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3	Variations of Arctic winter ozone from the LIMS Level 3 dataset
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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
- 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
- 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
- the lower mesosphere. Daily, synoptic maps of V6 ozone and temperature illustrate that they are
- an important aid in interpreting satellite limb-infrared emission versus local measurements,
- 29 especially when they occur during dynamically active periods of northern hemisphere winter.
- 30 We then show a sequence of V6 maps of upper stratospheric ozone, spanning the minor
- stratospheric warmings of late January and early February 1979. The map sequence of V6
- 32 geopotential height reveals how ozone was changing in the vortex and at the centers of adjacent
- anticyclones. We also report on zonal variations of the tertiary ozone maximum of the upper
- 34 mesosphere and its associated temperature fields during winter. These several examples provide
- a guide to researchers for further exploratory analyses of middle atmosphere ozone from LIMS.

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37 1 Introduction and objectives

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 middle atmosphere transport processes at high latitudes. 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





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49	Current research is focusing on the 3-dimensional character of ozone in the upper stratosphere
50	and mesosphere, based on more recent satellite datasets. Several studies report on how
51	temperature and ozone vary in association with sudden stratospheric warming (SSW) events (de
52	la Camara et al., 2018; Kim et al., 2020; Shams et al., 2021). Manney et al. (1995) and Harvey et
53	al. (2008) report on the development of low ozone pockets (LOPs) in the region of the Aleutian
54	anticyclone during winter. Siskind et al. (2005; 2021) report on the occurrence of a mesospheric
55	cooling associated with SSWs and on the role of gravity waves for modeling ozone in the upper
56	mesosphere. Chandran et al. (2013) developed a climatology of the Arctic elevated stratopause.
57	Sofieva et al. (2021) analyzed a multiyear dataset for regional trends in stratospheric ozone since
58	2001. LIMS provides similar data on ozone from an earlier decade for further comparisons.

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The LIMS profiles have been retrieved with an improved Version 6 (V6) algorithm. Those V6 60 (or Level 2) profiles were archived in 2008 and include ozone, temperature, and GPH that extend 61 62 from 316 hPa to ~ 0.01 hPa. The co-located V6 profiles of water vapor (H₂O), nitric acid vapor (HNO₃), and nitrogen dioxide (NO₂) extend through the stratosphere. Lieberman et al. (2004) 63 analyzed the V6 temperature profiles and found evidence for non-migrating tides, due to the 64 interaction of the diurnal tide and planetary zonal-wave 1, especially in late January 1979. Holt 65 66 et al. (2010) analyzed the descent of V6 NO₂ from the lower mesosphere to within the polar stratospheric vortex, where it interacts with ozone. Remsberg et al. (2013) assimilated V6 ozone 67 profiles in a reanalysis model and gained improved estimates of column ozone, especially in 68 Arctic winter. Such reanalysis studies assimilate temperature and ozone profiles within a model 69 70 framework. However, the models only approximate the effects of small-scale variations, so it is also useful to consider observed variations of the LIMS parameters without resort to a model. 71 72 Keep in mind that smaller-scale atmospheric variations also contribute to the analyzed intermediate and large-scale fields from V6. This paper provides some examples of the larger-73 74 scale variations of Arctic ozone, temperature, and GPH.

