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Variations of Arctic winter ozone from the LIMS Level 3 dataset

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19

20 **Abstract**

21 The Nimbus 7 limb infrared monitor of the stratosphere (LIMS) instrument operated from
22 October 25, 1978, through May 28, 1979. Its Version 6 (V6) profiles and their Level 3 or zonal
23 Fourier coefficient products have been characterized and archived in 2008 and in 2011,
24 respectively. This paper focuses on the value and use of daily ozone maps from Level 3, based
25 on a gridding of its zonal coefficients. We present maps of V6 ozone on pressure surfaces and
26 compare them with several rocket-borne chemiluminescent ozone measurements that extend into
27 the lower mesosphere. We illustrate how the synoptic maps of V6 ozone and temperature are an
28 important aid in interpreting satellite limb-infrared emission versus local measurements,
29 especially when they occur during dynamically active periods of northern hemisphere winter. A
30 map sequence spanning the minor stratospheric warmings of late January and early February
31 characterizes the evolution of a low ozone pocket (LOP) at that time. We also present time
32 series of the wintertime tertiary ozone maximum and its associated zonally varying temperatures
33 in the upper mesosphere. These examples provide guidance to researchers for further
34 exploratory analyses of the daily maps of middle atmosphere ozone from LIMS.

35

36 **1 Introduction and objectives**

37 The historic Nimbus 7 Limb Infrared Monitor of the Stratosphere (LIMS) experiment provided
38 data on middle atmosphere ozone from October 25, 1978, through May 28, 1979, for scientific
39 analysis and for comparisons with atmospheric models (Gille and Russell, 1984). Ozone is an
40 excellent tracer of stratospheric transport in the high latitude stratosphere. As an early example,
41 Leovy et al. (1985) showed how daily maps of the LIMS ozone fields correlate well with
42 geopotential height (GPH) fields on the 10-hPa pressure surface. They also reported on the
43 rapidly changing effects of wave activity on ozone, which led to a better understanding of
44 stratospheric transport processes within models. Hitchman et al. (1989) also analyzed the
45 temperature fields from LIMS and reported on Arctic observations of an elevated stratopause in
46 late autumn to early winter that they associated with momentum forcings from gravity waves.

47

48 Current research focuses on the 3-dimensional character of ozone in the upper stratosphere and
49 mesosphere, based on more recent satellite datasets. Several studies consider how temperature
50 and ozone vary in association with sudden stratospheric warming (SSW) events (Smith et al.,
51 2009; de la Camara et al., 2018; Kim et al., 2020; Shams et al., 2021). Manney et al. (1995) and
52 Harvey et al. (2008) describe the development of low ozone pockets (LOPs) in the region of the
53 Aleutian anticyclone during winter. Siskind et al. (2005; 2021) explain the occurrence of a
54 mesospheric cooling associated with SSWs and the role of gravity waves for modeling ozone in
55 the upper mesosphere, respectively. Chandran et al. (2013) provide a climatology of the Arctic
56 elevated stratopause, and Sofieva et al. (2021) analyze for regional trends in stratospheric ozone.
57 Smith et al. (2011; 2018) report on monthly changes of the tertiary ozone maximum at high
58 latitudes of the upper mesosphere during winter.

59

60 The LIMS (Level 2) profiles were retrieved with an improved Version 6 (V6) algorithm. They
61 were archived in 2008 and include ozone, temperature, and GPH that extend from 316 hPa to
62 ~0.01 hPa. Co-located V6 profiles of water vapor (H_2O), nitric acid vapor (HNO_3), and nitrogen
63 dioxide (NO_2) extend through the stratosphere. Lieberman et al. (2004) analyzed the V6
64 temperature profiles and found evidence for non-migrating tides in the mesosphere, due to the
65 interaction of the diurnal tide and planetary zonal-wave 1, especially in late January 1979. Holt
66 et al. (2010) analyzed the descent of V6 NO_2 from the lower mesosphere to within the polar
67 stratospheric vortex, where it interacts with ozone. Remsberg et al. (2013) assimilated V6 ozone
68 profiles in a reanalysis model and gained improved estimates of column ozone, especially in
69 Arctic winter. Such reanalysis studies assimilate temperature and ozone profiles within a model
70 framework. However, the models only approximate the effects of small-scale variations, so it is
71 also useful to consider observed variations of the LIMS parameters without resort to a model.
72 Keep in mind that smaller-scale atmospheric variations also contribute to the analyzed
73 intermediate and large-scale fields from V6. This paper further explores several instances of
74 those larger-scale variations of Arctic ozone, temperature, and GPH.

75

76 The SPARC Data Initiative (SPARC-DI) includes monthly zonal averages of V6 ozone up to the
77 0.1-hPa level (see Tegtmeier et al., 2013; SPARC, 2017; and Remsberg et al., 2021). In Section

78 2 we show January zonal averages of V6 ozone and temperature profiles that extend even higher
79 or to near the mesopause. The V6 Level 3 (map) product provides a 3-dimensional context for
80 those zonal mean data. Daily V6 maps are also an aid in interpreting individual V6 profiles
81 versus correlative data, especially during dynamically disturbed periods. Specifically, in Section
82 3 we compare several nighttime V6 ozone profiles with those obtained with a rocket-borne
83 chemiluminescent technique (Hilsenrath et al., 1980). Those profile comparisons are for
84 December 15 and for January 27 and 28, when the temperature and ozone fields were affected by
85 planetary wave forcings. There is a corresponding cooling and variations of ozone in the winter
86 lower mesosphere associated with the warming in the upper stratosphere. Section 4 presents
87 variations of ozone and GPH at northern extratropical latitudes during the minor SSW events of
88 late January and early February 1979, as a complement to the more comprehensive findings of
89 Harvey et al. (2008) on the occurrence of LOPs within anticyclones determined from satellite
90 solar occultation data. Section 5 considers the variability of the tertiary ozone maximum in the
91 upper mesosphere during that same period, as an adjunct to monthly zonal average values
92 reported by Smith et al. (2018). Section 6 notes that the maps of V6 ozone contain more details
93 about the gradients of atmospheric ozone during disturbed periods, but also cautions users about
94 occasional, pseudo-ozone features in the tropical lowermost stratosphere. Section 7 concludes
95 that the V6 Level 3 product represents an important resource for studies of the effects of
96 transport and chemistry on Arctic ozone.

97

98 **2 Characteristics of V6 Level 3 data**

99 *2.1 LIMS measurements and analyses*

100 Nimbus 7 was in a near-polar orbit, and LIMS made measurements at ~1 pm local time along its
101 ascending (A or south-to-north) orbital segments and at ~11 pm on its descending (D or north-to-
102 south) segments. A-D time differences are of the order of 10 hours at most latitudes because
103 LIMS viewed the atmosphere 146.5° clockwise of the spacecraft velocity vector, as seen from
104 above. The A-D differences narrow from 10 to about 6 hours from 60°N to 80°N, due to the
105 orbital geometry of Nimbus 7. The V6 processing algorithm accounts for low-frequency
106 spacecraft motions that affect the LIMS view of the horizon. As a result, its measured radiance
107 profiles are well registered in pressure-altitude (Remsberg et al., 2004). Retrieved V6 ozone,

108 temperature, and GPH profiles extend from 316 hPa to ~0.01 hPa and have a vertical point
109 spacing of ~0.88 km with an altitude resolution of ~3.7 km. Retrieved profile pairs are spaced
110 every 144 km along the orbital track or at every 1.3°, but closer together at the high, turn-around
111 latitudes of the orbital viewing geometry (Remsberg et al., 1990). LIMS made measurements
112 with a duty cycle of about 11 days on and 1 day off over its planned observing lifetime. The
113 LIMS algorithms (Remsberg et al., 2007) do not account for non-local thermodynamic
114 equilibrium (NLTE) effects in ozone (Solomon et al., 1986; Mlynczak and Drayson, 1990) and
115 in CO₂ (Edwards et al., 1996; Manuilova et al., 1998), so there are positive biases in the retrieved
116 V6 ozone throughout the mesosphere during daylight. However, the V6 nighttime ozone is more
117 nearly free of NLTE effects below about the 0.05-hPa level, except at times of SSWs (see e.g.,
118 Funke et al., 2012).

119

120 A sequential-estimation (SE) algorithm was used to generate daily, zonal Fourier coefficients
121 (zonal mean and up to six cosine and sine values or 6-zonal wavenumbers) for Level 3 at every
122 2° of latitude and at up to 28 vertical levels (Remsberg and Lingenfelter, 2010). The V6 SE
123 algorithm uses better estimates of data uncertainty and its zonal wave coefficients have a
124 memory of ~2.5 days, or about half that of the SE algorithm used by Remsberg et al. (1990).
125 The SE analysis is insensitive to the very few large, unscreened ozone profiles values found in
126 the lower stratosphere, as noted in Remsberg et al. (2013, their Fig. 1a). The SE algorithm
127 combines the coefficients from both the separate A and D orbital segments and effectively
128 interpolates the profile data in time to provide a continuous, 216-day set of daily zonal
129 coefficients versus pressure-altitude at 1200Z for each of the retrieved LIMS parameters.

