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1 **Polar ozone response to energetic particle precipitation over**
2 **decadal time scales: the role of medium-energy electrons**

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9 **Key Points:**

- 10 • Simulations (147 years) with new medium-energy electron forcing analyzed for
11 chemical responses
- 12 • Middle mesospheric ozone is reduced by up to 20% on average by inclusion of
13 MEE forcing
- 14 • Upper stratospheric ozone varies by up to 7% in the SH due to energetic particle
15 precipitation

Abstract

One of the key challenges in polar middle atmosphere research is to quantify the total forcing by energetic particle precipitation (EPP) and assess the related response over solar cycle time scales. This is especially true for electrons having energies between about 30 keV and 1 MeV, so-called medium-energy electrons (MEE), where there has been a persistent lack of adequate description of MEE ionization in chemistry-climate simulations. Here we use the Whole Atmosphere Community Climate Model (WACCM) and include EPP forcing by solar proton events, auroral electron precipitation, and a recently developed model of MEE precipitation. We contrast our results from three ensemble simulations (147 years) in total with those from the fifth phase of the Coupled Model Intercomparison Project (CMIP5) in order to investigate the importance of a more complete description of EPP to the middle atmospheric ozone, odd hydrogen, and odd nitrogen over decadal time scales. Our results indicate average EPP-induced polar ozone variability of 12–24% in the mesosphere, and 5–7% in the middle and upper stratosphere. This variability is in agreement with previously published observations. Analysis of the simulation results indicate the importance of inclusion of MEE in the total EPP forcing: In addition to the major impact on the mesosphere, MEE enhances the stratospheric ozone response by a factor of two. In the Northern Hemisphere, where wintertime dynamical variability is larger than in the Southern Hemisphere, longer simulations are needed in order to reach more robust conclusions.

1 Introduction

Variation in solar ultraviolet (UV) radiation is considered to be the main source of solar driven decadal variability in the stratosphere, influencing the ozone budget and radiative heating in the middle atmosphere [Gray *et al.*, 2010]. There is now growing evidence that solar driven energetic particle precipitation (EPP) is another important source for stratospheric variability [Seppälä *et al.*, 2014; Matthes *et al.*, 2017]. Auroral electron precipitation provides direct forcing at polar thermospheric altitudes (above about 100 km), while solar proton events (SPE) and medium-energy electron (MEE) precipitation generate excess ionization in the polar middle atmosphere (between about 30–80 km). This leads to significant changes in the neutral atmosphere through the formation of odd nitrogen (NO_x) and odd hydrogen (HO_x) [Jackman *et al.*, 2001; Verronen *et al.*, 2011; Funke *et al.*, 2011; Andersson *et al.*, 2012; Fytterer *et al.*, 2015; Arsenovic *et al.*, 2016]. Enhanced production

48 of NO_x and HO_x affects stratospheric and mesospheric ozone (O_3) [Verronen *et al.*, 2006;
49 Seppälä *et al.*, 2007; Jackman *et al.*, 2008; Andersson *et al.*, 2014a], which then has the
50 potential to further influence atmospheric dynamics [Langematz *et al.*, 2005; Baumgaert-
51 ner *et al.*, 2011]. Simulations and analysis of meteorological data have given indications
52 of chemical-dynamical coupling linking the initial EPP-induced response to changes in the
53 lower atmosphere, and ground-level climate variations on a regional scale [Lu *et al.*, 2008;
54 Seppälä *et al.*, 2009; Baumgaertner *et al.*, 2011; Rozanov *et al.*, 2012; Seppälä *et al.*, 2013].
55 It is possible that the impact of EPP on regional climate variability may be comparable or
56 even exceeds the effects arising from solar UV variations [Rozanov *et al.*, 2005; Seppälä
57 and Clilverd, 2014].

58 One of the outstanding challenges in understanding EPP impact on the atmosphere
59 is the role of MEE in the total EPP forcing and the related atmospheric and climate re-
60 sponse. There has been a persistent lack of an adequate description of MEE ionization
61 in atmospheric simulations due to issues in the satellite-based precipitating flux observa-
62 tions [Rodger *et al.*, 2010a]. We know from satellite-based OH observations that there is
63 a direct mesospheric response to MEE at geomagnetic latitudes between about 55 and 75
64 degrees [Verronen *et al.*, 2011; Andersson *et al.*, 2012; Andersson *et al.*, 2014b; Zawedde
65 *et al.*, 2016]. Observations have further shown the resulting effect on mesospheric ozone,
66 both in day-to-day changes during MEE events, and in longer-term variability [Verronen
67 *et al.*, 2013; Andersson *et al.*, 2014a].

68 A major open question concerns the magnitude of the EPP-driven response in strato-
69 spheric ozone over decadal time scales [Sinnhuber *et al.*, 2006]. In order to have an im-
70 pact, NO_x produced in the mesosphere-lower-thermosphere (MLT) region must be trans-
71 ported down to the upper stratosphere inside the polar vortex during wintertime when it
72 is not destroyed by photolysis. NO_x descent has been observed during many winters [Cal-
73 lis and Lambeth, 1998; Siskind *et al.*, 2000; Randall *et al.*, 2009; Päivärinta *et al.*, 2013]
74 and satellite data analysis has shown that NO_x descent occurs practically every winter, in
75 both hemispheres, with significant inter-annual variability seen especially in the Northern
76 Hemisphere (NH) [Seppälä *et al.*, 2007; Funke *et al.*, 2014a,b]. Capturing the observed
77 magnitude of the NO_x descent has been difficult to simulate in models due to incomplete
78 EPP forcing source producing the NO_x including, perhaps most importantly, the missing
79 MEE ionization.

80 On a year-to-year basis, understanding the response of stratospheric ozone to the de-
81 scending NO_x has been challenging because of the relatively large overall ozone variabil-
82 ity due to atmospheric dynamics [Pääviranta *et al.*, 2013]. Nevertheless, from observations
83 we know that polar upper stratospheric ozone can be depleted locally by 40–60% during
84 winters of exceptionally strong NO_x descent [Randall *et al.*, 1998; Randall *et al.*, 2005].
85 A recent study using satellite data between 1979 and 2014 has revealed a long-term re-
86 sponse of Southern Hemispheric (SH) stratospheric ozone to EPP activity, with an aver-
87 age ozone depletion of about 10–15% at 30–45 km altitude in late winter [Damiani *et al.*,
88 2016]. Fytterer *et al.* [2015] used a shorter time period of observations (2005–2010) and
89 reported a 5–10% depletion of SH polar ozone at 25–50 km over the winter months.

90 Up to now there are been few simulations including MEE in some form [Codrescu
91 *et al.*, 1997; Semeniuk *et al.*, 2011], but most recently Arsenovic *et al.* [2016] examined
92 the MEE effect on the polar atmosphere using a chemistry-climate model. Although their
93 MEE ionization data set restricted the simulated time period to just eight years, they nev-
94 ertheless reported substantial MEE effects on polar stratospheric ozone and subsequently
95 on atmospheric dynamics. However, for more general conclusions a multi-decadal time
96 series of simulations is needed.

97 Here we use the Whole Atmosphere Community Climate Model (CESM1(WACCM))
98 to study the polar atmosphere response to EPP over decadal timescales. We present an ex-
99 tended simulation time series of 147 years (3×49 years ensemble of runs) which gives our
100 results good statistical robustness. To complete the EPP forcing over the whole time se-
101 ries, we introduce to WACCM the new state-of-the-art MEE precipitation model which
102 is part of solar forcing recommendation for the sixth phase of the Coupled Model Inter-
103 comparison Project CMIP6 [van de Kamp *et al.*, 2016; Matthes *et al.*, 2017]. The big open
104 questions we wish to address concern the magnitude and detectability (e.g., statistical ro-
105 bustness) of EPP-driven signals in multi-decadal time series. These signals are currently
106 unknown because most previous MEE studies have been restricted to time periods of ~10
107 years or less. Thus, our study is an important contribution to the MEE research, and EPP
108 research in general.

109 Note that we contrast our results to the simulations from the fifth phase of the Cou-
110 pled Model Intercomparison Project (CMIP5) reported by Marsh *et al.* [2013] which were
111 used for the fifth Intergovernmental Panel on Climate Change (IPCC) Assessment Report.