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76 The SPARC Data Initiative (SPARC-DI) includes monthly zonal averages for the chemical 77 species from V6 (see Tegtmeier et al., 2013; SPARC, 2017; and Remsberg et al., 2021). In Section 2 we show January zonal averages of V6 ozone and temperature profiles that extend 78 even higher or to near the mesopause. The V6 Level 3 (map) product provides a 3-dimensional 79 context for those zonal mean data. Daily V6 maps are also an aid in interpretating individual V6 80 81 profiles versus correlative data, especially during dynamically disturbed periods. Specifically, in Section 3 we compare several nighttime V6 ozone profiles with those obtained with a rocket-82 borne chemiluminescent (CHEM) technique (Hilsenrath et al., 1980). Those profile comparisons 83 are for December 15 and for January 27 and 28, when the temperature and ozone fields were 84 affected by planetary wave forcings. There is a corresponding cooling and variations of ozone in 85 the winter lower mesosphere associated with the warming in the upper stratosphere. Section 4 86 presents variations of ozone and GPH at northern extratropical latitudes during the minor SSW 87 events of late January and early February 1979, as a complement to the more comprehensive 88 89 findings of Harvey et al. (2008) on the occurrence of LOPs within anticyclones based on solar occultation satellite data. Section 5 gives some details on the variability of the tertiary ozone 90 maximum in the upper mesosphere during that same period, as an adjunct to the monthly zonal 91 average values reported by Smith et al. (2018). Section 6 considers the variability of V6 ozone 92 93 within the lowermost extratropical stratosphere, but also cautions users about occasional, pseudo-ozone features in the tropics. Section 7 concludes that the V6 Level 3 product represents 94 an important resource for further studies of the effects of transport and chemistry of middle 95 96 atmosphere ozone.

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98 2 Characteristics of V6 Level 3 data

99 2.1 LIMS measurements and analyses

Nimbus 7 was in a near-polar orbit, and LIMS made measurements at ~1 pm local time along its ascending (A or south-to-north) orbital segments and at ~11 pm on its descending (D or north-to-south) segments. A-D time differences are of the order of 10 hours at most all latitudes because LIMS viewed the atmosphere 146.5° clockwise of the spacecraft velocity vector, as seen from above, or 33.5° counterclockwise from its negative velocity vector. The A-D differences narrow from 10 to about 6 hours from 60°N to 80°N, due to the orbital geometry of Nimbus 7. The V6





processing algorithm accounts for low-frequency spacecraft motions that affect the LIMS view 106 of the horizon. As a result, its measured radiance profiles are registered well in pressure-altitude 107 (Remsberg et al., 2004). Retrieved V6 ozone, temperature, and GPH profiles extend from 316 108 hPa to ~ 0.01 hPa and have a vertical point spacing of ~ 0.88 km with an altitude resolution of 109 \sim 3.7 km. They occur every \sim 1.6 degrees of latitude along orbits, and LIMS made measurements 110 with a duty cycle of about 11 days on and 1 day off over its planned observing lifetime. The 111 LIMS algorithms (Remsberg et al., 2007) do not account for non-local thermodynamic 112 equilibrium (NLTE) effects in ozone (Solomon et al., 1986; Mlynczak and Drayson, 1990) and 113 in CO₂ (Edwards et al., 1996; Manuilova et al., 1998), so there are positive biases in the retrieved 114 V6 ozone throughout the mesosphere during daylight. However, the V6 nighttime ozone is 115 essentially free of NLTE effects below about the 0.05-hPa level. 116

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A sequential-estimation (SE) algorithm generated daily, zonal Fourier coefficients (zonal mean 118 and up to 6-wavenumber, sine and cosine values) for Level 3 at every 2° of latitude and at up to 119 28 vertical levels (Remsberg and Lingenfelser, 2010). The V6 SE algorithm uses better 120 estimates of data uncertainty and a shorter relaxation time of ~2.5 days for the zonal wave 121 122 coefficients, as compared with the earlier algorithm of Remsberg et al. (1990). The SE analysis is also insensitive to the very few large, unscreened ozone profiles values found in the lower 123 stratosphere, as noted in Remsberg et al. (2013, their Fig. 1a). The SE algorithm combines the 124 coefficients from both the A and D orbital segments and effectively interpolates the profile data 125 in time to provide a continuous, 216-day set of daily zonal coefficients versus pressure-altitude 126 for each of the retrieved LIMS parameters. Those combined (A+D) Level 3 coefficients are the 127 basis for a gridding of synoptic maps at 1200Z of ozone and related parameters. Note that Level 128 3 also contains coefficients from its separate A and D profiles; their 'zonal mean' values 129 correspond to the local time-of-day of their respective measurements. The Level 3 data are in 130 131 ASCII format for easy access and use.