130

131 *2.2 Monthly average V6 data*

132 One can generate monthly average distributions from the daily Level 3 files of temperature,
133 GPH, and species (ozone, H₂O, HNO₃, and NO₂); zonal averages for the V6 species were
134 supplied to SPARC-DI (SPARC, 2017; Hegglin et al., 2021). Tegtmeier et al. (2013) compared
135 the V6 monthly ozone distributions with ones from other satellite-based, limb sensors and
136 reported good agreement throughout the stratosphere. Although the species cross sections for

137 SPARC (2017) extend only up to the 0.1-hPa level (~64 km), V6 average ozone extends higher
138 or to about 0.015 hPa (~75 km). Figure 1 shows the latitude-pressure cross section for January
139 from just the descending (D) orbital profiles, which avoids the larger NLTE biases that affect
140 daytime ozone in the mesosphere. Stratospheric ozone mixing ratios in Fig. 1 have largest
141 values at about 10 hPa near the Equator (> 9.2 ppmv), and they decrease sharply above and
142 below that level. Maximum mixing ratios for the middle to high latitudes occur between 3 to 5
143 hPa, due to the larger zenith angles and longer paths of the ultraviolet light for production of
144 atmospheric ozone. There is a nighttime ozone minimum of ~ 1.2 ppmv across most latitudes of
145 the middle mesosphere. A tertiary ozone maximum is present in the upper mesosphere near the
146 winter day/night terminator zone in the LIMS measurements for January (at about 67°N), in
147 accordance with the interpretation of Marsh et al. (2001). The location (~ 0.02 hPa) and
148 magnitude (~ 3.5 ppmv) of the NH maximum are somewhat higher and larger than those reported
149 by Smith et al. (2018, their Fig. 4) from more recent satellite datasets. On the other hand, while
150 the V6 ozone poleward of $\sim 50^\circ\text{S}$ is also from descending orbital profiles, it corresponds to
151 daylight conditions at the high southern latitudes in January. Thus, the decrease of mesospheric
152 V6 ozone at 0.1 hPa and poleward of 50°S in Fig. 1 indicates merely a change from night to day
153 values and agrees with findings of Lopez-Puertas et al. (2018).

154

155 Radiances from two $15\text{-}\mu\text{m}$ CO_2 channels are used for retrievals of V6 temperature versus
156 pressure or $T(p)$, and they are free of NLTE effects below about the 0.05-hPa level (~70 km)
157 (Lopez-Puertas and Taylor, 2001). To first order, the V6 $T(p)$ retrievals account for the effects
158 of horizontal temperature gradients in the stratosphere (Remsberg et al., 2004). Single profile
159 root-sum-squared (or RSS) errors for $T(p)$ vary from 1 K at 10 hPa to ~ 2.5 K in the upper
160 mesosphere, but they do not include possible temperature gradient errors. RSS error from $T(p)$ is
161 the primary source of bias error for ozone, growing to about 16% in the middle mesosphere
162 (Remsberg et al., 2021, Table 1). Random errors become large for single ozone profiles in the
163 upper mesosphere. As a complement to the V6 ozone of Fig. 1, we show the descending
164 (\sim nighttime) V6 $T(p)$ distribution for January in Figure 2, which extends to near the 0.01-hPa
165 level. The large-scale features of the $T(p)$ distribution compare well with climatological values
166 from the late 1970s (Fleming et al., 1990), having a maximum value of about 285 K at the SH

167 high latitude stratopause and minimum values of < 200 K at the tropical tropopause and near the
168 summer mesopause. There is also some elevation of the Arctic zonal-average stratopause.

169

170 Figure 3 shows the monthly-average, zonal (wave) standard deviations (SD) about daily zonal
171 means of the combined-mode (A+D) V6 ozone for January, where the SD values are derived
172 from the zonal-wave amplitudes of V6 Level 3. There are relatively small SD values at low
173 latitudes from 7 to 10 hPa; it is assumed that they are a result of smaller-amplitude Kelvin and
174 Rossby-gravity waves. Effects of more vigorous, planetary wave activity are most apparent at
175 high northern latitudes of the stratosphere during winter. Gravity waves also contribute to SD in
176 the uppermost mesosphere (Siskind et al., 2021). Ozone shows little zonal variation in the SH
177 upper stratosphere of Fig. 3, due to constraints on the upward propagation of planetary waves
178 through the summer zonal easterlies (Andrews et al., 1987). SD values near the tropical
179 tropopause are due mostly to residual effects of emissions from thin cirrus and represent spurious
180 ozone variations (see Section 6).

181

182 **3. V6 comparisons with rocket-borne chemiluminescent ozone measurements**

183 In this section we consider V6 comparisons with three nighttime, rocket-borne chemiluminescent
184 ozone soundings of Hilsenrath (1980)—one at White Sands, NM, (32.4°N , 253.5°E) on
185 December 15, 1978, and two more at Poker Flat, AK, (65.1°N , 212.5°E) on the successive days
186 of January 27 and 28, 1979. The estimated total, rocket ozone error is 14% (precision plus
187 accuracy), according to Hilsenrath and Kirschner (1980).

188

189 Ozone comparisons for December 15 are in Figure 4 (top); we plot every other V6 profile and
190 those four profiles have spacings of 2.6° in latitude. The short-dashed V6 profile is at 29.2° , and
191 the long-dashed profile is at 37.2° . The solid curve is the V6 profile at 31.8° (at 0611Z) or
192 closest to the rocket sounding from White Sands (at 0541Z). Horizontal bars on the profiles are
193 estimates of ozone error; they overlap between V6 and rocket, except in the upper stratosphere.
194 LIMS ozone is larger than rocket ozone in through the upper stratosphere. The corresponding

195 V6 ozone map at 4.6 hPa in Fig. 4 (bottom) reveals an ozone maximum just south of White
196 Sands (WS—blue dot), along the descending orbital segment of the satellite at (6°N, 265°E—
197 white dot) or viewing in the NNW direction toward White Sands. Note that while zonal
198 variations in the map are from a gridding of the Level 3 coefficients (2° latitude and 5.625°
199 longitude), there is no smoothing of the gridded field in the meridional direction; there is good
200 continuity across latitudes, nonetheless. The rocket profile is a local measurement and has a
201 vertical resolution that ranges from 1.5 km at 60 km to 0.1 km at 20 km; the nearby V6 profiles
202 have a lower vertical resolution of ~3.7 km and are an average over the finite horizontal length
203 (~300 km or ~3° latitude) of the LIMS tangent layer. There is an ozone maximum along the
204 LIMS view path just to the south of White Sands, which may account for the profile differences.
205 We also note that the ozone field of two days earlier has the region of sharp gradients positioned
206 over White Sands with ozone at only 8 ppmv. Thus, an ozone field that varies in both space and
207 time can lead to additional uncertainties for comparisons of the localized rocket and limb-
208 viewing satellite profiles in Fig. 4.

209

210 Because V6 ozone is obtained from retrievals of the measured V6 ozone radiance profiles, the
211 LIMS retrieved temperature profile must be representative of the atmospheric state for the
212 forward model of ozone radiance. Figure 5 (top) shows the corresponding temperature
213 comparisons between V6 and a separate rocket Datasonde instrument. Agreement between them
214 is very good throughout the upper stratosphere, indicating that the temperature variations are
215 well determined along the LIMS view path for the forward radiance calculations of V6 ozone
216 and that the retrieved V6 ozone should be nearly unaffected by temperature bias error. The map
217 of V6 temperature (Fig. 5—bottom) shows zonal variations on December 15, although their
218 meridional gradients are relatively weak above White Sands. Conversely, the ozone profiles
219 agree well near 0.68 hPa in Fig. 4, where there are apparent biases between the T(p) profiles.
220 There are significant horizontal gradients near White Sands in the maps of T(p) at 0.68 hPa, but
221 not in ozone (not shown). In fact, the V6 ozone field at that level has a nearly constant value,
222 and ozone is less sensitive (by half) to changes in T(p) at 0.68 hPa than at 4.6 hPa (Remsberg et
223 al., 2007). Co-location is more important for the V6 versus rocket comparisons of T(p) than of
224 ozone in the lower mesosphere.

