Response to anonymous referee #1

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Firstly, we thank the Anonymous Referee #1 for the useful and valuable comments, which provided insights that helped significantly to improve the paper. Secondly, we would like to stress that this publication really provides new findings, since the trend study of the GROMOS dataset has not been independently published yet. Steinbrecht *et al.* (2006) has used the GROMOS data mixed with the lidar data at Hohenpeissenberg to calculate the ozone trends from 1997 to 2005. In the WMO (2014) the GROMOS data (1994 to 2012) is used together with the Payerne station data, as far we can understand from Chapter 2 of WMO (2014). In addition, the trend profile of the microwave instruments is hidden by other lines in Figure 2-11 of WMO (2014). The important overview on trends in WMO (2014) cannot be a substitute for detailed trend studies of station data. Our study presents an harmonisation of the GROMOS dataset along with a trend estimation of the stratospheric ozone profiles from 1997 to 2015, calculated through a new robust method of trend estimation (von Clarmann *et al.*, 2010). We have revised the manuscript by following each one of your suggestions. This document includes all of your reported issues, as well as our responses in how we have addressed them.

General comments

1. **Comments from the referee:** There is no mention, where the data/homogenized time series from the GROMOS spectrometer are available. These data are probably available through the Network for the Detection of Atmospheric Composition, so this should be mentioned. Also it should be made clear which version of the data-set is available there - preferably the newest and most homogeneous version.

   Author's response:
   The harmonised GROMOS data since 1994 are available through the Network for the Detection of Atmospheric Composition Change (NDACC) on the NDACC public website at:
   http://ftp.cpc.ncep.noaa.gov/ndacc/station/bern/hdf/mwave/

   Author's changes in the manuscript: The ozone radiometer GROMOS is part of the NDACC~~.~~, hence our more than 20 years harmonised time series are available via http://ftp.cpc.ncep.noaa.gov/ndacc/station/bern/hdf/mwave/

2. **Comments from the referee:** Why is there no time series comparison with the NDACC lidar data from nearby stations at Hohenpeissenberg and Haute Provence? There is also no comparison with any satellite records, although many satellite ozone

records are availabe, e.g. during the 2009 to 2011 period when the two backend spectrometers were run in parallel.

Author's response:

In Studer *et al.* (2013, ACPD) one can find a time series comparison with coinciding lidar measurements from the Observatoire Haute Provence (OHP), France, also with satellite measurements including MIPAS onboard ENVISAT, SABER onboard TIMED, MLS onboard EOS Aura and ACE-FTS onboard SCISAT-1; aditionally, with ozonesondes launched from Payerne, Switzerland. The result being that the mean relative differences of GROMOS FFTS and these independent instruments are less than 10% between 50 and 0.1 hPa.

Author's changes in the manuscript: Ozone time series from the GROMOS microwave radiometer were used for comparisons with lidar, ozonesondes and collocated satellite observations and for detection of long-term trends (Dumitru *et al.*,2006; Steinbrecht *et al.*, 2006; Steinbrecht *et al.*, 2009; Keckhut *et al.*, 2010;van Gijsel *et al.*, 2010; Studer *et al.*, 2013; Delcloo and Kreher, 2013).

3. **Comments from the referee:** The multilinear fit residuals (middle panel in Fig. 6) are almost screaming for a parabolic trend in Eq. 1 (linear and quadratic terms in t). *Why has that not been tried? I think this should really be tested. It would be one "novel" aspect from the paper.*

Author's response:

With all due respect to the referee, we felt that there are no physical reasons for a parabolic trend estimation of 17 years of stratospheric ozone profiles. Actually there is a polynomial of third degree in the residuals but a geophysical interpretation is questionable.

Author's changes in the manuscript: No changes.

Specific comments

1. **Comments from the referee:** Pg. 16373, around line 20: I think the most important reference, Newchurch et al., JGR, 2003 is missing here. Please add.

Author's response:

No comments.

Author's changes in the manuscript: From the late 1990s, there were some measurements and model calculations indicating a turnaround in the decreasing ozone, suggesting that the negative ozone trends in the stratosphere would level out or even become positive (~~Huang *et al.*, 2014).~~Newchurch *et al.*, 2003).

2. **Comments from the referee:** Pg. 16374, line 21: What is meant by harmonic variation? Probably the annual cycle and its harmonics (12 months, 6, 4, 3 and 24 months). Why not say so?

