Response

We thank the referees for their additional comments, which are addressed below.

Furthermore, after taking a close look at the paper with the modification made, we made some additional changes to the discussion (section 4) for the part between lines 684 and 711. Parts of this section were moved to either section 3.1 or the discussion section, and the remainder of the part between lines 684 and 711 has been adjusted for readability and consistency.

Referee #1

We have included/modified the suggested technical corrections.

Referee #2

- We modified the discussion and conclusions so that it should be clear now where the "30-60%" originates from (table 7) and how to interpret those numbers.
- We rephrased the last sentence of the Abstract, explaining what we mean when we talk about "choices":

"... uncertainties in parameters and independent variables and choices in defining the optimal time period and area for calculating the ozone record and the independent variables ..."

Note that it is important to realize that both uncertainties in parameters and choices in time period and area over which to average hamper detection of recovery.

- We modified the discussion, giving less prominence to the notion that for some certain regressors extending the period does not always lead to higher significance. This discussion was originally added to show that all uncertainties discussed in the paper leave (quite) some room for interpretation and that not all results conform to prior expectations. It is therefore tempting to focus only on favorable results confirming prior expectations while neglecting unfavorable results, so we wanted to make sure that the unfavorable results (although minor) are mentioned.
- We added a sentence noting that Solomon et al. [2005] show that there appear to be some volcanic effects in height resolved ozone during the ozone hole season, but that these effects are too small in magnitude and vertical extend to leave a detectable impact in total ozone. When looking superficially there still appears to be some change in total ozone related to volcanic activity, but Poberaj [2011] show that these are more likely related to (coincidental) dynamical effects than to volcanic aerosols. Our study as well as those of Knibbe et al. [2014] confirm the lack of volcanic signal in total ozone. We made sure that in the discussion our results only apply to total ozone, not height resolved ozone.
- Adding the finding that we do find positive post-break trends in ozone (1.66 to 4.74 DU/year; 95% CI) is an important finding. It is one of the requirements for detection of ozone recovery, and despite uncertainties with regard to trend significance ALL trends in ozone are positive. There was no a priori reason to assume that had to be the case, and it is consistent with various other studies noting that although trends may not necessarily be statistically significant, they tend to be positive. No changes made.
- See also second comment. We added the explanation in the Abstract of what hampers detection (uncertainties in data AND choices for area and time period over which to average). That way the Abstract and Conclusions are consistent.
- Line 767-769: deleted the sentence. It is now implicitly included in the previous section.
- Added a sentence to both the discussion and conclusions section about the relation between statistical significance and length of the record noting that our results suggest that with continued extension of the total ozone record and using multi-variate regressions detection of Antarctic ozone hole recovery may be reached before 2020.

This is what our findings imply (see table 7). Currently (2010 or 2012) we are around 50% statistical significance. Our results also indicate that for each year added the significance increases by approximately 10 % (order of magnitude). Although just a ball-park estimate, it suggests that by 2020 you may expect to have passed the 95% confidence level.

- Caption changed. Table 5 shows the results for the time series ending in 2010 for all break years (97, 98, 99). Also adjusted the description of table 5 in the discussion section.

Referee #3

General comment & discussion on trend error calculation

Thanks to the extensive response of referee #3 we now finally understand where the differences stem from with regard to the trend error calculations in Kuttippurath et al. [2013] and ours. In essence there are different aspects relevant for the trend errors.

(A) The EESC regression parameter (C5 in referee #3 comment) has an associated error

(B) Linear regression of EESC multiplied with regression parameter (C5) pre-break and post-break have their own associated errors

(C) Residual of the multi-variate regression

In Kuttippurath et al. [2013] equations (2) and (3) of the #R3 report are used for calculating pre-break and post-break ozone trends. Equations (4) and (5) of the #R3 report are used to calculate the pre-break and post-break ozone trend errors.

In particular, the ozone trend error is calculated as:

$$\frac{dO_3}{dt}_{\substack{error\\time\ period}} = EESC_{error} \times \frac{dEESC}{dt}_{time\ period}$$

For trend error calculations the pre-break and post-break linear trends in the EESC must be calculated. These linear trends have their own errors because of the fact that the shape of the pre-break and postbreak EESC curve are not completely linear.

In our paper we calculate trend errors based solely on this last step, which results in fairly small trend errors.

If we calculate the trend error based on the method in Kuttippurath et al. [2013] we find order of magnitude similar values similar to those of kuttippurath et al. [2013], and indeed the post-break trend error is proportional to pre-break trend error via the ratio of the pre-break and post-break trends in EESC. See table values in bold.

Both methods describe two different error sources and should be taken into account together. For the case discussed here the error associated with the EESC regression dominates the error from the linear regression on both pre-break and post-break trends. See table values in bold + italics

	Kuttippurath et al. [2013]		Kuttippurath et al. [2013] Th		This study
Period	EESC	PWLT	EESC	PWLT	
1979-1999	-4.50 ± 0.65	-5.02 ± 1.11	-5.39 ± 0.22	-5.66 ± 1.03	
			-5.39 ± 0.97		
			-5.39 ± 0.99		
2000-2010	1.11 ± 0.16	2.91 ± 2.73	1.04 ± 0.12	3.30 ± 2.85	
			1.04 ± 0.19		
			1.04 ± 0.22		

However, after further consideration of the method applied in Kuttippurath et al. [2013] we argue that the method applied there is mathematically not justified and leads to conceptual problems. Underlying reason is that the error associated with the EESC regression is only valid for the entire EESC time series, not parts of it, as outlined below.

Consider the following hypothetical situation with the following EESC shape: the pre-break increase is as the current EESC but the post-break changes is flat (zero). According to equation (5) of the #R3 report and the equation here above, the associated ozone trend error would then be zero regardless of the regression parameter (C5).

This is counter-intuitive: in case of similar noise levels, one would expect that the smaller the trend, the larger the trend error. Or at least a non-zero trend error.

The EESC regression error applies to the entire record and should be considered as such. Because of the particular shape of the EESC (increase-decrease) the regression error will be largely determined by corresponding long term changes in ozone (decrease-increase), not the year-to-year changes in ozone. However, for the two separate periods one would expect the trend errors to also be dependent on the year-to-year variations in ozone (the sign change in EESC which dominates the fit with the total ozone record does not matter anymore in case of the two separate periods). Hence, the post-regression separation in two periods should be considered very carefully.

This is highlighted by the very different trend errors of the PWLT fit (see table above). Whereas the EESC-associated error is considerably smaller for the post-BREAK period compared to the pre-BREAK period, the PWLT trend errors show exactly the opposite: the pre-BREAK trend error is much smaller than the post-break trend error. The latter is more intuitive: the pre-BREAK period is longer and the trend is larger, which should results in smaller trend errors.

In summary, we argue that we think the calculation of trend uncertainty in Kuttippurath et al. [2013] is not justified, and that as far as we are concerned it is unclear how to actually do that (other than *a posteriori* applying a sort of piece-wise linear trend calculation, which then should be included in the regression to start with rather than be applied *a posteriori*). Trend errors as derived from the method applied in Kuttippurath et al. [2013] are counter intuitive: one would expect larger trend errors for the post-BREAK period, not smaller. The PWLT errors do are consistent with this notion. If the goal is to determine pre-break and post-break trends and errors, then the fit parameter should consist of two separate parts, as is done when including the PWLT in the regression.

We have modified sections 3.1 and the discussion in section 4, as well as table 3 according to the discussion above. Note that we have not referred to this in the in the abstract or the conclusions, as that would put too much emphasis on it and there were already sufficient arguments to argue that use of EESC in the regression is not preferred.

Specific comments

Line 28: included

Line 34-35, changed to "the onset of recovery of the Antarctic Ozone Hole"

Line 70-71, changed to "Both studies do not consider the effect of deterministic variations in ozone on estimating when the onset and complete recovery of the Antarctic ozone hole recovery occur."

Line 202-203. This is a tricky point. Salby et al. [2012] indeed only identify 2002 as an outlier. However, Weber et al [2003, 2011] and updates in the upcoming WMO 2014 ozone assessment report indicate that in both hemispheres there are outlier years that 'skew' the statistics.

We suggest to modify the text and make it more specific and use somewhat less strong wording.

"The Arctic and Antarctic may behave very similar [Weber et al., 2003; 2011] or much less similar [Salby et al., 2012]. This is because the notion of hemispheric similarities in how the EP flux affects ozone depletion so far may be biased by a few outlier years (2002 and 2006 for the SH, 1996, 2010 and 2011 for the NH)."

Line 515-517. Section between lines 511-517 was completely changed.

"Note that there is slight difference in the 1979-1999 trends for the period ending in 2010 and 2012 because of the difference in total record length, which results in slightly different regression coefficients. However, calculating trend errors for the EESC-based pre-BREAK and post-BREAK trends in ozone using the EESC regression error as done in Kuttippurath et al. (2013) is not justified. The trend errors depend on the actual trend values themselves (see table 3): the EESC-fit based post-break trend error is much smaller than the pre-BREAK trend. In the hypothetical case of no (zero) trend the trend error would also be zero, which would be physically unrealistic. The PWLT on the other hand shows opposite differences in trend errors: the post-BREAK trend error is much larger than the pre-BREAK trend error, conform expectations."

Line 560-562. Correct, should be table 3. We did an additional check on all table and references to them in the paper as there had been a change in table numbering between versions.

Line 563-565. See discussion above about trend errors.

Line 626. Figure 3 should be figure 4 (middle panel). The lower panel of figure 6 also shows a tri-modal distribution. Text adjusted accordingly.

Line 638. Text adjusted accordingly.

Line 652. Text adjusted accordingly.

Line 653-654. See discussion above about different approaches on trend error calculations. Answer: the EESC fit uncertainty will be largely determined by the long ter changes in ozone (decrease-increase), not the year-to-year variations. However, when separating the decreasing and increasing part of the EESC,

the year-to-year variations should be considered as the sign change in long term ozone changes do not matter anymore.

Line 707-711. Reading the comment, we think that the referee actually refers to the previous section (696-706). This section was replaced by the next statement, which is less pessimistic and does more justice to our analysis. It shows that there is a balance: a gain in confidence by removing some deterministic variations in ozone, but with some adverse effects as the multi-variate regression introduces new uncertainties. The section then ends on a positive note (lines 707-711):

"Furthermore, this implies that with continued extension of the total ozone record detection of Antarctic ozone recovery may be reached before 2020 using multi-variate regressions. Note that although in total the number if statistically significant trends increases with record length, this is not necessarily always the case (compare Table 5 and Table 6 – thus BREAK-2010 vs. 1998-2012 trends).