225

226 The two comparisons above Poker Flat, AK, occurred at the time of a stratospheric, zonal wave-
227 1 warming event. Leovy et al. (1985) provide a detailed discussion of the advective changes for
228 ozone in the middle stratosphere during January 1979. Figure 6 (top) shows three V6 ozone
229 profiles from along an ascending orbital segment on January 27. The LIMS instrument was
230 viewing from its satellite location (80.7°N, 113°E) at 2204Z, and the rocket ozone launch was
231 two hours earlier or at 2005Z at a solar zenith angle of 84° or near the terminator; there is good
232 agreement of the structure between them, even in the mesosphere. A second rocket launch
233 followed at 0833Z of January 28 (Hilsenrath, 1980). Since the separate V6/rocket ozone and
234 T(p) comparisons are similar for the two days, Fig. 6 contains results for January 27 only. The
235 rocket sounding recorded two ozone maxima, one near 15 hPa and another at about 0.6 hPa. The
236 ozone maximum at about 15 hPa is primarily due to advection of ozone of higher mixing ratios
237 from lower latitudes just prior to the warming event. The local maximum at 0.6 hPa was
238 unexpected, based on findings from a larger set of rocket ozone soundings. There is a relative
239 minimum for both V6 and rocket ozone through the upper stratosphere, although V6 ozone is
240 larger. The map of V6 ozone at 4.6 hPa in Fig. 6 (bottom) indicates that the rocket measurement
241 occurs at the center of the minimum, whereas the V6 profiles are averages across it. The ozone
242 profiles in Fig. 6 (top) indicate the relative minimum in a low-ozone pocket (LOP) that extends
243 from about 7 hPa to 2 hPa.

244

245 Figure 7 (top) shows the V6 temperature profile comparisons; T(p) from the Datasonde has more
246 vertical structure, as expected from a localized measurement. V6 T(p) values reach a maximum
247 of order 250 K at about 3 to 4 hPa. They agree reasonably with the Datasonde values, given that
248 there is significant horizontal structure in the temperature field surrounding Poker Flat. The
249 apparent V6 minus Datasonde bias of order 5 K at 3 hPa ought to lead to a V6 minus rocket
250 ozone bias of -40%, according to error estimates for retrieved V6 ozone. However, Fig. 7
251 (bottom left) indicates that LIMS was viewing Poker Flat across an area of higher temperatures,
252 such that it is likely that there is a spatial mismatch for V6 and Datasonde T(p) values. The
253 much smaller and positive ozone differences in Fig. 6 support that likelihood. There may also be
254 co-location differences between the rocket temperature and ozone soundings in this instance.

255

256 Figure 7 also shows a map of NH GPH at 4.6 hPa on January 27 for comparison with the ozone
257 map in Fig. 6. Lowest ozone values are in the polar vortex, where the GPH field is asymmetric
258 about the Pole. A second, low value of ozone is associated with the anticyclone over the
259 Alaskan sector. One can determine horizontal winds from gradients of GPH on the 4.6-hPa
260 surface and thereby estimate the transport of ozone to first order. Qualitatively, the direction and
261 strength of the large-scale transport follows from the character of the cyclonic and anticyclonic
262 features on the GPH map. The large-scale cyclonic circulation about the vortex transports air
263 from middle latitudes to across the Pole on January 27. The vortex region has low ozone and is
264 relatively cold, whereas stratospheric temperatures over Alaska show a maximum (the SSW),
265 and the rocket profile above Poker Flat, AK, was near the center of the anticyclone and in the
266 region of relatively low ozone (or LOP).

267

268 Ozone is an approximate tracer of transport processes and reveals dramatic changes with altitude
269 associated with this SSW event, even through the winter lower mesosphere. As an example,
270 Figure S1 (in Supplemental Materials) shows a concurrent cooling at 0.46 hPa above the Alaskan
271 anticyclone on January 27, where the co-located ozone field exhibits a local maximum. There is
272 also a major temperature increase above the polar stratospheric vortex over northern Europe at
273 0.46 hPa, or where ozone values remain low. In summary, Figs. 4 through 7 and S1 indicate the
274 utility of daily maps from LIMS for analyses of the ozone fields during dynamically disturbed
275 conditions.

276

277 **4. Variation of a low ozone pocket (LOP) from LIMS Level 3**

278 The polar vortex on January 27 was located over northern Europe and Asia; it was centered off
279 the Pole because of effects of large-scale, planetary waves in the development of the SSW
280 (Andrews et al., 1987, Chapter 6). In this section, we show sequences of polar plots of both
281 stratospheric GPH and ozone for February 1979. Manney et al. (1995) and Harvey et al. (2004,
282 2008) provide comprehensive analyses about the occurrence of polar anticyclones and their
283 associated LOPs from studies of GPH and ozone fields from several different satellites. They

284 determined the extent and character of the polar vortex based on meteorological data from the
285 UK Met Office or as obtained from relatively low vertical resolution radiance profiles from
286 operational, nadir temperature sounders. The V6 GPH profiles are derived from and have the
287 same vertical resolution as the T(p) profiles. Manney et al. (1995) showed that water vapor is a
288 useful tracer of the meridional transport of air, and the V6 H₂O fields at 6.8 and 10 hPa indicate
289 that low latitude air was transported to the region of the LOP in late January. But the V6 H₂O
290 fields are noisy at 4.6 hPa (not shown). Even so, the V6 Level 3 ozone, T(p), and GPH data
291 offer useful details about the occurrence of LOPs in the upper stratosphere.

292

293 Harvey et al. (2004) reported that LOPs occur nominally at about the 5-hPa level. Accordingly,
294 the three panels of Figure 8 show three daily NH maps of V6 GPH from February 3 to February
295 17 at 4.6 hPa; each successive map is spaced one week from the previous one. This sequence
296 shows that both the vortex and anticyclone weaken during the three weeks following January 27
297 at this level. The vortex re-centers on the Pole by February 17, and the anticyclone is nearly
298 absent at 4.6 hPa following the two minor warming events. The map sequence of GPH indicates
299 that there were significant changes in the horizontal transport of ozone in late January/early
300 February. The corresponding three panels of ozone in Figure 9 show the further evolution of
301 ozone, following that of January 27 (in Fig. 6). Even though the anticyclone had weakened
302 during the first week, there was a deepening of the LOP from January 27 to February 3 and a
303 filling of it thereafter.

304

305 Was there some chemical loss of ozone from January 27 to February 3 in the region of the LOP?
306 Morris et al. (1998) and Nair et al. (1998) conducted model calculations to show how that could
307 happen. Ozone reactions are affected by changes with latitude of solar insolation, temperature,
308 and loss via NO_x. Nair et al. (1998) reported on the effect of a decrease in the production of
309 ozone for the development of LOPs, as air parcels in the middle stratosphere move from low to
310 high latitudes or to higher solar zenith angles in winter. Remsberg et al. (2018) analyzed air
311 parcel trajectories that included chemistry, and they showed that there was some loss of ozone in
312 the middle stratosphere, due to reactions with NO_x. However, Holt et al. (2012) analyzed V6

313 NO₂ in the winter polar vortex, and they did not find enhanced values at 4.6 hPa due to energetic
314 particle precipitation (EPP) by late January.

315
316 Figure 10 (left) is a map of the V6 descending orbital (nighttime) NO₂ for January 27 at 4.6 hPa.
317 Based on the corresponding map of GPH in Fig. 7, one can trace the horizontal advection of high
318 NO₂ toward higher latitudes and toward the polar vortex as well as the advection of low NO₂ out
319 of the vortex and about the anticyclone. Fig. 10 (right) is a map of HNO₃ at 4.6 hPa. It has a
320 weak, relative maximum above the anticyclone that appears as a residual from the advection of
321 much higher vortex values from several days before. A closer inspection in time reveals that the
322 NO₂ values in the LOP were a bit lower by January 31, when ozone had already declined to near
323 its February 3 value. Thus, while an excess of HNO₃ in the region of the LOP is consistent with
324 a conversion of NO₂ to HNO₃ above the isolated anticyclone, there is no clear evidence from the
325 V6 map products that such chemistry led to significant changes in the ozone. One must conduct
326 trajectory studies that include chemistry and that rely on the V6 species profiles as input for
327 better, quantitative estimates. Unfortunately, the profiles of V6 NO₂ and HNO₃ become noisy in
328 the upper stratosphere. At a minimum though, one can follow the evolution of the LOP using the
329 daily maps of V6 ozone and GPH.

330

331 **5. Variations of the tertiary ozone maximum**

332 Smith et al. (2018) describe the changing monthly, zonally averaged character of the wintertime
333 tertiary ozone maximum of the polar upper mesosphere. They point out that the low latitude
334 edge of the tertiary ozone maximum is where HO_x radicals and the chemical loss of ozone due to
335 reactions with them are reduced. V6 ozone radiance profiles have low signal-to-noise in the
336 upper mesosphere; the precision estimate is 0.32 ppmv for retrieved ozone profiles. We show a
337 map in Figure 11 of the combined V6 ozone for December 15 at 0.022 hPa (~72 km), where its
338 distribution in the subpolar region is based on fewer than 13 zonal coefficients because some
339 profiles do not extend to that pressure altitude. The corresponding map of temperature is also in
340 Fig. 11, and one can see that there is significant non-zonal structure in its field at the latitudes
341 where ozone is enhanced. While both V6 ozone and temperature are not highly accurate due to

342 NLTE effects in the upper mesosphere, their maps reveal significant relative spatial structures
343 indicating advective transport and its likely effects on ozone.