112 The CMIP5 simulations, which include no MEE forcing, are freely available to the com-
113 munity and are widely used. It is very important to establish if a lack of MEE forcing in
114 those simulations (and simulations by other modeling groups for CMIP5) leads to an error
115 in determining the chemical response to external solar and geomagnetic forcing. Thus our
116 results have great significance for any researcher analyzing the solar signal in the CMIP5
117 simulations.

118 **2 Modeling and analysis methods**

119 WACCM is a chemistry-climate general circulation model with vertical domain
120 extending from the surface to 5.9×10^{-6} hPa (~ 140 km geometric height). The stan-
121 dard horizontal resolution used is 1.9° latitude by 2.5° longitude. The representation of
122 WACCM physics in the MLT and simulations of the atmospheric response to solar and
123 geomagnetic forcing variations are described by *Marsh et al.* [2007]. Details of recent
124 centennial-scale coupled simulations using the current version of WACCM (version 4) and
125 an overview of the model climate is presented by *Marsh et al.* [2013]. The chemistry mod-
126 ule in WACCM is interactive with the dynamics through transport, radiative transfer and
127 exothermic heating. Photochemistry associated with ion species (O^+ , NO^+ , O_2^+ , N_2^+ , N^+)
128 is part of the standard chemistry package. For EPP, the standard model uses a lookup ta-
129 ble parameterization for ionization-driven HO_x production, based on the work of *Solomon*
130 *et al.* [1981]. For NO_x , it is assumed that 1.25 N atoms are produced per ion pair with
131 branching ratios of 0.55/0.7 for $N(^4S)/N(^2D)$, respectively [*Porter et al.*, 1976; *Jackman*
132 *et al.*, 2005].

133 Except for the inclusion of MEE in the EPP forcing (described in the next para-
134 graph) the coupled model simulations presented here were set up identically to the CMIP5
135 simulations [for full details, see *Marsh et al.*, 2013]. We utilize the free-running dynam-
136 ics version of the model (compset "B55TRWCN") that includes active ocean and sea ice
137 components at 1° resolution. An ensemble set of three simulations was performed with all
138 observed forcings between 1955–2005. An ensemble of three was chosen to reduce the ef-
139 fects from internal variability in the model in our analysis. The observed forcings include
140 changes in surface concentrations of radiatively active species, daily solar spectral irradi-
141 ance, volcanic sulfate heating, and the Quasi-Biennial Oscillation (QBO). The initial con-
142 ditions for 1955 for all model components were taken from a single historical simulation
143 (1850-2005), in an identical manner to the CMIP5 simulations. Energetic particle forcing

144 due to solar proton events (SPE) and auroral electron (AE) precipitation was included in
 145 the original CMIP5 simulations, hence the difference between the CMIP5 and our simu-
 146 lations is the addition of the new MEE forcing, as described below. The three ensemble
 147 members of simulations (49 years each) result in a total of 147 years for our analysis.

148 The key feature in our simulations is that we have improved the EPP forcing in
 149 WACCM by introducing 30–1000 keV radiation belt electron precipitation using the APEEP
 150 model of *van de Kamp et al.* [2016]. Note that *van de Kamp et al.* [2016] presents two
 151 versions of the MEE precipitation model depending on the geomagnetic activity index
 152 used to determine the MEE variation. Here we utilize the version driven by the A_p index,
 153 from now on referred to as the APEEP model for " A_p -driven Energetic Electron Precipi-
 154 tation". In the 30–1000 keV energy range, electrons provide a major ionization source at
 155 60–90 km altitude, directly affecting mesospheric chemistry. APEEP is a proxy model,
 156 driven solely by the observed geomagnetic A_p index. In the model, A_p defines the level of
 157 magnetospheric disturbance and the location of the plasmapause, both of which are needed
 158 to calculate precipitating electron fluxes in 16 geomagnetic latitude bins between 45° and
 159 72° for each hemisphere. The daily, zonal mean fluxes of precipitating electrons from the
 160 APEEP model were used to calculate atmospheric MEE driven ionization rates [see *van de*
 161 *Kamp et al.*, 2016, for details] which were then included in WACCM. The long-term ion-
 162 ization data sets from the APEEP model are available back to 1850 as an official part of
 163 the solar forcing recommendation for the CMIP6 simulations [*Matthes et al.*, 2017]. The
 164 same ionization data set as described by *Matthes et al.* is used here.

165 Figure 1 (top panel) shows the time series of monthly mean APEEP ionization in
 166 the NH at about 77 km altitude (1.7898×10^{-2} hPa). This correspond to the altitude where
 167 HO_x production in the WACCM simulations maximizes when APEEP is included. Over-
 168 all, the APEEP ionization exhibits a considerable variability during all five solar cycles
 169 (SC19–SC23) with the strongest and most frequent ionization increases occurring during
 170 the declining phase of the solar cycle, in accordance with peaks in geomagnetic activity
 171 levels (not shown). In the APEEP model the electron flux characteristics are identical in
 172 the NH and SH, so that the ionization rates only have differences arising from different
 173 atmospheric conditions. The largest observed NH/SH differences are related to the lon-
 174 gitudinal distribution of fluxes [*Andersson et al.*, 2014b], due to variations in the strength
 175 in the geomagnetic field. Those longitudinal variations are not considered when the zonal
 176 mean APEEP model is used. For the MEE energy range, these differences primarily arise

177 during quiet geomagnetic conditions where weak diffusive scattering processes dominate,
 178 but the magnitude of electron precipitation is very low [e.g. *Rodger et al.*, 2013, Figure 4,
 179 upper panels]. During disturbed conditions, when the magnitudes are 1-2 orders higher,
 180 strong diffusion dominates [e.g. *Horne et al.*, 2009] and no significant differences are ex-
 181 pected with longitude or hemisphere [e.g. *Rodger et al.*, 2013, Figure 4, lower panels]. As
 182 such we expect any error in the modeling caused by using the same fluxes for NH and SH
 183 to be small compared to the overall uncertainties in the APEEP flux model.

184 From now on the WACCM simulations with the APEEP ionization will be referred
 185 to as "MEE_CMIP5" to highlight the addition of MEE forcing to simulations which are
 186 otherwise identical to the CMIP5 simulations. We first contrast the MEE_CMIP5 with the
 187 original CMIP5 simulations (from now on called "REF_CMIP5") and calculate the dif-
 188 ference in HO_x , NO_x and O_3 concentrations. The purpose of this comparison is to get
 189 an overall picture of the impact that including the APEEP ionization has. We will focus
 190 this first part of the analysis on the SH, with the more detailed analysis for both hemi-
 191 spheres in the second part. A monthly mean analysis is made for three selected sets of
 192 years: CASE 1 includes all years (147 altogether from all three 49-year ensemble mem-
 193 bers), CASE 2 includes only the years with high APEEP ionization (36 years in total),
 194 CASE 3 includes only the years with low APEEP ionization (33 years in total). The selec-
 195 tions are based on annual mean APEEP ionization as shown in Table 1. In the top panel
 196 of the Figure 1, red and blue indicate CASE 2 and CASE 3, respectively. The years are
 197 also listed in Table 1.

198 In the second part of the analysis we focus on the decadal variability due to EPP
 199 from SPE, AE, and MEE during winter (NH: December-January-February/DJF. SH: June-
 200 July-August/JJA) – this is when the EPP-driven *in situ* effects are expected to be the most
 201 pronounced. We contrast winters of high and low EPP forcing in the MEE_CMIP5 and
 202 REF_CMIP5 ensembles separately. The analysis is made for two selected sets of years:
 203 1) high wintertime (DJF/NH and JJA/SH) APEEP ionization at 77 km altitude (51 years
 204 in the NH, 48 in the SH), and 2) low wintertime APEEP ionization at 77 km altitude (51
 205 years in the NH, 45 in the SH), based on three-month averages of APEEP ionization. In
 206 Figure 1b (NH) and Figure 1c (SH), colors indicate the winter months of high (red) and
 207 low (blue) APEEP ionization levels. The corresponding years are also listed in Table 2.

208 The above selections were made with the aim to simultaneously a) contrast the ex-
 209 tremes of the high and low APEEP ionization periods in order to identify potential re-
 210 sponses and b) keep the number of years in the sets as large as possible to allow for ro-
 211 bust statistical conclusions. Later, in Section 3, we will discuss how these selections affect
 212 our results. Note that although our selections are based on the APEEP ionization levels,
 213 using the geomagnetic A_p index (which drives both APEEP and AE in WACCM) instead
 214 would lead to very similar year groups. As an indicator of statistical robustness, we have
 215 included the 90% and 95% confidence levels in the figures. These were calculated using
 216 Student's t-test. However, as pointed out e.g. by *Ambaum* [2010], this is not a quantitative
 217 test of significance of our results: a low confidence level does not necessarily imply that
 218 the results have no physical meaning.

