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### Key Points:

- Satellite data show strongly depleted HCl and enhanced ClO and ClONO<sub>2</sub> in the southern midlatitude lower stratosphere through much of 2020
- The unprecedented midlatitude chlorine repartitioning likely arose from heterogeneous activation on widespread Australian wildfire smoke
- The sustained midlatitude chlorine activation was far weaker than in typical winter polar vortices, inducing at most minor changes in ozone

### Supporting Information:

Supporting Information may be found in the online version of this article.

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## Prolonged and Pervasive Perturbations in the Composition of the Southern Hemisphere Midlatitude Lower Stratosphere From the Australian New Year's Fires

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**Abstract** The 2019/2020 Australian New Year's wildfires injected record amounts of smoke and biomass burning products into the lower stratosphere. The Aura Microwave Limb Sounder (MLS) tracked the evolution of distinct plumes of fire-influenced air as they rapidly spiraled up to the mid-stratosphere. In the months following the fires, smoke spread throughout the Southern Hemisphere (SH) stratosphere. We contrast the evolution of the SH midlatitude lower stratosphere in 2020 with the 17-year MLS record. Long after the coherent plumes dispersed, data from MLS and other satellite instruments show unprecedented persistent and pervasive depletion in HCl (50%–60% below climatology) and enhancements in ClO and ClONO<sub>2</sub> that were not transport related; peak anomalies occurred in mid-2020. We conclude that the observed perturbations likely arose from heterogeneous chlorine activation on widespread smoke particles. The sustained chlorine activation was far weaker than in typical winter polar vortices, inducing at most minor changes in ozone.

**Plain Language Summary** In late December 2019/early January 2020, during the Black Summer fire season in Australia, an intense outbreak of a number of fire-induced and smoke-infused thunderstorms occurred, collectively known as the Australian New Year's event. Multiple plumes of polluted air were rapidly lofted from the surface and deposited in the lower stratosphere (above 14 km altitude). The largest of the plumes of biomass burning pollutants were tracked by the Microwave Limb Sounder (MLS) on NASA's Aura satellite for several months as they circled the globe. In addition to those distinct confined plumes, smoke from the fires permeated the Southern Hemisphere (SH) lower stratosphere. Well after the discrete plumes dispersed, MLS and other satellite instruments observed widespread and long-lasting perturbations in the composition of the SH midlatitude lower stratosphere. In particular, unprecedented depletion in the main stratospheric chlorine reservoir species (HCl) was accompanied by enhancements in another chlorine reservoir (ClONO<sub>2</sub>) as well as the ozone-destroying form of chlorine (ClO), with maximum departures from average measured in mid-2020. These anomalous signatures likely arose from chemical processing on smoke particles. The enhancement in the ozone-destroying form of chlorine, far weaker than that in winter polar regions, had only minor effects on ozone.

## 1. Introduction

Intense heating by severe wildfires can trigger vigorous smoke-infused thunderstorms, termed pyrocumulonimbus (pyroCb), that rapidly loft polluted air from the surface to the lower stratosphere (e.g., Fromm et al., 2010; Peterson et al., 2021). The Aura Microwave Limb Sounder (MLS) provides profiles of a suite of atmospheric trace gases, including ozone, water vapor, reservoir and reactive chlorine species, and several markers of biomass burning pollution. Although MLS had observed significant composition anomalies caused by pyroCbs in the upper troposphere and lower stratosphere in the past (e.g., Pumphrey et al., 2011, 2021), the stratospheric impacts of previous events were eclipsed by those from the Australian bush fires of late 2019/early 2020, known as the Australian New Year's pyroCb complex (ANY). MLS measurements were used to track the injection and subsequent ascent of multiple ANY plumes, the most dense of which contained exceptional enhancements in

water vapor and biomass–burning products ( $\text{CO}$ ,  $\text{HCN}$ ,  $\text{CH}_3\text{Cl}$ ,  $\text{CH}_3\text{CN}$ , and  $\text{CH}_3\text{OH}$ ) in the lower and middle stratosphere, accompanied by values of stratospheric species ( $\text{O}_3$  and  $\text{HNO}_3$ ) more typical of tropospheric air (Kablick et al., 2020; Khaykin et al., 2020; Schwartz et al., 2020). The main plume remained observable by MLS for  $\sim 110$  days, circling the globe twice while ascending to  $\sim 35$  km. The dynamics of the rapidly rising confined air masses were investigated by Allen et al. (2020) and Lestrelin et al. (2021).

