**Atmospheric
Chemistry
and Physics
Discussions**

Interactive comment on "On the use of the correction factor with Japanese ozonesonde data" *by* G. A. Morris et al.

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Response to reviewer 2. Reviewer comments in italics.

General Remarks:
Although, the present study is based on a comprehensive analysis, discussion and conclusions drawn, my major criticisms deal with the methodology followed in chapter 2 and chapter 3. Here the paper lacks on structure and clarity. My criticisms focus on the fact why the authors still analyse the use of a CF based on the residual ozone column estimations by applying the constant mixing ratio (CMR) method instead of deriving residuals from satellite observations (e.g. McPeters et al., 1997, 2007). The CMR-method assumes a residual column with constant mixing ratio equal to the measured

values at the top of the sonde profile. This assumption is quite outdated now, as satellite observations have shown that the ozone mixing ratio declines above about 35 km altitude. At this point the present manuscript needs substantial improvement before publication in ACP (see my specific remarks).

We completely agree with the reviewer's comment that the constant mixing ratio (CMR) assumption is not the best approach to correcting the column derived from the integrated ozonesonde profile measurements. However, this was not our choice. Rather, the constant mixing ratio column adjustment for the above balloon-burst column is the method that the Japanese station ozonesonde files placed in the WOUDC database indicate. We double-checked this fact by performing our own calculation of the constant mixing ratio adjustment, which showed good agreement with the above balloon-burst column found in the WOUDC data files. If this problem truly is the one that most concerns the reviewer with respect to publication of our manuscript, it means that we failed to communicate clearly in the original manuscript this point. To that end, we have revised to the manuscript to clearly indicate that we do not endorse the use of the CMR assumption, but that it appears to have been employed regularly for the data in the WOUDC archive. You can find these changes in the abstract and in the Introduction in the revised manuscript.

Specific Remarks

Abstract
Generally okay, just revisions are needed corresponding the changes made in the paper based on my comments listed below.

We have made appropriate changes.

1 Introduction
Authors should more and clearly distinguish between the here used KC79/96 ozone sonde types and other existing types of ozone sondes such as the ECC-type that is used world wide by more than 90% of operational sounding stations or the BM-type.

KC79/96 sondes have their own specific characteristics which can be very different from ECC or BM. The reader should be aware of this from the very beginning that any conclusions drawn are only applicable on KC79/96 ozone sonde data. From historical perspective more correct is that the BM-type was first developed then CI and finally in 1969 the ECC (see WMO, 2011).

We have added some text in the introduction to clearly indicate that the CI design was used almost exclusively by the Japanese stations. The reference to the WMO (2011) document appears already. These modifications appear in the Introduction of the revised text.

2 Column measurements
This chapter is rather long, not well structured and the methodology followed should be improved:
a.) The comparison with satellites (TOMS and OMI should be moved to the end of the chapter).

It is not clear to us why we need to reorder this section in its present, revised form. If the reviewer and editor feel that the section could be clearer after reordering, we will revisit this issue, but we feel that at present, the manuscript is clear and the order logical.

b.) The chapter should start with clear definitions of CF and including the two approaches of determining the residuals (constant mixing ratio =CMR or satellite climatology=SC) and discuss briefly the pros and cons.

We have expanded the Introduction, where the topic first appears, and describe there, in some detail, the two approaches to computing the residual columns. Since this discussion appears before the column measurement discussion, we need not reorder the section.

c.) Then present time series of CF determined with CMR method and relative deviation

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of it when using SC-method.

We prefer the current order. By talking about the disagreement between the satellite columns and the Japanese sounding station columns first, we identify the apparent need for the use of a CF to achieve better agreement with correlative observations. We follow that with an examination of the relative merits of the two approaches (now clearly defined in the Introduction) for calculating the residual and demonstrate the superiority of the balloon burst approach with respect to the Japanese sounding data.

d.) In addition Figure 2 gives a good quantification of the differences between both, CMR and SC methods. The simple CMR assumption is actually not true, on the contrary it is well know from literature, for example SPARC-IOC-GAW 1998 assessment, that above about Z=35 km the ozone mixing ratio is even declining such that CMR-residuals certainly tends to be overestimated.

We agree. We cited the original paper by Dobson (1973) which recommended that the CMR assumption only be applied from the burst altitude up to ~11 hPa (~30 km), a comment that already appears in Section 2 of our manuscript.

e.) Therefore Figure 3 should be extended with curves presenting also the CFs based on SC-residuals in order to show in how far these have any altitude dependence on bursting point criteria or not.