On the other hand, the trend significance level if significant trends is generally between 2σ and 3σ (not shown), indicating that a considerable amount of variability is not accounted for in the regression. Our analysis also hows that detection of the 2^{nd} stage of ozone recovery based on just one arbitrary selected (set of) regressor – ozone record combination(s) does not reflect the structural uncertainties present in the underlying data.

Nevertheless, the appearance of larger groups of statistically significant results occurring for longer time series and a certain persistence among ozone scenarios and EP flux scenarios shows that these type of analyses are capable of removing deterministic variations in average ozone, and that with increasing length of the post-break period more statistically significant results can be expected."

Line 724-727: see discussion above. Now that we full understand what was done in Kuttippurath et al. (2013) we argue that that method does not take the fit residuals into account.

Line 735. The larger range of trends in the PWLT compared to the EESC fits is indicative of the limited use of the EESC – its post-peak trend is largely determine by the pre-defined EESC shape and thus does not do justice to all uncertainties. This is what we refer to as a lack of flexibility when using the EESC to determine post-BREAK ozone trends.

Line 774. Suggestion to change to "are expected", rather than "may" or "will", because we expect that future updates of this analysis provide better clues about the onset of recovery.

Table 5. Table caption modified (ending 2010).

Line 977. Typical maximum wind speed numbers added (20-30 m/s)

Line 982. Typo corrected.

Line 1000-1026.

Figures 6-7. Suggestion included.

1	Tracing the second stage of Antarctic ozone hole recovery with a "big data"
2	approach to <u>multi-multi</u> variate regressions
3	
4	A.T.J. de Laat, R.J. van der A, M. van Weele
5	
6	Royal Netherlands Meteorological Institute, De Bilt, The Netherlands.
7	
8	

9 Abstract

10

This study presents a sensitivity analysis of <u>multi-multi</u>variate regressions of recent
springtime Antarctic vortex ozone trends using a "big data" ensemble approach.

Our results indicate that the poleward heat flux (Eliassen-Palm Flux) and the effective chlorine loading explain, respectively, most of the short-term and long-term variability in different Antarctic springtime total ozone records. The inclusion in the regression of stratospheric volcanic aerosols, solar variability and the Quasi-Biennial Oscillation is shown to increase rather than to decrease the overall uncertainty in the attribution of Antarctic springtime ozone because of large uncertainties in their respective records.

19 Calculating the trend significance for the ozone record from the late 1990s onwards 20 solely based on the fit of the effective chlorine loading is not recommended, as this does 21 not take fit residuals into account resulting in too narrow uncertainty intervals, while the 22 fixed temporal change of the effective chlorine loading does not allow for any flexibility 23 in the trends.

24 When taking fit residuals into account into a piecewise linear trend fit, we find that 25 approximately 30-60% of the regressions in the full ensemble result in a statistically 26 significant positive springtime ozone trend over Antarctica from the late 1990s 27 onwardstto 2010 or 2012. Analysis of choices and uncertainties in time series show that, 28 depending on choices in time series and parameters, the fraction of statistically significant 29 trends in parts of the ensemble can range from negligible to a complete 100%. We also 30 find that, consistent with expectations, the number of statistically significant trends 31 increases with increasing record length.

32	Although our results indicate that the use <u>multi-multi</u> variate regressions is a valid
33	approach for assessing the state of Antarctic ozone hole recovery, and it can be expected
34	with increasing record length results will move towards more confidence in recovery,
35	uncertainties in parameters and independent variables and choices in uncertainties in
36	choices defining the optimal time period and area for calculating the ozone record and the
37	independent variables currently do not yet support formal identification of the onset of
38	recovery of the Antarctic Ozone Hole.

40 **1. Introduction**

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An important question in 21st century ozone research is whether the ozone layer is starting to recover as a result of the measures taken to reduce emissions of Ozone Depleting Substances (ODS) as agreed on in the Montreal Protocol [UNEP, 2012] and its subsequent amendments and adjustments.

46 The World Meteorological Organization has defined three different stages of ozone recovery [WMO, 2007]. The first stage consists of a slowing of ozone depletion, 47 48 identified as the occurrence of a statistically significant reduction in the rate of decline in 49 ozone due to changing stratospheric halogens. The second stage revolves around the 50 onset of ozone increase (turnaround), identified as the occurrence of statistically 51 significant increases in ozone - above a previous minimum value - that can be attributed 52 to declining stratospheric halogens. Note that what is meant by "statistically significant" 53 is not specified. Finally, the third stage is the full recovery of ozone from ODSs, 54 identified as when the ozone layer is no longer affected by ODSs, or alternatively, once 55 stratospheric ozone levels have returned to pre-1980 values.

The first stage of ozone recovery has already been identified in observations to have occurred roughly in the late 1990s [WMO 2007, 2011]. The third stage is not expected to occur until somewhere halfway the 21st century or later [WMO, 2011]. The spatial distribution of total ozone after the third stage probably differs somewhat from the pre-1980 distribution due to climate change – in particular changes in the stratospheric chemical composition and temperature structure [Bekki et al., 2011, and references therein]. 63 As far as the second stage of ozone recovery is concerned, it has recently been argued 64 that a statistically significant increase in ozone beyond a minimum and attributable to 65 decreases in ODSs can be identified for the Antarctic ozone hole [Salby et al., 2011, 66 2012; Kuttippurath et al., 2013; Knibbe et al, 2014]. To some extent this is surprising as it 67 has long been thought that identification of the second stage of ozone recovery could only 68 be expected after 2020 [e.g. Newman et al., 2006; Eyring et al., 2007]. Those estimates 69 were based on (model) simulations of ozone from which it is calculated when the ozone 70 trend from a certain starting year onwards would qualify for "statistically significant", or 71 in other words, would emerge from the year-to-year natural variations in ozone ("noise"). 72 Such methods implicitly assume Both studies do not consider the effect of deterministic 73 variations in ozone on estimating when the onset and complete recovery of the Antarctic 74 ozone hole recovery occur. that ozone variations around the trend are not deterministic 75 (random).

76 However, it has also long been established that many stratospheric ozone variations are 77 in fact deterministic. Various processes have been identified that affect stratospheric 78 ozone variability in the Southern Hemisphere on an inter annual interannual basis, like 79 volcanic aerosols [Telford et al., 2009], the Southern Annular Mode (SAM) [Thompson 80 and Wallace, 2000; Jiang et al., 2008], the poleward heat flux or Eliassen-Palm flux (EP 81 flux) [Randel et al., 2002], solar variability [Soukharev and Hood, 2006], and the Quasi 82 Biennial Oscillation (QBO) [Jiang et al., 2008]. If the physics and chemistry are 83 sufficiently understood, it might be possible to filter out part of the ozone variations from 84 the ozone records by means of a multi-variaetey regression, resulting in a smoother ozone 85 record for which trend significance might be reached earlier. This approach, in essence, forms the basis of the suggested identification of the second stage of ozone recovery reported by Salby et al. [2011, 2012], Kuttippurath et al. [2013] and Knibbe et al [2014]. However, none of these studies did systematically consider the uncertainties in the proxies that were selected for the regressions. In addition, no motivation or discussion was provided for the choice of a specific ozone record, *e.g.* a consideration of taking annual, seasonal, and/or monthly means of total ozone, and the integration over a chosen spatial domain.

93 Hence, we want to address the following question in this study: Is the suggested 94 detection of the second stage of ozone recovery robust when uncertainties in the 95 regression parameters and for different selected ozone records are taken into account? This question is approached here with combined multiple scenario – Monte Carlo 96 97 ensemble simulations using the same regression methodology as presented in Kuttippurath et al. [2013] but by inclusion of various uncertainties leading to a large 98 99 ensemble of different regressions. We analyze this "big data" ensemble for robustness of 100 the individual regressions.

101 Kuttippurath et al. [2013] considered different Antarctic vortex definitions and thus 102 different vortex ozone records. They found that regression results were not very sensitive 103 to the Antarctic vortex definition. Hence, we decided to use September-November 104 Antarctic vortex core (poleward of 70°S) average total ozone column based on the Multi 105 Sensor Reanalysis (MSR; van der A et al. [2010]), also because from a practical point of 106 view this definition does not require additional information about the location of the 107 vortex edge. The selected regressors are the SAM, solar flux, QBO, EP flux, stratospheric 108 volcanic aerosols and the Equivalent Effective Stratospheric Chlorine (EESC), similar to 109 Kuttippurath et al. [2013]. The EESC can be used to estimate ozone trends. Kuttippurath 110 et al. [2013] also calculated Piece Wise Linear Trends (PWLT) for estimating ozone 111 trends as alternative for the EESC-based ozone trends, an approach we will follow here as 112 well.

113 In this paper, we extend the analysis by introducing both several differing scenarios for 114 the ozone record and regressor records of the EP flux, volcanic aerosols, and EESC. 115 Monte Carlo variations were applied to the regressor records of the solar flux, QBO, 116 SAM by adding random variations. While we focus on parameter uncertainties in this 117 study, additional uncertainties do exist, for example with respect to possible time lags 118 between regressors and the ozone record. The resulting ensemble of regression results 119 provides a big data pool of about 23 million different regressions that is analyzed in terms 120 of probability distributions of the explanatory power of the regressions (\mathbb{R}^2), the ozone 121 trends and corresponding ozone trend uncertainties, and the regression coefficient values 122 quantifying the dependence of ozone on a particular regressor. We also investigate if 123 some way of optimization is possible for the chosen scenarios, and we discuss the 124 likelihood of detection of the second stage of ozone recovery within the context of all 125 uncertainties presented. Note that the uncertainties discussed here differ from formal 126 errors that come with a standard multi-multivariate regression. Also note that we 127 implicitly assume that the relation between the independent variables and ozone is linear, 128 even though the relation may very well be non-linear. The latter will to some extent be 129 considered in our study and is part of the discussion of the results, but the issue of non-130 linearity of the regressor-ozone relation is not addressed in detail, in particular because, as will be shown, for many regressors the non-linearity of its relation with ozone isinsufficiently characterized, or even unknown.

This paper is organized as follows. Section 2 describes the observational datasets used and the ozone and regressor scenarios or Monte Carlo simulations performed. Section 3 discusses the probability distributions of the explanatory power of the regressions, trends and regression values, including how the distributions depend on scenarios or Monte Carlo results. Section 4 discusses the question of detection of the second stage of ozone recovery, and in section 5 everything is wrapped up and some conclusions are drawn.