Author's response:

No comments.

Author's changes in the manuscript: The regression model consists of an axis intercept, a linear trend, ~~harmonic variation~~sine waves, and several proxies.

3. **Comments from the referee:** Pg. 16376, line 11: "than" should be replaced by "as"

   Author's response:

   No comments.

   Author's changes in the manuscript: A sample of a calibrated ozone spectrum is given in Figure 2. It shows the ozone line recorded by the FFTS on the same winter morning ~~than~~ as the FB spectrum (Figure 1).

4. **Comments from the referee:** Pg. 16376, line 13: Is there a "spectrum" missing at the end?

   Author's response:

   No comments.

   Author's changes in the manuscript: The integration time is 30 minutes and no frequency binning is applied in the blue curve whereas the red line represents the 15 MHz frequency binned spectrum.

5. **Comments from the referee:** Pg. 16377, line 13, 14: I do not understand the second part of that sentence? Why does the temperature at 2.5 km "exponentially approach" the surface temperature? Overall temperature at the surface and at 2.5 km will be highly correlated (except for diurnal cycle, or temperature inversion situations). This sentence should be reworded.

   Author's response:

   T_{mean} depends on the temperature profile ($T(z)$) as well as on the absorption profile $\alpha(z,\nu)$ at a specific frequency. Since the number density is highest at low altitudes and the absorption is highest near the ground, T_{mean} has a value close to the temperature of the lower troposphere. And it is defined,

   $$T_{mean} = \frac{\int_{z_1}^{z_t} T(z)\alpha(z,\nu)e^{-\tau(z,\nu)}dz}{\int_{z_1}^{z_t} \alpha(z,\nu)e^{-\tau(z,\nu)}dz} \quad (1)$$

   Author's changes in the manuscript: T_{mean} ~~is the temperature at approximately 2.5 km altitude of the actual temperature profile approaching exponentially the surface value (Peter, 1997).~~ depends upon the temperature profile as well as on the absorption profile at a specific frequency. Since the number density is highest at low altitudes and the absorption is highest near the ground, T_{mean} has a value close to the temperature of the lower troposphere (Ingold *et al.*, 1998).

6. **Comments from the referee:** Pg. 16379, around line 10: I would argue that 5% difference are not a "small" bias, when you try to analyse 3% per decade trends. As mentioned, I find it disappointing that no other instruments (lidars, satellites) were used as independent references.

   Author's response:

   As we highlighted previously, mean relative differences of GROMOS-FFTS and independent data sets (lidar, satellites and ozonesondes) are less than 10% between 50 and 0.1 hPa.

Author's changes in the manuscript: The purpose of the harmonisation is to correct this ~~small~~ bias between both spectrometers, by using the data from FFTS as reference.

7. **Comments from the referee:** Pg. 16380, line 14: Where do the 7.2 and 8.4 months come from? Please mention (e.g. 8.4 and 24 months are the annual modulation sidebands of a 28 month QBO)

   Author's response:
   7.2 and 8.4 months are the annual modulation sidebands of 18 and 28 months oscillation respectively, both related with QBO.

   Author's changes in the manuscript: No changes.

8. **Comments from the referee:** Pg. 16380, line 24: Where does the error covariance matrix come from? Are the diagonal elements from your error/ uncertainty considerations in Section 5? Where do the off-diagonal elements come from? Are they from the various lag-auto-correlations of the residuals after the regression? Are they significant? How big is the effect of the off-diagonal elements? Please make this part a lot clearer.

   Author's response:
   The diagonal elements of the error covariance matrix are the uncertainty of the ozone monthly mean profiles, described in Section 5 Uncertainty considerations. The off-diagonal elements are initially set zero. In a second iteration, the correlation coefficients between each data-point and its nth neighbour are estimated from the fit residuals, and an additional error term is built, based on these covariances, describing the deficiency of the multi-parametric model chosen. This error term (in terms of covariance matrix) is scaled according to chi-square statistics and added to the initial measurement error covariance matrix.