219 3 Results

220 3.1 MEE direct effects in the mesosphere

221 The monthly mean impact of the APEEP ionization on SH polar mesospheric HO_x
 222 ($\text{OH} + \text{HO}_2$), NO_x ($\text{NO} + \text{NO}_2$) and O_3 is shown in Figure 2 (VMR, volume mixing ra-
 223 tio) and Figure 3 (corresponding %^o-changes). In Figure 3, the relative difference is ex-
 224 pressed in percents of the REF_CMIP5 VMR. Both figures show results that were aver-
 225 aged zonally, and over the magnetic latitudes 60–90°S. The results are shown as functions
 226 of time (month) and altitude.

227 For each species, the month-altitude impact patterns are similar for the three sets
 228 of years, while the magnitude of the response, and the extent of the 90% and 95% confi-
 229 dence regions clearly depend on the level of APEEP ionization and the number of years
 230 included in the sets. As expected, these confidence regions are most extended for CASE 1,
 231 which includes the largest number of years. For all the species, the magnitude of the re-
 232 sponse is largest for the high APEEP ionization years (CASE 2) and smallest for the low
 233 APEEP ionization years (CASE 3), as expected. In CASE 3, there is a clearly different
 234 NO_x response above 80 km during the summer months (Figure 2, mid-right panel). How-
 235 ever, this response is in the region of lesser statistical robustness and thus could be caused
 236 by background variability.

237 For the high APEEP ionization years (CASE 2), HO_x enhancements of up to 0.6 ppbv
 238 (increase of 20% from REF_CMIP5) are seen during May–July at altitudes between 65

239 and 85 km. When considering CASE 1 (all years) and CASE 3 (low APEEP ionization
 240 years), the VMR response is smaller than for CASE 2 but the magnitude of the changes
 241 produced still exceeds 10%. Outside of these months, the HO_x increases between 60 and
 242 90 km, where the largest concentrations of HO_x are observed in general, are very small.
 243 At altitudes <60 km and >90 km, where the HO_x background is very small, MEE results
 244 in a small reduction. Note that above 90 km there would be an HO_x increase, rather than
 245 decrease, if we also included atomic hydrogen in HO_x (not shown). Thus, the decrease
 246 seen in our plots at these altitudes indicates a change in HO_x partitioning towards H,
 247 caused by the extra production of atomic oxygen by MEE and reactions such as O + OH
 248 → O₂ + H.

249 For NO_x, the APEEP-driven VMR increase peaks at 80–100 km, where it is seen
 250 throughout all seasons. This is consistent with the APEEP ionization typically peaking
 251 around 90 km [*van de Kamp et al.*, 2016]. For the years of high APEEP ionization (CASE 2),
 252 the VMR response reaches 200 ppbv in June–July and is smallest in December–January
 253 (20–50 ppbv). At lower altitudes, there is a clear seasonal cycle with a 20–30 % increase
 254 down to stratopause level **focused on** winter months when NO_x is descending inside the
 255 polar vortex. Above 100 km, NO_x decreases but relatively this effect is very small and not
 256 statistically significant. These effects are similar for the other sets of years, albeit smaller
 257 in magnitude especially for low APEEP ionization (CASE 3).

258 As seen in Figure 3, the NO_x percentage response patterns are quite different from
 259 those of VMR shown in Figure 2. The relative increase is largest during the summer due
 260 to the lower natural NO_x background values, exceeding 200% for CASE 2. During mid-
 261 winter, when NO_x is already enhanced due to AE and descent, the APEEP ionization
 262 leads to an increase of over 20% in the average mesospheric NO_x.

263 For O₃, the VMR response pattern below 85 km is similar to that of HO_x inverted
 264 (so that high HO_x correlates with low O₃), but shifted to lower altitudes and covering a
 265 wider range of altitudes. From March to September, ozone decreases at 60–80 km by up
 266 to 0.2–0.3 ppmv depending on the CASE, with strongest and most extended response seen
 267 for years of high APEEP ionization (CASE 2). Around its secondary maximum (at 90–
 268 100 km), ozone has a response which during spring and autumn months reaches magni-
 269 tudes similar to those seen at lower altitudes. However, in the context of total O_x (O +
 270 O₃) the magnitude of the effect is small because at these altitudes atomic oxygen concen-

271 tration is several orders of magnitude larger than that of ozone. In fact, if we plotted O_x
 272 instead of ozone, we would see an increase rather than a decrease at the secondary maxi-
 273 mum. This is caused by extra atomic oxygen production by MEE. Thus the decrease seen
 274 in ozone indicates a change in the O_x partitioning towards O. In percentage, the meso-
 275 spheric O_3 response is seen during all but the mid summer months and it is strongest
 276 in spring and autumn periods, varying between 10% and 30% in February–October in
 277 CASE 2. The equinox pattern does not coincide with the HO_x increase, indicating that
 278 the NO_x enhancements could have an additional effect on HO_x partitioning and ozone de-
 279 pletion [Verronen and Lehmann, 2015] and could modulate the formation of the tertiary
 280 ozone maximum [Sofieva *et al.*, 2009]. On the other hand, during mid winter the polar
 281 night covers a larger area over the polar cap. Thus the effect of ozone-depleting catalytic
 282 cycles, which depend on solar illumination, should be diminished leading to a smaller
 283 MEE response. The percentage difference is also affected by the background amount of
 284 ozone which is generally higher during winter and results in a smaller relative response.

285 Although not shown, the magnitude of the NH response of mesospheric HO_x and
 286 ozone is very similar to that presented for the SH. For NO_x , the maximum wintertime en-
 287 hancement is somewhat smaller and less pronounced than in the SH, which corresponds to
 288 larger dynamical variability in the NH, including the more frequent occurrence of Sudden
 289 Stratospheric Warming events [Päivärinta *et al.*, 2013]. For all the species, the month-
 290 altitude response patterns in the NH are very similar to those in the SH, except that the
 291 maximum percentage change in ozone peaks in the mid winter instead of the autumn
 292 months, possibly an indication of the earlier formation of the polar vortex in the SH.

293 **3.2 MEE indirect effects in the stratosphere**

294 Figure 4 shows the monthly mean APEEP impact on NO_x and O_3 in the SH polar
 295 stratosphere and lower mesosphere (15–65 km) as %-change (like Figure 3, but lower al-
 296 titude range). The electron energy range used in the APEEP ionization model provides
 297 direct forcing only at altitudes above 60 km, so the stratospheric response is entirely due
 298 to a) transport of APEEP- NO_x from above, b) chemical-dynamical coupling, or c) combi-
 299 nation of a and b. A tongue-like structure of excess APEEP- NO_x descends from the lower
 300 mesosphere starting in autumn, causing an ozone decrease in the stratosphere. The mag-
 301 nitude of the response is largest for the years of high APEEP ionization (CASE 2) and
 302 smallest for the low APEEP years (CASE 3). Because there is no direct MEE effect in the

303 stratosphere, the early winter increase around 30 km must be related to descending NO_x ,
 304 some of which remains over the summer months. Note that a similar early winter EPP
 305 effect also appears to be present in NO_x experimental observations [Funke *et al.*, 2014a,
 306 Figure 9].

307 The descending APEEP- NO_x reaches altitudes as low as 30 km by November with
 308 the maximum increase being 10–20% depending on the CASE. Corresponding ozone
 309 decreases of 5–8% are seen at altitudes between 30–50 km in all CASES. For CASE 1
 310 and 2, part of the stratospheric ozone response (a decrease) is within the 90-95 % sig-
 311 nificance region. In CASE 3, none of the ozone response below 50 km is statistically ro-
 312 bust, which may indicate a larger variation in percentages for CASE 3, probably due to
 313 the lower average background ionization in this case. Nevertheless, stratospheric NO_x and
 314 ozone are affected in years of low APEEP ionization even though the direct APEEP forc-
 315 ing is restricted to altitudes above 60 km. Above 55 km, the direct effect of the ozone
 316 response (see previous section) is influenced by both HO_x and NO_x increases.