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140 **2. Multivariate regression parameter uncertainties**

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142 Online data sources of the ozone observation records and applied regressors can be 143 found in Table 1.

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145 **2.1 Method**

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A common method for analyzing total ozone records is the use of a multi-multivariate linear regression, a method that we will use in this paper as well. The goal of the method is to attribute both inter annualinterannual as well as decadal variations in the ozone record to processes that are expected or known to affect the total ozone record (Kuttippurath et al. [2013], and references therein). In the regression, the total ozone variability (Y) as a function of time (t) is expressed as

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154	Y(t) = K	(Constant)
155	$+ C_1 \text{HF}(t)$	(Poleward Heat Flux or Eliassen-Palm (EP) flux)
156	$+ C_2$ SAM (t)	(Southern Annual Mode index)
157	+ $C_3(SF \times QBO)(t)$	(Solar Flux \times QBO index)
158	$+ C_4 \operatorname{Aer}(t)$	(Stratospheric Aerosol optical depth)
159	+ C_5 Trend(t)	(Total ozone trend)
160	$+ \epsilon(t)$	(Total ozone residual)

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In which *t* is the time from 1979 to 2010 or 2012, K is a constant and regression coefficients C_1 to C_5 are the regression coefficients for the respective proxies. The ozone trend (C_5) can be related to the time-dependent equivalent effective stratospheric chlorine loading (EESC) or a piecewise linear trend (PWLT) before and after a predefined break year. The PWLT regressions are calculated by including two linear terms in the regression: the first term is a linear fit for the entire time window, the second term is a linear term only for the years after a chosen break year [Kuttippurath et al., 2013].

169 The analysis of regression results will focus on two parameters that have previously 170 been used in papers investigating Antarctic ozone recovery [Yang et al., 2008; Salby et 171 al., 2011, 2012; Kuttippurath et al., 2013; Knibbe et al., 2014]: the serial correlation R 172 between the regression-based 'reconstructed' ozone record and the observations, and the 173 post-break trends and trend significance. Since the focus of our paper is to investigate 174 trend significance, not specifically what parameters can best explain Antarctic ozone, we 175 will only look in some detail at the usefulness of certain regressors. However, our 176 analysis does provide indications of what are more and less useful regressors.

In sections 2.2 to 2.7 the uncertainty in each of the proxies that is used as a regressor is discussed. These uncertainty ranges determine the spread in the ensemble that is used in the "big data" analysis. A summary of the regressor uncertainties and how they are incorporated in this study can be found in Table 2. The solar flux and QBO are combined into one proxy as discussed in section 2.3.

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183 **2.2 Poleward heat flux (EP flux)**

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185 Figure 1 shows the poleward heat flux, here represented by the (vertical) EP flux 186 [Andrews et al., 1987] at the 70-hPa level and averaged poleward of 40°S for the 187 combined months of August and September, as well as the average EP flux available for 188 a given year for a variety of datasets. Note that the datasets do not all completely overlap 189 in time. Before 2000 there are considerable differences between the datasets. After 2000 190 these differences are smaller, which to some extent is traced to the lack of ERA40 data 191 beyond 2001 and lack of JRA data beyond 2004. The lower panel shows the relative 192 differences between the five datasets and their mean. The standard deviation of all data is 193 7.65%, but from 2000 onwards only 2.67%.

Another source of uncertainty in the use of the EP flux as proxy is the choice of the time window over which the average EP flux is calculated. This choice depends on what is thought to be the relationship between variations in EP flux and ozone depletion. The basic theory states that the poleward movement of stratospheric air is proportional to the strength of the residual mean stratospheric circulation (Brewer-Dobson circulation), which in turn is driven by the poleward eddy heat flux. The poleward eddy heat flux is 200 expressed by the upward component of the Eliassen-Palm flux that measures the upward 201 transport of momentum by planetary waves [Andrews et al., 1987; Salby et al., 2012, and 202 references therein]. Planetary wave activity in the Northern Hemisphere affects Arctic 203 Polar vortex stability and thus Arctic ozone depletion. However, to what extent this is 204 similar in the Southern Hemisphere is still a topic of debate. The Arctic and Antarctic 205 may behave either very similarly [Weber et al., 2003; 2011] or not-much less similar 206 [Salby et al., 2012]. This is because the notion of hemispheric similarities in how the EP 207 flux affects ozone depletion so far is-may be heavily-biased on-by only one a few outlier 208 years (2002 and 2006 for the SH, -1996, 2010 and 2011 for the NH).

209 Current research efforts try to gain a better understanding of the physical and 210 photochemical mechanisms by which the heat flux and planetary wave action affects 211 Antarctic stratospheric ozone. A recently proposed mechanism [de Laat and van Weele, 212 2011] involves a pre-conditioning of Antarctic inner stratospheric vortex air whereby 213 stratospheric temperatures affect PSC formation which in turn affects the buildup of a 214 halogen reservoir that later during Austral spring changes the rate of catalytic ozone 215 destruction. This preconditioning mechanism explains some years with anomalous ozone 216 depletion, but not all. For example, during Austral winter 2013 the Antarctic vortex 217 remained largely undisturbed – opposite to 2010 and 2012, see de Laat and van Weele 218 [2011] and Klekociuck et al. [2011], thus allowing for widespread PSC formation and 219 pre-conditioning the inner vortex air for efficient ozone depletion. However, from the 220 start of Austral spring 2013 (halfway August) onwards the Antarctic stratospheric vortex 221 got disturbed by planetary wave activity. As a result, the amount of springtime ozone 222 depletion remained below what has been experienced during previous years with similar preconditioning. This suggests that there are multiple pathways as well as complicated interactions between chemistry and physics that can lead to reduced Antarctic springtime ozone depletion. Hence, it is unclear which regressor or regressors could act as proxies for these complex processes.

A further complicating factor is the disintegration of the Antarctic vortex, which is again controlled by planetary wave activity [Kramarova et al., 2014]. The stability of the vortex determines how long the ozone depleted inner-vortex air remains intact after photochemical ozone depletion ceases during Austral spring. Variations in the duration of Antarctic vortex stability introduce variations in the Antarctic total ozone record which are not related to variations in photochemistry.

233 We attempt to reflect these issues in our uncertainty range for the proxy used to account 234 for the EP flux variations in multivariate regressions. Salby et al. [2011, 2012] and 235 Kuttippurath et al. [2013] use the August – September mean EP flux poleward of 40°S 236 and at the 70-hPa level, the baseline also used in this study. Weber et al. [2011] uses the 237 100-hPa poleward heat flux rather than the 70-hPa heat flux and the average over the 238 region between 45°S and 75°S rather than between 40°S and 90°S. They further show that 239 there is no particular favorable wintertime month or period from the perspective of 240 Antarctic springtime ozone depletion over which to average the EP flux. Hence there is a 241 certain arbitrariness involved in selecting the optimum EP flux averaging period and 242 region.

For our study we define eight different EP flux scenarios, using different periods, latitudes and heights (see Table 2), all based on the ECMWF ERA Interim dataset. Performing the same exercise as in Figure 1 for these eight scenarios, the standard deviation of the EP flux time series is 21.5%. This is considerably larger than the
variability among the same EP fluxes of the different reanalysis datasets discussed above.
Thus, the uncertainty in EP flux estimates largely originates in using different periods,
latitudes and/or heights for which the EP flux is calculated, rather than in the use of
different reanalysis datasets to calculate the same EP flux.

- 251
- 252
- 253 2.3 The mixed solar-QBO index
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255 In Kuttippurath et al. [2013] the effects of solar variability and QBO variability are 256 combined into one proxy. As explained in Holton and Tan [1990], in studying high-257 latitude variability and trends the QBO and solar effects cannot be considered separately. Whereas the solar influence modifies tropical stratospheric ozone and dynamics, the 258 259 transport of the solar signal to higher/polar latitudes depends on the phase of the QBO. 260 As a result, solar effects on winter polar Antarctic stratospheric temperatures also depend 261 on the phase of the QBO [Labitzke, 2004]. If the QBO is westerly (easterly), stratospheric 262 temperatures vary in phase (out of phase) with solar activity. It has been proposed by 263 Haigh and Roscoe [2006] and Roscoe and Haigh [2007] to combine the QBO and solar 264 activity into a new regression index that takes this effect into account:

Solar – QBO index =
$$(Solar - S_m) \times (QBO - Q_m)$$

In which S_m is the mean of the solar flux and Q_m the midpoint of the QBO range. However, as Roscoe and Haigh [2007] note, this new index is rather sensitive to the choice of S_m and Q_m , in particular as the index is by construction the product of two 269 anomaly fields, and thus sensitive to sign changes. In addition, the choice of S_m and Q_m is 270 also arbitrary. Roscoe and Haigh [2007] solve this by selecting averages for which the 271 best total ozone column regression results are obtained. However, the best regression 272 results may not necessarily mean that the regressor is the best representation of the 273 underlying physical mechanism, in particular as regression results also depend on other 274 proxies and in principle there can be a cancellation of errors from different proxies in the 275 regression. Thus, the sensitivity of the combined solar-QBO index on the calculation 276 method of the anomalies must be further investigated.

277 Figure 2 shows the resulting solar flux - QBO index time series, given various 278 assumptions in its calculation. Clearly there is a considerable variability in the index 279 values. The lower plot shows that the variability for every single anomaly varies by \pm 280 200%. This is rather large compared to the estimated uncertainties in both individual 281 solar flux and QBO proxies. Hence, using a combined solar flux – QBO proxy introduces 282 a considerable amount of additional uncertainty. For the uncertainty range in our 283 regressions we construct 100 Monte Carlo time series in which for each single solar-flux 284 QBO index value random Gaussian noise is added with an amplitude of 200% of the 285 index value.

Note that the uncertainties in the individual QBO and solar flux proxies are much smaller than the uncertainty in the combined solar flux – QBO index which is relevant for high-latitude trends (see supplementary information for a separate discussion of the solar flux and QBO index).

290

291 **2.4 Southern Annular Mode**

The Southern Annular Mode (SAM) is a widely used index that reflects the zonal symmetry of the tropospheric circulation in the Southern Hemisphere. The symmetry of the Southern Hemisphere circulation has long been identified as an important mode of variability of the Southern Hemisphere climate. A positive index is characterized by anomalously high surface pressure at mid-latitudes and anomalously low surface pressure at latitudes closer to Antarctica.

