Response to Referee #1

This is the response of Irina Petropavlovskikh on behalf of all authors (AC) to the comments of Referee 1 (R1).

Major comments:

R1: Section 4.1 is not clear enough. The authors state that NAO and NIÑO3.4 indexes are correlated with TOC, but no evidences are presented in the article. Only visual correlations are mentioned. What about statistical correlation? Furthermore, Figs. 3 and 4 are difficult to read. Please, clarify the y-axes for ELOs and EHOs.

AC: Following the referees suggestion we will provide additional statistics and their discussion in the revised version of the manuscript. In both winter and spring the NAO Index correlates negatively with the frequency of EHOs and positively with the frequency of ELOs, indicating an increase (decrease) in the frequency of high ozone events during a negative (positive) NAO phase and vice versa for low ozone events, manifested also in the seasonal means (See Table R1 below, which will be included in the revised manuscript). Further differences in the occurrence and detection frequency of NAO fingerprints are captured in the correlation analysis, with overall stronger correlations during winter than spring. This is further explored in Figure R1 (which will be included in the revised manuscript), which shows the fraction of EHOs and ELOs during wintertime positive and negative NAO events: more (less) EHOs during NAO- (NAO+) phases and vice versa for ELOs. Correlation analysis between ENSO and column ozone (or the frequency of EHOs and ELOs) is less conclusive than that for the NAO, which is attributed to the rather small number of strong ENSO events.

Table R1: Correlation of the NAO-Index and the average number (#) of EHOs and ELOs and mean column ozone (TOC) on seasonal basis.

	Correlation with NAO Index		
season/station	**# EHOs**	**# ELOs**	**Mean TOC**
DJF			
BISMARCK	-0.44	0.40	-0.53
BOULDER	-0.26	0.29	-0.34
CARIBOU	-0.53	0.33	-0.61
WALLOPS ISLAND	-0.28	0.38	-0.52
NASHVILLE	-0.53	0.33	-0.48
MAM			
BISMARCK	-0.21	0.22	-0.29
BOULDER	-0.10	0.10	-0.21
CARIBOU	-0.16	0.10	-0.21
WALLOPS ISLAND	-0.10	0.23	-0.21
NASHVILLE	-0.27	0.10	-0.20

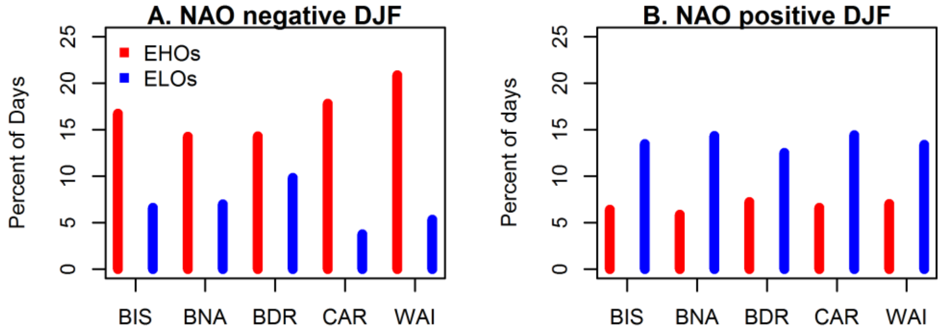

Figure R1: Average fraction of days identified as EHO and ELO during (a) negative and (b) positive NAO phase in winter (DJF) season.

We will update the caption for Figures 3 and 4 (and corresponding supplemental figures) to explain the y-axes, e.g. for Figure 3: 'Fingerprints' of the NAO and ENSO as detected for Boulder in the seasonal frequency time series of EHOs (right axis, top to bottom) and ELOs (left axis, bottom to top) for (A) winter (DJF), and (B) spring (MAM). Bottom panels (C) and (D) show 'fingerprints' in seasonal mean column ozone. Filled circles denote visible 'fingerprints' and crosses denote not visible 'fingerprints'. NAO positive (negative) phase is indicated for winter in red (blue) and for spring in orange (light blue), ENSO positive phase is indicated for winter (spring) in green (light green).