Apart from the swiftly ascending discrete plumes, smoke lofted either directly within the ANY pyroCbs or by energetic deep convection downwind from the fires permeated the Southern Hemisphere (SH) lower stratosphere, reaching polar latitudes by February (Hirsch & Koren, 2021; Yu et al., 2021) and attaining an altitude of 22 km by March (Peterson et al., 2021; Yu et al., 2021). Long after the distinct ANY plumes dispersed, unprecedented perturbations in the composition of the SH midlatitude lower stratosphere remained evident, with the most dramatic signatures of anomalous behavior seen in  $\text{HCl}$  and  $\text{ClONO}_2$  (the main reservoirs of chlorine in the stratosphere) and  $\text{ClO}$  (the main form of ozone–destroying reactive chlorine). We focus here on these prolonged and pervasive perturbations. We contrast the evolution of the SH midlatitude stratosphere in 2020 with the 17–year MLS record and with that in the Northern Hemisphere (NH) in 2019, when the eruption of Raikoke (Kloss et al., 2021b) injected a roughly comparable amount of volcanic aerosol into the lower stratosphere at a corresponding latitude during the equivalent season.

## 2. Data and Methods

We primarily use Aura MLS measurements (version 5, v5; described in detail by Livesey et al., 2020). MLS  $\text{ClO}$  has non–negligible biases at the lowest retrieval levels (68–147 hPa); as no nighttime  $\text{ClO}$  was measured at the SH midlatitudes considered here,  $\text{ClO}$  biases are mitigated by taking ascending ( $\sim 2$  p.m. at these latitudes) minus descending ( $\sim 2$  a.m.) differences as recommended by Livesey et al. (2020) for non–polar regions. MLS  $\text{N}_2\text{O}$  is subject to a negative drift that, while partially alleviated in v5, remains about  $-3\%$ /decade (Livesey et al., 2021). Accordingly, the MLS  $\text{N}_2\text{O}$  anomalies presented below have been “detrended” by removing a linear fit to the daily values across the Aura mission at each gridpoint.

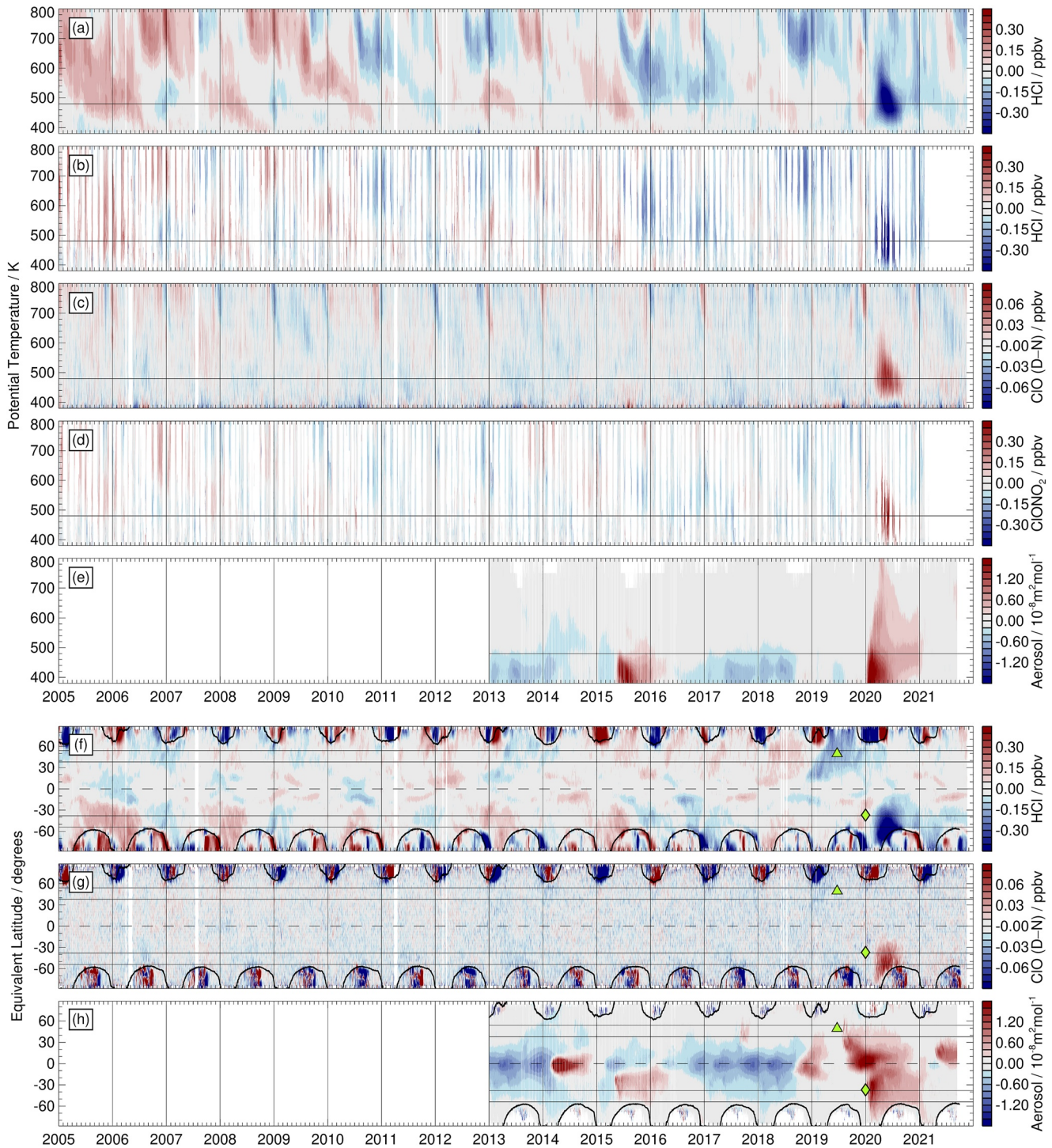
We supplement the MLS analysis with version 4.1 measurements from the Atmospheric Chemistry Experiment–Fourier Transform Spectrometer (ACE–FTS, Boone et al., 2020). For aerosols, we use version 2.0 extinction coefficient profiles obtained by the Ozone Mapping and Profiler Suite Limb Profiler (OMPS–LP) at 869 nm, as recommended for tracking events such as volcanic eruptions or pyroCb injections (Taha et al., 2021). We also use meteorological fields (temperatures, potential vorticity (PV)) from the Modern Era Retrospective Analysis for Research and Applications Version 2 (MERRA–2, Gelaro et al., 2017).