317 To consider the robustness of the ozone response in the middle atmosphere, Fig-
 318 ure 5 shows a statistical analysis of the wintertime APEEP impact on ozone, both in VMR
 319 and percentages, as a function of altitude. The responses were averaged over SH polar lat-
 320 itudes of 60–90°, and over the months of June to August in the ensembles. The month
 321 selection covers the period of strongest, most robust ozone response in the stratosphere (as
 322 seen in Figure 4). The graphs also include the standard error of the mean (SEM) of the
 323 difference, calculated as

$$SEM = \sqrt{\frac{STD_1^2 + STD_2^2}{n}} \quad (1)$$

324 where STD_1 and STD_2 are yearly standard deviations of the MEE_CMIP5 and REF_CMIP5
 325 simulations, respectively, and n is the number of years.

326 At mesospheric altitudes, ozone loss is connected directly to APEEP ionization and
 327 the resulting HO_x increase, and this response is generally very robust. This is demon-
 328 strated through the SEM being clearly smaller than the magnitude of the response. In
 329 the stratosphere, the decrease in ozone is caused by the descent of APEEP- NO_x and is
 330 strongly affected by dynamical variability. At 30–50 km, the SEM becomes comparable
 331 to the magnitude of the response. The SEM increases with decreasing number of included
 332 years, thus the ozone response is clearly most robust for CASE 1 which includes all years.

333 For years of high and low APEEP ionization, the response exceeds the SEM above 40 km
 334 and at 30–40 km, respectively.

335 **3.3 Decadal variability due to EPP in mesosphere and stratosphere**

336 In this section, we will investigate the variability of HO_x, NO_x, and ozone by ana-
 337 lyzing the differences between the responses for high and low EPP ionization winters as
 338 listed in Table 2. Figures 6 and 7 present the wintertime HO_x and ozone variability at
 339 altitudes between 70 and 80 km for the NH and the SH, respectively.

340 Results from MEE_CMIP5 (panels a and c) show clear differences between high
 341 EPP and low EPP winters in both hemispheres. At geomagnetic latitudes directly affected
 342 by radiation belt electrons (55–72°), there is up to 15% more HO_x in high EPP winters
 343 (panel a). The zonal asymmetry seen in the HO_x distribution is caused by different illumi-
 344 nation conditions over the affected geomagnetic latitudes, i.e. at lower geographic latitudes
 345 the higher level of solar-driven water vapor photodissociation leads to higher amounts of
 346 background HO_x and smaller EPP response in relative terms. The strongest ozone vari-
 347 ation coincides with the largest HO_x variation, with ozone decreases of about 8% in the
 348 NH and 10% in the SH.

349 On the other hand, the results from REF_CMIP5 (panels b and d), which does not
 350 include direct APEEP ionization in the mesosphere, are clearly different. Here, the NH
 351 HO_x and ozone generally lack a clear correlation pattern. In the SH in the REF_CMIP5,
 352 around 10% increase in HO_x is seen at high geomagnetic latitudes, higher than the outer
 353 radiation belt latitudes (panel 7b), during high EPP winters. This is likely caused by a
 354 combination of production due to SPEs and changes in HO_x partitioning due to increased
 355 NO_x [Verronen and Lehmann, 2015]. In this case the corresponding ozone decrease is less
 356 than 5% and is outside of the 90% confidence limit (panel 7d).

357 Figures 8 and 9 present the NO_x and ozone variability (%) in the stratosphere–lower
 358 mesosphere at high polar latitudes in the NH (≥70°) and SH (≥60°), respectively. In the
 359 NH, a smaller latitude range was used because the area of the polar vortex (which we
 360 wanted to cover in wintertime) is typically smaller there than in the SH. Note, however,
 361 that the results for 70–90°N (shown in Figure 8) are very similar to those for 60–90°N
 362 (not shown). Both Figures 8 and 9 display the full 12 month progression, with winter
 363 months placed in the middle of the x-axis to ease comparison.

364 In the NH (Figure 8) the dynamical variability is much stronger than in the SH and
 365 includes sudden stratospheric warmings [*Päivärinta et al.*, 2013]. As a result the response
 366 to MEE is less pronounced than in the SH (Figure 9) [*Funke et al.*, 2014a,b]. Although
 367 individual winters may show strong NO_x descent, the signal becomes less clear when av-
 368 eraged over decadal time scales, even when APEEP ionization is included. As a result of
 369 the dominating dynamical variability in the NH the timing of the descent can also vary
 370 from year-to-year much more than in the SH, which easily leads to smearing of the signal
 371 when averaging. We note that the early winter NO_x enhancement signal in both experi-
 372 ments is due to the so-called Halloween SPEs in 2003.

373 In the SH (Figure 9), the NO_x difference between high and low EPP winters is clear
 374 in both MEE_CMIP5 and REF_CMIP5 simulations. The difference shows a pattern of de-
 375 scending NO_x from early winter (April) to early summer (December) with and without the
 376 APEEP ionization. The inclusion of the APEEP ionization significantly adds to this NO_x
 377 variability - the highest variability goes from 50% to 70%. For the MEE_CMIP5 results
 378 in Figure 9a, the NO_x increase during High EPP forcing at 30–50 km is between 40–70%.
 379 The corresponding REF_CMIP5 signature (Figure 9b), which is due to the descent of AE-
 380 produced NO_x, is between 30% and 50%.

381 Stratospheric ozone loss coincides with the NO_x descent in both Figures 9c and 9d.
 382 During high EPP and from early winter (April) to early summer (December), there is up
 383 to 7% and 2% less ozone at 25–50 km with and without APEEP, respectively. Although
 384 the response patterns are similar, in the MEE_CMIP5 results the effect is much stronger
 385 and statistically significant. As a clear pattern in both simulations, the ozone depletion
 386 persists throughout the summer, descending in altitude and decreasing in magnitude with
 387 time, with final remnants seen until early next winter at about 25 km. The fact that the
 388 late summer signal seems to be more robust in the REF_CMIP5 simulation could be sim-
 389 ply caused by internal model variability. The increase of ozone peaking at about 30 km in
 390 August-October, is caused by the enhanced NO_x converting active chlorine and bromine
 391 to their reservoir species, which leads to less ozone loss by catalytic reactions [*Jackman*
 392 *et al.*, 2009].

393 When considering the difference between high and low EPP years in the MEE_CMIP5
 394 simulation, in the mesosphere the HO_x and ozone signal is strong and only weakly depen-
 395 dent on the number of years included in the analysis (not shown). By using stricter selec-

396 tion criteria, leading to a smaller number of winters with larger differences in EPP forcing,
397 the HO_x and ozone response gets consistently stronger for the latitudes affected by outer
398 radiation belt electrons. However, this is not the case when considering the stratospheric
399 difference. The selection criteria are much more a critical issue, e.g., as reducing the num-
400 ber of years results in an ozone response which is not necessarily stronger but quickly be-
401 comes statistically less robust (i.e. does not reach the 95% confidence level). For example,
402 this happens in the SH when the total number of years is reduced from 50 to about 30.
403 This indicates that a time series of considerable length, extending over several decades, is
404 needed to robustly identify the signal.

405 In our analysis, we are implicitly assuming the 147 individual years as samples of
406 the same population. If the response is not invariant over the timeseries, it would add to
407 the variance and lead to an underestimate of the statistical significance of the response to
408 MEE and EPP in general. And if there are any large trends, we could be overestimating
409 the background variability, which would in fact make the response harder to detect. The
410 fact that we still see a statistically significant response implies that the signal is probably
411 stronger and more robust rather than the other way around. It also shows that the signal
412 could be detectable in a real, observational timeseries rather than in an idealized constant
413 forcing scenario, for example.

414 **4 Discussion**

415 Our results can be compared to previous studies although it should be carefully
416 noted that these typically consider only a portion of our 147-year (3×49 year ensemble)
417 time series due to, e.g., limited availability of experimental data and/or forcing data for at-
418 mospheric simulations. Overall, there is a qualitative agreement with previous simulation
419 studies and satellite-based observations which suggested a clear EPP-driven impact and an
420 important role for MEE in the polar middle atmosphere.

421 Our results on the APEEP ionization impact on mesospheric HO_x and O₃ are in
422 very good agreement with satellite observations. The magnitude of our simulated HO_x
423 responses (0.3–0.6 ppmv) as well as their spatial distributions are similar to the results
424 based on satellite data analysis [Andersson *et al.*, 2014b; Zawedde *et al.*, 2016]. Also the
425 magnitude of our simulated mesospheric ozone variability over decadal time scales agrees
426 well with observations [Andersson *et al.*, 2014a]. This seems to indicate that the level of

427 the APEEP forcing, which directly affects the mesosphere in our simulations, is reasonable
428 – at least in the middle and upper mesosphere where the APEEP ionization peaks.

