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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Drivers of variations in the vertical profile of ozone over Summit Station, Greenland: An analysis of ozonesonde data" *by* Shima Bahramvash Shams et al.

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Responses to anonymous reviewer #1:

We are thankful to the anonymous reviewers for their comments. We believe that our responses to these comments have significantly improved the manuscript. Below are our detailed responses. Note that any citations that appear in our responses have references in the bibliography of the manuscript. All updated parts are highlighted in the revised manuscript.

Rev_1: Interactive comment on "Drivers of variations in the vertical profile of ozone over

Summit Station, Greenland: An analysis of ozonesonde data" by Shima Bahramvash Shams et al. Anonymous Referee #1 Received and published: 25 October 2018

Shima Bahramvash Shams et al. analyse ozonesonde data from Summit Station, Greenland to determine the primary drivers of ozone variations at this station by using a stepwise multiple regression analyses. They find that the QBO has the strongest influence for ozone variations over Greenland. This influence is not as strong found over other Arctic stations and thus the authors suggest that Greenland may be particularly sensitive to the QBO. This is a quite interesting study, but it lacks in presentation. I would suggest major revisions before this study can be considered for publication in ACP. I give detailed comments and suggestions for improvements below.

Response: We thank Reviewer #1 for such a thorough review and for helpful suggestions that have greatly improved the manuscript. Based on the Reviewer's comments and questions, we have added three additional high-latitude Arctic stations to this study (Ny-Ålesund, Alert, and Eureka). We also extended the time series by an additional 7 months. (Unfortunately, the ozonesonde launches were discontinued at Summit Station, Greenland in the summer of 2017.) Instead of using climatological data above the ozonesonde profiles, we now use ozone retrievals from the Microwave Limb Sounder (MLS) satellite instrument. The time period of our study is thus constrained by both the ozonesonde record at Summit and the availability of MLS data. The merged ozone profiles are of higher quality than those used in our original manuscript. We investigate the variation of seasonal cycle at these stations. These updates to our study allow us to now identify drivers of ozone variations that are common at the sites. The manuscript now examines the important drivers for different layers in the atmosphere and for the total column ozone for a particular sector of the Arctic. Thus, the Reviewer's comments have allowed us to expand the scope of the study, and we believe that this has greatly improved the manuscript.

General comments:

So far, the results are mainly presented in a descriptive manner than really explaining something. Furthermore, the entire analyses is described and documented in much more detail than actually necessary for a research paper. 1) Therefore, I would first of all suggest to reduce the number of tables and figures to what is really needed. a. For example, the few values for latitude, longitude and altitude of the stations could be easily (if not already done so) given in the main text.

Response: Table 1 has been deleted.

b. Tables 2 and 3 could be provided (if necessary) in a supplement. The numbers from Table 4 which are important could be given in the main text of the manuscript or in the respective figures.

Response: Table 2 has been deleted. We decided to keep Table 3 because we feel that it's important to list the source of each proxy dataset. Figures 3, 4, 6 and 7 have been removed. Overall, we reduced the number of figures from 12 to 8, and we have included new version figures of some figures based on our updated analysis.

2) How can you justify an extrapolation to 60 km? From Figure 2 one can see that there are generally no ozonesondes measuring above 40 km, except the 2 balloons that measured up to 50 km that were removed from further analysis due to erroneous readings from the pressure sensor. That means extrapolation is done over an altitude range of at least 20 km. How can you be sure that your results are not just reflecting the climatological changes and not the actual changes? Why don't you extrapolate the profiles up to 30-40 km as it would match the typical maximum altitude for the measurements of the ozonesondes?

Response: Based on this Reviewer's comments and questions, we have improved our dataset substantially by using ozone retrievals from the Microwave Limb Sounder (MLS) to provide ozone profiles up to 60 km, instead of our previous use of the ozone climatology. This has improved the quality of ozone profiles that have low burst heights by using actual measurements of stratospheric ozone that are nearly coincident in both

space and time with the ozonesondes. It should be noted that using MLS retrievals is the primary reason for differences in our current analyses versus our original analyses over Summit Station.

"We use retrievals from the MLS (version 4.2) above the maximum height of each ozonesonde up to 60 km. MLS is an instrument on the Aura spacecraft that uses microwave emission to measure atmospheric composition, temperature and cloud properties (Waters et al. 2006). Ozone retrievals from MLS are available continuously since 2004 over the Arctic with overpasses over these sites every few days. The standard MLS ozone product, which is retrieved from spectra with frequency 240 GHz, is used in this study. The column value uncertainty (σ) is 2 to 3% (Livesey et al., 2017). The vertical resolution of the MLS profiles is from 100 to 22 hPa is 2.5 km and increases to 3 km in both the lower and upper stratosphere (Livesey et al., 2017). The MLS ozone products have previously been used in analyses of polar ozone loss (Manney et al. 2011; Kuttippurath et al. 2012; Wohltmann et al. 2013; Livesey et al. 2015; Strahan & Douglass 2018)" "MLS has high uncertainty in the lower atmosphere, thus, to minimize the uncertainty in the calculation of TCO, ozonesondes that reached a maximum height of greater than 12 km were used in this study; profiles with maximum heights below 12 km were eliminated from further analysis. The fraction of TCO below 12 km ($\sim$200 hPa) at these sites is about 13-17%." "Ozone profiles in this study are constructed by merging the ozonesondes up to the burst altitude (Figure 2) and then using the MLS profiles up to 60 km. The merged profiles are generated only if an MLS ozone profile is within a 2° by 2° latitude/longitude grid cell around each station and within 4 days of the ozonesonde launch. The majority of the merged profiles are generated using MLS data on the day of the launch or within one day of the launch. Figure 3 shows the difference of TCO from the merged ozone profile versus TCO from MLS only at all stations. This shows that MLS mostly overestimates the ozone in the lower atmosphere at all stations. Thus, the merged profile dataset minimizes the uncertainty in ozone at these sites by using the more accurate ozonesonde data for as much of the lower atmosphere as possible."