344

345 Figures S2 and S3 in the Supplemental Materials show additional panels at 0.022 hPa of ozone
346 and temperature, respectively, for January 13, February 10, and March 1. Elevated values of
347 ozone occur at higher latitudes on February 10 and March 1 than on December 15 and January
348 13, which is consistent with the more northward position of the terminator away from winter
349 solstice and the consequent effects for the chemical loss of ozone. The temperature fields are
350 also perturbed on January 13 and February 10, but they are more nearly zonal by March 1.
351 However, there are meridional gradients of temperature on all three days in the region of the
352 tertiary ozone maximum. On January 13 there is also a well-defined mesospheric vortex in GPH
353 (not shown), and the highest values of ozone correlate reasonably with it. The vortex is most
354 disturbed and tertiary ozone maximum has largest values on February 10, perhaps in response to
355 the upward propagation of wave activity following the minor SSW of late January.

356

357 Figure 12 shows time series of peak zonal mean ozone at 0.022 hPa and its latitude location for
358 each week from November through mid-March. The separate time series are for peak ozone
359 (bottom two series) and their latitude locations (top two). Dashed red curves represent zonal
360 mean results for combined (A+D) ozone; solid black curves are results for nighttime (D) only.
361 Blue horizontal lines represent the approximate latitude position of the terminator. Peak
362 nighttime ozone values are based on just the 'zonal mean' and the cosine and sine coefficients
363 for waves 1 and 2 because not all profiles reach to the 0.022-hPa level. Peak ozone occurs at
364 lower latitudes ($\sim 65^\circ\text{N}$) in December, increasing to $\sim 75^\circ\text{N}$ in early November and early February
365 and to near 80°N by early March. The latitude time series of peak ozone values is reasonably
366 coincident with the changing location of the terminator. Peak combined (A+D) ozone increases
367 slowly from a minimum of 2.2 ppmv in November to 3.6 ppmv in late February and March.
368 Descending (or nighttime only) ozone varies from 3.3 ppmv in November, to ~ 4.5 ppmv in
369 January, to a maximum of 6.3 ppmv in mid-February, and then declining to 3.5 ppmv by mid-
370 March, although the time series shows rather large variations. Those maximum V6 values are

371 larger than reported by Lopez-Puertas et al. (2018), perhaps due to biases from V6 T(p) and/or
372 ozone at 0.022 hPa.

373

374 The increasing V6 ozone in Fig. 12 during winter disagrees with that of Smith et al. (2018), who
375 found decreasing ozone in February. They reported that, in most years, there is a slow descent of
376 relatively dry air into the vortex region in late autumn and early winter in the upper mesosphere,
377 and that the reduction in water vapor implies that there are fewer HO_x radicals for the destruction
378 of ozone near the terminator zone, leading to accumulations of ozone. However, there were two
379 minor warmings and associated lower mesospheric cooling events during late January and early
380 February 1979 (Hitchman et al., 1989). The enhanced V6 ozone of February 1979 follows those
381 SSW events. It may be that there were wave-driven disturbances and a dissipation of their
382 energy in the upper mesosphere at higher latitudes at that time (e.g., Siskind et al. 2005). One
383 should be able to gain more information about the evolution of the tertiary ozone maximum in
384 the winter of 1978-79 from the daily maps of V6 ozone, T(p), and GPH (as in Figs. S2 and S3).

385

386 **6. Other aspects of V6 Level 3 ozone**

387 The combined (A+D) Level 3 coefficients are the basis for a gridding of daily synoptic maps at
388 1200Z of ozone and related parameters. The Level 3 product also contains coefficients from its
389 separate A and D profiles; their ‘zonal mean’ values correspond to the local time-of-day of their
390 respective measurements. Remsberg et al. (2007) noted that maps from V6 reveal more details
391 about the variations of ozone. In Figure S4 of the Supplemental Material we compare a map of
392 V6 ozone at 10 hPa on 27 January with a similar map for V5 of Leovy et al. (1985). The ozone
393 gradients are more pronounced with V6 than with V5 at both the subtropical and vortex edges of
394 the ozone field. The V6 maps make use of all profiles along the orbit, and the SE mapping
395 algorithm was applied to them every 2° of latitude. However, the tighter gradients were also
396 achieved with the V6 algorithm because it has a relaxation time (or memory) that is half that of
397 V5. This means that the V6 maps are more representative of the rapidly changing atmospheric
398 ozone fields on that day. Similar version differences are evident throughout winter, when the so-
399 called ‘stratospheric surf zone’ develops and expands (Leovy et al., 1985).

400

401 Significant exchanges of air and ozone occur from the extratropical stratosphere to the
402 troposphere in winter and spring (Gettelman et al., 2011). There are large zonal variations about
403 the daily zonal means of ozone in the Arctic region of the lower stratosphere in Fig. 3. There are
404 similar variations in GPH (and derived winds) and in zonal wave activity that lead to ozone
405 transport. Zonal variations are resolved in the daily ozone maps down to the 146-hPa level.
406 Notably, Shepherd et al. (2014) integrated the V6 monthly zonal mean ozone above the
407 tropopause and subtracted it from observed total ozone, as part of their assessment of long-term
408 trends of tropospheric ozone from models. Their determination of extratropical tropospheric
409 ozone based on LIMS agrees with that obtained from other ozone datasets.

410

411 There is also a relative excess of SD ozone values in Fig. 3 centered at 68 hPa at tropical
412 latitudes, and similar anomalies occur in other LIMS months (not shown). As an example,
413 Figure 13 shows a map of V6 ozone at 68 hPa (~18 km) on December 15, to give more insight
414 about the source of the tropical variations. Ozone mixing ratio values in Fig. 13 are of order 2 to
415 3 ppmv at high latitudes, becoming much smaller in the subtropics. However, there is also an
416 unexpected, high value of 2 to 3 ppmv at about 15°N, 150°E. Limb measurements in the ozone
417 channel include radiance effects from cirrus particles that can occur along the tangent view path,
418 although the retrieved ozone mixing ratio profiles were screened of those effects to first order
419 (Remsberg et al., 2007). Even so, we note that ozone is easily affected by any excess radiance
420 because of highly non-linear effects for retrievals of ozone in the lower stratosphere. It is very
421 likely that the anomalous ozone at 68 hPa is a result of residual effects from subvisible cirrus,
422 which is nearly ubiquitous over the western tropical Pacific region (see SPARC, 2006, Fig. 1.8).
423 While individual V6 ozone profiles may include such spurious features in the tropics, the Level 3
424 ozone product at 68 hPa is affected mainly when there is an organized convection and outflow of
425 air that persists for several days. The adjacent map of ozone at 46 hPa appears unperturbed in
426 that region (not shown), and tropical ozone at 100 hPa approaches zero. There are much smaller
427 anomalies in maps of nitric acid, as its mixing ratio retrieval is very nearly linear. Anomalies are
428 also not so apparent in maps of V6 H₂O at 68 hPa because the cloud screening algorithm for H₂O
429 accounts for the larger vertical field-of-view and extent in altitude for measurements in the water

430 vapor channel of LIMS. To summarize, one must be mindful that the Level 3 product may
431 indicate excess, but spurious ozone at 68 hPa in the tropics.

432

433 7. Conclusions

434 This report provides guidance to researchers for their use of the LIMS V6 Level 3 product and
435 for their generation of daily gridded distributions of its temperature, ozone, and GPH on pressure
436 surfaces. H₂O, NO₂, and HNO₃ are also available for the stratosphere from the Level 3 product.
437 The V6 dataset represents an early baseline for considering possible changes in the middle
438 atmosphere from 1979 to today and into the future. LIMS made measurements at a time when
439 stratospheric effects from volcanoes were minimal and when catalytic effects of chlorine on
440 ozone were relatively small. Accordingly, Stolarski et al. (2012) found small, but significant
441 changes in the distribution of upper stratospheric ozone for recent decades compared with 1978-
442 1979. The LIMS measurements were taken near solar maximum and when atmospheric
443 concentrations of the greenhouse gases (GHG), CO₂, CH₄, and CFCs, were smaller than today.
444 Middle atmosphere T(p) distributions were warmer in 1978-1979.

445

446 The LIMS measurements in the winter Arctic region occurred when there was a lot of wave
447 activity for the transport and mixing of ozone. As a result, ozone varied dramatically in winter,
448 particularly during times of stratospheric warming events. There was a so-called Canadian
449 warming in early December 1978, two minor SSW events in late January and early February,
450 and a final warming in late February 1979. We showed V6 comparisons with temperature and
451 ozone profile data obtained using rocket borne Datasonde and chemiluminescent instruments,
452 and we pointed out how an examination of changes in their nearby fields is valuable for the
453 interpretation and validation of V6 profiles against those correlative measurements. The Level 3
454 dataset provides daily details on variations of ozone with latitude, longitude, and altitude, along
455 with related variations in temperature, geopotential height, NO₂, and HNO₃. We noted also that
456 there are instances of spurious, excess ozone from the Level 3 coefficients at 68 hPa in the
457 tropics but not in the extratropical stratosphere.