299 The SAM used in this study is derived from the National Oceanic and Atmospheric 300 Administration (NOAA). It is based on Empirical Orthogonal Functions (EOF) applied to 301 the monthly mean National Centers for Environmental Prediction and National Center for 302 Atmospheric Research (NCEP/NCAR) reanalysis [Kalnay et al., 1996] 700-hPa height 303 anomalies poleward of 20°S for the Southern Hemisphere, with the seasonal cycle being 304 removed. The monthly SAM index is constructed by projecting the daily and monthly 305 mean 700-hPa height anomalies onto the leading EOF mode. Both time series are 306 normalized by the standard deviation of the monthly index (1979-2000 base time period). 307 Since the leading pattern of SAM is obtained using the monthly mean height anomaly 308 dataset, the index corresponding to each loading pattern becomes one when it is 309 normalized by the standard deviation of the monthly index.

However, there is no unique SAM index due to the existence of different meteorological datasets and different methods to quantify the symmetry of the Southern Hemisphere circulation. Kuttippurath et al. [2013] use the AntArctic Oscillation (AAO) index, which is in fact a certain choice of SAM index. A study by Ho et al. [2012] provides a comprehensive analysis of eight different SAM indices. Their analysis shows that the correlation (\mathbb{R}^2) between the indices varies between 0.45 and 0.96 for seasonal values and 0.73 and 0.96 for monthly values. This corresponds with random (Gaussian) variations between 20-100% (root-mean-square value). For most of the indices the correlation is better than 0.75. As a point of reference, adding random Gaussian noise of 50% to a time series of a parameter and calculating its correlation with the original time series results to a correlation (\mathbb{R}^2) of almost 0.8.

For the uncertainty analysis we construct 100 Monte Carlo time series in which for each
single SAM index value Gaussian noise is added with – to be on the conservative side an amplitude of 100% of the index value.

324

325 **2.5 EESC loading**

326

327 Uncertainties in the estimates of the EESC loading originate from two factors: the mean 328 age-of-air, which reflects how fast stratospheric halogen concentrations decline due to 329 transport velocity of halogen poor tropospheric air from the tropical stratosphere to the 330 polar stratosphere, and the so-called 'fractional release', the rate with which Ozone 331 Depleting Substances (ODSs) release chlorine and bromine in the stratosphere. ODSs 332 typically have not yet been dissociated when they enter the stratosphere at the tropical 333 tropopause, and thus have fractional release values of zero. After transiting through the 334 upper stratosphere, the ODSs in an air parcel get fully dissociated due to their exposure to 335 energetic radiation and the fractional release values get close to 1.0 [Newman et al., 336 2007].

337 To complicate matters, the mean age-of-air in the stratosphere is not a constant but 338 varies with latitude, height and season [Stiller et al., 2008]. On average, the age-of-air 339 increases with height, *i.e.* it takes longer for tropospheric air to travel higher in the 340 stratosphere, and the age-of-air also increases towards the poles because of the time it 341 takes for air to travel from the tropical "source" region to higher latitudes. In the 342 Antarctic vortex regions there is a strong seasonal dependence of the age-of-air due to the 343 isolation of inner vortex air during Austral winter and spring, while upper stratospheric 344 and mesospheric air slowly descends in the Antarctic vortex. The descending air is 345 particularly "old" air and causes strong vertical gradients in the age-of air in the 346 wintertime polar vortex. Stiller et al. [2008; their figure 7] show that the age-of-air almost 347 triples going up from 15 km (θ = 400 K; age-of-air ~ 4 years), to 20 km (θ = 400 K; age-348 of air ~ 7 years), to 25 km (θ = 600 K; age-of-air ~ 9 years), to finally 30 km (θ = 750 K; 349 age-of-air ~ 11 years). How to account for this variability in a regression is unclear, but it 350 is unlikely that one age-of-air value can be attributed to the total ozone column.

Moreover, ozone variability in the Antarctic vortex is determined by different processes at different altitudes. Halogen related ozone depletion typically maximizes between 15-20 km altitude (~ 100-50 hPa, US Standard atmosphere 1976; $\theta = 400-500$ K), whereas the effect of vortex stability on ozone depletion is seen predominantly between 20-30 km altitude (~50-10 hPa; $\theta = 500-750$ K) [de Laat and van Weele, 2011]. Thus, total ozone columns observations which are vertically integrated amounts of ozone are being affected by different processes at different altitudes.

The age-of-air may also not be constant over the time period over which ozone trends are determined. Due to a changing climate the stratospheric circulation may speed up [*e.g.* Engel et al., 2009; Bunzel and Schmidt, 2013], causing a decrease in the age-of-air
with increased warming, which obviously then depends on the exact warming. This
introduces yet another uncertainty for the periods from 1979 to 2010 or 2012 that are
considered in this study.

The age-of-air uncertainties do not manifest themselves as a random process, which would make it useful for applying a Monte Carlo method, but as a structural uncertainty, *i.e.* the entire EESC shape would change for different parameter settings. Such uncertainty could be captured by applying a parametric bootstrap rather than a Monte Carlo approach. However, such parametric approach would also not suffice because we use total column observations and we know that ozone at different altitudes would be affected by different parameter values.

A pragmatic approach with regard to the sensitivity of the regression to EESC values is testing the robustness of the regression results as a function of the assumed EESC time evolution. For the uncertainty analysis we assume three different EESC scenarios with an age-of-air of 2.5, 4 and 5.5 years and a half-width of, respectively, 1.25, 2 and 2.75 years. Largest differences between the three scenarios are in their post-peak trend in EESC (see later on in Figure 3).

377

378 **2.6 Volcanic aerosol.**

379

380 Aerosols from sufficiently strong volcanic eruptions can reach the stratosphere and 381 affect stratospheric ozone chemistry. In particular strong eruptions occurring in the 382 tropics can have long lasting effects on stratospheric ozone. Aerosols reaching the tropical stratosphere are slowly transported towards middle and high latitudes. It can take up to a decade before the stratosphere is cleared from volcanic aerosols [Vernier et al. 2011; Solomon et al., 2011]. Volcanic eruptions at middle and high latitudes have much shorter lasting effects. These aerosols enter in the descending branch of the stratospheric circulation and will be relatively quickly removed from the stratosphere.

388 The short-term effect of stratospheric volcanic aerosols is heating of the stratospheric 389 layer which affects stratospheric ozone in the tropical belt. The dominant long-term effect 390 of stratospheric volcanic aerosols on global and polar ozone is however the increase in 391 aerosol surface area density and subsequent heterogeneous ozone loss. Model simulations 392 of volcanic aerosol effects on stratospheric ozone suggest that in particular under cold 393 conditions (high latitude, wintertime, lower stratosphere) total ozone columns can be 394 reduced by up to 10-15 % [Rozanov et al., 2002]. During other seasons, total ozone 395 column depletion by volcanic aerosols is of the order of a few percent.

396 Since 1979 two major tropical volcanic eruptions have affected stratospheric ozone: El 397 Chichón, Mexico, in 1982, and Pinatubo, Philippines, in 1991. Although the total amount 398 of stratospheric aerosols by both eruptions has been characterized relatively well, there 399 appear to be considerable uncertainties associated with the time evolution of the aerosol 400 amounts in the Southern Hemisphere. A brief and incomplete survey of a latitudinal 401 volcanic aerosol radiative forcing data [Ammann et al., 2003] and a global volcanic 402 aerosol proxy record [Crowley and Unterman, 2012] as well as the standard volcanic 403 aerosol index used in Kuttippurath et al. [2013] – aerosol optical depth, Sato et al. [1993] 404 and updates, available via NASA GISS - all show that there are large differences 405 between the El Chichón aerosol peak relative to the Pinatubo peak. Large differences are 406 seen in global, hemispheric and Southern Hemisphere (Antarctic) aerosol amounts as 407 well as differences in the exact timing of the peak aerosols [Sato et al., 1993; Ammann et 408 al., 2003]; Crowley and Unterman, 2012]. The El Chichón aerosol peak relative to the 409 Pinatubo peak for high Antarctic latitudes can be similar [Ammann et al., 2003], about 410 three times smaller [Sato et al., 1993] to (globally) eight times smaller [Crowley and 411 Unterman, 2012]. The Pinatubo peak aerosol in the Southern Hemisphere was about half 412 the size of the global-mean Pinatubo peak [Ammann et al., 2003].

Kuttippurath et al. [2013] shift the Southern Hemisphere aerosol data by six months to account for the transport of aerosols. Although they report that the six month shift results in the best statistics, the analysis presented in the previous paragraph shows that the effect of the shift is relevant for the shape of the volcanic aerosol changes, but does not introduce variations as large as the other variations in volcanic aerosol indices. Given that a time shift is included in the 6 volcanic aerosol scenarios defined above, we do not add additional time shifts in the aerosol record.

420 We define six volcanic aerosol scenarios that reflect the uncertainty in the volcanic 421 stratospheric aerosol records. Base scenario is the scenario used in Kuttippurath et al. 422 [2013] which in turn uses the NASA GISS stratospheric aerosol record. A second 423 scenario is with the Pinatubo aerosol curve scaled so that the maximum matches the El 424 Chichón aerosol peak, the Pinatubo curve maximum is 2.5 times the El Chichón peak, 425 and the Pinatubo curve maximum is five times the El Chichón peak. The uncertainty in 426 timing of the Southern Hemispheric aerosol peak is considered by a shift of the El 427 Chichón peak one year back compared to the Pinatubo peak and a shift of the Pinatubo 428 peak one year back compared to El Chichón peak.

429

430 **2.7 Ozone Scenarios.**

431

432 It is *a priori* unclear what would be the most appropriate ozone scenario to use in the 433 regression. Both Salby et al. [2011, 2012] and Kuttippurath et al. [2013] use the 434 September-November three-month averaged total ozone record. However, as discussed in 435 the introduction, different processes affect ozone during different time periods. Studies in 436 the literature use very different time periods for averaging ozone to investigate Antarctic 437 ozone trends. We define eight different ozone scenarios to reflect the ozone records used 438 in literature (see also de Laat and van Weele [2011]), using the MSR dataset [van der A 439 et al., 2010]. The MSR is a 30-year total O₃ column assimilation dataset for 1979–2008 440 based on a total of eleven satellite instruments measuring total O_3 columns – including 441 SCIAMACHY - that were operating during various periods within these 30 years. For the period 2009-2012 the MSR dataset was extended with assimilated SCIAMACHY and 442 443 GOME-2 total ozone column data. Apart from the September-November three-month 444 averaged total ozone record we also use averages of total ozone over the month of 445 September, the month of October, the two-month period September-October, a very long 446 period (19 July -1 December), a very short 10-day period (21 -30 September), the 447 period 7 September – 13 October, and a year-dependent "worst" 30-day period (30-day 448 average with the largest Ozone Mass Deficit).