3) In this study the results for only one station are presented, but it would be more interesting to have such an analyses for several stations that can be compared to each other. As it is done now (with just mentioning the other two stations that however cannot be analysed because they do not provide measurements for all seasons) it is in my opinion not sufficient.

Response: We agree with this suggestion. Therefore, we have included data from Alert and Eureka, Canada and Ny-Alesund, Svalbard. This allows us to compare the seasonal cycles and investigate the drivers of ozone variations for an important sector of the Arctic. This has greatly improved our paper and made it much more relevant.

"Summit Station, Greenland (72°N, 39°W), Ny-Ålesund, Svalbard (79°N, 12°E), Alert, Canada (82°N, 62°W), and Eureka, Canada (70°N, 86°W) are chosen as the study sites for this research because there is a long history of ozonesonde observations at these locations. Figure 1 shows the locations of these stations in the Arctic." "Summit Station ozone measurements were started in February 2005 and continued until the summer of 2017. The other stations have longer datasets, but in this study, twelve annual cycles from February 2005 through February 2017 are studied to have a consistent dataset at all stations. The time period is also constrained by the availability of MLS data, which is available since 2004. The ozonesonde profiles from Summit Station are available from NOAA's Earth System Research Laboratory, while the profiles from the Canadian stations and Ny-Ålesund can be found at the World Ozone and Ultraviolet Radiation Data Centre (WOUDC). The ozonesondes used here utilize electrochemical concentration cells (ECC) (Komhyr, 1969), manufactured by either Science Pump for Ny-Ålesund or Environmental Science (EN-SCI) for Summit, Alert and Eureka. The ozonesondes at Ny-Ålesund, Alert and Eureka used a sensing solution of neutral buffered 1% potassium iodide, while the ozonesondes at Summit used a reduced (1/10) buffer concentration. The data records of the Canadian sites have recently been re-evaluated (Tarasick et al., 2016), as has the Summit record (Sterling et al., 2017). Based on the ozone sensor response time of 25-40 s (Smit and Kley,

1998), and assuming a typical balloon ascent rate of 4-5 m/s, the ozonesondes have a vertical resolution of about 100-200 meters. The measurement precision is ±3-5%, and the overall uncertainty in ozone concentration in ppmv is about ±10% up to 30 km (Komhyr, 1986; Komhyr et al., 1989; Kerr et al., 1994; Johnson et al., 2002; Smit et al., 2007; Deshler et al., 2008, Deshler et al., 2017)."

4) What is the reason for the QBO influence over Greenland? If you cannot find such an influence over other Arctic stations, how can you then be sure that this is not just a measurement artifact? This really needs some more discussion, analyses and scientific explanations to be sure that there really is a connection.

Response: We also agree with this suggestion. As a result of these questions and those from Reviewer #2, we have substantially changed our approach for selecting important proxies/drivers. We now use the stepwise multiple regression to choose drivers that are common to 3 or more of the 4 sites, which increases confidence that the specific proxies are important in this sector of the Arctic. We then use the important proxies in final regression models for each site. As before, we discuss the contributions of each of the proxies and how they affect ozone concentrations. This provides a much better physical understanding of how these parameters affect Arctic ozone.

"We use a two-step approach to first determine the important drivers of ozone variations at the four high-latitude Arctic sites, then use these to develop models that explain the ozone variations. Stepwise multiple regression analysis is performed to determine significant proxies that affect ozone variations over the four sites. If a proxy is chosen at three or more of the four sites, then it is considered an important contributor in this sector of the Arctic. A final regression model is then fit to each time series. The final model is successful in identifying proxies that explain a significant portion of the ozone variance in the deseasonalized time series, with 90% of the models having R2 >= 70% and 40% with R2 >= 80%. The tropopause pressure, equivalent latitude at 370 K and the QBO are important drivers between the surface and 10 km. The QBO, eddy heat flux and the volume of polar stratospheric clouds are important in the lower stratosphere, while the equivalent latitude at 550 K, eddy heat flux, and the volume of polar stratospheric clouds strongly influence the middle and upper stratospheric ozone. The final regression model explains over 80% of the variance in the time series of total column ozone at the four sites. The contribution from the important drivers is greatest at Summit Station, Greenland in the troposphere (21%) and middle stratosphere (32%). In general, the important drivers explain the greatest variance at all the sites in the middle stratosphere, which is the region of the atmosphere that has the least variance explained by the seasonal cycle. Interestingly, the Arctic Oscillation, solar flux and El Niño-Southern Oscillation are not important for ozone variations in this sector of the Arctic. "

Specific comments:

M1) P1, L12: Here you state that "12 years" of data were used, but later you always state "11 years" of data were used. Response: We have corrected all occurrences to 12 years. We actually added 7 additional months to the duration of the study to analyze 12 complete cycles of ozone data at all of the sites. We are constrained to this time frame by the availability of both the Summit ozonesondes and the MLS satellite data.

M2) P1, L15: Extended to 60 km from which altitude? How many km are extrapolated? Response: We no longer extrapolate any data in the creation of the ozone profiles. The profiles are now a merger of ozonesonde data from the lower atmosphere (up to the burst height) and MLS retrievals in the upper atmosphere. Figure 2 shows the histogram of the maximum height of ozonesondes at all stations.

M3) P2, L14-15: This sentence could be misleading and should thus be rephrased. The reason why there is less photochemical loss of O3 is because if temperatures are warmer, there are less PSCs and if there are less PSCs there are less surfaces for heterogeneous reactions that convert the inert reservoir species into reactive species that destroy ozone. Response: This has now been clarified in the text. "The warmer temperatures are associated with less occurrence of polar stratospheric clouds (PSCs)

and consequently less heterogeneous reactions on the surface of the PSCs, which lead to less photochemical ozone loss in the stratosphere"

M4) P2, L17-18: This only holds with the current ozone loading. When the chlorine concentrations decline, there will be no longer massive ozone destruction due to chlorine. Thus, this sentence should also be rephrased. Response: We agree with the Reviewer's comment, but we are not sure which sentence to rephrase on lines 17 and 18 based on this comment.