The reviewer makes an excellent observation, and in this regard, we were not as thorough in our original submission. We have now created a figure parallel with our original Figure 3 and based upon the CFs we calculate using the balloon burst climatology. We have added this new figure to the paper as Figure 3b (it appears attached to this response as Figure 1) and relabeled the original figure as Figure 3a. It is clear that by using the balloon burst climatology, lower burst altitude data can be meaningfully compared with correlative observations, a important conclusion that did not appear in the original manuscript, but that appears in our revised version. This conclusion provides even more of an endorsement for everyone using the balloon burst climatology rather

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than a constant mixing ratio assumption for computing the above burst altitude column corrections.

f.) Knowing that CFs based on CMR-method will tend to be lower than CF's based on SC-method. This is certainly the case with KC79/96 sonde types which both tend to measure more ozone in the upper part of the profile (See e.g. JOSIE 1996/2000 and BESOS 2004 results) such that residuals estimated with CMR get even more enhanced.

We concur, and we strongly recommend not using CMR to estimate the above burst column. Nevertheless, the CMR estimate is used widely among the stations that submit their data to the WOUDC archive, and the community of data users should be reminded of the limitations of this approach, as this study, we believe, clearly points out.

g.) At present it is not clear in how far the results presented in the Figures 4 and 5 may be biased by the criterion of only using data with Zburst >= 32 km.

We have reproduced Figures 4 and 5 (not shown) using a minimum burst altitude of Zburst >= 26 km instead. The figures appear to be nearly identical to those using the higher altitude burst criterion. We also have reproduced Tables 4 and 5 using a minimum burst altitude of Zburst >= 26 km (not shown). Only one entry in the entire Table 4 changes by an amount greater than one standard deviation. No entries in Table 5 change by a statistically significant amount. Thus, we can say that the choice of the burst altitude for this analysis is NOT the primary factor in the trends observed.

Questions remain in how far CFs based on SC method show a similar behavior or less sensitive to this criterion. The statistics presented in Table 4 & 5 and discussed extensively at pages 15605/6 are these still relevant when one would apply CF's based on SC method or getting different?

These questions are also good ones. From our original Figure 2, it is apparent that the use of the balloon burst climatology improves the CF distributions (i.e., move the

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distributions closer to 1.0), but the shapes of the distributions remain unchanged. Furthermore, we have reproduced Figure 4 using CFs calculated from the balloon burst climatology (Figure 2 in this response). This figure closely resembles the original Figure 4 from the paper, so the trends are still present. That means that the above-burst column corrections themselves are not the cause of the trends.

We have added Table 4b (Figure 3 in this response) to the paper with the trends computed using the columns computed with the balloon-burst climatology, and we have added some text to the discussion. While these trends tend to be smaller than those found with the CMR columns, they remain statistically significantly different from 0. Thus, the above-burst columns are not the primary factor in the trends.

We have also computed Table 5 (Figure 4 in this response) based on the balloon burst CFs and found nearly identical results. Thus, it appears that the method has little impact on the main points that we are trying to make in this paper.

h.) These questions should be addressed by the authors before they start to discuss the impact on any ozone trend.

Done.

i.) After this the comparison with TOMS and OMI satellites (Figure 1 and Table 3) should be presented and discussed.

Again, this point gets to reorganizing the paper, which we would prefer not to do. We believe that the current sequencing communicates our points clearly, but if the editor feels the paper could be substantially improved by a reorganization as suggested by this reviewer, we are willing to attempt it with additional time.

3 Surface measurements
The comparison can be only made in a qualitative way. To compare near surface

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ozone measurements made on different locations is difficult and certainly not straight forward as has been done in the present study because the origin of air masses at the sounding and co-located surface site can be very different and strongly influenced by local pollution.

While we do not disagree with the reviewer's point, we would point out that the large number of comparisons most likely mitigate the influence of local sources. Variation in wind direction at the sampling locations relative to the various sources should prevent any one local source from dominating the distribution. Further, as we acknowledge in the paper, the surface stations are separated by a few to as large as 160 km from the ozonesonde launch sites.

The Japanese surface measurements reported here are therefore not only dealing with ozone but with the sum of all oxidative (i.e. odd oxygen = O3 + NO2 + NO3 etc.) compounds. Near surface OX measurements can have at least significant NO2 contributions of 5-10 ppbv or even more depending on strength of nearby pollution. Hereby emitted NO reacts with O3 into NO2 that reacts with OH into HNO3 that acts as a net O3 sink in polluted and urbanized regions. Quantitative comparisons of O3-sonde measurements with OX surface measurements are meaningless: Figure 5 can only give qualitative indications about the shift towards 0 line when no CF applied. Table 6 should be omitted.