R1: A deeper discussion should be performed in section 4.3. The authors attribute to the large spatial distance the absence of fingerprints during NAO and ENSO events at the five sites. I partially agree with the explanation of regional effects. As Figure 6 is, practically, limited to certain months, I think more analysis should be performed for particular events. Is there any difference (meteorological, synoptic,...) between those events discernible at the five sites and those only visible in northern sites or the eastern sites,...?? In this sense, are these differences in line with the results shown of Table 3? Some sites present good relationships in winter or spring for the extreme influence, are these similarities translated to the fingerprints during certain events?

AC: Correlations of daily TOC among sites are rather noisy (local effects, temporal lags due to dynamics), vertical investigations are limited by the absence of vertically resolved ozone profiles, and seasonal comparisons between years with 'fingerprints' and without are restricted to a small sample size (i.e., few missing fingerprints on a site basis). Nevertheless a comparison of CDFs on site basis between neighboring years with 'fingerprints' and without reveals the absence of high or low ozone events associated with the NAO or ENSO (see Figure R2, which will be included in the supplemental material of the revised manuscript). Thus instead of individual effects, we quantify the overall contribution of extremes to seasonal mean column ozone by calculating the influence of ELOs and EHOs at each site.

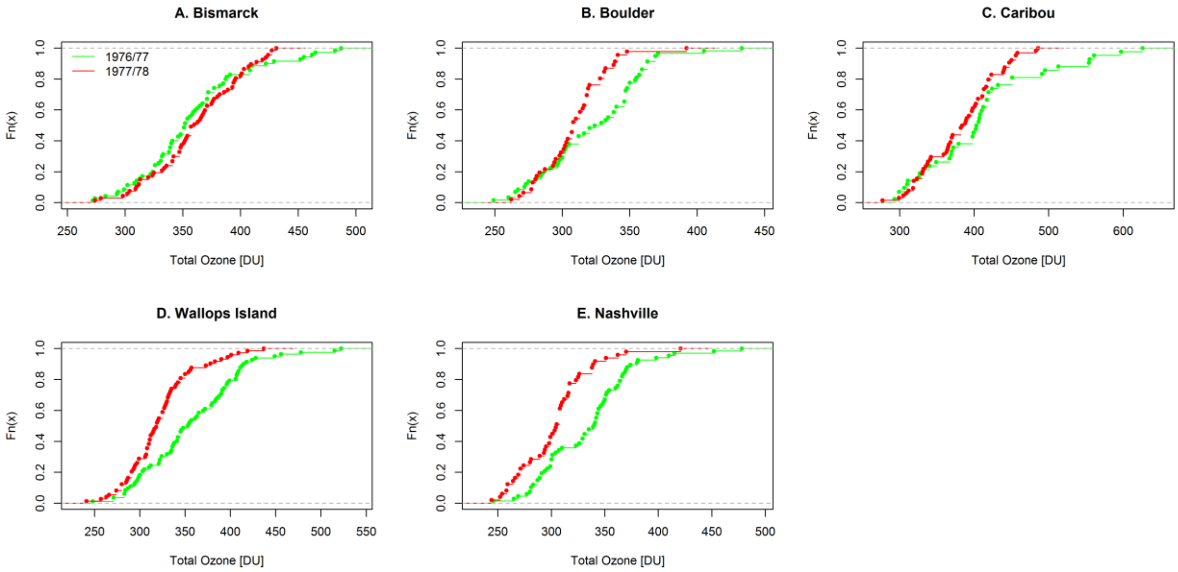

Figure R2: Empirical cumulative distribution functions (ecdf) of total ozone in winter 1976/77 (green) and 1977/78 (red). In 1976/77 fingerprints of the negative NAO phase have been detected at all sites (increased frequency of high TOC values, i.e. EHOs), while a fingerprint for the negative NAO phase in 1977/78 has been detected only at site Bismarck, i.e. reduced frequency of high ozone events at all other sites.