M5) P2, L26: Remote sensing instruments → from space or ground? Please be more precise. Response: We referred to both ground-based and space-borne instruments. Most of ground-based instruments are transmission Fourier-transform spectrometers and many satellite instruments, such as OMI, need sunlight to function. We have clarified this in the text.

"This situation is exacerbated by both the lack of high temporal observations at high latitudes, as well as the difficulty of making quality measurements during winter; many ground-based and spaceborne remote sensing instruments for measuring ozone depend on solar radiation"

M6) P2, L27-29: I do not agree to this statement. There are plenty of satellite measurements providing global daily measurements of ozone. Not all satellite instruments are dependent on solar radiation. There are many satellite instruments that are capable of measuring during polar winter.

Response: We understand that there are measurements from satellite instruments that operate in the Arctic winter, such as MLS. We have updated this sentence to describe MLS, because we now use MLS profiles in this study.

"This situation is exacerbated by both the lack of high temporal observations at high latitudes, as well as the difficulty of making quality measurements during winter; many ground-based and spaceborne remote sensing instruments for measuring ozone depend on solar radiation (Bowman, 1989; Hasebe, 1980; Vigouroux et al., 20008; 2015). The Microwave Limb Sounder (MLS) is a spaceborne instrument that measures atmospheric emission, which makes it capable of retrieving ozone over the Arctic (Waters et al. 2006). This capability motivates using MLS retrievals for analysis of stratospheric ozone in Arctic (Manney et al. 2011; Kuttippurath et al. 2012; Wohltmann et al. 2013; Livesey et al. 2015; Strahan & Douglass 2018)."

M7) P3, L19: This needs some more explanation. How is a climatology used to create a vertical profile?

Response: This section has been deleted and replace with text explaining our use of MLS data.

"The Microwave Limb Sounder (MLS) is a spaceborne instrument that measures atmospheric emission, which makes it capable of retrieving ozone continuously since 2004 over the Arctic (Waters et al. 2006). This capability motivates using MLS retrievals for analysis of stratospheric ozone in Arctic (Manney et al. 2011; Kuttippurath et al. 2012; Wohltmann et al. 2013; Livesey et al. 2015; Strahan & Douglass 2018)."

M8) P4, L1: for analyses? Do you mean for these analyses? Response: The sentence has been deleted in the revised version.

M9) P5, L4: Column abundance in the vertical profile sounds a bit weird. Do you mean here "the total amount of ozone in the vertical column"? Response: Yes. We have corrected this.

"The total amount of ozone in the vertical profile is a useful parameter for understanding ozone variations in the atmosphere."

M10) P5, L16: What is the purpose of the extrapolation? To fill the missing levels or to extend the column? Response: As mentioned above in M2, we no longer extrapolate any profile data. Thus, the referring sentence has been removed.

M11) P5, L22-24: I would not put the text into brackets. I would suggest to either

remove the brackets or to put the text into a footnote. Response: This entire paragraph has been deleted.

M12) P5, L25. What do you mean with "is used to 60 km"? Response: This sentence has been deleted.

M13) P6, L34: What do you mean with "using data from the dates of ozonesondes"? Do you mean for the same dates as the ozonesonde measurements? Please rephrase accordingly. Response: Yes, this is what we mean. We have clarified this in the text.

"The monthly averaged values for TP and AO are calculated using data for the same dates as the ozonesonde launches for each station."

M14) P7, L1: The given list of sources rather gives the links to the "data" used for the "proxies" than to the "proxies" themselves. Response: Most of these entries are actually direct links to data that we used for the proxies. In the text we updated the sentence to "data sources and weblinks". "Table 1 lists the data sources and weblinks of these proxies."

M15) P7, L10: What do you mean with forward selection? Response: "Forward selection" is a common technique for predictor selection that is describe in Wilks (2011). Wilks states that "stepwise regression" and "forward selection" are synonymous in the statistical literature.

M16) P7, L29: Minimal in four months? In which month do you get the absolute minimum? Response: This sentence has been removed because Figure 6 has been deleted from the manuscript.

M17) P8, L15: In Fig 7b the minima are not that apparent? Why? Further, it would be worth (especially in Fig 7a) to mark these low areas during these winters for better visibility. Response: Figure 7 has also been deleted from the manuscript.

M18) P8, L26: Also here the text should be either given without parentheses or as a footnote. Response: Done.

“Note that the tropopause is low in the Arctic, so the layer from the ground to 10 km represents primarily values in the troposphere, but also contains some ozone from the lowest portion of the stratosphere. However, we refer to this layer here as the “troposphere” for convenience.”

M19) P9, L8: Winter accumulation? Accumulation of what? Of ozone? Response: Yes, we were referring to ozone accumulation. This has been clarified in the text.

“This pattern represents the well-known springtime maximum in the Arctic, which is caused by winter ozone accumulation that occurs before ozone is transported to the troposphere (Rao, 2003; Rao et al., 2004; Staehelin et al., 2001).”

M20) P9, L18: What is FTS? Has the abbreviation been introduced? Response: As we no longer use the retrieved ozone from FTIR at Ny-Alesund and Kiruna, this sentence has been deleted.

M21) P10; L3: “12 years” or “11 years”? The number of years used in not the same throughout the manuscript. Response: See M1 above. This has been corrected.

M22) P10, L22: Positive of negative → What do you mean? positive “and/or” negative Response: This should have read “positive or negative”. This has been corrected.

“We also list the sign of the slope of the regression fit of each proxy in Table 3 to the left of the R2 value (except for the QBO because this proxy involves multiple terms); the sign of the slope indicates positive or negative correlation between the proxy and the residual time series.”