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133 2.2 Monthly average V6 data





One can generate monthly average distributions from the daily Level 3 files of temperature, 134 135 GPH, and species (ozone, H₂O, HNO₃, and NO₂); zonal averages for the V6 species were supplied to the SPARC Data Initiative or SPARC-DI (SPARC, 2017; Hegglin et al., 2021). 136 Tegtmeier et al. (2013) compared the V6 monthly ozone distributions with ones from other 137 satellite-based limb sensors and reported good agreement throughout the stratosphere. Although 138 the species cross sections of SPARC-DI (2017) extend only up to the 0.1-hPa level (~64 km), V6 139 average ozone extends higher or to about 0.015 hPa (~75 km). Figure 1 shows the latitude-140 pressure cross section for January from just the descending (D) orbital profiles, which avoids the 141 NLTE biases from the daytime ozone. Stratospheric ozone mixing ratios in Fig. 1 have largest 142 values at about 10 hPa near the Equator (> 9.2 ppmv), and they decrease sharply above and 143 below that level. Maximum mixing ratios for the middle to high latitudes occur between 3 to 5 144 hPa, due to the larger zenith angles and longer paths of the ultraviolet light for production of 145 atmospheric ozone. There is a nighttime ozone minimum of ~ 1.2 ppmv across most latitudes of 146 the middle mesosphere. A tertiary ozone maximum is present in the upper mesosphere near the 147 day/night terminator zones of the LIMS measurements for January (~50°S and ~65°N), in 148 accordance with the interpretation of Marsh et al. (2001). The location (~ 0.02 hPa) and 149 150 magnitude (~3.5 ppmv) of the NH maximum agree with those reported from subsequent satellite studies by Smith et al. (2018, their Fig. 4). On the other hand, while the V6 ozone poleward of 151 $\sim 60^{\circ}$ S is also from descending orbital profiles, it corresponds to daylight conditions at the high 152 southern latitudes in January. Thus, the decrease of mesospheric V6 ozone at 0.1 hPa and 153 154 poleward of 60°S in Fig. 1 indicates merely a change from night to day values.

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156 Radiances from two 15-µm CO₂ channels are used for retrievals of V6 temperature versus

pressure or T(p), and they are also free of NLTE effects below about the 0.05-hPa level (~70 km)

158 (Lopez-Puertas and Taylor, 2001). Retrievals for T(p) in the stratosphere account for the effects

- 159 of horizontal temperature gradients to first order. Single profile errors for T(p) range from 1 K at
- 160 10 hPa to greater than 2 K in the upper mesosphere. Systematic errors from T(p) are the primary
- source of bias error for ozone, which grow to of order 16% in the middle mesosphere (Remsberg
- 162 et al., 2021, Table 1). As a complement to the V6 ozone of Fig. 1, we show the descending
- 163 (~nighttime) V6 T(p) distribution for January in Figure 2, which extends to near the 0.01-hPa





- 164 level. The large-scale features of the T(p) distribution compare well with climatological values 165 from the late 1970s (Fleming et al., 1990), having a maximum value of about 285 K at the high 166 latitude, SH stratopause and minimum values of < 200 K at the tropical tropopause and near the 167 summer mesopause. There is also an apparent elevation of the Arctic zonal-average stratopause.
- 168
- Figure 3 shows average, zonal (wave) standard deviations (SD) about the daily zonal mean of the 169 combined-mode (A+D) V6 ozone for January, where the SD values are also part of the LIMS 170 SPARC-DI product. There are relatively small SD values at low latitudes from 7 to 10 hPa; it is 171 assumed that they are a result of both Kelvin and gravity wave activity. Effects of more 172 vigorous, planetary wave activity are most apparent at high northern latitudes throughout the 173 stratosphere and in the upper mesosphere. Ozone shows little zonal variation in the SH upper 174 175 stratosphere of Fig. 3, due to constraints on the upward propagation of waves through the summer zonal easterlies. SD values near the tropical tropopause are due mostly to residual 176
- effects of emissions from thin cirrus and represent spurious ozone variations (see Section 6).
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179 **3.** V6 comparisons with rocket-borne chemiluminescent ozone measurements