R1: I miss more comments about the interesting topic of double tropopauses (e.g., Randel et al., 2007;Pan et al., 2009). Previous studies have shown that intrusions of subtropical air above the extratropical tropopause produce a decrease in the ozone levels (e.g., Castanheira et al. 2012), and being more frequent with NAO positive phases (e.g., Mateos et al., 2014). It is not necessary a complete analysis of these events (maybe, it is beyond the aims of the article), but these events should be more clearly mentioned in the discussion since they are clearly latitudinal dependent and can play a notable role in the differences among the five sites.

AC: We agree with the referee that the subject of double tropopauses (DTROP) and their influence on column ozone received increasing attention in recent years. While a detailed event based analysis is beyond the scope of the present work, we agree that a section detailing the relationship between DTROP events and TOC and their relation to the 5 US sites will strengthen the manuscript. We will include the following section in the revised version of the manuscript:

'Several studies have linked the occurrence of multiple tropopauses to Rossby wave breaking events along the subtropical jet (Homeyer and Bowman, 2013, and references therein), and to associated tropospheric intrusions (e.g., Pan et al., 2009); climatological maxima in multiple tropopause occurrence have been linked to observed changes in vertical profiles of satellite-observed trace gases that are consistent with air from the tropical tropopause layer being drawn into the region between the two tropopauses; specifically, climatological ozone mixing ratios in midlatitude multiple tropopause regions are substantially lower than those in regions with a single tropopause (Schwartz et al., submitted). Schwartz et al. estimated that in NH winter midlatitudes, when multiple tropopauses are most common, climatological ozone values can be as much as 20% lower that they would be without multiple tropopauses. These results are consistent with the observed association of lower column ozone with multiple tropopauses (e.g., Castanheira et al., 2012;Mateos et al., 2014). Mateos et al. (2014) also noted more common occurrence of such tropospheric intrusion events during NAO positive phases, suggesting a role for dynamical modes such as NAO and ENSO in modulating multiple tropopause occurrence and thus their corresponding effects on ozone.

In addition, there is a maximum in multiple tropopause occurrence frequency over the US in winter and spring, extending poleward from the region where upper tropospheric jets are most common (e.g., Manney et al., 2014). Boulder, Nashville, and Wallops Island are near the latitude of maximum multiple tropopause occurrence just poleward of the subtropical upper tropospheric jet, while Bismarck and Caribou are at the northern edge of the region of enhanced multiple tropopause activity (Manney et al., 2014), and are thus less frequently affected by processes in multiple tropopause regions.'

Minor comments:

R1: Abstract. "from the five US sites...". I understand that no more sites have ozone records since 1960 in the US. Is this true?

AC: Correct. The five sites analyzed in this study are the only ones that provide continuous total ozone observations back to the 1960s within the continental US.

R1: Figure 1. Although the coordinates of each site are indicated in the figure, I'd appreciate if the map is geo-localized.

AC: We will update Figure 1 according to the referee's suggestion for the revised version of the manuscript.

R1: Figure 2. Please, add some discussion about this figure. For instance, the authors can discuss the latitudinal or longitudinal dependence of the threshold values among the five sites, and others.

AC: Additional discussion of Figure 2 will be provided in the revised manuscript as given below 'Thresholds for ELOs and EHOs, as well as long-term monthly mean values for the five US sites, are shown Figure 2. Here the well-known seasonal cycle with ozone minima in fall and maxima in spring, as well as the latitudinal dependence of total ozone mean values and thresholds (i.e., higher TOC at northern sites (Bismarck and Caribou) due to transport of ozone rich air from high latitudes) is visible.'

R1: Tables 1 and 2. More statistical information should be given in these Tables. Although the standard error is given for each trend, I'd appreciate the knowledge of, e.g., the p value. Maybe it is possible to reduce these two tables to only one. With one table is easier to compare the two periods analyzed.