458

459 We displayed evidence of a low ozone pocket (LOP) during the minor SSW of late January
460 above the Aleutian anticyclone, and we followed its evolution into mid-February. The V6
461 nighttime ozone is relatively accurate through the mesosphere in Arctic winter. We provided
462 time series of the wintertime, tertiary ozone maximum of the upper mesosphere from V6 data.
463 Its ozone reached maximum values in February, perhaps as a response to enhanced wave activity
464 in the mesosphere following several SSW events. Together with V6 maps of T(p) and GPH, one
465 may explore further the daily evolution of that ozone maximum throughout the NH winter of
466 1978-1979.

467

468 **Data Availability**

469 The LIMS V6 Level 3 product is at the NASA EARTHDATA site of EOSDIS and its website:
470 https://disc.gsfc.nasa.gov/datacollection/LIMSN7L3_006.html (Remsberg et al., 2011). The
471 SPARC-Data Initiative data are located at <https://doi.org/10.5281/zenodo.4265393> (Hegglin et
472 al., 2021). We acknowledge the individual instrument teams and respective space agencies for
473 making their measurements available, and the Data Initiative of WCRP's (World Climate
474 Research Programme) SPARC (Stratospheric Processes and their Role in Climate) project for
475 organizing and coordinating the compilation of the chemical trace gas datasets used in this work.

476

477 *Author Contributions.* ER wrote the manuscript and prepared the figures with contributions from
478 his co-authors. EH provided his rocketsonde data on ozone and temperature along with their
479 error estimates.

480

481 *Competing interests.* The authors declare no competing interests for this study.

482

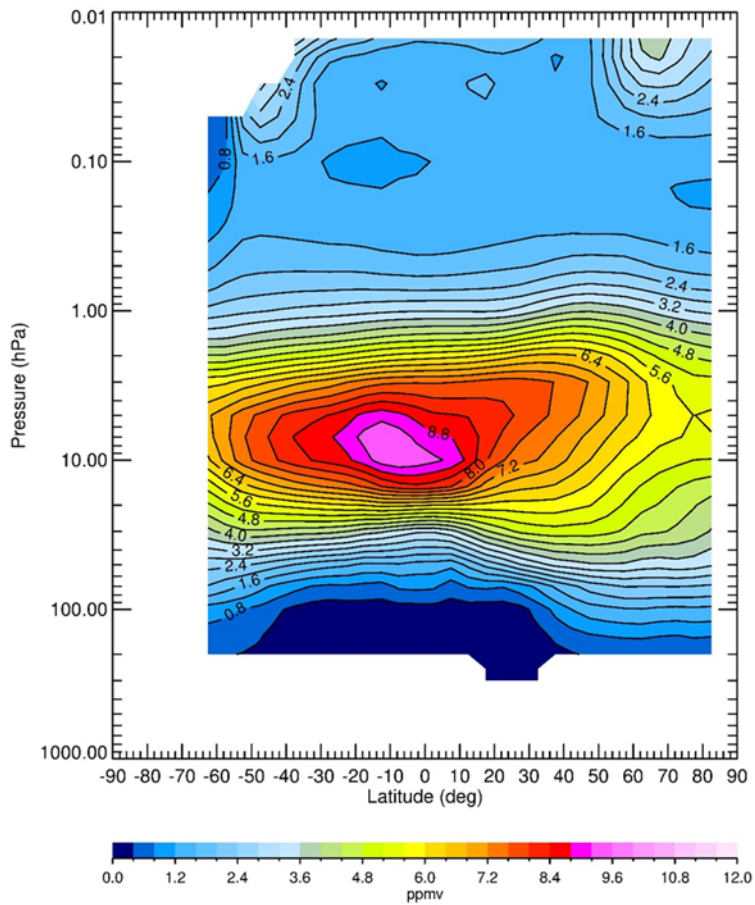
483 *Acknowledgements.* The authors appreciate John Gille and Jim Russell III and members of the
484 LIMS Science Team for their leadership in the development of the LIMS instrument and for their
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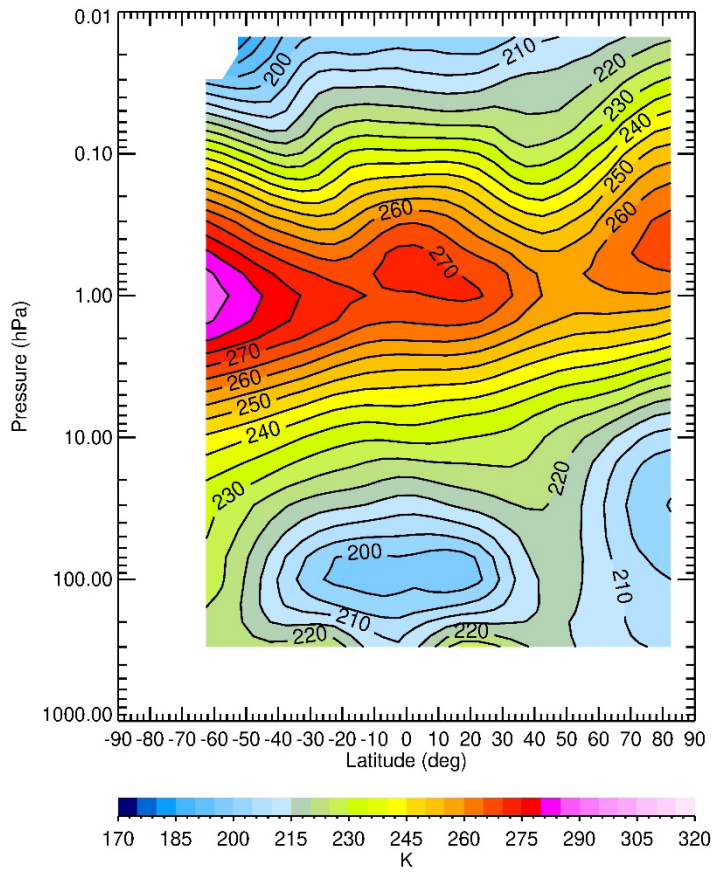
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495

496 Figure 1—LIMS V6 Level 3 monthly zonal mean ozone for descending-mode only (or nighttime
497 equatorward of $\sim 55^\circ\text{S}$) for January 1979. Contour interval (CI) is 0.4 ppmv.

498

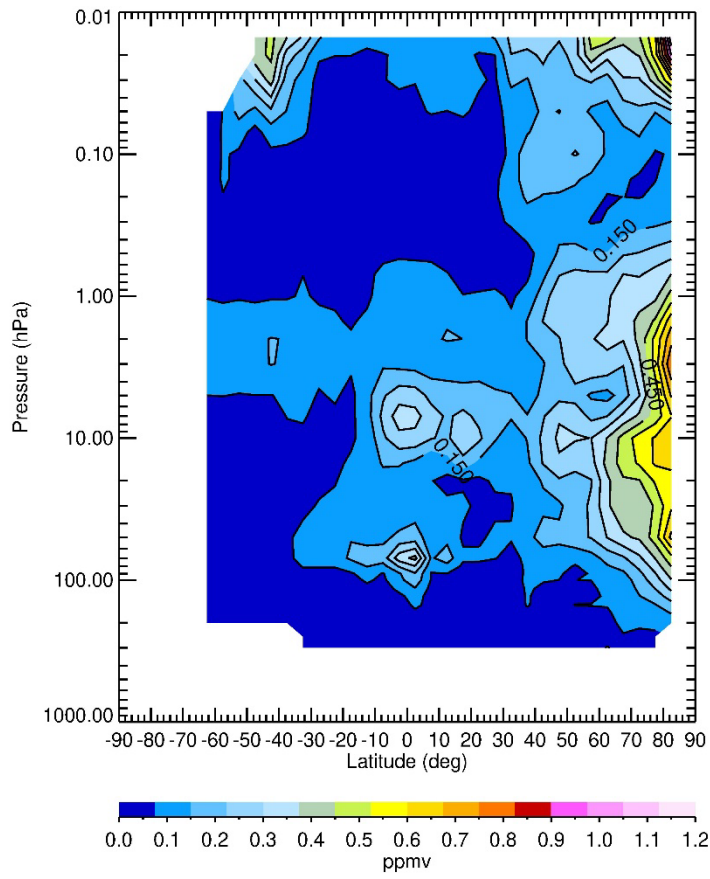


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501 Figure 2—Zonal average, descending-mode, temperature for January 1979. CI is 5 K.