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

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Ozone comparisons for December 15 are in Figure 4, where the four V6 profiles are spaced about 2.6° in latitude along the descending orbital segment with the LIMS instrument viewing in the NNW direction. The short-dashed V6 profile is at 29.2°, and the long-dashed profile is at 37.2°. The location of the rocket ozone sounding lies midway between them. Horizontal bars on the profiles are estimates of ozone error; they overlap between V6 and CHEM, except in the upper stratosphere. However, the corresponding V6 polar ozone map at 4.6 hPa reveals a region





192 of elevated ozone near White Sands (blue dot) on that date. Note that while zonal variations in the polar plot are from a gridding (2° latitude and 5.625° longitude) of the Level 3 coefficients, 193 there is no smoothing of the gridded field in the meridional direction; there is good continuity 194 across latitudes, nonetheless. The CHEM profile is a local measurement and has a vertical 195 resolution that ranges from 1.5 km at 60 km to 0.1 km at 20 km; the nearby V6 profiles have a 196 lower vertical resolution of ~ 3.7 km and are an average over the finite horizontal length (~ 300 197 km or $\sim 3^{\circ}$ latitude) of the LIMS tangent layer. Thus, an ozone field that varies in both space and 198 time may lead to a somewhat reduced quality for comparisons between the localized rocket and 199 limb-viewing satellite profiles in Fig. 4. 200

201

Because V6 ozone is obtained from retrievals of the measured V6 ozone radiance profiles, the 202 203 LIMS retrieved temperature profile must be representative of the atmospheric state for the forward model of ozone radiance. Figure 5 shows the corresponding temperature comparisons 204 between V6 and the correlative rocket Datasonde instrument (*). Agreement between them is 205 very good throughout the upper stratosphere, which means that the retrieved V6 ozone is nearly 206 207 unaffected by temperature bias error. Temperatures do not agree as well in the lowermost mesosphere. The map of V6 temperature in Fig. 5 clearly indicates that there are significant 208 variations in the temperature field at 0.68 hPa near 35°N on December 15. Still, there is little 209 evidence of disagreement between V6 and CHEM ozone in Fig. 4 at 0.68 hPa, indicating that the 210 211 temperature variations are determined well along the LIMS view path for the forward radiance calculations of ozone. 212

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214 The two late January comparisons occurred at the time of a stratospheric, zonal wave-1 warming 215 event and during a time of advection of air from lower latitudes to near the Pole (Leovy et al., 1985). Figure 6 shows three co-located V6 ozone profiles from along a nearby descending 216 orbital segment at ~2204Z (nighttime). The LIMS instrument was viewing from its sub-satellite 217 location of 80.5°N, 130°E, and the CHEM rocket launch was two hours earlier or at 2005Z; there 218 is good agreement for ozone between them, even in the mesosphere. A second rocket launch 219 followed at 0833Z of the next day. Since the V6/CHEM ozone and T(p) comparisons are similar 220 for the two days, we show results for January 27 only. The CHEM sonde recorded two ozone 221





- 222 maxima, one at about 0.6 hPa and another near 15 hPa, and the V6 profiles of Fig. 6 also have 223 maxima at those levels plus an ozone minimum at about 3 hPa. Agreement is good between the profiles, although the CHEM profile has more vertical structure. The ozone maximum at about 224 15 hPa is primarily due to advection of ozone of higher mixing ratios from lower latitudes just 225 prior to the warming event. Leovy et al. (1985) provide a more detailed discussion of the
- 226
- advective changes for ozone in the middle stratosphere during January 1979. 227
- 228

Figure 7 shows the V6 temperature profile comparisons; T(p) from the Datasonde also has more 229

vertical structure, as expected from a localized measurement. Although there is significant 230

horizontal structure in the temperature field surrounding Poker Flat, the V6 temperature profiles 231

agree reasonably with the Datasonde values and indicate that retrieved V6 ozone again has very 232

little bias due to temperature. T(p) values reach a maximum of order 250 K at about 3 to 4 hPa, 233

or where the ozone profiles in Fig. 6 reveal a relative minimum within a low-ozone pocket 234