M23) P10, L24: Time trends? Do you mean time series or trends? Response: We actually mean the “trends in the time series” over the 12-year period. This has been corrected. “The time trends were included in the regression analysis by using Ak = 1 in Eq. 1.”

M24) P11, L19-20: That is to simplify and could be misleading. The photochemical loss ozone is less than temperatures are warmer, because PSC will not form that are a

necessary requirement for the processes leading to ozone depletion. Response: This has been corrected.

“In general, higher stratospheric temperatures in the Arctic lead to less PSCs, which result is less photochemical loss of ozone.”

M25) P12, L4: Please rephrase “changes in final model”. Response: That sentence has been removed.

M26) P12, L10: Also this sentence is formulated in that they that it could easily be misunderstood. The reactions involving the surfaces of Polar Stratospheric Clouds lead to ozone loss not the PSC itself. Response: This has been corrected.

“Heterogeneous reactions on the surfaces of the polar stratospheric clouds contribute to ozone depletion (Rex et al, 2004; Brunner et al., 2006).”

M27) P12, L16: Two times “layer”, thus one “layer” is obsolete. Response: This sentence has been removed.

M28) P12, L22: Caused in part? Isn’t it mostly this process? What other processes are responsible for the seasonal cycle? Response: We have deleted “in part”.

“The Brewer-Dobson circulation is one of the most important processes of ozone transport from the Tropics to the Arctic (Staehelin et al., 2001). The seasonal cycle of ozone in the extratropics is modulated by this circulation (Fusco and Salby, 1999).”

M29) P13, L1: This sentence should be rephrased. Not the equivalent latitude itself affect the ozone. It is just a different way for analysing/presenting the data and if course of the data is plotted on equivalent latitudes instead of latitude, the distribution or profile looks a bit different. Response: We have corrected this.

“Potential vorticity (PV) also affect the ozone concentration. Equivalent latitude (EQL) is an index estimated based on PV that is indicative of ozone (air parcel) transportation on an isentropic level of potential temperature (Danielsen, 1968; Butchart & Remsberg, 1986; Allen & Nakamura, 2003)."

M30) P14, L13: What is GrIS? Please clarify. Response: "GrIS" stands for Greenland Ice Sheet and is defined on page 3 line 17.

"Summit Station is located in central Greenland atop the Greenland Ice Sheet (GrIS) and is the drilling site of the GISP2 ice core."

M31) P14, L17: In global? But in your study only local ozone concentrations are considered. What do the here presented results mean for the ramifications of the Montreal Protocol? Response: This is a valid point. We have deleted this sentence.

Technical corrections: T1) P1, L11: there are few. . .. → there are only few. . ... Response: Done.

"Understanding variations in atmospheric ozone in the Arctic is difficult because there are only a few long-term records of vertical ozone profiles in this region."

T2) P1, L21: due primarily → primarily due Response: Sentence has been removed.

T3) P2, L13: weaken → "weak" or "weakening of the vortex" Response: Done.

"This creates a weakening of the vortex that increases the transport of relatively warm, ozone-rich air into the Arctic (Holton and Tan, 1982)."

T4) P2, L19: arctic atmospheres → Arctic atmosphere Response: Done.

"Although there is strong observational evidence to support this teleconnection between the tropical and Arctic atmosphere, a complete theoretical explanation has proved difficult (Anstey and Shepherd, 2014)."

T5) P2, L23: remove space between parentheses and reference. P2, L28: remove space between parentheses and reference. Response: Done.

"Furthermore, these effects depend on location and can also affect different portions of the atmosphere (Staehelin et al., 2001; Rao, 2003; Rao et al., 2004; Yang et al., 2006;

Vigouroux et al., 2008; 2015)."

T6) P2, L31: for validation → for the validation Response: Done.

"These instruments can be launched year-round and can provide valuable information for the validation of remote sensing instruments aboard satellites."

T7) P3, L5: evaluate → validate Response: Done.

"Ozonesonde profiles from various Arctic stations have been used to study the climatology of the ozone cycle (Rao et al., 2004), the vertical distribution of ozone and its dependence on different proxies (Rao, 2003; Tarasick, 2005; Kivi et al., 2007; Gaudel et al., 2015), trends and annual cycles of ozone (Christiansen et al., 2017), the variability of ozone due to climate change (Rex, 2004), ozone loss and relation to dynamical parameters (Harris et al., 2010), the difference of ozone depletion in the Arctic and Antarctic (Solomon et al., 2014), and to validate other sensor measurements (McDonald et al., 1999; Vigouroux et al., 2008; Ancellet et al., 2016)."

T8) P3, L24: I would suggest to write instead of "this research" rather "this study" or "this research study". Response: Done.

"These drivers are then used to create final models of ozone variations. Section 5 presents the conclusions of this research study."

T9) P4, L4: remove space between parentheses and reference. Response:Done.

"The ozonesondes used here utilize electrochemical concentration cells (ECC) (Komhyr, 1969), manufactured by either Science Pump for Ny-Ålesund or Environmental Science (EN-SCI) for Summit, Alert and Eureka."

T10) P4, L8: ozonesonde → ozonesondes Response: This sentence has been deleted.

T11) P4, L9: then an → then with an Response: This sentence has been deleted.

T12) P4, L27: profiles has significant missing → the profiles has a significant amount of missing values Response: This paragraph has been removed to minimize unnecessary information.

T13) P5, L5: defined using → defined by Response: Done.

“The ozone column density is traditionally defined by the Dobson Unit (DU), which is the thickness of a compressed gas in the atmospheric profile in units of 10 μm at standard temperature and pressure, 1 DU is equivalent to 1 milli-atm-cm, or 2.69 x 1016 molecules/cm2.”

T14) P5, L6: thickness of compressed → thickness of a compressed gas Response: Done.

“The ozone column density is traditionally defined by the Dobson Unit (DU), which is the thickness of a compressed gas in the atmospheric profile in units of 10 μm at standard temperature and pressure, 1 DU is equivalent to 1 milli-atm-cm, or 2.69 x 1016 molecules/cm2.”