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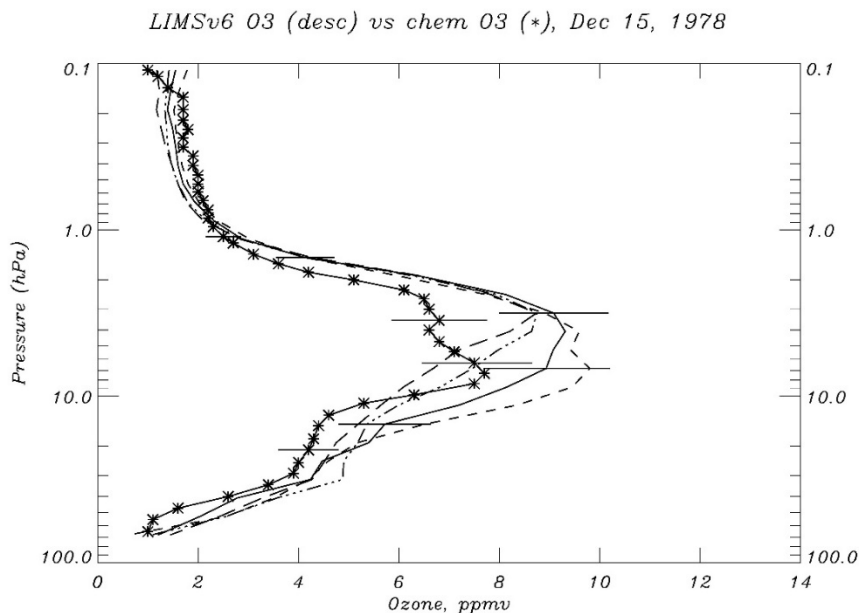
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506 Figure 3—Zonal standard deviation about average (A+D) zonal mean ozone for January 1979.

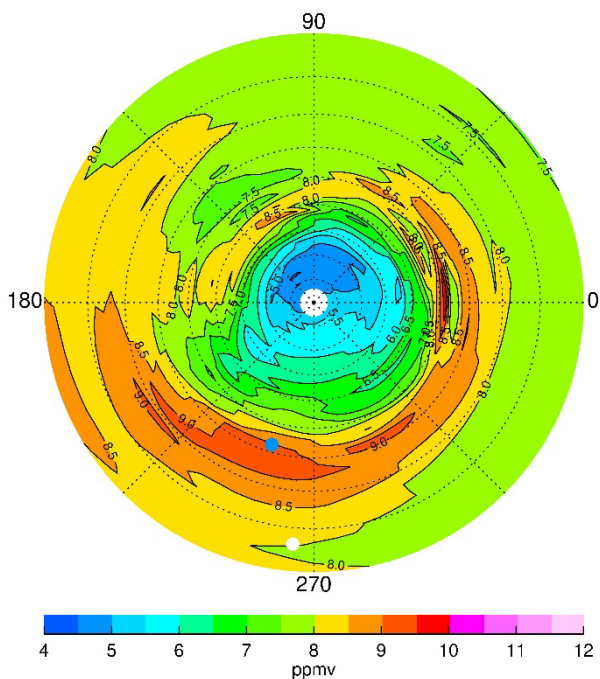
507 CI is 0.075 ppmv.

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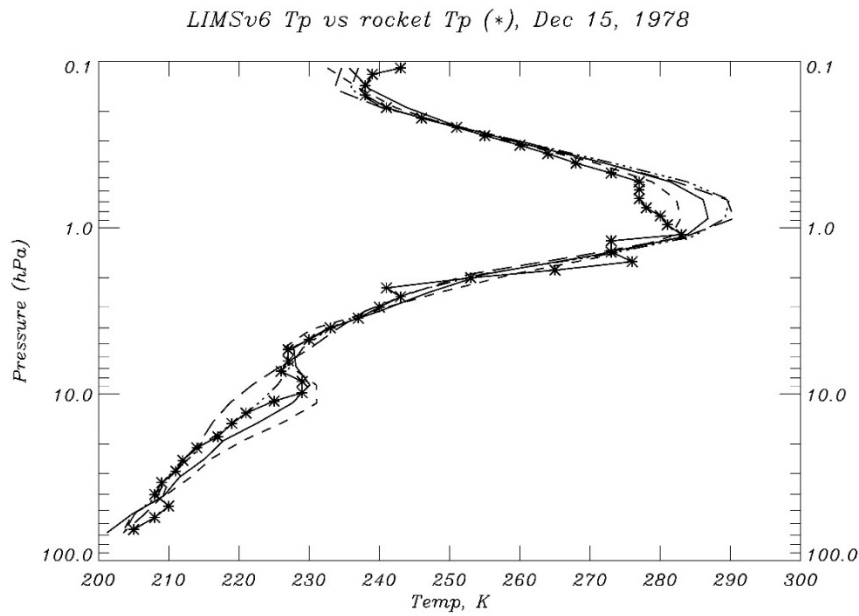
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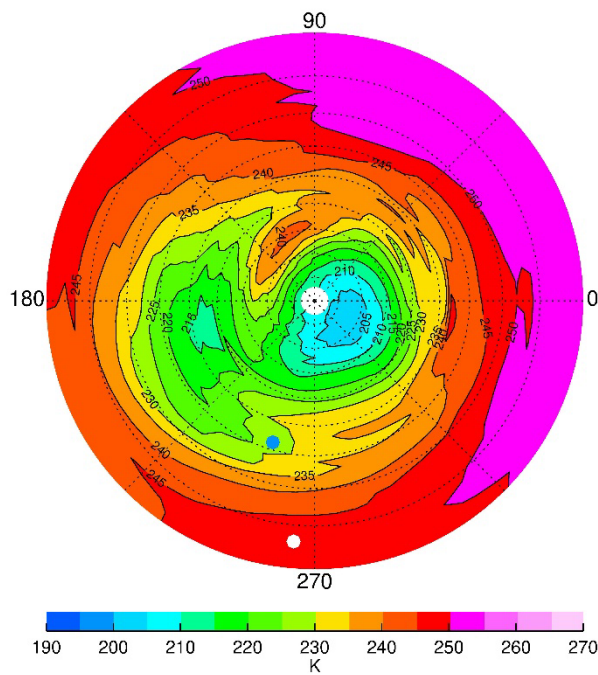
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512 Figure 4—(top) Profiles of V6 ozone (at 0611Z) versus rocket chem ozone (* at 0541Z) on
513 December 15. The four V6 profiles have separations of 2.6° latitude, and the solid curve (at
514 31.8°N) is closest to White Sands (WS, 32.4°N). Horizontal bars are ozone errors. (bottom) NH
515 V6 ozone at 4.6 hPa; Greenwich (0°E) is at right, and CI is 0.5 ppmv. Latitude (dotted circles) is
516 every 10°. Satellite location is white dot (6°N, 265°E), and WS is blue dot.

517



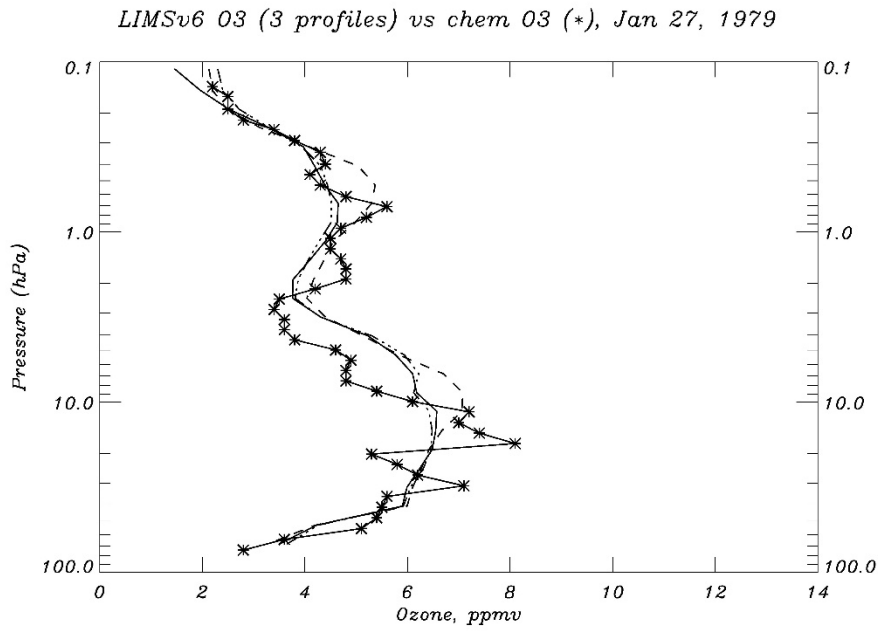
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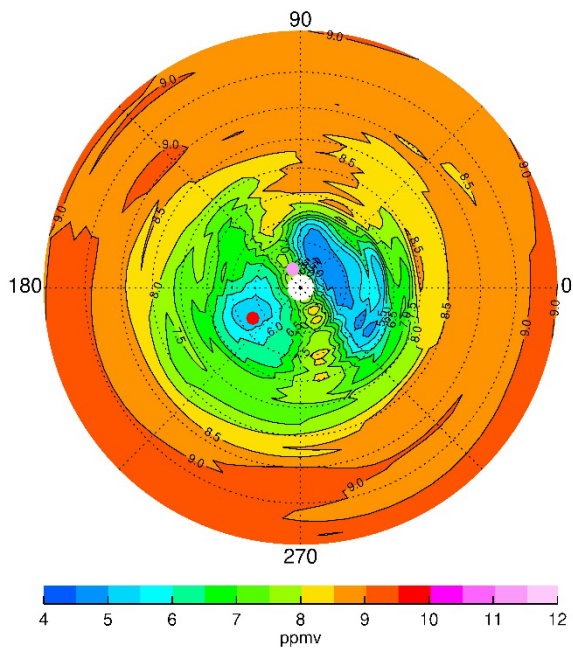
520 Figure 5—(top) Profiles of V6 temperature compared with Datasonde values (*) on December
521 15. The four V6 profiles are separated as in Fig. 4, where the short-dashed curve is for 29.2° and
522 the long-dashed curve is for 37.2°. (bottom) NH V6 temperature distribution at 4.6 hPa; CI is 5
523 K, and satellite location is white dot and White Sands is blue dot.