   Author's changes in the manuscript: ~~, with the squared standard errors of the monthly means as the diagonal terms ([?]). The autocorrelations among data points are considered in the covariance matrix, and a model error component is assessed iteratively and added to the covariance matrix in order to account the autocorrelative nature of the atmosphere and to get realistic error estimates.~~ . The diagonal elements of the error covariance matrix are the uncertainty of the ozone monthly mean profiles, described in the following section. The off-diagonal elements are initially set zero. In a second iteration, the correlation coefficients between each data-point and its nth neighbour are estimated from the fit residuals, and an additional error term is built, based on these covariances, describing the deficiency of the multi-parametric model chosen. This error term (in terms of covariance matrix) is scaled according to chi-square statistics and added to the initial measurement error covariance matrix.

9. **Comments from the referee:** Pg. 16381 1st paragraph: As mentioned, the shape of the residuals in Fig. 6 (middle panel) suggests strongly that a quadratic term should also be included/ tested in Eq. 1.

   Author's response:
   As previously mentioned, we think that there is no physical argument for adding

a quadratic term in the trend model. The shape of the residuals at 10 hPa shows some short term anomalies.

Author's changes in the manuscript: No changes.

10. **Comments from the referee:** Pg. 16381, lines 7-8: "Most ... can be explained". Please be more quantitative. What fraction of the variance is explained? What are typical values for R^2?

    Author's response:
    No comments.

    Author's changes in the manuscript: The residual is within 0.5 ppmv except for some particular cases~~. Most of the ozone variations can be explained by the fitted proxies~~ , maybe due to some short term anomalies.

11. **Comments from the referee:** Pg. 16382: Eqs. 2 and 3 are not correct. There needs to be a square root taken from $\frac{1}{n-1}\sum$ on the right side.

    Author's response:
    No comments.

    Author's changes in the manuscript:

$$\sigma = \frac{1}{n-1}\sum_{i=1}^{n}(x_i-)^2 \sqrt{\frac{1}{n-1}\sum_{i=1}^{n}(x_i-\bar{x})^2} \quad (2)$$

$$SEM = \frac{\sigma}{\sqrt{DGF}} = \frac{1}{(n-1)\sqrt{DGF}}\sum_{i=1}^{n}(x_i-)^2 \frac{1}{\sqrt{(n-1)DGF}}\sqrt{\sum_{i=1}^{n}(x_i-\bar{x})^2} \quad (3)$$

12. **Comments from the referee:** Pg. 16383, last paragraph: Why are the data from 1994 to 1997 not used? This should be mentioned, and should be explained.

    Author's response:
    As we mentioned on page 16383 line 25, the selection of the time interval is based on the assumption that 1997 is the turn-around year of EESC. Therefore, we decided to discard the firsts years of GROMOS measurements in order to not bias the trend result by starting before the EESC peak at mid-latitudes.

    Author's changes in the manuscript: No changes.

13. **Comments from the referee:** Pg. 16384, line 14: Which studies? The ones below? Please reword/ clarify.

    Author's response:
    No comments.

    Author's changes in the manuscript: On the other hand, other recent studies (Eckert *et al.*, 2014; Vigouroux *et al.*, 2015; Harris *et al.*, 2015; and references therein) have found positive but not significant trend in our location. But we have to be careful about these discrepancies since it could arise from differences in treatment and propagations of uncertainties, selection of data, ozone measurement techniques, statistical approach, latitudinal and altitudinal extent and/or the time period covered in the trend study~~(Eckert *et al.*, 2014; Vigouroux *et al.*, 2015; Harris *et al.*, 2015; and references therein)~~.

14. **Comments from the referee:** Pg. 16385, line 17 to page 16386, line 10: This discussion has little to do with the presented GROMOS data and is not supported by anything else presented in the paper. As such is speculative and I strongly suggest that it should simply be deleted.

    Author's response:

    The aim of this discussion is to present to the reader a wider view of stratospheric ozone, considering the ozone photochemistry and transport processes along with anthropogenous and natural changes of the global circulation system. Changes in the position and strength of the circulation cells may induce regional changes in ozone. A discussion of this topic is important for our article since it underlines the need for reliable monitoring and detection of ozone trends over the next decades.

    Author's changes in the manuscript: No changes.

15. **Comments from the referee:** Pg. 16386, lines 11 to 16: What is the explanation for the declining ozone trend in the mesosphere? Please add an explanation.

    Author's response:

    Unfortunately no explanation so far have been given in the literature for such a decline of ozone in the mesosphere though several other authors observed the same effect as mentioned in Section 6: Results and discussion. The mesospheric ozone trend would be an interesting topic for a simulation study with a chemistry climate model such as WACCM.