T15) P5, L11: Letters and numbers should put in the according sub and subscripts. Response: This sentence has been removed.

T16) P5, L16: What do you mean with “appreciable ozone”. Wouldn’t the right wording be “applicable” or “measurable”. Response: “appreciable ozone” is what we mean. The phrase refers to having enough ozone to be measured and therefore significant ozone concentration to contribute to the total column ozone.

“Merged ozonesondes up to 60 km provide an appropriate dataset to integrate over all layers of the atmosphere that contain appreciable ozone.”

T17) P6, L18: either “retrieved from” or “measured by”. Response: This sentence has been deleted.

T18) P6, L18-19: rephrase sentence to avoid repetition of “which”. Response: This

sentence has been deleted.

T19) P6, L24: in the section 4 → in section 4 Response: This sentence has been deleted.

T20) P6, L25: depend → depends Response: The sentence has been removed.

T21) P6, L28: Write either “below” or “section 4” Response: Done. “These proxies are then used to create a final model for PCO and TCO as described in section 4.4.”

T22) P6, L30: Reference should be given here without parentheses. Response: Done. “This list is similar to that used by previous studies (Brunner et al, 2006; Vigouroux et al, 2015) and contains variables that are known to influence ozone concentrations in the troposphere and stratosphere.”

T23) P7, L28: add comma after “(in DU)”. Response: This sentence has been deleted.

T24) P7, L29: in the ozone → in ozone Response: This sentence has been deleted.

T25) P9, L6: before ozone transport → before ozone is transported Response: Done. “This pattern represents the well-known springtime maximum in the Arctic, which is caused by winter ozone accumulation that occurs before ozone is transported to the troposphere (Rao, 2003; Rao et al., 2004; Staehelin et al., 2001).”

T26) P9, L19: March-September → March to September Response: This sentence has been deleted.

T27) P9, L20: January-November → January to November Response: This sentence has been deleted.

T28) P11, L29: influence → influenced Response: This sentence has been deleted.

T29) P12, L21: impacts → is responsible for Response: This sentence has been deleted.

T30) P12, L29: in upper stratosphere → in the upper stratosphere Response: This sentence has been deleted.

T31) P13, L9: found insignificant → found to be an insignificant Response: This sentence has been deleted.

T32) P14, L14: remove space between parentheses and reference. Response: This sentence has been deleted.

T33) P28, l4: with → which Response: sentence has been rephrased.

"Proxies that improve the R2 by at least 1% and that have a p-value equal or less than 0.05 are added to model."

——————————— Responses to anonymous reviewer #2: Rev_2: The abstract summarizes well the content of the manuscript. It is a regression analysis of a 12-year ozone time series at Summit, Greenland. That's it. There are a number of flaws in this work.

Response: We agree with the Reviewer that it is difficult to draw firm conclusions about Arctic ozone from data at a single station. Thus, in addition to using the ozone profiles from Summit Station, Greenland, we have added ozonesonde data from Alert and Eureka, Canada and Ny-Alesund, Svalbard to our study. This has allowed us to analyze the seasonal cycle and identify drivers of ozone variations that are common at the sites. The manuscript now examines the important drivers for different layers in the atmosphere and for the total column ozone for a particular sector of the Arctic. Thus, the Reviewer's comments have allowed us to expand the scope of the study, and we believe that this has greatly improved the manuscript.

Rev_2_M1) First, the column time series is built using an extrapolation method described as "robust" in the abstract (what does it mean here?). The altitude reached by the balloons does not exceed about 20-25 km during winter (see Figure 3). The only information used to extrapolate to 60 km is climatological. One can extrapolate in various ways, but is still left with climatological values above 20-25 km. Since a signifi-

cant part of the total ozone column is climatological, some of the interannual variability signal is lost. They are essentially analyzing a vertically truncated time series. Sometimes, there is a hint of contradiction: It is stated that Ân ÌĄ Figure 4 shows that these methods agree well for most of the year", giving the impression the extrapolation methods give similar results. But, then, "The lack of an absolute reference for stratospheric ozone over Greenland make it difficult to choose which method is best. Therefore, the average of the four methods is used for subsequent analysis in this study. Âz ÌĞ There is no justification or validation about the overall extrapolation. The reconstructed ozone column should be evaluated against independent data, which brings us to another critical point. They justify the use of this balloon dataset by claiming that "during winter; many remote sensing instruments for measuring ozone depend on solar radiation". However, several satellite instruments do not rely on sunlight, notably MLS. Why not combine/use these well-established datasets directly, or at least use them to evaluate the ozone column reconstructed from climatological extrapolation?

Response: We agree with the Reviewer's comments. In response to the concerns of Reviewer #1 and to answer Reviewer #2's questions, we have eliminated the use of the climatology for constructing the ozone profiles. Instead we have improved the dataset substantially by using ozone retrievals from the Microwave Limb Sounder (MLS) to provide profiles up to 60 km. This has greatly improved the quality of profiles above the burst height of the ozonesondes because we now use MLS data that is nearly coincident in both space and time with the ozonesondes.

Changes are similar to 2nd major comments of reviewer one.

Rev_2_M2) Second, the multiple regression analysis (MLR) is not clear to me. If I have understood correctly, Table 5 shows the fraction of observed variance explained by a number of proxies. No error bars are provided. I keep telling my students that numbers without error bars do not mean much. In addition, one of the hypotheses in MLR is that proxies are not correlated (aliasing issue). The problem is that some of proxies can sometimes be correlated in short time series, even if they are not physically correlated. In addition, some of the proxies used here are physically correlated (even if in a short time series, they might not be), for example VPSC and EHF or the influence of solar variability on high altitude ozone being dependent on the phase of the QBO etc... There is the need for assessing properly the effects of these possible correlations on the results and to estimate error bars. I would recommend not to use the standard errors (not reliable for short time series) but rather a Monte Carlo approach.