429 In the SH upper stratosphere we found an EPP-driven decadal variability of up to
430 70% in NO_x and up to 7% in ozone. The magnitude of the ozone response is within but
431 at lower end of the 5–15% range of response obtained from satellite data analysis [*Fyt-*
432 *terer et al.*, 2015; *Damiani et al.*, 2016] and the 3–20% range from previous simulations
433 [*Baumgaertner et al.*, 2011; *Semeniuk et al.*, 2011; *Rozanov et al.*, 2012]. Compared to
434 previous work our study uses fully time-dependent EPP forcing and provides the longest
435 analyzed time series so far, extending almost five solar cycles, giving us better statistical
436 robustness and allowing for more general conclusions.

437 The MEE ionization, which directly affects the polar mesosphere, has been a major
438 source of uncertainty in the EPP forcing used in earlier simulations. As our results now
439 indicate, simulations using the APEEP model generally agree better with the observed
440 ozone response, in both the mesosphere and the stratosphere. As the comparison to the
441 earlier CMIP5 simulations (without MEE) shows, the decadal polar ozone response de-
442 pends very much on MEE, and any analysis based on those CMIP5 simulations will sig-
443 nificantly underestimate the EPP signal. In the forthcoming CMIP6 simulations, it is likely
444 that the situation will drastically improve as the APEEP model is part of the official solar
445 forcing recommendation.

446 The amount of the descending EPP- NO_x is clearly important for the magnitude of
447 the stratospheric ozone response. In WACCM, underestimation of polar mesospheric NO_x
448 has been reported, likely caused by some combination of missing *in-situ* production by
449 EPP and also weak transport of NO_x from the lower thermosphere [*Randall et al.*, 2015].
450 Further model development is needed to better simulate dynamically perturbed winters
451 and improve the mesosphere-to-stratosphere descent in high-top models such as WACCM
452 [*Funke et al.*, 2017]. MEE is included in our simulations through the APEEP model. This
453 work is therefore a significant contribution towards understanding the importance of the
454 missing MEE. It is likely that the production and transport of lower thermospheric NO_x
455 is the primary remaining issue leading to any NO_x underestimation. It should be noted
456 that in the WACCM simulations of *Randall et al.* [2015] and *Funke et al.* [2017] the model
457 dynamics were nudged to the MERRA reanalysis data, and these studies considered just
458 two individual, highly-disturbed NH winters. Therefore, as we are using WACCM with

459 free-running dynamics and consider a time series of 147 years for both hemispheres, those
460 previously reported NO_x issues should not be critically affecting our results. Additional
461 adjustment of EPP- NO_x may also be achieved by including the lower ionospheric (D-
462 region) chemistry which is shown to increase the production in the mesosphere [Anderson
463 et al., 2016]. One might also consider the inclusion of relativistic electron precipita-
464 tion (>1 MeV) which would be expected to directly impact stratopause altitudes. Finally,
465 enhanced eddy diffusion in the mesosphere-lower thermosphere region would increase
466 the transport of auroral NO_x into the mesosphere and below, which seems to yield bet-
467 ter agreement with observations [Meraner and Schmidt, 2016][Matthes et al., 2017, Fig-
468 ure 13].

469 5 Conclusions

470 Here we have introduced long-term MEE forcing to the Whole Atmosphere Com-
471 munity Climate Model (CESM/WACCM). We simulated EPP-driven variability, including
472 the new MEE forcing, in polar ozone over a period of 147 years (3-member ensemble of
473 49-year simulations). The results were compared with those from the CMIP5 climate sim-
474 ulations in order to study the contribution of the additional MEE forcing. The main results
475 can be summarized as follows.

- 476 • EPP-driven variability in mesospheric HO_x and ozone is clear in both hemispheres:
477 the ozone difference between high and low EPP winters varies from 8% to 10% at
478 70–80 km (less ozone when EPP is high).
- 479 • Stratospheric ozone response is distinct in the SH: EPP-driven ozone variability of
480 2–7% is seen down to about 25–35 km.
- 481 • The contribution of MEE is very important to the total EPP-driven response. In
482 the mesosphere, there is either a small or no clear response in HO_x and ozone
483 without the inclusion of direct ionization by MEE. In the stratosphere, inclusion of
484 MEE enhances the response in NO_x and ozone by a factor of about two.
- 485 • Our study indicates that in order to assess the indirect EPP effect in the strato-
486 sphere in a robust way, multi-decadal simulations are needed to overcome the levels
487 of dynamical variability in the model.

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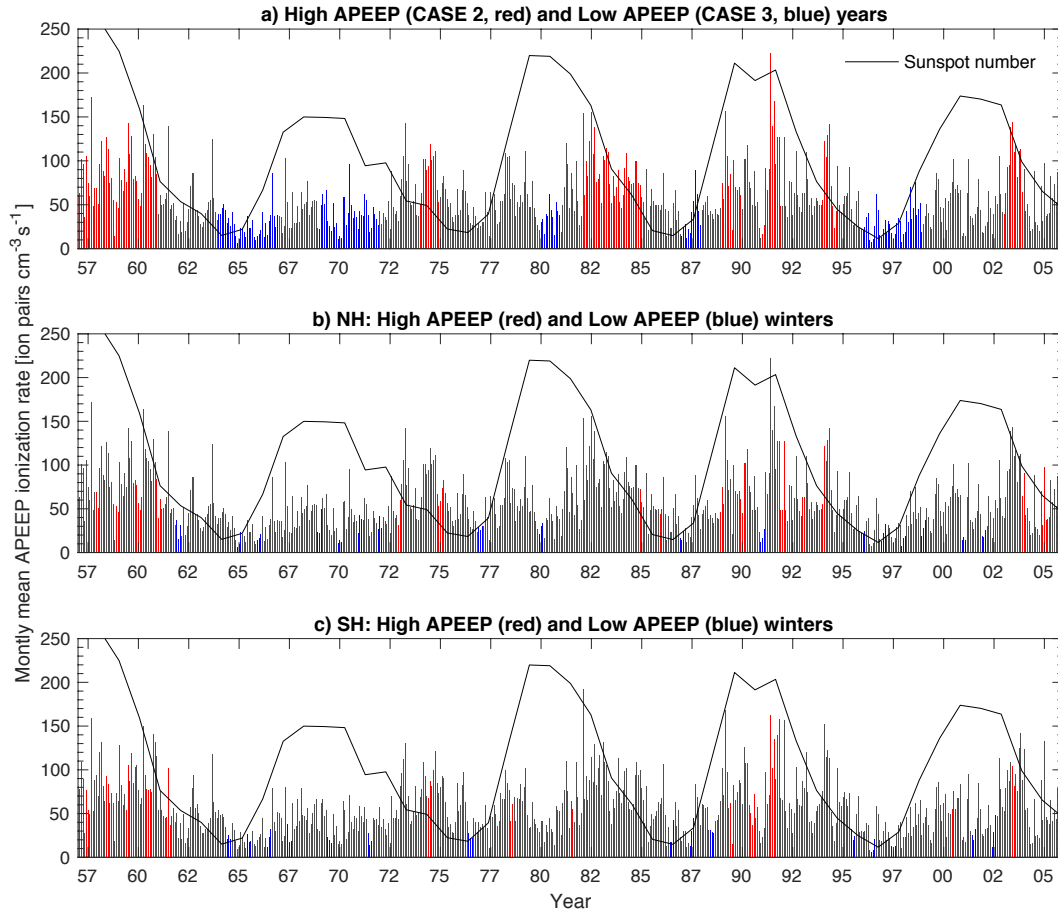
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689 **Table 1.** Selected sets of years for the analysis of the impact due to the APEEP ionization. The selection cri-
 690 teria for CASE 2 and 3 **are** based on the annual mean ionization rate at ≈ 77 km altitude (1.7898×10^{-2} hPa).
 691 This produces two groups of years that are roughly the same size but have a clear separation in average ioniza-
 692 tion rate levels.

