. We re-analyzed data to look into the contribution of individual proxies separately for the summer and spring seasons. Explained variance in ozone in layers 1, 2+3, 4 and 5 is increased during spring when ENSO is added to the statistical model composed of Solar, QBO and AOD (see New Table 2). Further improvement (from 57 to 62% in layer 5 and from 44 to 50 % in layer 6) is found when model includes other variables (SOR, QBO, AOD, EqLat 850 K and ENSO). However,

C1393

at the end, most of the improvement in explained variability is found when SAM or HF10 proxy is used in place of the ENSO. At the same time, summer ozone variability is improved by including ENSO into model (2.4 vs 1.1) in layers 2+3, 4 and 5. It is also similar to the results of the fit with EqLat850K in place of ENSO, which could mean that EqLat 850K represents variability that is driven by ENSO.

RC: Page 398, lines 10-11: Have we been shown any V8 data? Why is this statement here?

AC: This sentence is deleted.

RC: Page 398, lines 17-18: Not according to Table 2. Although that is what one would expect, since each proxy added adds one additional degree of freedom to the fit.

AC: We have improved this table. Each proxy is cross-checked for correlation before it is used in the model.

RC: Page 400, line 27 - page 401, line 8: Why this discussion? 2006 doesn't appear especially low in Figure 6.

AC: This has been corrected.

RC: Pages 401-402: CUSUM analysis is not so well-known that it can be introduced with just a reference to Newchurch et al.. It should be explained. In any case, I'm not sure that this is an improvement over the straightforward method of showing that the data fit better to Polar 2.

AC: The CUSUM method has been used in several papers and helps to compare the rate of ozone recovery with two curves. It is based on the linear fit of data up to 2001, and then integrating the residual of the fit after 2001, which should smooth out the variability in the data and help to assess whether the change in the data follows slope of the Polar 2 or Polar3 EESC curves. The other method used in paper, that is, comparing the explained variance in ozone data after adding proxies to the model, is summarized in Table 2.

C1394

RC: Page 405, lines 10-17: I don't understand this argument.

AC: This sentence has been deleted.

RC: Page 406, lines 1-4: Really? In what way did you "verify" this?

AC: We have verified that there is a good fit by 850K by examining the correlation of EqLat with ozone at three levels (520, 850, 1300K) (see new Table 2). Improvement of the statistical model fit to ozone data is found after the EqLat proxy is added. The EqLat provides an indication of the change in the location of the polar vortex relative to the station location. The long-term trend of the EqLat proxy (at 1300 and 850 K) is shown in Figure 10 (new Figure 7).

RC: Page 407 lines 14-17: This difference is only 0.5%, and the losses are more than 20%.

AC: The upper stratospheric ozone depletion (layer 8, 4 hPa) prior to 2001 is estimated at  $\sim 1.2$  % per year (Table 4). For the 2001-2011 time period, ozone trend is found as small positive and only  $\sim 0.2\%$  per year or about 1 % over 5 years, which is comparable to 0.7 % change in ozone due to Solar cycle spectrum that is proposed in the referenced paper.

RC: Page 430: "Time series of monthly ozone variability..."? I think these are annual averages.

AC: This has been corrected. "Time series of annual and seasonal averages ozone variability. ..."

RC: I would suggest dropping Figures 2, 3a, 9 and 11.

AC: This has been corrected as suggested.

---

Interactive comment on Atmos. Chem. Phys. Discuss., 13, 379, 2013.