449

- 450 **2.8 Other uncertainties**
- 451

452 Kuttippurath et al. [2013] address two other important uncertainties for the 453 determination of the ozone trend. First, the area over which the ozone record is defined 454 (Inside Vortex, Equivalent Latitude 65°S-90°S, and Vortex Core). The area is important 455 for the absolute amounts of ozone depletion but Kuttippurath et al. [2013] show it is 456 much less relevant for the differences in trend. That is, the uncertainties in the estimated 457 linear trend dominate the uncertainties due to different areas over which the ozone 458 anomalies are calculated. A second uncertainty on their ozone trend derives from the use 459 of different ozone datasets (ground-based, TOMS/OMI and MSR). Also here the 460 uncertainties in the estimated linear trend dominate the uncertainties due to the different 461 data sets. Hence, we do not include these uncertainties in our analysis.

In addition, there are many studies trying to identify the moment where ODSs stop increasing and/or where ozone stops decreasing. The maximum ODSs appears somewhere between 1997 and 2000 (Newman et al., 2007), depending on geographical location and height. However, due to saturation effects – there are more than sufficient ODS present to destroy all Antarctic ozone – the moment where ozone starts to be affected by decreasing ODSs may actually be later (Kuttippurath et al., 2013; Kramarova et al., 2014).

The moment of a structural break in ozone based on observations indicates an early break around 1997 (Newchurch et al., 2003; Yang et al., 2008). However, some processes affecting stratospheric ozone vary on long time scales – solar effects and volcanic eruptions come to mind – which may affect the observations-based analysis of break points (Dameris et al., 2006). Note that we confirm this break year of 1997 based on a applying a break-point analysis algorithm to the MSR ozone record (not shown). Hence, we decided to use three different break years that have been identified and/or are mostcommonly used: 1997, 1998 and 1999.

477

478 **2.9 Selected uncertainties ranges and ozone record scenarios.**

479

480 Figure 3 shows the baseline regressor time series and the scenarios for ozone, the EP flux, 481 EESC loading and volcanic aerosols. A total of 100 different solar flux - QBO index and 482 SAM index time series are used to span their uncertainty range (not shown in Figure 3). 483 All scenarios and Monte Carlo results combined provide 11.5 million different choices 484 for the regressions $(100 \times 100 \times 8 \times 8 \times 6 \times 3)$; see Table 2). Ozone trends are calculated based 485 on the EESC loading or using a piecewise linear trend (PWLT) analysis. For the PWLT 486 ensembles the three different EESC scenarios are irrelevant. Instead, the sensitivity of the 487 regressions is tested using three different break years (1997, 1998 and 1999). In total we 488 analyze approximately 23 million 000-different trends using the EESC and PWLT 489 scenarios.

490 Note that by basing our analysis on both different ozone and EP flux scenarios certain 491 time-lag relations are taken into account. It should also be noted that the use of such a 492 wide range of scenarios indicates that much remains unclear about what best describes 493 Antarctic ozone depletion and the time-lag relations between ozone and explanatory 494 variables.

495

496 **3 Scenario analysis**

497

498 **3.1 Reproducing Kuttippurath et al., [2013].**

499

500 First a multi-multivariate regression is performed similar to Kuttippurath et al. [2013] 501 in which the MSR dataset is used within the Vortex core (70°-90°S). The results are 502 summarized in their Figure 5 and Table 4 which are duplicated here in Table 3 along with 503 the results from a multi-multivariate regression based on the same variables as used in 504 Kuttippurath et al. [2013].

Our results reproduce the results from Kuttippurath et al. [2013], although there are 505 506 minor differences in the absolute numbers, most likely related to differences in EP fluxes 507 [Jayanarayanan Kuttippurath, *personal communication*, September 2013]. The trends for 508 the periods 1979-1999 and for 2000-2010 are of comparable magnitude in both studies, 509 as well as the PWLT significance levels for the period 1979-1999 and the EESC trends 510 for both 1979-1999 and 2000-2010. The magnitude of the recovery for 2000-2010 based 511 on the PWLT is slightly larger, but also in our analysis the post 2010 linear trend in 512 ozone is significant beyond 2σ . For the correlation of the regression model with the ozone record we obtain a value of 0.87 (R^2) comparable to the 0.90 (R^2) reported in 513 514 Kuttippurath et al., [2013]. Thus, the results are sufficiently similar to proceed with 515 studying the effects of the uncertainties in regressors and ozone record scenarios on the 516 regression results. Note that there is slight difference in the 1979-1999 trends for the 517 period ending in 2010 and 2012 because of the difference in total record length, which 518 results in slightly different regression coefficients. Note that we calculate the pre break 519 and post-break EESC-based trends by applying linear regressions to the EESC curve 520 multiplied with the EESC regression coefficient for the pre-break and post-break time 521 periods. As a result, EESC-based trend errors are related to the non-linearity of the EESC
522 curve, and the trend errors differ for both the pre-break and post-break time periods. Our
523 EESC-based trend errors differ from those in Kuttippurath et al (2013), which lacks a
524 description of how EESC-based trend errors are calculated.

- 525 However, calculating trend errors for the EESC-based pre-BREAK and post-BREAK
- 526 trends in ozone using the EESC regression error as done in Kuttippurath et al. (2013) is
- 527 <u>not justified. The trend errors depend on the actual trend values themselves (Table 3): the</u>
- 528 EESC-fit based post-break trend error is much smaller than the pre-BREAK trend. In the
- 529 <u>hypothetical case of no (zero) trend the trend error would also be zero, which would be</u>
- 530 physically unrealistic. The PWLT on the other hand shows hand shows opposite
 531 differences in trend errors: the post-BREAK trend error is much larger than the pre-
- 532 BREAK trend error, conform expectations.
- 533

534 **3.2 Ozone record and regressor correlations.**

535

Before analyzing the ensemble of regression results it is important to investigate the correlations between the different regressors. If correlations between regressors are too large, they cannot be considered to be independent, and it should be decided which one to omit from the analysis, as the regression otherwise cannot separate which variability is related to which regressor. Furthermore, it is *a priori* useful to understand how regressors correlate with the ozone record, as a small correlation implies that a regressor can only explain a limited amount of ozone variability. 543 Table 4 shows the mean correlation between the different regressors and their 2σ spread 544 based on the ozone record and regressor selections and/or Monte Carlo results (SAM, 545 SF×QBO index). The EP flux correlates positively with the EESC and negatively with 546 the SAM and, to a lesser extent, also with the SF×QBO index. The other regressors do 547 not show significant cross-correlations. Only for a few individual ozone record scenarios, 548 regressor selections and Monte Carlo results cross-correlations are found to exceed 0.5. 549 The uncertainty in the correlations with the ozone records ranges between about 10% 550 and 20% for each of the regressors. Small cross-correlations between the regressors 551 however do not provide a justification for *a priori* omitting one of the regressors.