Satellite data are cast into the quasi–Lagrangian PV–based equivalent latitude ( $\text{EqL}$ ; Butchart & Remsberg, 1986) and potential temperature coordinate system to facilitate tracking stratospheric air motions and allow parcels with similar dynamical histories to be grouped together. For interpolation to isentropic surfaces, the aerosol data are first converted to cross section per mole of air ( $\text{m}^2\text{mol}^{-1}$ ), which, unlike the more commonly used extinction ( $\text{km}^{-1}$ ), is conserved under changes of atmospheric pressure in the absence of aerosol formation or loss. The conversion factor relating cross section to extinction is the local molar volume,  $RT/p$  ( $\text{m}^3\text{mol}^{-1}$ ), where  $R$  is the molar gas constant,  $T$  is temperature, and  $p$  is pressure. We use the remotely sensed optical properties directly without conversion to aerosol surface area or volume density, as would be necessary to use these data as inputs in chemical models (e.g., Hervig & Deshler, 1998; Kovilakam & Deshler, 2015).

## 3. Results

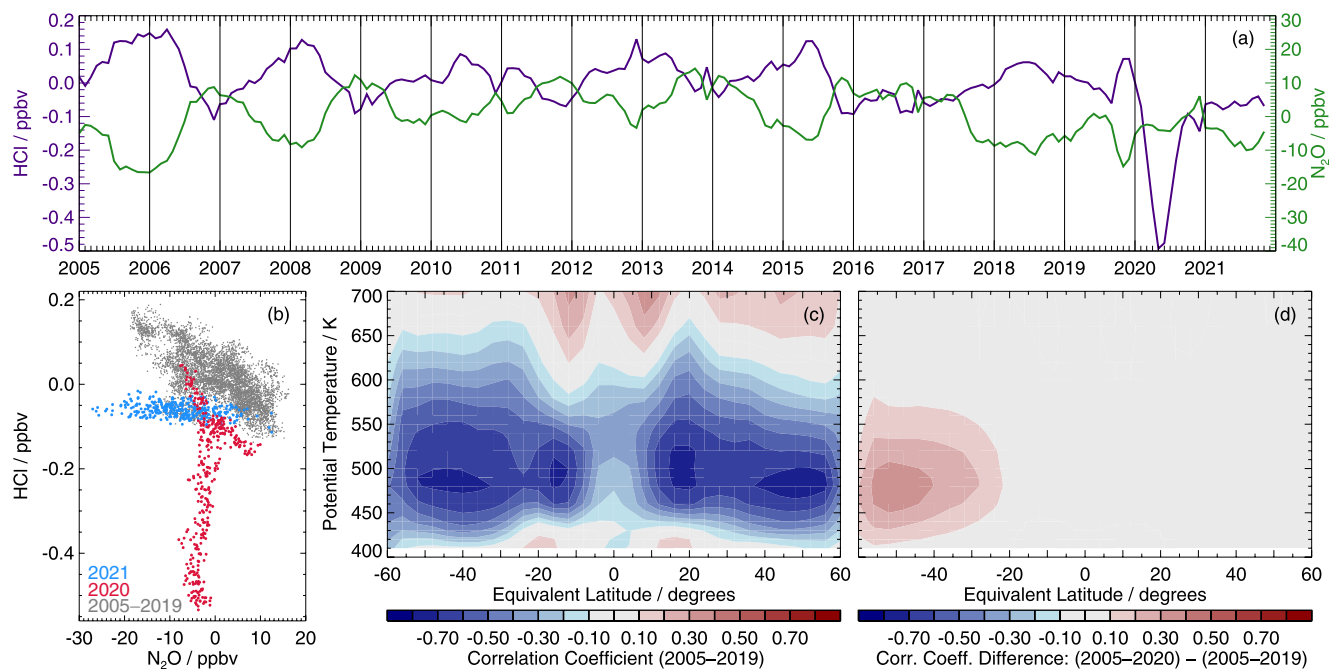
### 3.1. The Southern Hemisphere Midlatitudes in 2020