AC: We will update the Tables according to the referee's suggestion for the revised manuscript.

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Response to Referee #2

This is the response of Irina Petropavlovskikh on behalf of all authors (AC) to the comments of Referee 2 (R2).

We thank the referee for his/her positive judgment and valuable suggestions leading to an improved version of the manuscript. The individual points raised by the referee are addressed in the point to point reply below.

General comments

R2: 1. Abstract p. 21067, line 9-12: I object the statement: The Loess smoothed trend components show a decline of total ozone between the 1970s and 2000s and a "stabilization" at lower levels in recent years which is confirmed by linear trend analysis (see below).

AC: We will remove this statement from the manuscript. See more detailed discussion regarding STL-trend components following the referees' comments below.

R2: 2. p. 21073, line 12: "Our main interest in this study lies in the trend component of the STL-decomposition": I see the values using STL plots to illustrate the relations between ozone time series and fingerprints (such as in the figures 5 and S5) but the scientific interpretation of "trend component" of total ozone time series analysis is obscure to me (see below) as the procedure leads to smoothing over the effects of many different processes (see below)

AC: We agree with the referee that the STL-trend components are not reliable measures for TOC trends as they are rather TOC residual trends that are smoothed after the seasonal cycle is removed, which removes individual short-term variability associated with various meteorological effects. Throughout the paper STL-trend components are mainly used to provide a secondary assessment for 'fingerprints' of NAO and ENSO events corroborating the EVT analysis. Statistical trend calculations in the presented manuscript (time periods for which will be updated according to the referees' comment, see below) are solely based on linear regression models. For clarity we will remove any 'trend interpretation' of the STL-trends and use STL-trend components solely for demonstration of the 'fingerprint' detection.

R2: 3. p. 21075, line 13 "Fingerprints" of NAO and ENSO in the frequency distribution of extreme events: "The NAO fingerprints are in broad agreement with those for European sites and satellite data, and p. 21076, line 6 are in good agreement with findings for European sites and satellite data": please provide more specific statements

AC: We will update this section according to the referees' comment as given below:
'The NAO 'fingerprints' identified in the US column ozone records are in broad agreement with those for European sites and satellite data. Appenzeller et al. (2000) were among the first to report on the influence of the NAO on column ozone over Europe, based on their analysis of the world's longest total ozone record, Arosa, Switzerland. Rieder et al. (2010) extended these investigations toward low and high ozone values and Rieder et al. (2011) documented the influence of the NAO in its positive (reduced column ozone, reduced frequency of high ozone events) and negative phase (increased column ozone, increased frequency of high ozone events) for 5 European ground based sites in 1970-2010. These authors report a similar number of detected 'fingerprints' and occasional misses at individual sites due to local effects. Frossard et al. (2013) extended investigations to larger spatial scales

analyzing the NIWA assimilated total ozone data set in 1979-2007. These authors report that the spatial 'fingerprint' of the NAO is of similar spatial extent for both mean values and ozone extremes, but that the magnitude of influence on total ozone is larger for extremes than mean values. These results are in broad agreement with those presented here for the US long-term ozone records, documenting the significant influence of the NAO on column ozone variability throughout northern mid-latitudes.

R2: 4. p. 21077, line 12ff: I cannot follow the arguments: "show that the strong decrease during 1980s and 1990s came to a halt around the turn of the century": I see the Fig 5 for Boulder is a decreasing tendency until 1996 followed by sort of a "jump" and a new "decreasing tendency" staring again around 1997 which is similar for Wallops Island (Fig S5) whereas Bismark might show a tendency for an upward trend after 1993, less pronounced in Nashville. Because of the subjectivity of such statements I recommend to avoid STL plots for statements regarding "trend analyses" whereas the plots can be used to illustrate the relation between fingerprints and ozone time series. Additionally I find it difficult to interpret the "trend component" from STL
plots as the procedure basically "smoothes" over different processes, e.g. the Pinatubo effect.

AC: See earlier comment regarding STL-trend components.