Our Japanese co-author, Masayuki Takigawa, has provided the following clarification of the Japanese "Ox" data (in bold):

It seems the Ministry of the Environmental in Japan has defined "oxidants (Ox)" as an oxidizer which produces Iodine from Potassium Iodide solution. It includes ozone, Peroxylacetyl nitrates (PAN), and other oxidizer which is produced from photochemical reactions, but it excludes nitrous oxides (cf. http://www.env.go.jp/kijun/taiki.html, in Japanese).

Until 1997, oxidants were observed by using absorption spectroscopy which has weak sensitivity to PAN (Ox is nearly equal to O3). However, most of stations use UV absorption for the observation of "oxidants" now, but it observes ozone (Ox=O3).

So, you do not need to subtract NO2 from Ox for the estimation of O3. It seems some stations at Sapporo still uses absorption spectroscopy, but the fraction of PAN in Ox is negligibly small (0.8 ppbv for the average during 1976-1985).

Based on Takigawa-san's response, our comparison in the original version of the paper remains valid. We also note that the Hedo data are identified as O3. In all cases, the comparisons between the surface monitors and the sondes at the surface are improved by removing the CF from the profile.

Finally, we note that the surface monitor data is an important piece of the puzzle (absent additional profile intercomparisons) to support our suggestion that if CFs are applied, they probably should not be applied to the tropospheric portion of the profile. We cannot at present explain the reason for the need for a CF in the stratosphere but not in the troposphere, but that question is worthy of additional study.

We recognize that in the original version of the paper, the Japanese surface measurements were described in such a way that we understand why the reviewer made his/her comment. We hope that the discussion above has clarified the data. We have added text to the revised paper based on Takigawa-san's note as well.

4 Sapporo ozone sonde intercomparison
This comparison is most powerful comparison because it shows clearly the key issues on the use of CF and its impact on the different parts of ozonesonde profile. However, here should also be discussed the differences between CF based on CMR and SC method respectively.

Additional comparisons with other vertical ozone profiling instruments (Lidar, Micro-Wave) that are available at Tsukuba would increase statistics and are certainly very valuable for the paper.

While we do not disagree that such comparisons would be very valuable, the lack of their presence in this paper does not detract from our main result: the Japanese sonde stations display an unexplained trend in the correction factors applied to their ozone profiles. The fact that this data has been in the public domain for years and has been used in numerous studies argues for the publication of the present work in the short term. A follow-up study along the lines suggested by the reviewer would be valuable, but needs to be part of a separate manuscript to be submitted in the future. We would be delighted to collaborate with the reviewer if he/she has access to the particular data sets in mind for our use in such a study.