552

```
553 3.3 Trends.
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554

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Figure 4 shows the probability distributions of the ozone trends for $1979-Y_B$ and Y_B- 555 556 2012 periods, in which Y_B is the break year which can either be 1997, 1998 or 1999. For 557 the 1979-Y_B period the mean EESC trend is -5.56 DU/year (-4.00 to -7.06; 95% CI) and 558 the mean PWLT trend is -6.40 DU/year (-4.22 to -7.18; 95% CI). For the Y_B -2012 period 559 the mean EESC trend is ± 1.97 DU/year (± 0.84 to ± 3.32 DU/year; 95% CI), and the mean 560 PWLT trend is +3.18 DU/year (+1.66 to +4.74; 95% CI). 561 For the 1979-Y_B period the distributions of EESC and PWLT trends (top panel) are 562 rather similar, although the PWLT correlations show a larger peak towards high 563 correlations compared to the EESC correlations (bottom panel). However, for the Y_B-

565 trends show a tri-modal distribution, because only three different EESC curves were

2012 trends the probability distributions are very different (middle panel). The EESC

used. These three EESC curves differ predominantly in their post-1997 EESC trends (see also Figure 3). In addition, the tri-modal EESC trend probability distribution for Y_B -2012 (middle panel) shows that in the linear regression the EESC fit is determined by the 1979- Y_B period more than by the Y_B -2012 period, as the pre-break trend distribution does not show the same tri-modal shape. This is not surprising because the trends for the 1979- Y_B period are larger and cover a longer period than for Y_B -2012.

572 The correlations distributions (lower panel) are similar for the lowest and highest 573 correlations for both the EESC and PWLT regressions, but in the bulk of the distribution 574 the PWLT results in systematically higher correlations than the EESC regressions.

575 The upper two panels of Figure 4 also include the 1979-1999 and 2000-2012 PWLT 576 trends and 2σ errors as reported in Table <u>32</u>. The uncertainty range of the 2000-2012 577 PWLT trend in Table <u>32</u> and the range in Figure 4 are quite similar. However, the 578 uncertainty range of the 1979-1999 PWLT trend in Table <u>32</u> is considerably smaller. This 579 shows that uncertainties in the 1979-1999 ozone trends are larger than estimated by a 580 single regression estimated even though all 1979-1999 trends are statistically significant.

The auto-correlation of the ozone residuals is small (one-year lag values are approximately zero), indicating that the auto-correlation present in the ozone record (*e.g.* Fioletov and Shepherd, 2003; Vyushin et al., 2007) is related to some of the processes described by the regression parameters and are removed by the <u>multi-multi</u>variate regression. Auto-correlation thus does not have to be taken into account in the trend significance calculation.

587

3.4 Regression model performance: sensitivity to the independent variables

590 Sensitivities of the PWLT-based and EESC-based regressions to the ozone and EP flux 591 scenarios are shown in Figure 5. PWLT-based regressions show that the PWLT 592 distribution peak at high correlations is a consistent feature of different ozone records 593 (Sep-Nov, Sep-Oct, Sep, 7 Sep - 13 Oct, worst 30 days). Similarly, use of several different 594 EP fluxes also aligns with the PWLT correlation distribution peak, in particular the EP 595 flux scenarios that include both the August and September months. For ozone, 596 correlations get smaller for, respectively, the longest period (19 July -1 December), 597 shortest period (21-30 September) and October averages.

598 Figure 6 shows the probability distribution of volcanic aerosols for both the PWLT and 599 EESC regressions. Volcanic aerosols have little impact on the explanatory power of the 600 regression results, as already indicated by lack of correlation of this regressor with the 601 ozone record. The PWLT regression coefficient values show that the effect of volcanic 602 aerosols on total ozone can be either positive or negative, largely depending on the 603 assumed amount of Pinatubo aerosols relative to El Chichón aerosols, although the 604 distribution predominantly suggests positive regression values. The EESC regressions 605 show a similar sign dependence of total ozone on volcanic aerosol, but with no clear sign 606 of the regression value. None of other parameters (EP-FLUX flux scenario, Ozone 607 scenario) have a sign-dependent effect on the aerosol regression coefficient value for both 608 the EESC and PWLT scenarios. The strong sensitivity of the volcanic aerosol regression 609 value - including sign changes - to either aerosol or EESC scenario indicates that 610 including volcanic aerosols is not very important for the multivariate regression and 611 better should be excluded altogether from multi-multivariate regressions due to
612 insufficient information in the Antarctic total ozone record to constrain the total ozone –
613 volcanic aerosol relation.

For the solar flux – QBO index (Figure 7, panel A) we find no clear dependence of regression coefficient values on any of the scenarios or parameters. The probability distributions for both the EESC and PWLT regressions are very similar. Hence, like for volcanic aerosols, the solar-QBO parameter better should be excluded altogether from multi-multivariate regressions because the Antarctic ozone record also contains insufficient information to constrain the ozone – solar-QBO relation

The SAM regression coefficient values show a continuous random distribution while the overall dependence is predominantly negative (Figure 7, panel B). A positive phase of the SAM correlates with more ozone depletion than a negative phase of the SAM. This is a well-known two-way effect: tropospheric circulation changes affect Antarctic stratospheric ozone on the short term, while the long term changes in Antarctic ozone have affected the tropospheric circulation in the Southern Hemisphere [Kirtman et al.,

626 2013; IPCC AR5, Ch. 11, section 11.3.2.4.2 and references therein].-

627 -For the EP<u>-FLUX flux</u> the regressions show a positive dependence (Figure 7_{27} panel C) 628 and a similar distribution for both the EESC and PWLT regression.

629

630 **3.5 Optimal regressor and ozone record scenarios**

631

Based on the analysis of the entire ensemble presented here it might be possible to
choose an optimal set of regressors as well as an optimal ozone record scenario for
Antarctic ozone trend analysis. Volcanic aerosols (Figure 6), the QBO and the solar cycle

(Figure 7) are shown to have little effect on the regression and thus better should be
excluded. For the EP flux, it appears that including the months August and September
leads to a better fit (higher correlations; Tables <u>54</u> and <u>65</u>, Figure 5). For ozone, results
suggest that there is no clear optimal time-window over which to calculated average
ozone, but it appears that the period should not be too short, not be too long, and should
include September and preferably the first half of October (Tables <u>54</u> and <u>65</u>).

641 In addition, the use of three different EESC scenarios results in tri-modal distribution 642 features in several parameters (Figures 43, middle panel and 6, lower panel), suggesting 643 that care has to be taken with in particular the ozone trend values attributed to changes in 644 EESC²-s. Furthermore, the post-break trends are particularly sensitive to the choice in 645 EESC scenario (Figure 3). It could therefore be argued that using a PWLT for post-break 646 trend estimates is preferred over using the EESC-based post-break trend as its distribution 647 better reflects structural uncertainties in the regression and takes the regression residuals 648 into account for calculation of trend uncertainties.

Figure 8 illustrates what the best single regressions in the entire ensemble for all three regression models separately look like. The best EESC-regression correlation ($R^2 = 0.95$) was found for a case with Sep -Nov ozone, Jul-Aug EP flux and an EESC with an age-ofair of 4 years. For the best PWLT-regression correlation ($R^2 = 0.96$) these were the same with 1997 as optimal break year. Reason for the high explanatory power is that in all three cases the <u>specific</u>SAM anomalies align with strong ozone peaks whereas the solar flux – QBO index variations coincidentally align with the smaller ozone anomalies.

656

4. Discussion: Second stage of ozone recovery and trend significance.

659 Given the broad range of outcomes for the different types of regressions and regressors, 660 an important question is not only if ozone has started to increase after the late 1990s, but 661 also whether the trend is statistically significant and can be attributed to declining 662 stratospheric halogens, which is required by WMO for the second stage of ozone 663 recovery to be formally identified. Because the EESC curve-shape is prescribed, there is no degree of freedom allowing for different pre-break and post-break trends in the EESC 664 665 regression. As discussed in section 2, it is not clear *a priori* which EESC scenario is the 666 optimal choice or if it is even appropriate to use just a single EESC scenario. 667 Furthermore, as already discussed in section 3.1, the method for estimating the trend uncertainty of the post-BREAK trend for the EESC fit in Kuttippurath et al. [2013] is not 668 669 justified, and Hence, how to assign an overall-uncertainty to the EESC curve remains an 670 open question. Therefore, a better-more appropriate approach would be to investigate 671 whether the PWLT post-break trends are statistically significant as they use the ozone fit 672 residuals for their significance calculation.

Figure 9 shows the probability distribution of correlations (R^2) of the PWLT regression 673 674 models vs. ozone for the entire Monte Carlo dataset, as well as the fraction of post-break 675 PWLT trend estimates that are statistically significant (2σ) for both the periods ending in 676 2010 and 2012. This figure is comparable to Figure 4 (lower panel) and Figure 5, but 677 with larger correlation bins for visualization purposes. Results indicate that trends only 678 become statistically significant beyond a certain explanatory power of the regression 679 model. This is not surprising: only when ozone residuals after removing the regression 680 results are sufficiently small can the post-break trend become statistically significant. This automatically requires a high correlation between the ozone record and the selected regression model. The analysis here shows that statistically significant trends require a correlation (R^2) of at least approximately 0.60. Furthermore, only for high ozoneregression model correlations ($R^2 > 0.80$) the majority of trends become statistically significant. In addition, the level of significance for the statistically significant trends is in most cases still less than 3σ (not shown), indicating that a considerable amount of variability is not accounted for in the regression.

688 In section 3.5 the results of the ensemble were analyzed to determine optimal scenarios in terms of explanatory power (R^2) . However, the second stage of ozone recovery 689 690 requires also a statistically significant post-break year trend. We therefore analyzed the 691 percentage of statistically significant post-break trends in the ensemble for the PWLT-692 based regressions. We focus on the ozone record and EP flux scenarios as the 693 uncertainties associated with these two parameters are the most important ones, as 694 discussed before. Table 5 shows the percentage of regressions for each combination of 695 ozone record and EP flux scenarios that is statistically significant for the ozone records 696 ending in 2010. There are large differences in the fraction of statistically significant 697 PWLT-based trends, ranging from less than 0.1% (21-30 September average ozone) to a 698 complete 100% significance (September-October and October ozone, 45S-75S Aug-Sep 699 EP flux). Table 6 shows the same results as Table 5, but only for the break year 1997 and 700 the period ending in 2012. In this case there is a large number of ozone record - EP flux 701 scenario combinations with statistically significant trends. If we would consider only the 702 EP fluxes that include the August and September months, then with the exception of the 703 21-30 September time window nearly all trends are statistically significant.

704	Excluding the year 2002 from the regressions has a significant impact on the post-break
705	ordinary linear ozone trends without applying a multi-multivariate regression. However,
706	it hardly has any effect on the post-break ordinary linear trends when including the