Response: We note that the Reviewer found our discussion of the MLR analysis to be unclear. We have attempted to improve our description of the analysis and our overall approach in our edited manuscript. We believe that these changes have greatly improved the paper. Although stepwise multiple regression (SMR) is commonly used in studies of atmospheric ozone variations (Appenzeller et al, 2000; Brunner et al, 2006; Kivi et al, 2007; Vigouroux et al, 2015; Steinbrecht et al, 2017), we have extended this analysis using the additional Arctic sites. By studying multiple sites, we now identify drivers that are common across this sector of the Arctic, which provides additional confidence as to what physical mechanism affect the ozone variations. We have been upfront regarding the correlation between the variables by calculating and discussing the covariance matrix between all of the proxy variables. There are only two variables that have significant correlation between them: the eddy heat flux and the volume of polar stratospheric clouds. Both Brunner et al (2006) and Wohltmann et al (2007) reported this as well and elected to include both variables because they have fundamentally different physical characteristics. As expected, the equivalent latitude (EQL) at 370 K and at 550 K are also correlated, but we avoid any issue with our analysis by not including these variables together; EQL at 370 K is used in the lower two layers of atmosphere, while EQL at 550 K is used in the upper two layers. Because we have chosen to continue to use SMR (based on the multitude of past studies that also have), we have taken the Reviewer's suggestion and report the standard error of our results. These are now listed in the Supplementary Material.

"The seasonal cycles are also shown in Figure 6 as the green curves. The correlation

of determination (R2) values are shown above each panel and represent the variance of the original time series that is explained by the seasonal cycle. These values are also shown in Table 2 for comparison. The seasonal cycle explains over 50% of the variance in both the total and partial column ozone values at all stations except middle stratosphere at Summit Station (47%) and Ny-Ålesund (38%). The R2 value for TCO is highest at Eureka (0.80) and lowest at Ny-Ålesund (0.65). Because the seasonal cycle explains a high percentage of the ozone fluctuations over Eureka, this site may be less susceptible to dynamical and chemical perturbations compared to the other sites. By comparing the R2 values in the different atmospheric layers, we see that the middle stratosphere has the lowest R2 at all the stations except Eureka. This shows that the ozone in the middle stratosphere at these Arctic sites is more susceptible to perturbations than other layers." "By examining the difference between the original time series (black dots) and the seasonal cycles (green lines) in Figure 6, we can see that there is additional variance that remains unexplained. This is motivation to conduct the SMR analysis to identify the most important drivers that are common at the four Arctic sites. To accomplish this, the SMR analysis is performed on the deseasonalized time series" "Stepwise multiple regression (SMR) has been widely used in the past [e.g., Appenzeller et al. (2000); Brunner et al. (2006); Kivi et al. (2007); Mäder et al (2007); Vigouroux et al (2015)] for selecting important variables that affect ozone concentrations. Wohltmann et al (2007) explains some of the issues with using multiple regression to determine atmospheric ozone variations. However, this technique can be inaccurate if there is spurious correlation between the different variables and the deseasonalized ozone time series (Wohltmann et al, 2007). In this study, we use a combination of SMR and the "process-based" approach of Wohltmann et al (2007) to determine the important drivers of ozone variations at the Arctic sites. In particular, SMR is used to determine a set of physical parameters that are important at 3 or more of the sites and are, therefore, common drivers of ozone variations in the Arctic. These variables are then used to derive final models for PCO in each of the four atmospheric layers and for TCO at each site. This procedure then reduces the effect of spurious correlations between variables and deseasonalized ozone time series that is experienced when using SMR only." "The best proxy at each step in the analysis is the one with the largest R2 value, which is at least 1% higher than R2 of previous step. These fits must also have a p-value less than 0.05 to be considered in the analysis. Table 3 summarizes the results for each time series. (More detailed information, such as the regression slopes and standard errors of the slope, can be found in Tables S2-S5 of the Supplementary Material)." "The final set of drivers of Arctic ozone in each layer, as well as the TCO, are defined as those that are common among 3 or more sites based on the SMR analysis. These proxies are then used to create a final model for PCO and TCO as described in section 4.4." "To investigate how the proxies are correlated with each other (Appenzeller et al, 2000; Vigouroux et al, 2015), we calculated the covariance matrix for all combinations of the proxies used in the SMR model and found that most covariances are less than 0.30. However, two correlations were large: EHF-VPSC = 0.66, EQL_370K-EQL_550K = 0.58. In our final regression model EQL at 370 K is used for the troposphere and lower stratosphere, while EQL at 550 K is used for the middle and upper stratosphere. Both EHF and VPSC contributed in many layers and excluding one for the analysis did not significantly improve the contribution of the other. EHF and VPSC exhibit different physical characteristics and both influence stratospheric ozone significantly, so this justifies keeping both proxies in final regression models because both were selected as important (Brunner et al., 2006; Wohltmann et al., 2007). Figure 7 shows the results of the final regression model (red curves). The final models are calculated using Eq. 1, but now including the terms for the seasonal cycle and the important drivers identified in the SMR analysis and shown in Table 4. The final values of R2 for each layer at each station are shown in Table 2, along with the R2 values for the seasonal cycles and the SMR analysis. The improvement in the R2 values from final is shown asïĂăïĄĎ (and is simply the difference between the R2 values of the final model and the seasonal cycle model). Values of ïĄĎ that show improvement in R2 of greater than or equal to 20% are shown as bold values in Table 2. By comparing the values in Table 2 from the SMR and the final model, we can see that a majority