Figures 1a–1d show negative anomalies in  $\text{HCl}$  (from MLS and ACE–FTS) and positive anomalies in  $\text{ClO}$  and  $\text{ClONO}_2$  (from MLS and ACE–FTS, respectively) in the SH midlatitudes in 2020 that are unprecedented in the record. Anomalies are calculated by subtracting from the daily data in each  $\text{EqL}$ /potential temperature bin the corresponding climatological (2005–2020 for MLS and ACE–FTS, 2013–2020 for OMPS–LP) value for that day of year. All chlorine species depart from climatology from  $\sim 400$  to  $650$ – $700$  K, with peak anomalies at



**Figure 1.** Potential temperature/time sections of anomalies in the 38°S–54°S EqL band of (a) Microwave Limb Sounder (MLS) HCl, (b) Atmospheric Chemistry Experiment–Fourier Transform Spectrometer (ACE–FTS) HCl, (c) MLS CIO (day minus night), (d) ACE–FTS CIONO<sub>2</sub>, and (e) Ozone Mapping and Profiler Suite Limb Profiler (OMPS–LP) aerosol; the horizontal line marks 480 K. EqL/time series of MLS (f) HCl and (g) CIO (day minus night) and (h) OMPS–LP aerosol anomalies at 480 K. Black overlays identify the approximate boundary of the winter polar vortices (as defined by Lawrence et al., 2018). Solid horizontal lines mark 38° and 54° EqL in both hemispheres. Green diamonds (triangles) mark times/latitudes of ANY (Raikoke). Blank spaces represent data gaps.





**Figure 2.** (a) Time series of anomalies (calculated as in Figure 1) of HCl and detrended N<sub>2</sub>O (see Section 2) averaged over 38°S–54°S EqL at 480 K. For clarity monthly means are plotted. (b) Scatter plot of daily HCl and N<sub>2</sub>O anomalies in the 38°S–54°S EqL band at 480 K (gray dots; 2020 in red, 2021 in blue). (c) EqL/potential temperature cross section of the HCl/N<sub>2</sub>O correlation coefficient calculated from daily means over 2005–2019. (d) As (c), but showing the difference between the correlation coefficients calculated over 2005–2020 and 2005–2019.

~460–520 K (see also Figures S1a and S1b in Supporting Information S1). Figures 1f and 1g, Figures S1g and S1h in Supporting Information S1 show that, on isentropic surfaces near their peak, the perturbations in HCl and ClO rapidly extend from subtropical to polar EqLs. The enhancements in ClO and ClONO<sub>2</sub> are roughly coincident in space and time with the depletion in HCl, although the sparse data coverage of ACE–FTS precludes definitive determination of the timing and duration of the ClONO<sub>2</sub> anomaly. Consistent with previous results (Hirsch & Koren, 2021; Kablick et al., 2020; Khaykin et al., 2020; Yu et al., 2021), Figures 1e and 1h, Figures S1d and S1j in Supporting Information S1 show the rapid vertical and horizontal dispersion of ANY smoke, which reached potential temperatures as high as 800 K and pervaded much of the SH within weeks of the fires.