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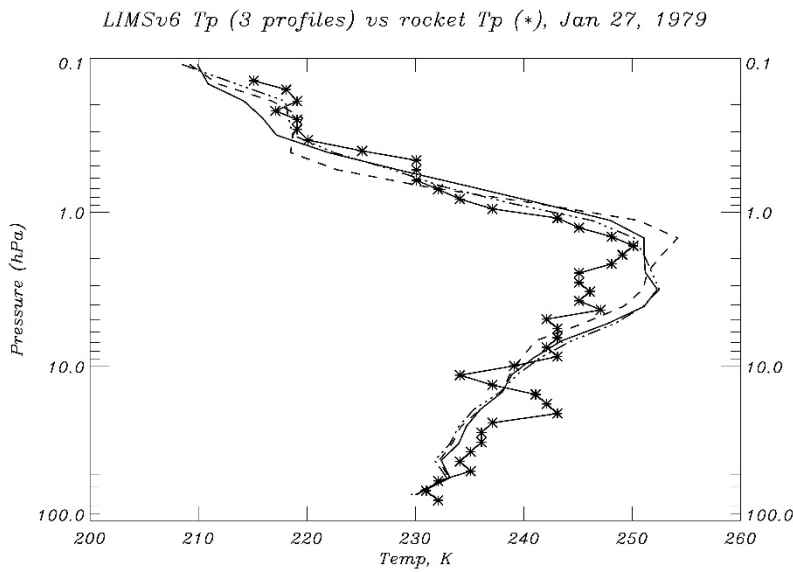
528 Figure 6—(top) As in Fig. 4, but for January 27, 1979, at Poker Flat, AK (65°N, 212.5°E);

529 (bottom) NH V6 distribution of ozone at 4.6 hPa, where CI is 0.5 ppmv. Latitudes (dotted

530 circles) are spaced every 10°; Poker Flat is red and satellite position (81°N, 113°E) is pink.

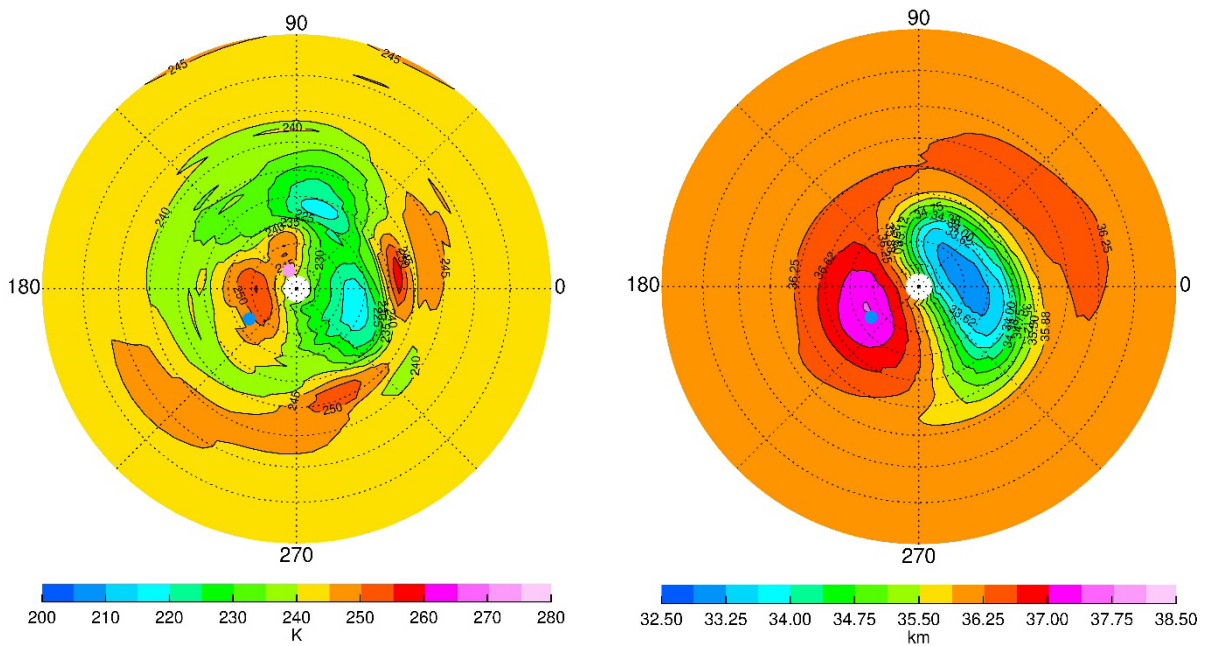
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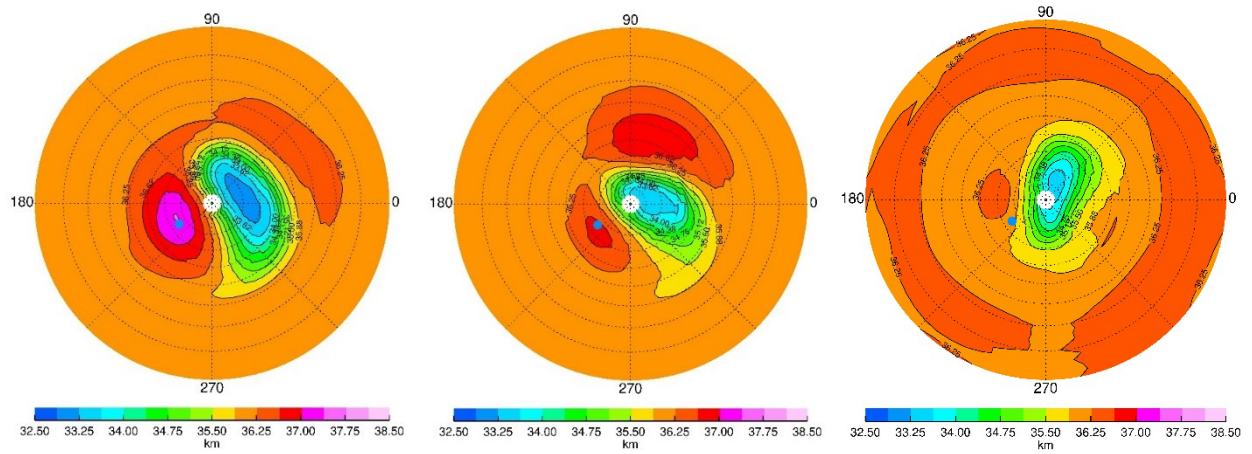


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537 Figure 7—(top) As in Fig. 5, but for January 27, 1979. (bottom-left) NH V6 temperature; CI is 5
 538 K. Poker Flat is blue and satellite position is pink. (bottom-right) V6 GPH; CI is 0.375 gpkm.

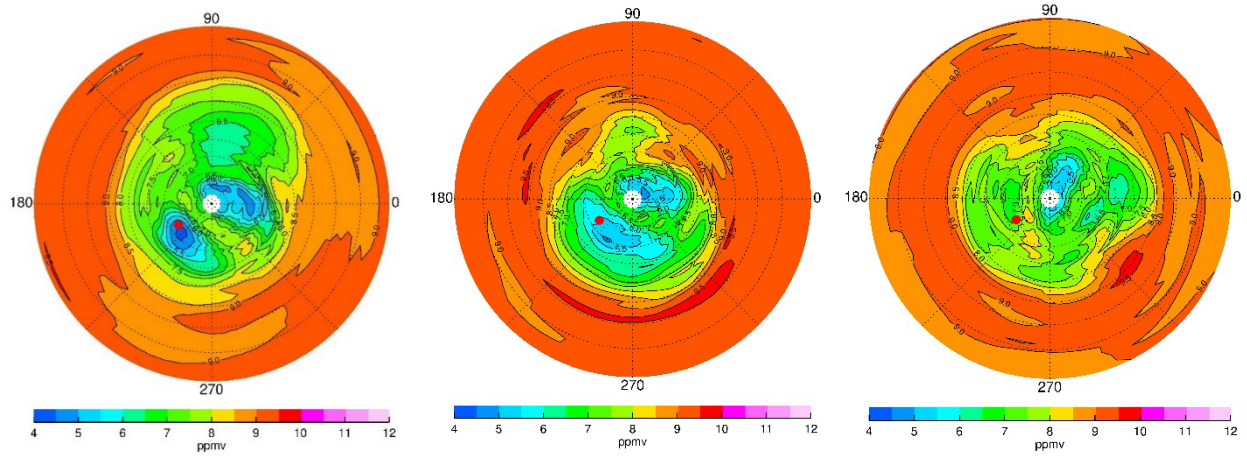
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541 Figure 8—NH V6 GPH at 4.6 hPa; CI is 0.375 gpkm. Poker Flat is blue dot. Panels are spaced
 542 one week apart; (left) February 3; (middle) February 10; and (right) February 17.