of the final models are within 1 to 2% of the SMR. This is similar to the conclusion of Wohltmann et al (2007) when they compared their process-based model to the SMR analysis performed by Mäder et at (2007). From this, we conclude that our choices of the important drivers of the PCO and TCO values at these Arctic sites are indeed capturing a significant amount of the variability in ozone. Furthermore, the elimination of certain variables from the final model seems justified. For instance, at Eureka the SMR found significant correlation between TP and middle stratospheric ozone and the EQL at 370K and upper stratospheric ozone, which is not seen at the other stations. Nevertheless, the final model explains about 80% of the variance. The final models provide significant improvement over the seasonal cycle model in all cases. In 80% of the cases, the R2 is improved by 10% or more, and 20% of the cases are improved by more than 20%. The final models at each site for TCO explain between 80% and 93% of the variance. The PCO values in the different altitude ranges are improved the most at Summit, with the largest improvements in the troposphere (21%) and the middle stratosphere (32%). In general, the largest improvement at all the sites was in the middle stratosphere. The final models for TCO at Alert, Eureka and Ny-Ålesund show comparable improvement between 13% and 15% with the largest improvements in the troposphere and middle stratosphere at all sites. From the results in Table 2, we conclude that we have identified the important physical drivers of ozone variations at these four Arctic sites and within this sector of the Arctic. As an example of this analysis, Figure 8 shows the time series of deseasonalized ozone and the selected proxies in middle stratosphere over Summit Station. The vertical dash lines show the extreme values of ozone variations, and how they coincide with extreme values in the different proxies. This provides confidence that our approach and the development of final models are identifying important physical processes that affect the ozone variations at these sites. Table 4 shows that TP, EHF, EQL, VPSC and the QBO are all important drivers of ozone variations at these sites, and that all of these proxies are necessary for a complete understanding the variations in total column ozone."

Rev_2_M3) Third, the outcome of the study does not warrant publication in a journal like ACP. Let's have a look at the abstract: "The monthly mean total column ozone reaches a maximum of about 400 DU in April, then decreases to minimum values between 275 and 300 DU in the late summer and early fall. The partial column ozone values peak at different times between late winter and early summer." There is nothing new about seasonal variations of polar column ozone. "There is a positive trend in the total column that is likely due to increases in springtime ozone, however, these trends are not robust given the short period of record". There is a trend but they don't know whether it is significant or indicative of something. At the end, I was wondering what a trend analysis of truncated 12-year time series at a specific location can bring? If the aim is to detect ozone recovery, why not use more data over the entire Arctic region? Then, the results of the MLR on individual proxies are presented in the abstract: "This analysis shows that the variations in total column ozone are due primarily to changes in the tropopause pressure, the quasi-biennial oscillation (QBO), and the volume of polar stratospheric clouds. The eddy heat flux is also important for variations in the partial column ozone in the different altitude regions." Again, it is not possible to see whether the results obtained at this station are significant because there are no error bars, no such results (with error bars) presented for other stations, no comparisons to satellite-based studies or ot other studies, explanations about what the results mean physically. The last sentence illustrates the level of analysis: "The importance of the QBO appears to be a unique characteristic for ozone variations over the Greenland Ice Sheet (when compared to other nearby Arctic Stations) and may be related to the fact that Greenland is particularly sensitive to the phase of the QBO." What does "unique" mean here when compared to 2 other stations only (whose results are not presented and discussed thoroughly)? For the authors, the fact that the QBO is an important driver in the LMR model "may be" related to the fact that Greenland ozone is sensitive to the phase of the QBO. It is self-explanatory, and a bit circular. The authors seem to doubt the MLR results. If ozone was not sensitive to the QBO, it should not appear as significant in the MLR analysis. And, vice-versa. The bottom line is that it is not possible to be conclusive with an MLR over a short time series at a single specific

station. This cannot tell us anything new and "robust" about polar ozone.

Response: The aim of this paper is not to detect ozone recovery, but rather to understand ozone variations and identify important drivers of ozone variations at high-latitude Arctic sites. We report the trends of the time series, but this is not our primary focus. Based on the suggestions of both Reviewers, we have improved the quality of the ozone profiles by using MLS retrievals above the ozonesonde data. Our new Figure 3 shows that the sonde data are quite important and correct a positive bias in the MLS data in the lower atmosphere. Thus, we feel that it is a significant contribution to include the more accurate in situ ozonesonde measurements in the analysis of Arctic ozone. Most of the total column ozone is actually measured by the ozonesondes, and the only layer of partial column ozone that is significantly affected by the MLS retrievals (as opposed to the ozonesonde data) is the upper stratosphere. So we believe that this manuscript is an important contribution because it merges the advantages of both the ozonesonde and the MLS data. As stated above, we agree with the Reviewer that drawing firm conclusions from data at one station is difficult. Therefore, as stated above in Rev_2_M2, we now identify drivers that are common at the sites. We agree with the Reviewer that our conclusion regarding the effect of the QBO on ozone over Summit, Greenland was weak. However, our new results indicate that the QBO is also important in the troposphere and lower stratosphere at the other Arctic sites that we considered. Table 4 now summarizes all of the drivers that are important at 3 or more sites. It can be seen that different sets of drivers are important in the troposphere (TP, EQL, QBO), the lower stratosphere (EHF, VPSC, QBO) and the middle and upper stratosphere (EHF, EQL, VPSC). We believe that these results present a more complete understanding of ozone variations in this sector of the Arctic, and that the Reviewer's suggestions have greatly improved the overall conclusions of the manuscript.