(LOP) that extends to other levels or from about 7 hPa to 2 hPa. 235

236

The polar vortex is located over northern Europe and Asia on January 27; it is shifted off the 237

Pole because of effects from large-scale, planetary waves on the development of SSWs 238

(Andrews et al., 1987, Chapter 6). The polar vortex region has low ozone and relatively cold 239

240 temperatures; stratospheric temperatures over Alaska have a relative maximum (the SSW). The

- sounding rocket profile from Poker Flat occurs near the center of an anticyclone and in the 241
- region of relatively low ozone. 242

243

Figure 8 shows that there is concurrent cooling at 0.46 hPa or above the Alaskan anticyclone, 244

245 and the corresponding nighttime (or D) ozone field exhibits a local maximum at that same level.

246 Conversely, there is a major temperature increase in the Arctic region above the polar

stratospheric vortex over northern Europe, where ozone values remain relatively low. Since 247

248 ozone is an excellent tracer of transport processes in Arctic winter, it can reveal dramatic

changes with altitude, associated with this SSW event. In summary, Figs. 4 through 8 show the 249

250 utility of daily maps from LIMS Level 3 for the validation and interpretation of the ozone fields





- during dynamically disturbed conditions. In the next section, we consider sequences of polar
- plots of both GPH and ozone from January 27 through February 17, 1979, to illustrate the value
- of the V6 Level 3 products for studies of ozone transport over time.
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255 4. Variations of upper stratospheric ozone during stratospheric warmings

Manney et al. (1995) and Harvey et al. (2004, 2008) contain comprehensive analyses about the 256 occurrence of polar anticyclones and their associated LOPs from analyses of GPH and ozone 257 fields from several different satellites. They determined the extent and character of the polar 258 vortex based on meteorological data from the UK Met Office and as obtained from relatively 259 low, vertical resolution radiance profiles from operational, nadir temperature sounders. Those 260 meteorological analyses extend through the stratosphere but only up to the lower mesosphere. 261 262 The V6 GPH coefficients extend through both the stratosphere and mesosphere, as derived from the V6 T(p) profiles that have a vertical resolution of order 3.7 km. Thus, the LIMS dataset 263 offers potentially more detail about the occurrence of LOPs. 264

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The first panel of Figure 9 shows a map of NH GPH at 4.6 hPa on January 27 for comparison 266 267 with the ozone map in Fig. 6 and the temperature map in Fig. 7. Lowest ozone values are in the polar vortex, which is asymmetric about the Pole. A second, relatively low value of ozone (or 268 LOP) is associated with the anticyclone over the Alaskan sector. One can determine horizontal 269 winds from gradients of GPH on the 4.6-hPa surface and thereby estimate the transport of ozone 270 271 to first order. Qualitatively, the direction and strength of the large-scale transport follows from 272 the character of the cyclonic and anticyclonic features on the GPH map. A large-scale cyclonic circulation about the vortex transports air from middle latitudes to across the Pole on January 27. 273

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The other three panels of Figure 9 are a sequence of three daily NH maps of GPH from February 3 to February 17; each successive map is spaced one week from the previous one. This sequence shows that both the vortex and anticyclone weaken during the three weeks following January 27





at this level. The vortex is re-centered on the Pole on February 17, and the anticyclone is nearly

absent at this level following the two minor warming events.