Although the concurrent changes in the major chlorine species, together with those in aerosol, strongly suggest heterogeneous chemical processing, we must consider the possibility that large-scale transport contributed to the unusually low HCl. Accordingly, we correlate the distribution of HCl with that of N<sub>2</sub>O, a long-lived tracer of dynamical processes in the stratosphere whose vertical and meridional gradients are opposite those of HCl. In addition, to ensure that the results are not skewed by residual uncorrected drift in the MLS N<sub>2</sub>O data (see Section 2), we examine CH<sub>3</sub>Cl, a relatively long-lived species that is also useful as a tracer of air motions (Santee et al., 2013). Outside the polar regions, where seasonal changes in HCl are controlled by both dynamical and chemical processes, the patterns in HCl are typically mirrored by converse patterns in the two transport tracers (cf. Figure 1f with Figures S2f and S2g in Supporting Information S1). A clear example of a dynamically induced feature in HCl is the negative anomaly throughout the NH in 2019; the anomalous transport, noted previously (Manney et al., 2020), is the subject of ongoing study. Time series and scatter plots of HCl and N<sub>2</sub>O anomalies in the SH midlatitude lower stratosphere show a strong anticorrelation over the MLS record except for 2020, when no comparable spike in N<sub>2</sub>O is observed to match the steep drop in HCl (Figures 2a and 2b). While the anomalies in CH<sub>3</sub>Cl do not perfectly track those of N<sub>2</sub>O, they exhibit the same general anticorrelation with HCl (Figures S3a, S3b in Supporting Information S1). The correlation coefficient calculated over the interval 2005–2019, which is strongly negative throughout most of the lower stratosphere (Figure 2c, Figure S3c in Supporting Information S1), is considerably weakened in the SH midlatitudes when the calculation is extended to encompass 2020 (Figure 2d, Figure S3d in Supporting Information S1). This weakening of the transport-controlled HCl/N<sub>2</sub>O and HCl/CH<sub>3</sub>Cl anticorrelation (whose month-to-month evolution in 2020 is shown in Figure S4 in Supporting

Information S1) provides convincing evidence that the striking features in the chlorine species in Figure 1 are primarily chemical, not dynamical, in origin. The hemispheric scale of the HCl depletion is illustrated in Figure S5 in Supporting Information S1.