    Author's changes in the manuscript: No changes.

16. **Comments from the referee:** Pg. 16391, line 6, Typo: temporal

    Author's response:

    No comments.

    Author's changes in the manuscript: Tiao, G. C., Reinsel, G. C., Xu, D., Pedrick, J. H., Zhu, X., Miller, A. J., DeLuisi, J. J., Mateer, C. L., and Wuebbles, D. J.: Effects of autocorrelation and ~~tempotal~~ temporal sampling schemes on estimates of trend and spatial correlation, J. Geophys. Res., 95, 20507-20517, doi:10.1029/JD095iD12p20507, 1990

17. **Comments from the referee:** Figure 4: I do not find this plot useful. Mostly it shows repetition of the well known annual cycle. To make this plot useful, it would be much much better to remove the annual cycle and show ozone anomalies, either as ppmv or as % deviation from the annual cycle. Please make a better plot.

    Author's response:

    The reason to include this harmonised 20 years time series of stratospheric ozone VMR profiles, is to show our harmonised data prior the trend analysis. This figure surely shows the annual cycle but also the increase (positive anomaly) of mid-stratospheric ozone in last years. Further, with this figure one can get a view of the maximum of ozone at our station and its evolution over the last two decades.

Author's changes in the manuscript: No changes.

18. **Comments from the referee:** Figure 7: I think this Figure could be improved a lot by not comparing apples (=%) and eggs (ppmv and days). The total uncertainty (blue line) should not be shown in ppmv, but also in percent. Then it becomes comparable with the estimated (thermal) observation noise (purple line), and with the observed uncertainty (black line). Instead of time lag (red line) the $1/\sqrt{DGF}$, converted also to % of the ozone profile, should be shown. I would expect that this atmospheric variability part would explain why the observed uncertainty is much larger than the thermal noise at levels between 100 hPa and 10 hPa. These changes would result in a much clearer plot.

Author's response:

We agree with the referee that all the uncertainties should be shown in percent. Regarding the monthly mean correlation length profile we believe that this should be shown in units of days. Anyway, even showing it in units of days one can easily estimate the DGF from the correlation length profile. For example, if the amount of measurements of GROMOS within a month is around 1300, and at 10hPa the time lag is around 2 days then the DGF are more or less 12.5. Following the same assumption, at 100 hPa (time lag around 5 days) the DGF are 4.8 and 81.25 at 1hPa (8 hours of time lag).

Author's changes in the manuscript: Figure ~~??~~ 1 shows the error budget used as input for the trend estimation model. The red line is an example of monthly mean correlation length profile, in day units, calculated for the time interval from October 2011 to October 2014. We can see the monthly autocorrelations of stratospheric ozone. The magenta line is the monthly mean observation error profile, from the thermal noise on the spectra, calculated for the same time interval. The ~~black~~ blue line is the estimated ~~systematic~~ instrumental error profile based on recent and past intercomparisons of coincident data from GROMOS, ozonesondes, nearby lidars and satellites. And the ~~blue line represents in ppm VMR~~ black line represents the total contribution of the uncertainty of GROMOS taking into account all the aforementioned errors.

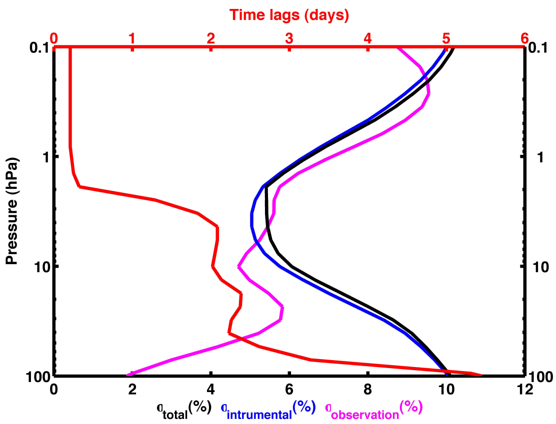

Figure 1: Uncertainty budget of GROMOS used in the trend analysis. The red line is an example of monthly mean correlation length profile, in day units, calculated for the time interval from October 2011 to October 2014. The magenta line is the monthly mean observation error profile, calculated for the same time interval. The ~~black~~ blue line is the estimated ~~systematic~~ instrumental error profile. And the ~~blue~~ black line represents ~~in ppm VMR~~ the total contribution of the uncertainty of GROMOS.