280

281 The map sequence in Fig. 9 also indicates that there are significant changes in the horizontal transport of ozone during this dynamically active period. Morris et al. (1998) conducted model 282 calculations to show that there can be chemical changes of ozone in the upper stratosphere at that 283 time. Ozone reactions are affected by changes with latitude in solar insolation, temperature, and 284 NO_x. As an example, Figure 10 (top left) is a map of the V6 descending (nighttime) NO₂ on 285 January 27, and it shows that air having larger NO₂ values was transported northward from 286 middle latitudes toward the anticyclone. Thus, it is likely that there is some loss of ozone due to 287 reactions with NO_x (and NO₂) in the middle stratosphere that is contributing to the LOP feature. 288 The other three panels of Figure 10 show the evolution of ozone for February 3 through 17, 289 following that of January 27 (in Fig. 6). Based on the GPH panels of Fig. 9, there was a 290 deepening of the LOP from January 27 to February 3, but a filling of it thereafter. Transport of 291 air from middle to higher latitudes decreased from February 3 to February 17, when the vortex 292 293 and anticyclone weakened. But the anticyclone also weakened from January 27 to February 3. 294 Thus, a chemical loss of ozone due to NO_x may also have occurred during that first week. Although we do not show a sequence of maps for other levels, the V6 Level 3 product indicates 295 significant variations in ozone, temperature, GPH, and NO₂ throughout the upper stratosphere. 296

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298 5. Variations of the tertiary ozone maximum

Smith et al. (2018) describe the changing monthly, zonally averaged character of the wintertime tertiary ozone maximum of the polar upper mesosphere. They point out that the low latitude edge of the tertiary ozone maximum is where HO_x radicals and the chemical loss of ozone due to reactions with them are reduced. The Level 3 ozone and temperature products provide daily, 3dimensional information about those processes. The top two panels of Figure 11 are of upper mesospheric ozone on the 0.022-hPa surface (~72 km) for January 13 and February 10, respectively. The bottom two panels show the variations of temperature on those same days.

306 First, consider the two ozone plots, which are from only the descending (or nighttime) orbital





- 307 segments. V6 ozone radiance profiles have low signal-to-noise in the upper mesosphere; the 308 precision estimate for retrieved ozone and its Fourier coefficients is 0.32 ppmv at 0.022 hPa. 309 Therefore, we show gridded ozone maps in Fig. 11 based on just the zonal mean and the sine and 310 cosine coefficients for waves 1 and 2. Elevated values of ozone occur at a lower latitude on 311 January 13 than on February 10, which is consistent with the slightly different location of the 312 terminator on those two days. One can also see that the tertiary maximum has a larger value on
- 313 February 10 than on January 13.

314

The bottom two panels of Fig. 11 are maps of temperature for January 13 and February 10, 315 respectively, for comparison with the ozone fields. The temperature maps are gridded from the 316 zonal mean and coefficients out to zonal wavenumber 6. There is significant zonal structure in 317 them, and temperatures are much warmer in the Canadian sector on January 13 than on February 318 10. There are meridional gradients of temperature on both days. On January 13 there is also a 319 well-defined mesospheric vortex (not shown), and the highest values of ozone correlate 320 reasonably with it; by February 10 the vortex was disturbed at this level. Recall that there were 321 322 two minor warmings and associated lower mesospheric cooling events during late January and early February. It is very likely that there were wave-driven disturbances in the upper 323 mesosphere during that time, due to filtering of gravity waves and their dissipation (e.g., Siskind 324 et al. 2005). We infer that the warmer temperatures of mid-January at this level led to a radiative 325 326 relaxation and a descent of relatively dry air into the vortex region. A reduction in water vapor will mean that there are fewer HO_x radicals for the destruction of ozone near the terminator zone, 327 leading to an accumulation of ozone by February 10. Although the seasonal evolution of the 328 tertiary ozone maximum is understood reasonably well (Smith et al., 2018), there is more 329 330 information about this ozone feature from the daily maps of ozone, T(p), and GPH from Level 3.