"Abstract. Understanding variations in atmospheric ozone in the Arctic is difficult because there are only a few long-term records of vertical ozone profiles in this region. We present 12 years of ozone profiles from February 2005 to February 2017 at four sites: Summit Station, Greenland, Ny-Ålesund, Svalbard, and Alert and Eureka, Nunavut, Canada. These profiles are created by combining ozonesonde measurements with ozone profile retrievals using data from the Microwave Limb Sounder (MLS). This combination creates a high-quality dataset with low uncertainties by relying on in situ measurements to the maximum altitude of the ozonesondes ($\sim$30 km) and satellite retrievals in the upper atmosphere (up to 60 km). For each station, the total column ozone (TCO) and the partial column ozone (PCO) in four atmospheric layers (troposphere to upper stratosphere) are analyzed. Overall, the seasonal cycles are similar at these sites. However, the TCO over Ny-Ålesund starts to decline two months later than at the other sites. In summer, the PCO in the upper stratosphere over Summit Station is slightly higher than at the other sites and exhibits a higher standard deviation. The decrease in PCO in the middle and upper stratosphere during fall is also lower over Summit Station. The PCO in the lower and middle stratosphere has a higher rate of increase in winter over Eureka. Trend analysis over the 12-year period shows significant trends in most of the layers over Summit and Ny-Ålesund during summer and fall. To understand deseasonalized ozone variations, we identify the most important dynamical drivers of Arctic ozone at each level. These drivers are chosen based on mutual selected proxies at the four sites using stepwise multiple regression analysis of various dynamical parameters with deseasonalized data. The final regression model is able to explain more than 80% of the TCO and more than 70% of the PCO in almost all of the layers. The regression model provides the greatest explanatory value in the middle stratosphere. The important proxies of the deseasonalized ozone time series at the four sites are: tropopause pressure and equivalent latitude at 370 K in the troposphere, the quasi-biennial oscillation in the troposphere and lower stratosphere, the equivalent latitude at 550 K in the middle and upper stratosphere and the eddy heat flux and volume of polar stratospheric clouds throughout the stratosphere."

Rev_2_M4) Fourth, there are too many unnecessary details provided in the text. Everybody knows how to calculate an ozone column from a profile, this could end up in an Annex. Some of the explanations are unclear and longwinded, some parts need to

be rephrased. All the authors should re-read very carefully the entire manuscript. In summary, as it stands, I find it very difficult to be positive. I find the manuscript very far from being acceptable for ACP. Even after tackling the flaws in the methodology, I can't see what the results can bring in terms of new knowledge.

Response: We agree with the Reviewer that we provided too many details in the original draft; Reviewer #1 mentioned this as well. Therefore, in the revised manuscript, we have removed many of the tables and figures. Most of the remaining figures and tables have been updated with results from the additional sites. We believe that the manuscript has been greatly improved by addressing the Reviewer's comments and questions with new results regarding the important drivers of variations in partial and total column ozone at these four high latitude Arctic stations.

"Conclusions. There is continuing debate as to what controls Arctic ozone, and what are the relative contributions of dynamics and photochemistry (Antsey and Shepard, 2014). Understanding what causes variations in Arctic ozone is particularly difficult because there are few long-term records of the vertical profile of ozone in this region. We present 12 years of vertical profiles of ozone over Summit Station, Greenland, Ny-Ålesund, Svalbard, and Alert and Eureka, Nunavut, Canada from February 2005 to February 2017. Ozone profiles are created by merging ozonesonde profiles with ozone retrievals from the Microwave Limb Sounder, creating profiles from the surface to 60 km. This dataset is of high quality because in-situ measurements of ozone are used in the lower atmosphere, which accounts for an overestimation of ozone in this region by MLS. On the other hand, the MLS ozone profiles are quite accurate (2-3%) in the stratosphere (above the maximum altitude reached by the ozonesondes) (Livesey et al., 2017). The analysis of the seasonal cycles at the different sites shows that they are, in general, similar, but that significant differences exist from site-to-site. The TCO exhibits maxima in spring and minima in fall at all the sites. The PCO in the upper stratosphere peaks in summer at all the sites, with slightly larger values at Summit for most months. In the middle stratosphere, the seasonal cycle at Ny-Ålesund is shifted later by about one month, giving a delayed buildup of ozone in spring and decay in summer. The lower stratosphere shows the most significant differences in the seasonal cycle at the four sites, with Summit Station exhibiting an earlier decay in ozone from March to July and Ny-Ålesund showing a delay in ozone decay in summer. The seasonal cycle of tropospheric ozone variations peaks around May for Alert, Eureka and Ny-Ålesund and in March at Summit; Summit also has significantly less ozone in the troposphere due to its high elevation. There are no significant trends in the multi-year annual TCO values at any of the sites. The most significant seasonal trends are seen at Ny-Ålesund with positive trends in the summer and negative trends in the spring and fall; negative trends are also seen at Summit in summer and fall. However, we acknowledge the large uncertainty associated with these trends due to the short period of study. The seasonal cycles at each site explain the majority of ozone fluctuations in the TCO and the PCO in most of the atmospheric layers. However, the seasonal model explained less variations in the middle stratosphere than other atmospheric layers, except over Eureka. We use a two-step approach to first determine the important drivers of ozone variations at the four high-latitude Arctic sites, then use these to develop models that explain the ozone variations. Stepwise multiple regression analysis is performed to determine significant proxies that affect ozone variations over the four sites. If a proxy is chosen at three or more of the four sites, then it is considered an important contributor in this sector of the Arctic. A final regression model is then fit to each time series. The final model is successful in identifying proxies that explain a significant portion of the ozone variance in the deseasonalized time series, with 90% of the models having R2 >= 70% and 40% with R2 >= 80%. The tropopause pressure, equivalent latitude at 370 K and the QBO are important drivers between the surface and 10 km. The QBO, eddy heat flux and the volume of polar stratospheric clouds are important in the lower stratosphere, while the equivalent latitude at 550 K, eddy heat flux, and the volume of polar stratospheric clouds strongly influence the middle and upper stratospheric ozone. The final regression model explains over 80% of the variance in the time

series of total column ozone at the four sites. The contribution from the important drivers is greatest at Summit Station, Greenland in the troposphere (21%) and middle stratosphere (32%). In general, the important drivers explain the greatest variance at all the sites in the middle stratosphere, which is the region of the atmosphere that has the least variance explained by the seasonal cycle. Interestingly, the Arctic Oscillation, solar flux and El Niño-Southern Oscillation are not important for ozone variations in this sector of the Arctic."

Please also note the supplement to this comment:
https://www.atmos-chem-phys-discuss.net/acp-2018-620/acp-2018-620-AC2-supplement.pdf

Interactive comment on Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2018-620, 2018.