R2: 5. p. 21078, line 18 ff: I cannot see the rational of the selection of the periods of linear trends analyses, namely 1970-2000 and 1990-2010: I don't believe that maximum ODS was "around 2000": What means "maximum ODS": emissions or EESC? To my knowledge EESC for mid-latitude was peaking around the middle of the last decade (1997 ?). The start of the second period in 1990 includes the low ozone values generally attributed to the volcanic eruption of Pinatubo and therefore these low values are expected to contribute to the upward trends. If a statement in connection with anthropogenic ozone destruction is attempted I recommend to use linear trend analysis for 1970-1995 and 1995-2010 leading to a more positive trend.

AC: We agree with the referees remarks regarding the time periods chosen for trend analysis. According to the referees suggestion we updated the calculations for the periods 1970-1995 and 1996-2010 in accordance with the EESC maximum for middle latitudes found in ~1996-97. While the magnitude of the trends is affected by this change the overall conclusions of the trend analysis are not. For 1970-1995, the period with almost linearly increasing ODS and significant mid-latitude ozone losses in the early 1980s and 1990s (following the El Chichon and Mt. Pinatubo eruptions), ozone trends vary between -2.8 and -4.8 percent per decade among the sites and seasons. All sites, except Caribou, show larger negative trends in spring compared to winter. For the more recent past, 1996-2010, we find positive trends at most sites, an anticipated result since stratospheric chemistry in this period is impacted by slowly but steadily declining ODS. Positive trends at the majority of sites indicate that ozone has stopped declining over the US, particularly during winter, suggesting that chemical depletion may have ceased. Nevertheless, since the trend estimates over the period 1996-2010 during spring typically do not exceed the standard errors, there is no clear evidence that significant ozone recovery has been identified yet. Additionally we will keep the tables presenting the trend analysis for individual sub-periods (1980-2000; 1990-2010 from the original manuscript) in the supplemental material to emphasize that although trends are mostly of the same sign they differ in magnitude and significance among seasons and time periods analyzed. The results presented here highlight the importance of a continued spatiallydistributed long-term ozone monitoring program to address future ozone changes and to detect and confirm the progress of ozone recovery in the context of the Montreal Protocol.

R2: 6. P. 21079, line 17.: I suggest to mention here again that the record low values in total ozone in northern mid-latitudes are commonly attributed to the effect of Pinatubo aerosols.

AC: We will update this section according to the referees' suggestion in the revised manuscript as given below:
'Turning now to the more recent past, i.e., the last two decades (Table 2), we find positive trends at most sites, an unsurprising result since the period with largest mid-latitude ozone losses in the early 1990s (i.e., following the Mt. Pinatubo eruption) has been followed by a period of slowly but steadily declining ODS.'

R2: 7. P. 21082, line 5-8: please explain how the changes in frequencies of ELOs and EHOs are connected with the expansion of tropical belt and the contraction of the northern polar band.

AC: ELOs are indicative of the extension of the subtropical jet to the north of the station, which brings tropical air masses with low ozone content, while EHOs are indicative of a descent of the polar jet toward the equator and advection of O_3-rich air masses from high latitudes. Thus the changing frequency of ELOs and EHOs is in agreement with the notion of the expansion of the tropical band and the contraction of the northern polar band (e.g., Hudson et al., 2006;Seidel et al., 2008).

References:

Appenzeller, C., Weiss, A. K., and Staehelin, J.: North Atlantic oscillation modulates total ozone winter trends, Geophysical Research Letters, 27, 1131-1134, 2000.

Frossard, L., Rieder, H. E., Ribatet, M., Staehelin, J., Maeder, J. A., Di Rocco, S., Davison, A. C., and Peter, T.: On the relationship between total ozone and atmospheric dynamics and chemistry at mid-latitudes - Part 1: Statistical models and spatial fingerprints of atmospheric dynamics and chemistry, Atmospheric Chemistry and Physics, 13, 147-164, 10.5194/acp-13-147-2013, 2013.

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