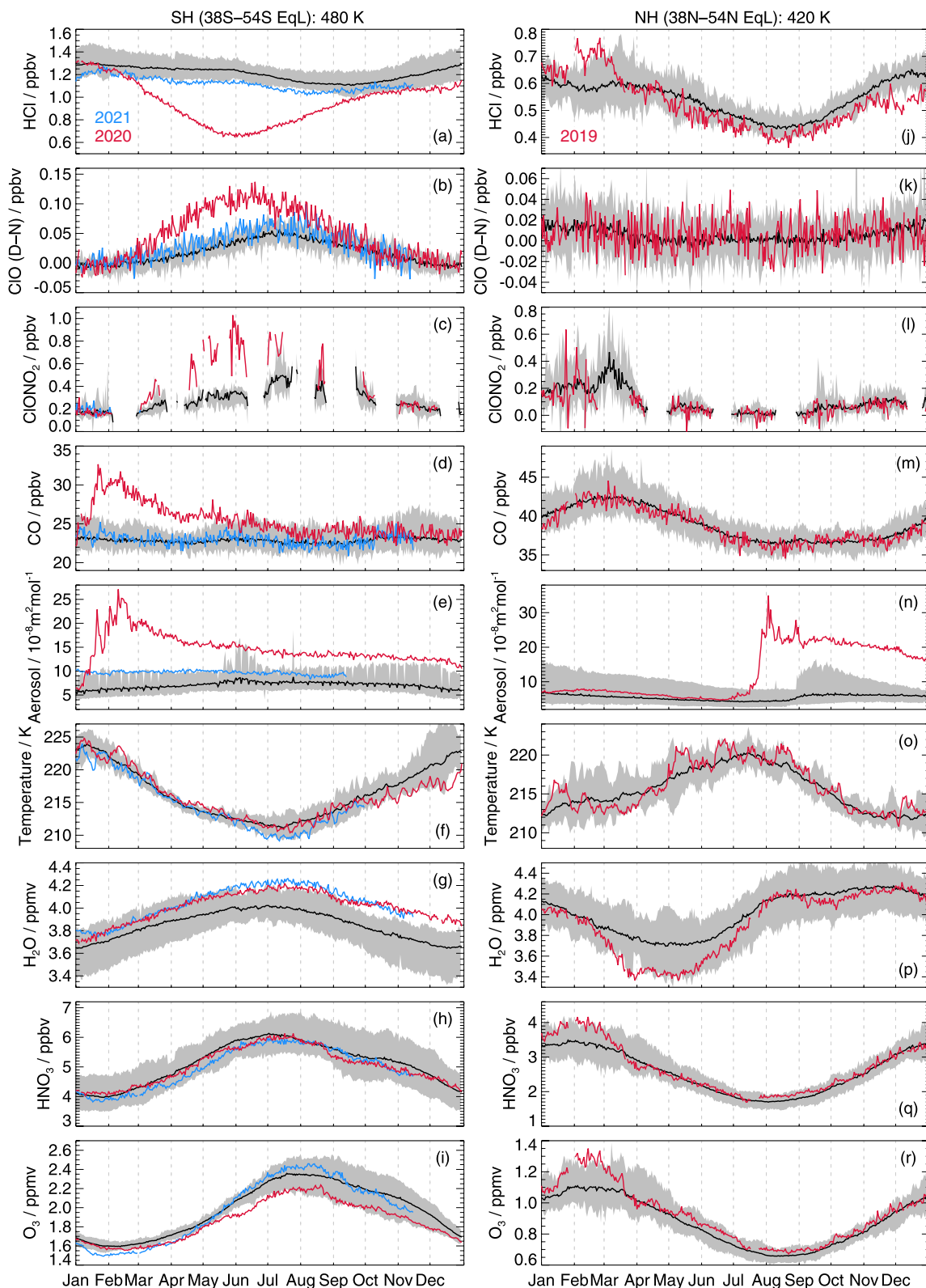
In Figure 3 we place the 2020 observations into climatological context and examine the evolution of chlorine partitioning following ANY in relation to fire emissions, temperature, and water vapor abundances. As in Figures 1a–1e, we focus on the 38°S–54°S EqL band, which was selected to highlight the SH midlatitude region where perturbations in the chlorine species are largest while mitigating the confounding influence of chemical processing inside the Antarctic winter polar vortex (Figures 1f and 1g, Figures S1g and S1h in Supporting Information S1). Although the HCl depletion and ClO enhancement at SH midlatitudes are stronger at 500 K and endure longer at 460 K (Figures S1a, S1b in Supporting Information S1), 480 K (~19.5 km in this EqL band in June) captures both the intensity and the persistence of the chlorine repartitioning.

Figure 3a shows that HCl mixing ratios at 480 K start to substantially deviate from climatology in late January, drop below any values previously observed by MLS in mid-February, then decrease fairly smoothly until reaching the deepest depletion in May/June, after which they gradually rebound while continuing to redefine the mission envelope through the end of the year. The slight dip in late 2020 and ongoing low values in 2021 are most likely dynamical in origin (Figure 2 and Figure S3 in Supporting Information S1). At the time of maximum deficit, HCl abundances are as much as 0.6 ppbv, or nearly 50%, below the climatological mean. The behavior of ACE–FTS HCl is very similar (Figure S6b in Supporting Information S1), though with a slightly larger maximum deviation from its climatology (0.7 ppbv, ~60%).

ClO likewise resets the MLS–measured maxima from early March through August, with values as much as 0.1 ppbv above average in May/June (Figure 3b); the enhancement then fades by October. Analysis of MLS observations of previous severe pyroCb events (Pumphrey et al., 2011, 2021) demonstrated that air masses with enhanced abundances of biomass burning pollutants may also register elevated ClO, but such apparent enhancements are retrieval artifacts induced by contamination of the MLS ClO spectral signature by methanol (CH<sub>3</sub>OH), a product of wildfires. Methanol was extraordinarily enhanced in the ANY plume, but with its relatively short lifetime, the signal decayed within a few weeks (Schwartz et al., 2020). Moreover, a secondary MLS ClO product retrieved from radiances in a different spectral band also indicates substantial enhancement in March through July (Figure S6d in Supporting Information S1); although of generally poorer quality than the standard ClO product (Livesey et al., 2020), the secondary ClO measurements are not subject to CH<sub>3</sub>OH interference. Thus, while high ClO values in early January (Figure 3b) are attributable to a contaminating signature from CH<sub>3</sub>OH that is not completely removed in day minus night differences, spectral interference from that molecule cannot account for the ClO enhancement in subsequent months.