C1395

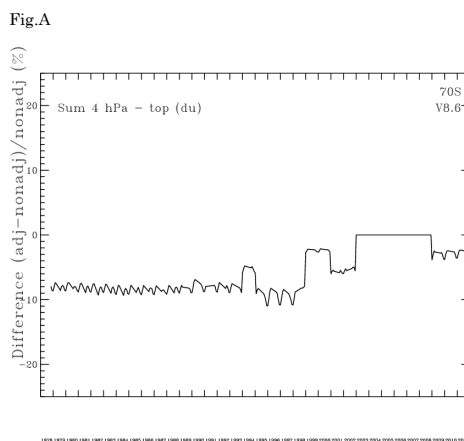


Fig. 1. Fig A

C1396

Pressure [hPa]	LAYER								
	1000	250	62.5	31.2	15.6	7.8	3.9	3.9	
<b>SPRING (SON)</b>	1	2+3	4	5	6	7	8	8+9+10	
1.0)SEASONAL VARIANCE [DU*2]	1.08	117.3	103.0	33.5	7.51	1.54	0.21	0.36	
1.1)SOR+QBO+AOD [%]	-49	-48	-41	-29	-25	-34	-28	-28	
2.1)SOR+QBO+AOD+EqL520K [%]	-58	-58	-51	-35	-25	-27	-13	-10	
2.2)SOR+QBO+AOD+EqL850K [%]	<b>-61</b>	<b>-71</b>	<b>-71</b>	<b>-57</b>	<b>-44</b>	<b>-60</b>	<b>-60</b>	<b>-58</b>	
2.3)SOR+QBO+AOD+EqL1300K [%]	-51	-49	-42	-37	-46	-55	-39	-36	
2.4)SOR+QBO+AOD+ENSO [%]	-56	-55	-47	-34	-25	-34	-28	-25	
2.5)SOR+QBO+AOD+SAM [%]	-56	-61	-63	<b>-60</b>	<b>-49</b>	-43	-35	-33	
2.6)SOR+QBO+AOD+HF10 [%]	-52	-54	-50	-43	-34	-38	-28	-28	
3.1)SOR+QBO+AOD+EqL850K+ENSO [%]	-56	-67	-69	-62	-50	-44	<b>-42</b>	<b>-41</b>	
3.2)SOR+QBO+AOD+EqL850K+SAM [%]	-56	-68	-74	-70	-56	-45	<b>-42</b>	<b>-41</b>	
3.3)SOR+QBO+AOD+EqL850K+HF10 [%]	-61	<b>-74</b>	<b>-80</b>	<b>-79</b>	<b>-64</b>	<b>-52</b>	-39	-39	
3.4)SOR+QBO+AOD+EqL520K+SAM [%]	-58	-63	-62	-56	-44	-35	-21	-19	
3.5)SOR+QBO+AOD+EqL1300K+SAM [%]	-57	-62	-62	-59	-54	-43	-28	-28	
3.6)SOR+QBO+AOD+EqL520K+HF10 [%]	<b>-73</b>	-71	-64	-49	-34	-32	-13	-13	
4.1)SOR+QBO+AOD+EqL850K+ENSO+HF10 [%]	-56	-68	-73	-69	<b>-57</b>	<b>-47</b>	<b>-42</b>	<b>-41</b>	
4.2)SOR+QBO+AOD+EqL850K+ENSO+SAM [%]	<b>-56</b>	<b>-69</b>	<b>-75</b>	<b>-71</b>	-56	-45	-42	-41	
<b>SUMMER (JFM)</b>	1	2+3	4	5	6	7	8	8+9+10	
1.0)SEASONAL VARIANCE [DU*2]	0.32	15.1	9.1	13.2	2.99	0.50	0.10	0.18	
1.1)SOR+QBO+AOD [%]	-20	-2	-25	-33	-35	-19	-12	-9	
2.1)SOR+QBO+AOD+EqL520K [%]	<b>-20</b>	-15	-39	-35	-35	-19	-18	-13	
2.2)SOR+QBO+AOD+EqL850K [%]	-17	-14	-41	<b>-42</b>	<b>-50</b>	<b>-36</b>	<b>-23</b>	<b>-22</b>	
2.3)SOR+QBO+AOD+EqL1300K [%]	<b>-20</b>	-16	-39	-35	-42	-33	-18	-18	
2.4)SOR+QBO+AOD+ENSO [%]	<b>-20</b>	-16	-41	-39	-37	-19	-18	-18	
2.5)SOR+QBO+AOD+SAM [%]	<b>-20</b>	<b>-22</b>	<b>-59</b>	-37	-35	-19	-18	-18	
2.6)SOR+QBO+AOD+HF10 [%]	-17	-13	-40	-32	-30	-19	-12	-13	
3.1)SOR+QBO+AOD+EqL850K+ENSO [%]	<b>-23</b>	-21	-43	<b>-42</b>	<b>-49</b>	-36	-23	<b>-22</b>	
3.2)SOR+QBO+AOD+EqL850K+SAM [%]	<b>-23</b>	<b>-24</b>	-57	-40	-48	-36	-23	<b>-22</b>	
3.3)SOR+QBO+AOD+EqL850K+HF10 [%]	<b>-23</b>	-19	-44	-37	-45	<b>-40</b>	<b>-29</b>	<b>-22</b>	
3.4)SOR+QBO+AOD+EqL520K+SAM [%]	-20	-22	<b>-56</b>	-36	-35	-19	-18	-18	
3.5)SOR+QBO+AOD+EqL1300K+SAM [%]	-20	-21	<b>-57</b>	-37	-44	-38	-23	<b>-22</b>	

$$^a \%z_1 = (VAR_{z1} - VAL_{z1}) / VAL_{z1} * 100$$

Fig. 2. Table 2a

C1397

Table 2b. Lag (month) derived for each proxy (columns) and for each layer (rows) for the model 1.1 in Table 2a (summer)

Umkehr Layer	SOR	QBO50	QBO30	AOD
1	3			
2+3				
4				9
5	3			
6				3
7	3	11	3	
8	3	11		
8+		11		

Table 2c, the same as 2b but for spring

Umkehr Layer	SOR	QBO50	QBO30	AOD
1	3	9	7	11
2+3	3	7	7	11
4	3	3	7	9
5	3	5	7	9
6	7	7	9	
7	5	9	3	
8	11	9	3	
8+	11	9	3	

Table 2

a) Summary of the explained variance for the statistical models fit separated into spring and summer season. Results are provided for Umkehr layers (columns) and for different statistical models that include several proxies (rows of the table). The first row of spring and summer season section of the Table 2 represents the variance of the residuals calculated between monthly averaged ozone time series and a statistical model that includes seasonal and EESC Polar 2 curve parameters (de-seasonalized and de-trended time series). This result is considered as a benchmark and is set at 100 %. Other rows show results for the fit of several statistical models that include additional proxies. Results are presented as percent reduction of the variance in the residuals relative to the first row. For example, the second row shows change in the variance of the residual for the model fit that included seasonal, EESC, SOR, QBO and AOD proxies as compared to the residual of the model that had only seasonal component and EESC proxy. Note, that negative values indicate improvement of the fit and the reduction in the unexplained variance (-10 means that the original ozone variability was reduced by 10 %). The boldface numbers in the table show the best fit of the

Fig. 3. Table 2b, 2c

C1398