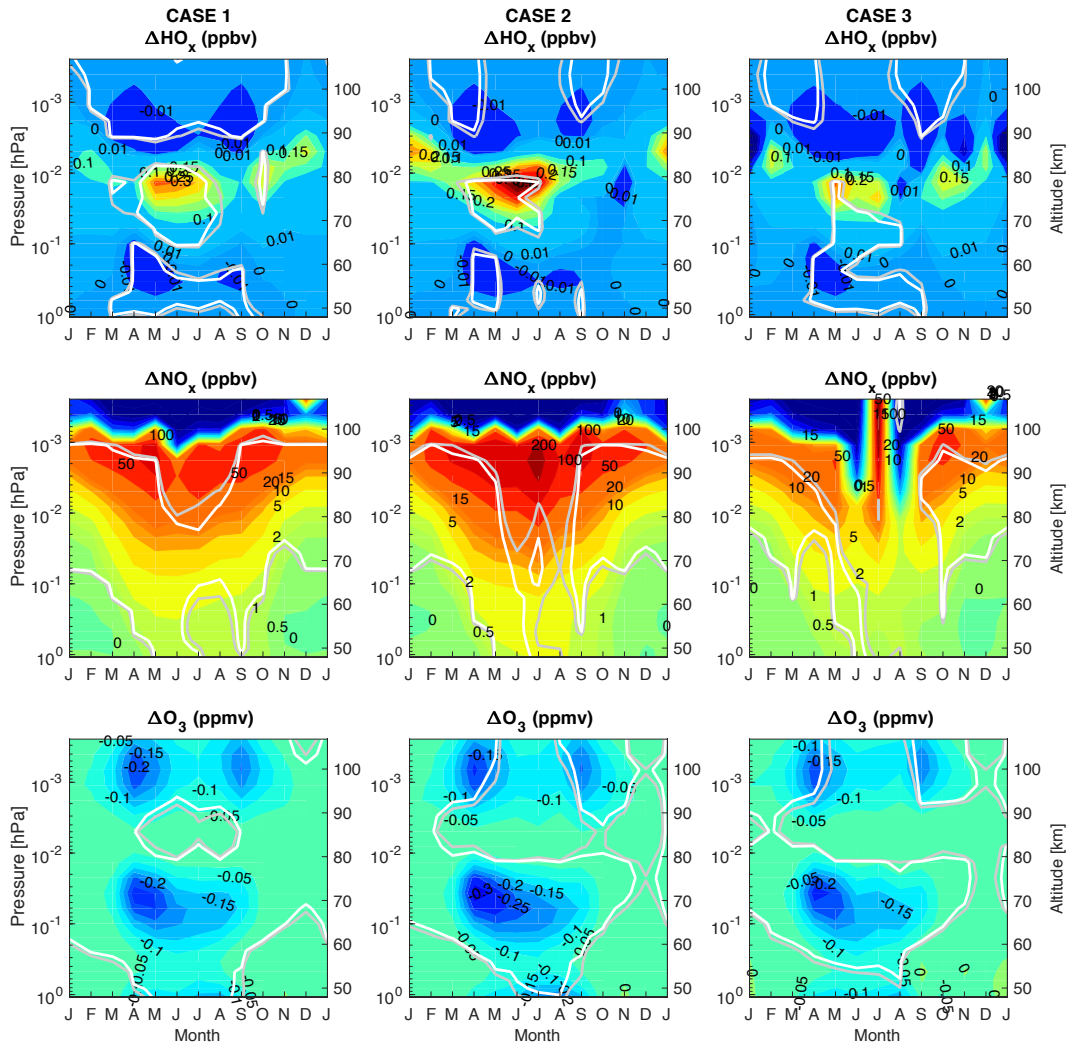
Set	Ionization rate		# years
	selection criteria	Years	
	[ion pairs $\text{cm}^{-3}\text{s}^{-1}$ @77km]		
CASE 1	–	All years: 1957-2005	147
CASE 2	annual mean > 75	1957-60, 1974, 1982-84, 1989, 1991, 1994, 2003	36
CASE 3	annual mean < 40	1964-66, 1969-71, 1980, 1987, 1996-98	33

693 **Table 2.** Selected sets of high and low EPP years for the analysis of EPP-driven variability in mesosphere
 694 and stratosphere. The selection limits are set at the median of the APEEP ionization at ≈ 77 km altitude
 695 (1.7898×10^{-2} hPa) over winter season ± 10 ion pairs/cm³/s, separately for the two hemispheres. For the NH,
 696 the years listed correspond to the year of the December e.g. DJF 1974 = December 1974 – February 1975.
 697 The number of years is the total from all ensemble members, i.e. three times the number of years listed.

Set	Ionization rate		# years
	selection criteria	Years	
	[ion pairs cm ⁻³ s ⁻¹ @77km]		
High NH	DJF > 55	1957-60, 1972, 1974, 1981-82, 1984-85, 1988-89, 1991-93, 2003, 2004	51
High SH	JJA > 50	1957-61, 1974, 1978, 1981-84, 1989-91, 2000, 2003	48
Low NH	DJF < 35	1961, 1964-65, 1968-71, 1976, 1979, 1986, 1990, 1995-98, 2000-01	51
Low SH	JJA < 30	1964-67, 1969, 1971, 1976, 1986-88, 1995-97, 2001-02	45



698 **Figure 1.** Monthly mean ionization rates at 77 km altitude and L-shell range 3.25–10 (magnetic latitude
 699 55–72°) from the APEEP model. The black line is the annual mean sunspot number (values given on y-axis)
 700 indicating the progression of the 11-year solar cycle. a) Red and blue bars indicate years of high MEE (CASE
 701 2) and low MEE (CASE 3) as in Table 1, respectively. b) Red and blue bars indicate high and low MEE win-
 702 ters in the Northern Hemisphere (see Table 2), respectively. c) Same as b) but for the Southern Hemisphere
 703 (see Table 2).



704 **Figure 2.** Monthly mean polar SH (60–90°S) HO_x (top, ppbv), NO_x (middle, ppbv) and O₃ (bottom,
 705 ppmv) composite difference "MEE_CMIP5 – REF_CMIP5". The data are from all ensemble members for
 706 CASE 1 (left panel, all years), CASE 2 (middle, High APEEP ionization), and CASE 3 (right, Low APEEP
 707 ionization). The gray and white contours represent the 90% and 95% confidence levels respectively. Note that
 708 winter months are in the middle of the x-axis.