543

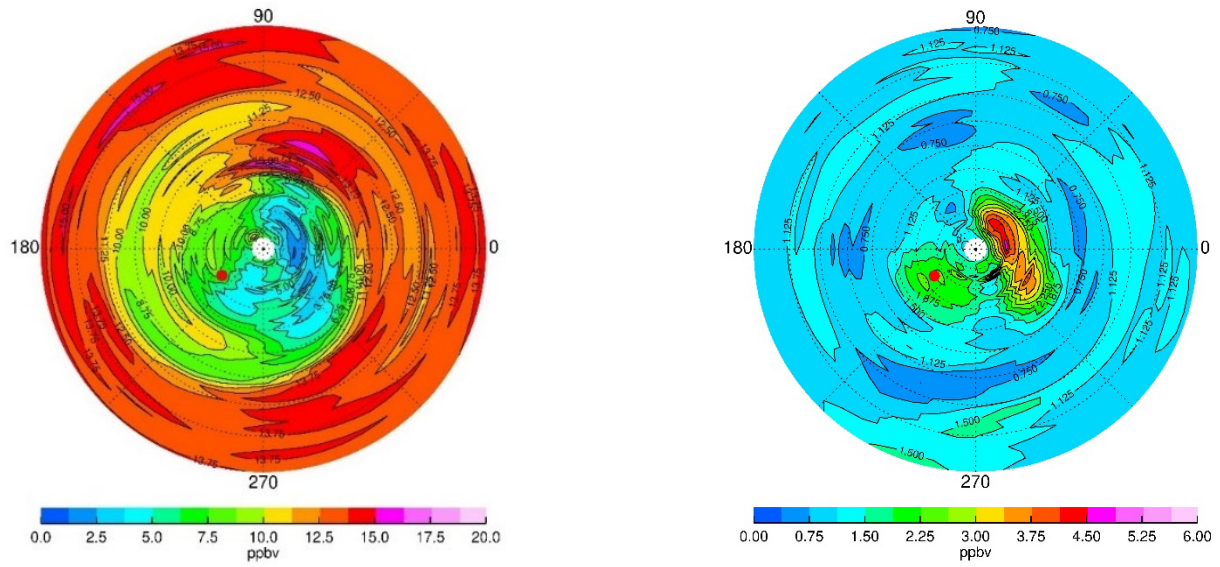


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546 Figure 9—Maps of ozone at 4.6 hPa (left) on February 3, (middle) on February 10; and (right) on
 547 February 17. CI is 0.5 ppmv and red dot is Poker Flat.

548



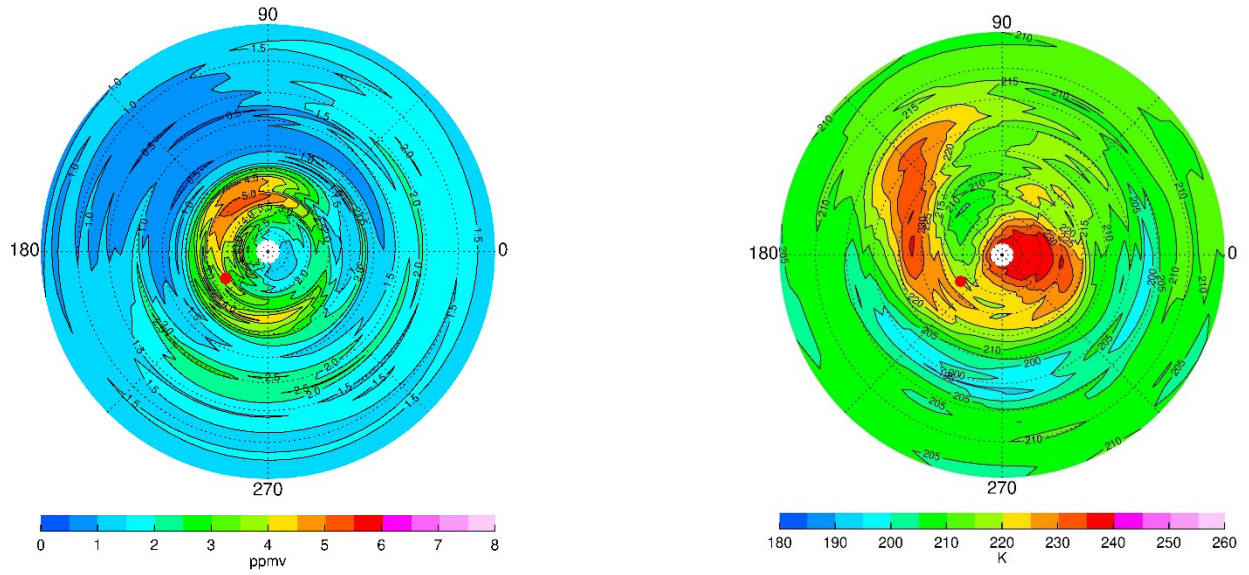
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561 Figure 10—(left) Nighttime NO₂ on January 27 at 4.6 hPa; CI is 1.25 ppbv. (right) HNO₃ at 4.6
 562 hPa; CI is 0.375 ppbv. Red dot is Poker Flat.

563

564



573

574 Figure 11—(top) NH distributions for December 15 at 0.022 hPa for (left) ozone and for (right)
 575 temperature; CIs are 0.5 ppmv and 5 K, respectively. Red dot denotes location of Poker Flat.

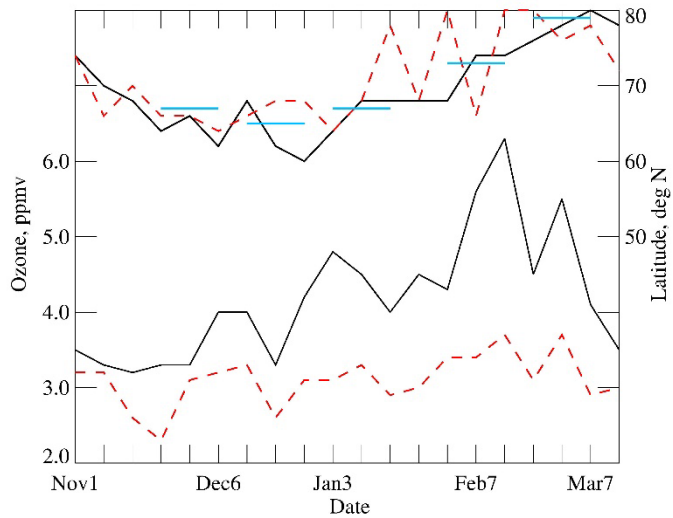
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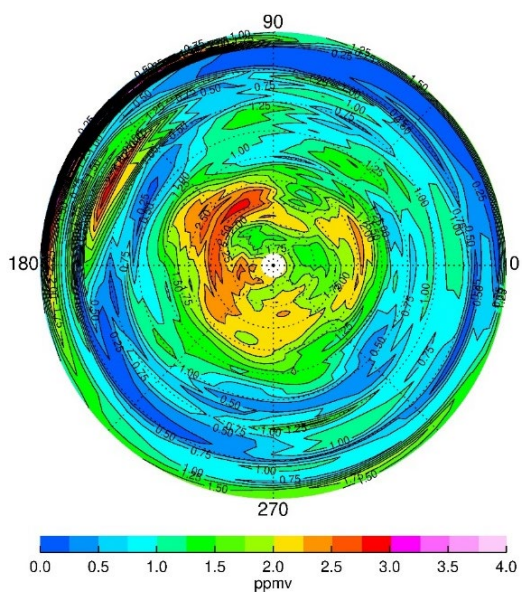


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582 Figure 12—Time series of peak V6 ozone (bottom two curves) and its latitude location (top two
583 curves) at 0.022 hPa. Dashed red curves are for combined ozone, while solid curves are for
584 descending (nighttime) ozone only. Horizontal blue lines indicate the latitude of the terminator.

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588

589 Figure 13—NH V6 combined (A+D) ozone distribution at 68 hPa for December 15, 1978. CI is
590 0.25 ppmv.

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