331

6. V6 ozone in the lowermost stratosphere

333 Significant exchanges of air and ozone occur from the extratropical stratosphere to the

troposphere in winter and spring (Gettelman et al., 2011). Figure 3 also showed that there are

335 large zonal variations about the daily zonal means of ozone in the Arctic region of the lower





stratosphere. There are similar variations in GPH (and derived winds) and in zonal wave activity
that lead to ozone transport. Zonal variations are resolved in the daily ozone maps down to the
146-hPa level. In fact, Shepherd et al. (2014) integrated the V6 monthly zonal mean ozone
above the tropopause and subtracted it from observed total ozone, as part of their assessment of
long-term trends of tropospheric ozone from models. Their calculation of extratropical
tropospheric ozone based on LIMS agrees with that obtained from other ozone datasets.

latitudes, and similar anomalies occur in other LIMS months, too (not shown). As an example, 344 Figure 12 shows an ozone map at 68 hPa (\sim 18 km) for the correlative measurement day of 345 December 15, 1978, to give more insight about the source of the tropical variations. Ozone 346 347 mixing ratio values in Fig. 12 are of order 2 to 3 ppmv at high latitudes, becoming much smaller in the subtropics. However, there is also an unexpected high value of 2 to 3 ppmv at about 15°N, 348 150°E. Limb measurements in the ozone channel include radiance effects from cirrus particles 349 that can occur along the view path, although the retrieved ozone mixing ratio profiles were 350 351 screened of those effects to first order (Remsberg et al., 2007). Even so, we note that ozone is 352 easily affected by any excess radiance because of highly non-linear effects for retrievals of ozone from the radiances in the lower stratosphere. It is very likely that the anomalous ozone at 68 hPa 353 is a result of residual effects from subvisible cirrus, which is nearly ubiquitous over the western 354

tropical Pacific region (see SPARC, 2006, Fig. 1.8).

356

357 While individual V6 ozone profiles may include such spurious features in the tropics, the Level 3 ozone product at 68 hPa is affected mainly when there is organized convection and outflow of air 358 that persist for several days. The adjacent map of tropical ozone at 46 hPa appears unperturbed 359 (not shown), and tropical ozone at 100 hPa approaches zero. There are much smaller anomalies 360 in maps of nitric acid, as its mixing ratio retrieval is very nearly linear. Anomalies are not 361 present in maps of V6 H₂O at 68 hPa because the cloud screening algorithm for H₂O accounts 362 for the larger vertical field-of-view and extent over altitude for the measurements of the water 363 364 vapor channel of LIMS. To summarize, one must be mindful that the Level 3 product may show pseudo ozone at 68 hPa in the tropics, but likely not in the extratropics. 365





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367 7. Conclusions on the utility of LIMS V6 Level 3 ozone

This report provides guidance to researchers for their use of the LIMS V6 Level 3 product and 368 369 their generation of daily gridded distributions of its temperature, ozone, and GPH on pressure surfaces. H₂O, NO₂, and HNO₃ are also available for the stratosphere from the Level 3 product. 370 The V6 dataset represents an early baseline for considering changes in the middle atmosphere 371 from 1979 to today and into the future. Atmospheric concentrations of the greenhouse gases 372 (GHG), CO₂, CH₄, and CFCs, were smaller in 1979 versus now. As an example, the LIMS 373 374 algorithm for retrieving T(p) profiles is based on a middle stratosphere CO₂ value of 327 ppmv, compared with \sim 415 ppmv in 2021. As a result, middle atmosphere T(p) distributions were 375 warmer in 1979, which affects both the chemistry and transport of ozone. LIMS also made 376 377 measurements at a time when stratospheric effects from volcanoes were minimal and when catalytic effects of chlorine on ozone were relatively small. Accordingly, Stolarski et al. (2012) 378 found small, but significant changes in the distribution of upper stratospheric ozone for recent 379 decades compared with 1978-1979. 380

381

The LIMS measurements in the winter Arctic region occurred when there was a lot of wave 382 activity for the transport and mixing of ozone. Ozone varies dramatically in winter, particularly 383 384 during times of stratospheric warming events. There was a so-called Canadian warming in early December 1978, two minor SSW events in late January and early February, and a final warming 385 in late February 1979. We showed V6 comparisons with temperature and ozone profile data 386 387 obtained using rocket-borne chemiluminescent (CHEM) techniques, and we pointed out how an examination of changes in their nearby fields are essential for the interpretation and validation of 388 389 V6 profiles against correlative measurements. The Level 3 dataset provides daily details on those variations in latitude, longitude, and altitude, along with related variations in temperature, 390 geopotential height, and NO₂. We noted also that there are instances of spurious, excess ozone 391 from the Level 3 coefficients at 68 hPa in the tropics but not in the extratropical stratosphere. 392 One may find that there are regional changes in ozone in recent decades compared with that in 393 394 1978-1979 from V6 (see, e.g., Sofieva et al., 2021).