5 Conclusions and recommendations
These should be revised and streamlined based on my comments made before.

We have revised our conclusions after addressing all of the comments from both reviewers.
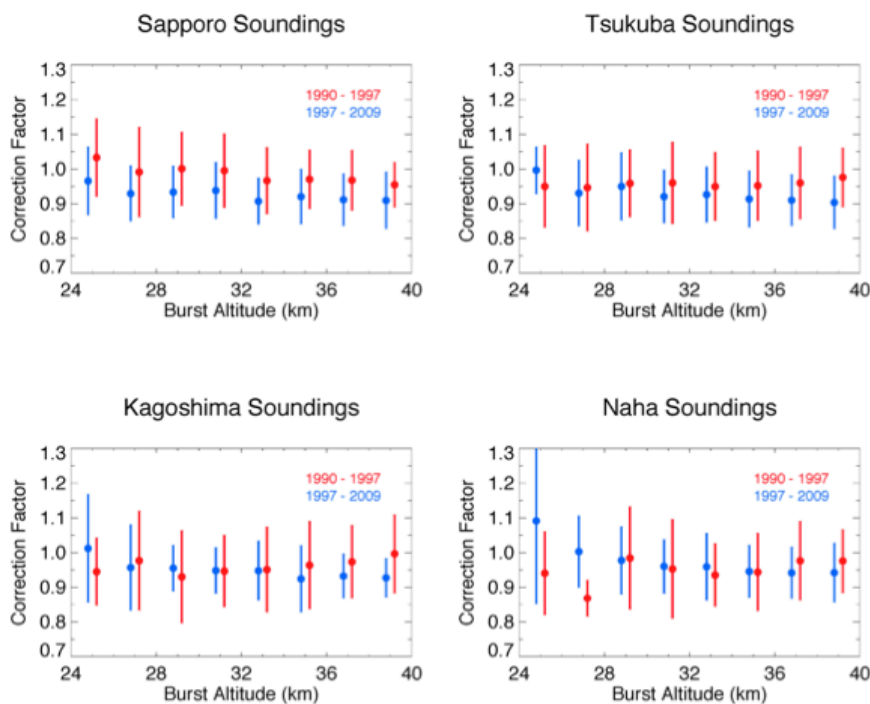
Please also note the supplement to this comment:
http://www.atmos-chem-phys-discuss.net/12/C10370/2012/acpd-12-C10370-2012-supplement.pdf
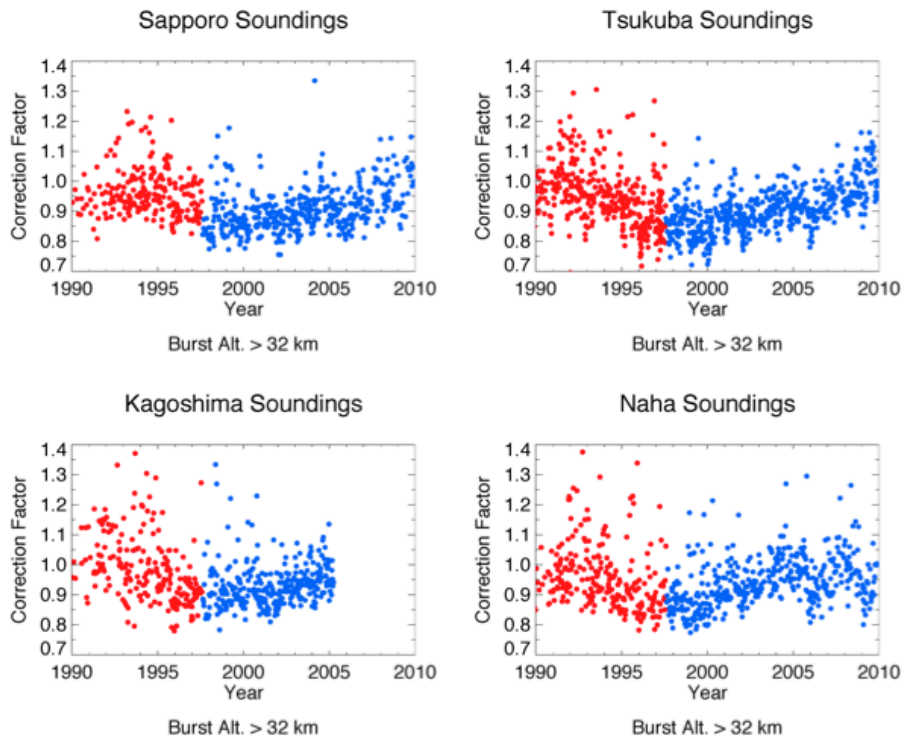
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Interactive comment on Atmos. Chem. Phys. Discuss., 12, 15597, 2012.

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Fig. 1. As in Figure 3 of the original manuscript, but the CFs are calculated on columns computed using the balloon burst climatology to compute the above-burst altitude columns.

Fig. 2. As in Figure 4 in the original manuscript but using the balloon burst climatology rather than the constant mixing ratio assumption.

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Site	1990 – 2010 (10^{-2} yr^{-1})	(KC-79) 1990 – 1997 (10^{-2} yr^{-1})	1993 – 1999 (10^{-2} yr^{-1})	(KC-96) 1999 – 2009 (10^{-2} yr^{-1})	Delta (10^{-2} yr^{-1})
Sapporo	− 0.136 (0.049)	−0.71 (0.23)	−1.76 (0.22)	0.94 (0.10)	2.69 (0.24)
Tsukuba	−0.050 (0.045)	−1.89 (0.18)	−1.86 (0.20)	1.338 (0.077)	3.19 (0.22)
Kagoshima	−0.596 (0.085)*	−2.13 (0.28)	−1.43 (0.28)	0.66 (0.21)*	2.08 (0.35)
Naha	0.025 (0.053)	−1.41(0.26)	−1.30 (0.24)	0.57 (0.11)	1.87 (0.27)
Overall	*−0.190 (0.030)*	*−1.54 (0.12)*	*−1.59 (0.12)*	*0.874 (0.067)*	*2.45 (0.14)*

Fig. 3. As in Table 4 of the original manuscript, but using the balloon burst climatology for the above burst altitude column.

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Station	CFs 1968 – 1975	CFs 1990 - 2002
Sapporo	0.980 (0.090)	0.929 (0.076)
Tsukuba	0.97 (0.11)	0.90 (0.10)
Kagoshima	0.94 (0.14)	0.94 (0.11)

Fig. 4. As in Table 5 of the original manuscript, but using the balloon-burst adjusted column rather than the constant mixing ratio assumption.