707	ordinary linear trend in the multi-multivariate regressions (not shown), indicating that the
708	multi-multivariate regression effectively removes the anomalous year 2002. Excluding
709	volcanic years from the regression had no significant effect on both the ozone trends
710	before and after the regression, consistent with our finding that there appears to be little
711	(direct) impact of volcanic aerosols on Antarctic springtime total ozone. Note that
712	Solomon et al. [2005] showed that volcanic effects can be seen in Antarctic ozone
713	profiles during the ozone hole season - in particular in the UTLS region, but that the
714	magnitude and vertical extent of the effects are too small to be detectable in total ozone
715	column variations.
716	Table 7 shows the number of significant trends as function of the length of the period
717	over which the trend is calculated. The number of significant trends varies between
718	approximately 30-60%, and that the number of significant trends further depends on the
719	choice of break year, with an overall increase in the number of statistically significant
720	trends increasing steadily with increasing length of the period over with trends are
721	calculatedThis is not surprising as the regression trend error decreases with increasing
722	number of points for which the trends are calculated (Supplementary Information
723	equation S2). Furthermore, this implies that with continued extension of the total ozone
724	record detection of Antarctic ozone recovery may be reached before 2020 using multi-
725	multivariate regressions. Note that although in total the number oif statistically significant
726	trends increases with record length, this is not necessarily always the case: -(For example
	1

727	by comparinge Table 5 and Table 6 – thus for it is conclude showsed that for some scenario
728	combinations the number of significant trends is larger for a (shorter) period (BREAK-
729	2010) than for the full period vs. 1998-2012-trends).
730	
731	Excluding the year 2002 from the regressions has a significant impact on the post-break
732	ozone trends themselves. However, it hardly has any effect on the post break trends from
733	the regressions (not shown), indicating effective removal of the anomalous year 2002
734	from the results. Excluding volcanic years from the regression had no significant effect
735	on both the ozone trends before and after the regression, consistent with our finding that
736	there appears to be little (direct) impact of volcanic aerosols on Antarctic springtime
737	ozone.
738	It is tempting to interpret, based on some selections of our results, that the significance
739	is sufficient for identification of the second stage of ozone recovery by 2012. However,
740	comparing Table 5 and Table 6 - thus 2000-2010 trends vs 1998-2012 trends - shows
741	that the longer period not always results in increased statistical significance. In particular,
742	the need to average ozone over longer periods of time may introduce long term changes
743	in average ozone that are not related to photochemical ozone destruction. Furthermore,
744	the t <u>Trend significanceothe</u> is generally between 2σ and 3σ (not shown), indicating that a
745	considerable amount of variability is not accounted for in the regression. In addition, our
746	analysis shows sthat detection of the 2 nd stage of ozone recovery based on just one more
747	or less arbitrary selected (set of) regressor - ozone record combination(s) does not reflect
748	the structural uncertainties present in the underlying data.

Nevertheless, the appearance of large<u>r</u> groups of statistically significant results occurring for longer time series and a certain persistence among ozone scenarios and EP flux scenarios; shows that these type of multivariate regression, preferably using piecewise linear analyses before and after a predefined break year, are capable of removing deterministic variations in average ozone, and that with increasing length of the post-break period more <u>and robust</u> statistically significant results can be expected.

755

756 **5. Conclusions**

757

The primary goal of this study was to investigate whether or not the 2nd stage of ozone recovery – a statistical increase in ozone attributable to ozone depleting substances – can be detected, given uncertainties in underlying data. A detailed sensitivity analysis of widely used <u>multi-multi</u>variate regression analysis of total ozone columns was presented focusing on Antarctic springtime ozone. By combining regressor scenarios and Monte Carlo simulations for various ozone record scenarios, a total of approximately 23 million different multivariate regressions were performed.

Our analysis shows that detection of the 2nd stage of ozone recovery based on one more
 or less arbitrary selected (set of) regressor – ozone record combination(s) does not reflect
 the structural uncertainties present in the underlying data.

768

Use of the post-break trends based on fitting the EESC to the total ozone record is not recommended, as these trends are solely based on the pre-defined EESC shape, and do not allow flexibility in the trend calculation while it is unclear how to assign uncertainties to the EESC-regression-based trends in total ozone. Because the resulting EESC fit based
trend uncertainties do not take the ozone fit residuals into account the EESC scenarios
result in overconfident ozone trend uncertainties, neglecting structural uncertainties and
sensitivity to the chosen scenario.

Our analysis <u>further</u> shows that the EP flux and the SAM effects are capable to explain significant parts of Antarctic ozone variations and the removal of these effects improves the analysis of recovery;, in contrast to the inclusion in the regressions of volcanic aerosols and the combined QBO/Solar flux index.

Consistent with expectations, we find a robust gradual small increase in Antarctic ozone since the late 1990s that can be attributed to decreases in ODS for selected combinations of regressors, although the magnitude of the increase is rather uncertain $(+1.66 \pm +4.74 \text{ DU/year}; 95\% \text{ CI})$. The trend significance shows a clear dependence on the length of the period over which the trend is calculated. The number of statistically significant trends in our ensemble varies between approximately 30-60%, depending on the length of the period, with an average of approximately 50%.

787 The limited information present in the Antarctic ozone record for volcanic aerosols 788 (essentially two isolated peaks) is consistent with Knibbe et al. [2014], who found little 789 evidence for volcanic effects on total ozone throughout the Southern Hemisphere. 790 Furthermore, Poberaj et al. [2011] also reported little impact of volcanic aerosols from 791 the Pinatubo eruption on Southern Hemispheric ozone, attributing it to dynamical 792 conditions favoring more poleward transport of ozone from the tropics and mid-latitudes 793 than usual, thereby "overcompensating the chemical ozone loss ... and reduce the overall 794 strength of the volcanic ozone signal".

The lack of correlation between Antarctic ozone and the solar-flux/QBO combined index was also found by Knibbe et al. [2014] for both Antarctic (and Arctic) ozone trends. This lack of QBO-solar signal in Antarctic springtime ozone – also e.g. reported in both Labitzke [2004] and Roscoe and Haigh [2007] - may be related to the dominance in absolute values of the ozone change of ozone depletion and vortex dynamics over potential indirect solar influences on Antarctic springtime ozone.

801 From our analysis it remains unclear what the appropriate time window would be over 802 which to average the ozone record and the EP flux. Results indicate that the best 803 regression occur for ozone averaged over a time window that includes the ozone hole 804 season - typically September and part of October. On the other hand, the time window 805 should also not extend far beyond the ozone hole season as more and more non-806 photochemical ozone variations are introduced in the averaged ozone with a longer 807 averaging period. Similarly, for the EP flux we find that including both the August and 808 September months result in the best regressions. However, the choice for using complete 809 calendar months is rather arbitrary, and better choices may exist which is here left for 810 future research.

The lack of a proper definition of appropriate time windows drives our recommendation that care has to be taken with drawing firm conclusions about Antarctic ozone recovery based on <u>multi-multi</u>variate regression of Antarctic vortex average ozone. <u>Given</u> <u>uncertainties in parameters and independent variables and choices in defining the optimal</u> <u>time period and area for calculating the ozone record and the independent variables Fit is</u> tempting to discard those results <u>in the full ensemble</u> that do not confirm our expectations, but without proper justification of what constitutes the best set of 818 independent explanatory variables – for example physically compelling arguments - there
819 is the danger of working towards an expected answer.

Another last finding is that Finally, a longer post-break period does not necessarily 820 821 always results in more significant trends, which provides yet another indication to remain 822 careful with drawing too firm conclusions from multivariate regressions. On the other 823 handdespite these uncertainties, our results indicate it can be expected that with extending 824 the ozone record and using a multi-multivariate regression method to remove well-825 selected non-ODS influences from the total ozone record – the second stage of recovery 826 of the Antarctic ozone-hole will-may be detectable before 2020. Future updates of the 827 analysis in this paper by extension of the present-day ozone records are expected will 828 rather soon provide indications whether this moment approaches fast or not.

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977**Tables**

EP flux

http://www.awi.de/en/research/research_divisions/climate_science/atmospheric_circulati

ons_old/projects/candidoz/ep_flux_data/

QBO

http://www.geo.fu-berlin.de/met/ag/strat/produkte/qbo/

Solar flux

 $\underline{ftp://ftp.geolab.nrcan.gc.ca/data/solar_flux/monthly_averages/solflux_monthly_average.t}$

<u>xt</u>

SAM

ftp://ftp.cpc.ncep.noaa.gov/cwlinks/

EESC

http://acdb-ext.gsfc.nasa.gov/Data_services/automailer/

Volcanic aerosol

http://data.giss.nasa.gov/modelforce/strataer/

Assimilated total ozone

http://www.temis.nl/protocols/O3global.html

978 **Table 1**. Data sources

regressor	variations
Average EP flux	- 70 hPa, 40°S-90°S, Aug-Sep (baseline)
- 8 scenarios	- 70 hPa, 40°S-90°S, Jul-Aug
	- 70 hPa, 40°S-90°S, Jul-Sep
	- 70 hPa, 40°S-90°S, Jul
	- 70 hPa, 40°S-90°S, Aug
	- 70 hPa, 40°S-90°S, Sep
	- 70 hPa, 45°S-75°S, Aug-Sep
	- 100 hPa, 40°S-90°S, Aug-Sep
Solar flux – QBO index	- Random variations in Solar flux – QBO anomalies
- 100 Monte Carlo series	- 200% Gaussian noise variations on single solar flux – QBO anomalies

SAM index - 100 Monte Carlo series	- 100% random error on annual mean SAM index values
EESC loading - 3 scenarios	- EESC shapes based on different age of air of 2.5, 4.0 and 5.5 years
Volcanic aerosol - 6 scenarios	 Baseline Volcanic Aerosol index (NASA GISS) Pinatubo peak scaled to El Chichón peak Pinatubo peak 2.5 times the El Chichón peak Pinatubo peak 5 times the El Chichón peak El Chichón peak shifted one year back compared to Pinatubo peak Pinatubo peak shifted one year back compared to El Chichón peak
Ozone record - 8 scenarios	 Sep-Oct-Nov average ozone (baseline) Sep-Oct average ozone Sep average ozone Oct average ozone 7 Sep - 13 Oct average ozone Very short 21-30 Sep average ozone Very long 19 Jul - 1 Dec average ozone "Worst" 30-day average ozone.

Table 2. Summary of the uncertainties for the proxies discussed in section 2.1 to 2.9 and

981 their inclusion in the regression analysis in this study.

	Kuttippı [20	urath et al. 13]	This study		
Period	EESC	PWLT	EESC	PWLT	
1979-	-4.50 ±	$-5.02 \pm$	-5.39 ± 0.9722	-5.66 ±	
1999	0.65	1.11		1.03	
2000-	1.11 ±	$2.91 \pm$	1.04 ± 0.1 <mark>9</mark> 2	3.30 ±	
2010	0.16	2.73		2.85	
1979-			-5.26 ± <u>1.02</u>	-5.75 ±	
1999			0.21	1.00	
2000-			1.09 ± 0. <u>23</u> 10	$3.28 \pm$	
2012				2.49	

Table 3. EESC-based Antarctic vortex core ozone trends and their 2σ trend uncertainties (DU/year) derived from <u>multi-multi</u>variate linear regression. The trends in ozone based on EESC regression are calculated by an Ordinary Linear Regression based of the predefined change in EESC multiplied with the EESC regression coefficient for the time period under consideration [*cf*. Kuttippurath et al., 2013]. The EESC trend is in pptv/year, the EESC regression coefficient is in DU/pptv, hence the trend in ozone is in DU/year, allowing direct comparison with the PWLT ozone trends (also in DU/year)