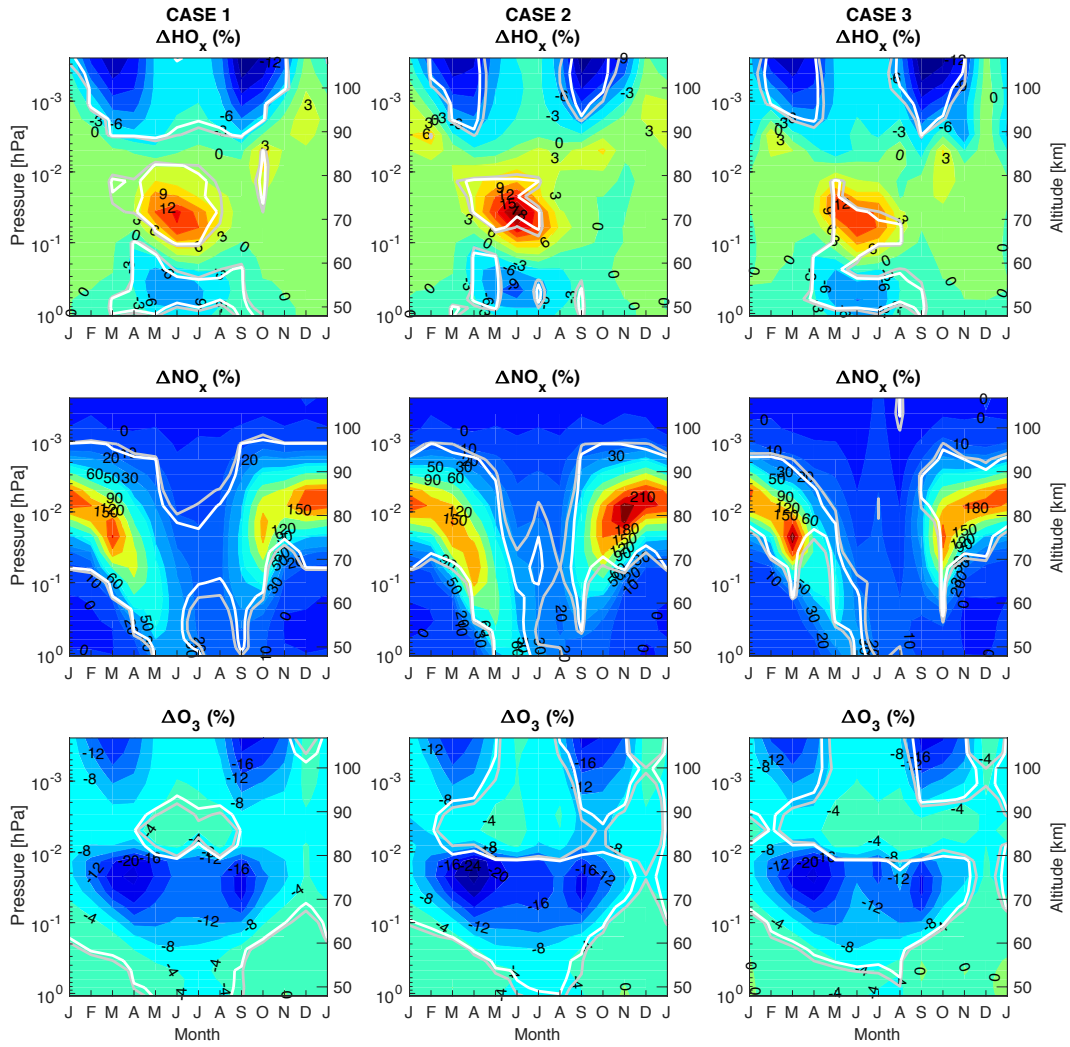
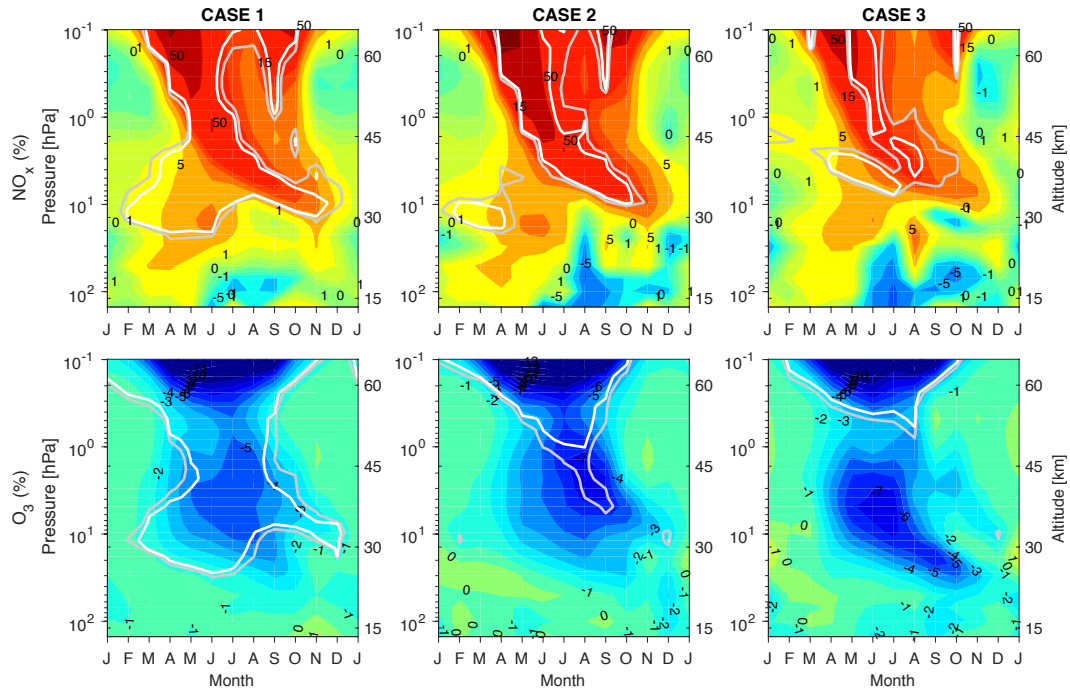
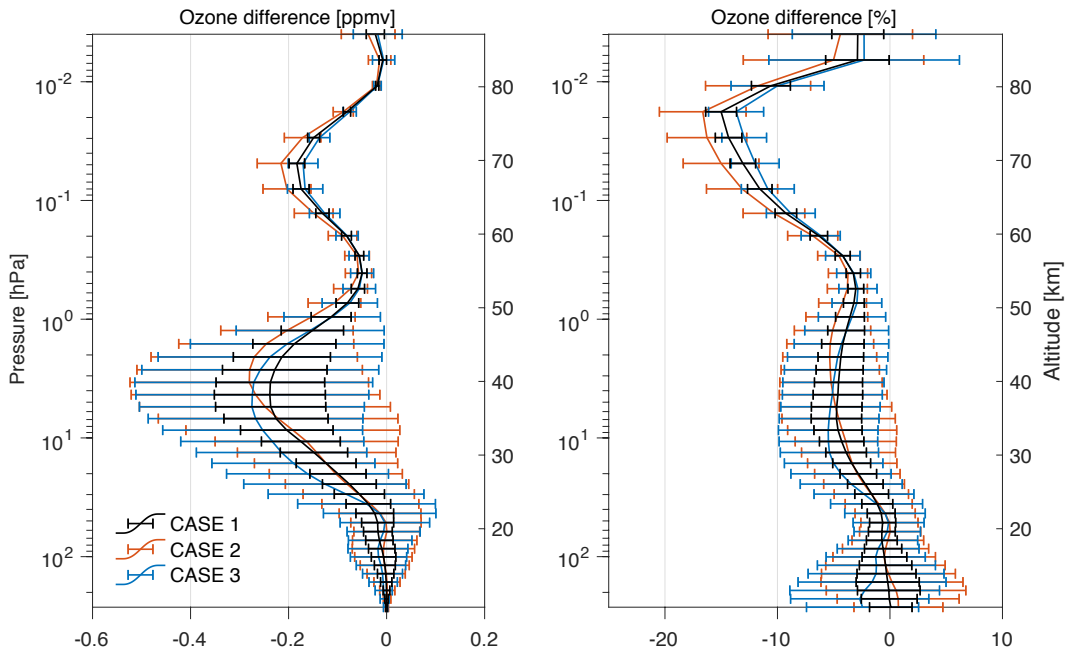


Figure 3. Same as Figure 2 but in relative to the REF_CMIP5 results (%-change).