Similarly, ClONO<sub>2</sub> values surpass those previously measured by ACE–FTS over the March–July period, with maximum anomalies greater than 0.6 ppbv on a few days (Figure 3c). Thus, the deficit in HCl is approximately balanced by increases in ClO and ClONO<sub>2</sub>, although the combined enhancements in the latter two diurnally varying species (sampled at different local times) occasionally exceed the MLS–observed (but not the ACE–observed) depletion in HCl. When this EqL band is sampled by the sparse solar occultation measurements, far fewer points are obtained than for a limb emission sounder with dense uniform coverage. Consequently, ACE–FTS averages have poorer precision and show larger day–to–day variability than those from MLS.

CO is included as a representative marker of biomass burning pollution lofted into the stratosphere; other fire emission products measured by MLS show similar, albeit noisier, evolution. After a brief initial pulse in early January indicative of the rapid ascent of the main ANY plume through this layer of the atmosphere due to solar heating of absorptive aerosol (Kablick et al., 2020; Khaykin et al., 2020; Schwartz et al., 2020), CO abundances then increase rapidly through the rest of the month and remain well outside the mission envelope into mid-May and above the climatological mean into October (Figure 3d, Figure S1c in Supporting Information S1). Kloss et al. (2021a) report a similar evolution in 15–day 30°S–60°S averaged MLS CO in 2020 compared to climatology at 68–215 hPa. The evolution of CO closely tracks that of OMPS–LP aerosol (Figure 3e, Figure S1d in Supporting Information S1); both are in line with the calculations of Yu et al. (2021), who found that substantial smoke–induced zonally averaged upward motion occurred in the SH midlatitudes while the aerosol was relatively dense in January and then subsided. Thus the onset of chlorine activation coincides with a steep increase in aerosol cross section, which by mid-February exceeds climatological mean values by a factor of ~4.



**Figure 3.** EqL–band (38°–54°) averages for the Southern Hemisphere at 480 K (a–i; 2020 in red, 2021 in blue) and the Northern Hemisphere at 420 K (j–r; 2019 in red) of Microwave Limb Sounder (MLS) (a, j) HCl and (b, k) ClO (day minus night); (c, l) Atmospheric Chemistry Experiment–Fourier Transform Spectrometer ClONO<sub>2</sub>; (d, m) MLS CO; (e, n) Ozone Mapping and Profiler Suite Limb Profiler aerosol; (f, o) Modern Era Retrospective Analysis for Research and Applications Version 2 temperature; and MLS (g, p) H<sub>2</sub>O, (h, q) HNO<sub>3</sub>, and (i, r) O<sub>3</sub>. Gray shading depicts the envelope of behavior and black lines the mean over 2005–2019.

Although aerosol gradually declines thereafter, the cross sections remain nearly twice the norm and above any previously measured at this EqL/potential temperature (apart from late May/June, when the eruption of Calbuco in 2015 briefly produced a bump in the OMPS–LP record) into 2021; the longevity of the signature is consistent with the stratospheric residence time of the smoke reported by Peterson et al. (2021). Measurements from the Stratospheric Aerosol and Gas Experiment III instrument on the International Space Station indicate similarly strong and long-lasting aerosol enhancements (Kloss et al., 2021a), as do those from the Cloud–Aerosol Lidar with Orthogonal Polarization (not shown).

Climatological temperatures generally decrease over the first half of the year, from ~224 K to ~210–212 K. In 2020, temperatures are ~1 K below average in late January, but they rise to ~2.5 K above average at the time of peak aerosol loading in February and remain slightly elevated until June (Figure 3f), reflecting the stratospheric warming caused by ANY smoke (Kablick et al., 2020; Khaykin et al., 2020; Rieger et al., 2021; Yu et al., 2021). Under normal stratospheric conditions, heterogeneous reaction rates are negligible at these temperatures. Water vapor is near its annual cycle minimum when chlorine repartitioning begins in 2020; however, 2020 H<sub>2</sub>O abundances are slightly (~0.05–0.15 ppmv) above average, reaching a maximum of ~4.2 ppmv in July and redefining the envelope (excluding 2021) thereafter (Figure 3g). The relatively moist conditions in the 2020 SH midlatitudes observed by MLS (and ACE–FTS) in much of the lower stratosphere (Figures S1f, S2d, and S2e in Supporting Information S1) are likely attributable to the wet phase of the horizontal tropical “tape recorder” in stratospheric water vapor (e.g., Rosenlof et al., 1997; Figure S2h in Supporting Information S1), although the record amount of H<sub>2</sub>O in the primary ANY plume (Schwartz et al., 2020) may have played a minor role right after the fires. HNO<sub>3</sub>, although a key participant in heterogeneous chemical processes, shows no notable deviations from climatology in most of 2020 in either MLS (Figure 3h) or ACE–FTS (not shown) measurements.