	EP- flux	EESC	AEROSOL	SF×QBO
SAM	-0.31 ± 0.27	-0.03 ± 0.17	-0.09 ± 0.19	-0.09 ± 0.29
SF×QBO	0.08 ± 0.28	0.07 ± 0.42	-0.02 ± 0.19	
AEROSOL	0.05 ± 0.17	0.03 ± 0.30		
EESC	0.25 ± 0.17			



EP flux Ozone	Aug - Sep	Jul - Aug	Jul - Sep	Jul	Aug	Sep	45°S-75°S	100 hPa
Sep - Nov	27.7	16.9	43.5	2.3	18.2	2.6	84.9	70.7
Sep - Oct	98.5	80.7	99.7	37.5	71.5	71.2	100.0	100.0
Sep	34.8	23.1	41.9	5.0	23.0	15.0	60.8	60.4
Oct	99.4	72.5	99.2	35.2	63.7	77.6	100.0	99.9
21 - 30 Sep	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	1.0	1.9
7 Sep – 13 Oct	54.3	19.5	55.2	5.3	17.8	24.4	92.1	90.7
Worst 30 days	87.3	52.9	94.2	18.8	42.2	53.6	99.6	99.1
19 Jul – 1 Dec	30.1	21.8	36.3	4.6	27.4	5.3	78.6	68.2
Table 5. Percent	age of s	tatistical	ly signit	ficant re	gressior	ns for ea	ch comb	ination of

and EP flux scenarios, as defined in section 2, based on the PWLT regression model for
the all-break years 1997, 1998, and 1999, and all ending in 2010. Each ensemble consists
of results of 180,000 single regressions (6 volcanic aerosol scenarios, 100 SAM and 100
QBO-solar index Monte Carlo runs, 3 break years). Numbers in bold are statistically
significant > 95%.

EP flux Ozone	Aug - Sep	Jul - Aug	Jul - Sep	Jul	Aug	Sep	45°S-75°S	100 hPa
Sep - Nov	99.9	10.7	92.6	0.1	36.3	29.5	100.0	100.0
Sep - Oct	100.0	52.2	100.0	4.2	73.4	100.0	100.0	100.0
Sep	100.0	40.3	99.5	2.0	67.5	96.4	100.0	100.0
Oct	100.0	12.1	98.0	0.6	27.2	98.1	100.0	100.0
21 - 30 Sep	0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	2.0	20.9
7 Sep – 13 Oct	100.0	10.0	97.7	0.8	19.6	98.4	100.0	100.0
Worst 30 days	100.0	20.0	99.4	1.2	29.6	99.7	100.0	100.0
19 Jul - 1 Dec	99.9	25.6	95.3	1.5	66.1	56.5	100.0	100.0



Start year	End year	Length (years)	significant trends
2000	2010	11	34.3%
1999	2010	12	47.8%
1998	2010	13	59.5%
all	2010		47.3%
2000	2012	13	39.0%
1999	2012	14	52.7%
1998	2012	15	60.7%
all	2012		50.5%

Table 7. Fraction of statistically significant trends (%) in all regression results for different break years, period lengths and different types of trend calculations. The start year and end year refer to the time period for which trends are calculated. The "all" start years refers to the statistics for all three start years scenarios combined.

1007 Supplementary information

1010 **QBO**.

1011

The Quasi-Biennial Oscillation (QBO) of the winds in the equatorial stratosphere has 1012 1013 been discovered in the 1950s through the establishment of a global, regularly measuring 1014 radiosonde network [Graystone, 1959; Ebdon, 1960]). The Free University of Berlin has 1015 compiled a long-term record from 1953 onwards of daily wind observations of selected 1016 stations near the equator. From these daily measurements monthly mean zonal 1017 components were calculated for pressure levels of 70, 50, 40, 30, 20, 15, and 10 hPa. For 1018 the period after 1979 only measurements from Singapore are used. The QBO data set is 1019 supposed to be representative of the equatorial belt since various studies have shown that 1020 longitudinal differences in the phase of the QBO are small [Hood, 1997]. It should be 1021 noted, however, that some uncertainties arose at higher levels during the early years from 1022 the scarcity of observations. More information on the original data and their evaluation 1023 can be found in Naujokat [1986].

1024 As proxy for the regressions we will use the 40-hPa QBO index, also used in 1025 Kuttippurath et al. [2013]. Salby et al. [2011, 2012] chose to use 30-hPa winds instead. 1026 Typical maximum zonal wind speeds are 20-30 m/s. -The relevance of the choice of 1027 QBO index will be evaluated later. Information on the uncertainties in the monthly QBO 1028 data is not available. One indirect method to estimate the uncertainties is by examining 1029 QBO index variability close to the maximum and minimum of the QBO cycles, where the 1030 QBO index values remains more or less constant for some months. Assuming that during 1031 the maximum or minimum in the QBO phase variations from month to month are indicative of uncertainties in the QBO, we come up with estimated uncertainties ofaround 1.5-2.0 m/s in the zonal mean wind speeds.

1034

1035 Solar flux

1036

Variations in incoming solar radiation – in particular the shorter ultraviolet wavelengths
– have an effect on stratospheric ozone [Haigh, 1996; McKormack and Hood, 1996;
Soukharev and Hood, 2006; Anet et al., 2013]. A standard proxy for variations in
incoming solar radiation in ozone regression studies is to use the monthly mean 10.7 cm
radio flux, as also used in Kuttippurath et al. [2013]. This data set was obtained via
NOAA/NESDIS/NGDC/STP.

1043 However, there are other solar activity proxies available. Ideally, in absence of true UV 1044 spectral measurements, one would like to use a proxy that is representative for solar 1045 activity at those wavelengths where stratospheric ozone formation occurs, which is of 1046 roughly between 200 and 300 nm. Dudok de Wit et al. [2009] tried to identify the best 1047 proxy for solar UV irradiance, and concluded that proxies derived from a certain 1048 wavelength range best represent the irradiance variations in that wavelength band. Thus, 1049 the 10.7-cm radio flux might not fully represent solar UV variability. Using the results 1050 from Dudok and de Wit et al. [2009] to analyze a set of seven solar activity proxies 1051 dating back to at least 1979 based on the solar2000 model and obtained from 1052 NOAA/NESDIS/NGDC/STP (F10.7, Lyman-alpha, E10.7, and the solar constant S), we 1053 will assume in our regressions that the uncertainty range associated with the solar proxy 1054 is approximately 15% of the root-mean-square of the anomaly values.

1056 Why do standard errors of an ordinary linear regression relative to the regression1057 slope not depend on the actual regression itself?

1058

1059 This analysis is based on the "Data Analysis Toolkit" document (chapter 10), written by 1060 Prof. James Kircher, Professor of Earth and Planetary Science at the University of

1061 California, Berkley and emeritus Goldman Distinguished Professor for the Physical1062 Sciences.

1063

1064 <u>http://seismo.berkeley.edu/~kirchner/</u>

1065

1066 The standard error of the regression slope **b** of an ordinary linear regression of two 1067 variables **x** and **y**, and the regression slope **b** itself can be written as:

1068

1069
$$s_b = \frac{b}{\sqrt{n-2}} \sqrt{\frac{1}{r^2} - 1}$$
 and $b = r \frac{S_y}{S_x}$ (S1)

1070 In which s_b is the standard error of the regression slope, **n** the number of data points of 1071 the variables **x** and **y**, **r** is the Pearson correlation coefficient between the variables **x** and 1072 **y**, and $S_{x,y}$ is the standard deviation of the variables **x** and **y**.

1073 For a statistically significant trend one generally defines that the trends (slopes) should 1074 exceed two times the standard error. Or, in other words, the standard error of the 1075 regression slope divided by the regression slope itself should be less than 0.5

1076 The standard error of the regression slope relative to the regression slope itself – which 1077 directly relates to statistical significance of the trend - becomes, based on the equation

1078 above:

1079
$$s_b / b = \frac{1}{\sqrt{n-2}} \sqrt{\frac{1}{r^2} - 1}$$
(S2)

- 1080 which only depends on the correlation between the variables \mathbf{x} and \mathbf{y} and the number of
- 1081 data points of variable **x** and **y** (record length).

1 Figures





Figure 1. Vertical Eliassen-Palm (EP; kg/s^2) flux at 70 hPa between 40°S and 90°S for 4 5 five different meteorological datasets for the period 1979-2012 averaged for the two 6 month period August-September: NCEP reanalysis 1979-2012, ECMWF ERA INTERIM 7 1979-2012, ECMWF ERA 40 1979-2001, Japan Reanalysis 1979-2005, ECMWF 8 operational analysis 1998-2012. Top panel shows the EP flux as function of time, 9 including the mean EP flux for each year based on all datasets. Bottom panel shows the 10 EP flux anomalies (%) of a given year as function of the mean EP flux (black dots in the 11 upper panel) for all meteorological datasets available for that year. The insert shows the 12 probability distribution of the relative anomalies. Data are obtained from the EP flux data 13 website of the Alfred Wegener Institute (AWI) for Polar and Marine Research in 14 Bremerhaven, Germany.



16 Figure 2. Time series of the combined Solar flux- QBO index (arbitrary units) (upper plot) and the index anomalies relative to the average of different possibilities to derive at 17 18 the index. The Solar flux ("S") and QBO ("Q") anomalies were calculated based both on 19 the average ("M") as well as the range of Solar flux and QBO values ("R", see section 2.5 20 for the explanations of the "range"), and for both the entire record of Solar flux and QBO 21 values (1947-2012 and 1953-2012, respectively; "1") as well as for the period 1979-2012 22 ("2"), resulting in a total of 16 combinations. The different colors denote the different 23 combinations.





Figure 3. Time series of regressors for the period 1979-2012. For ozone, EP flux, EESC and stratospheric aerosol all scenarios as defined in section 2 are included (indicated by the different colors). For the SAM and the solar flux - QBO index only the baseline time series is shown, and both indices – being unitless to start with - are scaled for proper comparison. Ozone values are in DU, EP fluxes are in kg/s, EESC values are in ppbv and stratospheric aerosol is in optical depth.





Figure 4. Probability distribution of ozone trends for the period 1979-break (upper plot)
and break-2012 (middle plot) as well as time correlations (R²) for the regression models

and the ozone record scenarios (lower plot). The colors indicate the distributions for the two different long-term ozone regressions (EESC, PWLT). Indicated in the figure are also the 0.5%-2.5%-mean-median-97.5%-99.5% probability values of trends and correlations. The vertical black lines in the upper two panels indicate the trend (solid) and 2σ errors (dotted) of the PWLT regression results of table 2 for the period 2000-2012.



Figure 5. Probability distribution of regression model – ozone scenario correlations as Figure 4, lower plot, for the PWLT and EESC regression model and sensitivity to the different ozone scenarios and different EP flux scenarios, indicated by the different colors. The blue and red outlines show the sum of all scenarios combined.



probability distribution AEROSOL EESC regression values & EESC scenarios



46 Figure 6. Upper panel: probability distribution of aerosol scenario regression coefficient 47 values of all PWLT regression results. Indicated in the figure are also the 0.5%-2.5%-48 mean-median-97.5%-99.5% probability values of trends and correlations. Included are 49 also the distributions for the different stratospheric aerosol scenarios, indicated by the 50 different colors. Lower panel: probability distribution of the aerosol regression coefficient 51 values of the EESC regression model results. Included are also the distributions for the 52 three different EESC age of air scenarios, indicated by the different colors. The blue and 53 red outlines show the sum of all scenarios combined.





Figure 7. Panel A: probability distribution of the solar flux – QBO index regression coefficient values of all EESC and PWLT regression model results. Panel B: probability distribution of the SAM index regression coefficient values of all EESC and PWLT regression model results. Panel C: probability distribution of the EP flux regression coefficient values of all EESC and PWLT regression model results. Indicated in the figure are also the 0.5%-2.5%-mean-median-97.5%-99.5% probability values of trends and correlations.



63198019851990199520002005201064Figure 8. Optimal regression model result for the EESC and PWLT and regressions

65 (upper panels, red line) as well as the corresponding ozone record scenario (upper panel,

- 66 black line). The ozone variations attributable to each are also shown. Ozone and ozone
- 67 anomalies are given in DU.



Figure 9. The probability distribution of regression model – ozone record scenario correlations (\mathbb{R}^2) as shown in Figure 5 for the PWLT regressions and the cumulative fraction of statistically significant (2σ) ozone trends for each correlation interval (red, right axis). The upper panel shows the distribution for the regressions ending in 2010, the lower panel for the regressions ending in 2012. See also Table 7.