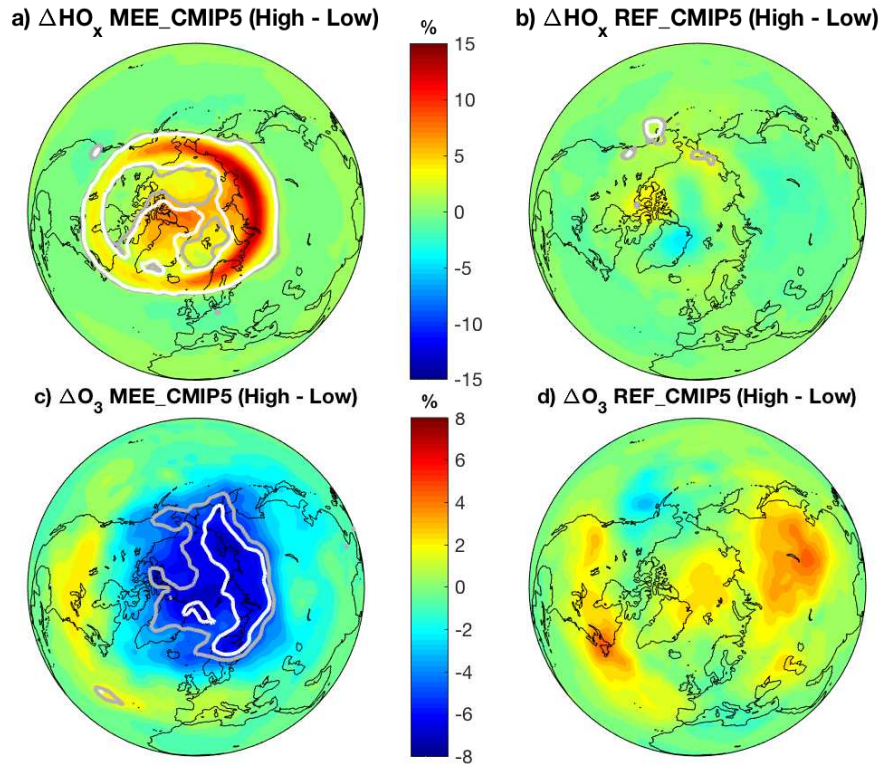
709



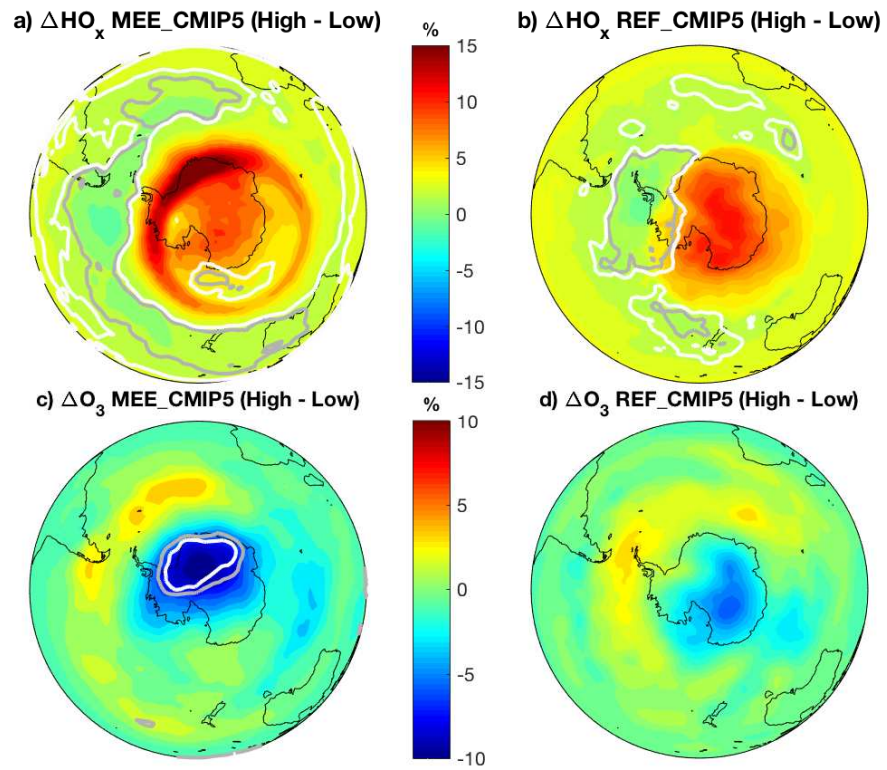
710 **Figure 4.** Monthly mean NO_x (top panels) and O_3 (bottom panels) response to the ionization from the
 711 APEEP model, calculated as percent of the composite difference "MEE_CMIP5 – REF_CMIP5". The data
 712 are from the SH, averaged over latitudinal range 60–90°S and over all ensemble members for CASE 1 (left, all
 713 years), CASE 2 (middle, High APEEP ionization), and CASE 3 (right, Low APEEP ionization). The gray and
 714 white contours represent the 90% and 95% confidence levels respectively.



715 **Figure 5.** SH winter (June–August) zonal mean O₃ response to the ionization from the APEEP model,
 716 calculated as difference between the MEE_CMIP5 and REF_CMIP5 simulations. The data were averaged
 717 over the latitudinal range 60–90°S and over all ensemble members. Horizontal bars indicate the standard error
 718 of the mean (SEM) of the difference (see text for details).

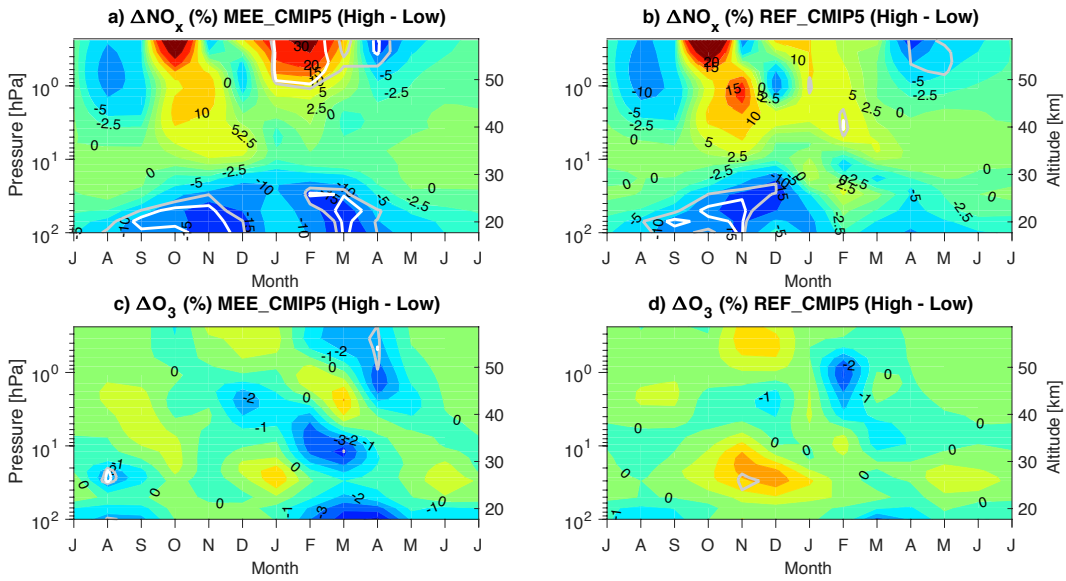


719 **Figure 6.** NH winter (December–January–February) "High EPP – Low EPP" composite HO_x (top) and O_3
 720 (bottom) %-differences for the MEE_CMIP5 simulation (left) and REF_CMIP5 simulation (right) in the upper
 721 mesosphere at 70–80 km altitude. The gray and white contours represent the 90% and 95% confidence levels,
 722 respectively. For list of years in each composite group see Table 2.

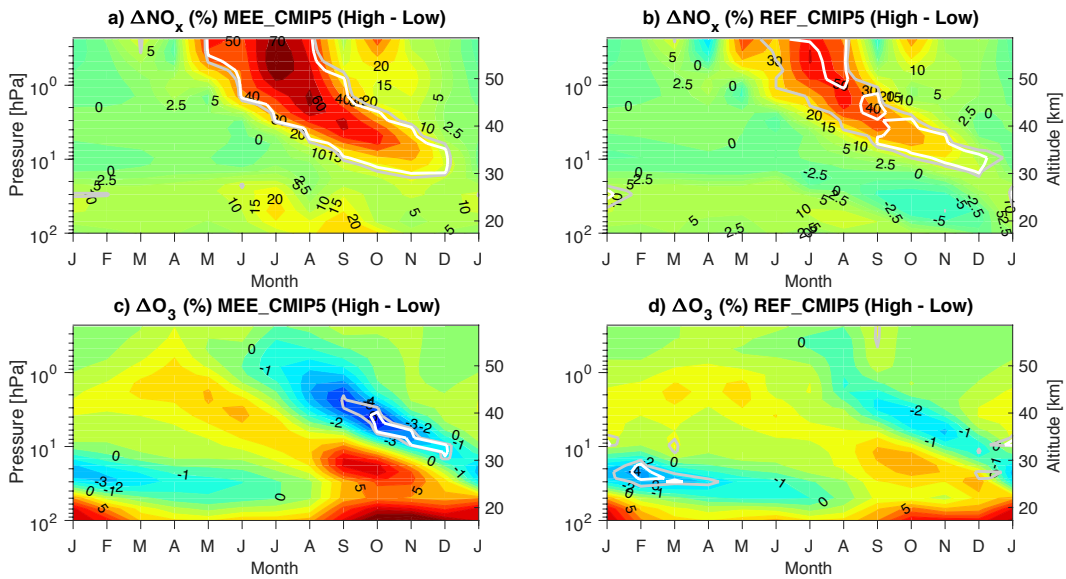


723

Figure 7. As Figure 6, but for SH winter (June–July–August).



724 **Figure 8.** Monthly mean NH polar (70° – 90° N) EPP-driven NO_x (top) and ozone (bottom) variability:
 725 "High EPP – Low EPP" (shown as %-difference). Left: MEE_CMIP5 simulation. Right: REF_CMIP5 sim-
 726 ulation. The gray and white contours represent the 90% and 95% confidence levels, respectively. For list of
 727 years in each composite group see Table 2. The early winter NO_x enhancement visible on both experiments is
 728 a result of the Halloween 2003 SPEs being included in the total EPP forcing for "High EPP" years. Note that
 729 winter months are in the middle of the x-axis to ease comparison with Figure 9.



730 **Figure 9.** As Figure 8 but for the SH (60° – 90° S). For list of years in each composite group see Table 2.