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- V6 nighttime ozone is accurate up through the middle mesosphere, and surface maps of the large-scale variations of ozone are relatively accurate through the upper mesosphere. Daily ozone and temperature maps reveal zonal wave features in the region of the wintertime, tertiary ozone maximum of the upper mesosphere. Together with V6 maps of T(p) and GPH, one may explore the daily evolution of that ozone maximum throughout the NH winter of 1978-1979.
- 401

402 Data Availability

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

411 Author Contributions. ER wrote the manuscript and prepared the figures with contributions from

- his co-authors. EH provided his rocketsonde data on ozone and temperature along with theirerror estimates.
- 414

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

416

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430 Figure 1—LIMS V6 Level 3 monthly zonal mean ozone for descending-mode only (or nighttime

431 equatorward of ~55°S) for January 1979. Contour interval (CI) is 0.4 ppmv.





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





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439 Figure 3—Zonal standard deviation about the average of (A+D) ozone for January 1979.







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Figure 4—(top) Profiles of V6 ozone compared with CHEM sonde ozone (*) on December 15.

The four V6 profiles are separated by about 2.5° latitude on the descending orbit. Horizontal

bars are estimates of ozone error. (bottom) NH V6 ozone distribution at 4.6 hPa; Greenwich

448 $(0^{\circ}E)$ is at right, and contour interval (CI) is 0.5 ppmv. Latitudes (dotted circles) are spaced

449 every 10° . The blue dot denotes White Sands.





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453 Figure 5—(top) Profiles of V6 temperature compared with Datasonde values (*) on December

454 15. The four V6 profiles are separated as in Fig. 4, where the short-dashed curve is for 29.2° and

the long-dashed curve is for 37.2°. (bottom) NH V6 temperature distribution at 0.68 hPa;

456 contour interval is 5 K, and blue dot denotes White Sands.





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

462 (bottom) NH V6 distribution of ozone at 4.6 hPa; CI is 0.5 ppmv and red dot is Poker Flat.





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Figure 7—(top) As in Fig. 5, but for January 27, 1979. (bottom) NH V6 temperature at 4.6 hPa
on January 27; Contour interval is 5 K. Blue dot is location of Poker Flat.





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474 Figure 8----(left) Temperature at 0.46 hPa for January 27, 1979, for comparison with Fig. 7.

475 (right) Ozone at 0.46 hPa for comparison with Fig. 6. Contour interval for T(p) is 5 K and for

476 ozone is 0.25 ppmv. Red dot is location of Poker Flat.







Figure 9-- NH V6 GPH at 4.6 hPa; contour interval (CI) is 0.375 gpkm. Blue dot is location of
Poker Flat. Panels are spaced one week apart; (top left) January 27; (top right) February 3;
(bottom left) February 10; and (bottom right) February 17.

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Figure 10—Maps for 4.6 hPa of (top left) nighttime NO₂ on January 27, CI is 1.25 ppbv and red
dot is Poker Flat; (top right) ozone on February 3; (bottom left) ozone on February 10; and
(bottom right) ozone on February 17; CI is 0.5 ppmv. Ozone color bar applies to the bottom two

494 panels, as well.







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Figure 11—(top) Distributions of NH ozone at 0.022 hPa on January 13 (left) and February 10
(right), respectively; contour interval is 0.5 ppmv. (bottom) NH temperature at 0.022 hPa on

502 January 13 (left) and February 10 (right); contour interval is 5 K.



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Figure 12—NH V6 ozone distribution at 68 hPa for December 15, 1978. Contour interval is
0.25 ppmv.





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