Modeling studies exploring the potential for chemical ozone loss outside the winter polar regions (e.g., Anderson et al., 2012; Bregman et al., 2002; Robrecht et al., 2019; Solomon et al., 1997; von Hobe et al., 2011) have shown that heterogeneous reaction rates are strongly dependent on temperature, water vapor abundances, and aerosol surface area and particle properties. Airborne in situ measurements provided evidence of chlorine activation on volcanic or background sulfate aerosol or cirrus ice clouds in the midlatitude and tropical near–tropopause region (e.g., Borrmann et al., 1997; Keim et al., 1996; Thornton et al., 2007; von Hobe et al., 2011). However, the ClO enhancements in those cases occurred under considerably colder and moister conditions than those reported here. Even if some local temperatures were lower than the EqL–band averages in Figure 3f, they are unlikely to have dipped into the 200–205 K range needed to initiate heterogeneous processing even for moderately (3–10×) enhanced (volcanic) aerosol or convectively injected high (10–20 ppmv) H<sub>2</sub>O (e.g., Robrecht et al., 2019). Moreover, it was far drier in the SH midlatitudes in 2020 than the conditions often assumed in modeling investigations; also, MLS observed H<sub>2</sub>O abundances as high or higher in other years with no appreciable perturbation in chlorine species. Our results, therefore, indicate that the key driver in 2020 was the presence of an extraordinary amount of particulate surface area from the ANY fires. Using a coupled aerosol–climate model, Yu et al. (2021) found that ANY smoke was composed of 2.5% black carbon, with the remainder mainly organic carbon. While such particles initially have very different physical properties than sulfate aerosols, polar stratospheric clouds (PSCs), or cirrus ice (the surfaces typically associated with heterogeneous chlorine activation), over time they may become coated with sulfuric acid, as has been shown to occur on stratospheric volcanic ash (Pueschel et al., 1994) or mineral dust (Korhonen et al., 2003). That the largest anomalies in the chlorine species were observed several months after the peak in aerosol cross section may reflect not only the timing of minimum temperatures and maximum H<sub>2</sub>O values, but also the timescales over which a substantial fraction of smoke particles in a broad swath of the SH stratosphere acquired a sulfate coating. Despite the continuing large aerosol loading, HCl then began to “recover” at a time when it normally decreases and while temperatures were still seasonally low and H<sub>2</sub>O abundances high (Figures 3a, 3e, 3f, and 3g). We note that preliminary simulations using state-of-the-art chemical models do not reproduce the observed features (S. Strahan, M. Chipperfield, D. Kinnison, *personal communications*, 2021). As discussed by Murphy et al. (2021), organic–sulfate particles from the troposphere differ from sulfuric acid aerosols of stratospheric origin in several ways (e.g., size, acidity, water content), and the effects of those differences on heterogeneous chemistry are poorly understood. Further analysis and modeling, beyond the scope of this paper, is necessary to elucidate the particular particle properties that enabled efficient heterogeneous processing under the prevailing conditions.

The midlatitude chlorine activation in 2020 was not as complete as that observed in the winter polar regions, where vortex-averaged HCl and ClONO<sub>2</sub> values approach zero and ClO values exceed 1 ppbv (e.g., Santee et al., 2008). In the core of the cold polar vortex once sunlight returns, high values of ClO coincide with low values of ClONO<sub>2</sub>; in the “collar” regions around its edge, ClO is deactivated to form ClONO<sub>2</sub>. In contrast, in the sunlit midlatitude lower stratosphere in 2020, enhancements in ClO and ClONO<sub>2</sub> coexisted for several months. We conjecture that as chlorine was continuously converted from the reservoir species to ClO through ongoing heterogeneous reactions (specifically,  $\text{HCl} + \text{ClONO}_2 \rightarrow \text{Cl}_2 + \text{HNO}_3$ ), HCl, initially the more abundant reactant, steadily declined, whereas ClONO<sub>2</sub> was recycled. That is, heterogeneous and photolytic loss of ClONO<sub>2</sub> were counterbalanced by reformation through reaction of ClO with NO<sub>2</sub>. The latter process depends on the continuing availability of NO<sub>2</sub>, produced through photolysis of HNO<sub>3</sub>. In the winter polar regions, sequestration of HNO<sub>3</sub> in PSCs keeps NO<sub>2</sub> concentrations low, inhibiting ClONO<sub>2</sub> regeneration. At higher temperatures, however, while some of the HNO<sub>3</sub> produced through chlorine activation reactions stays bound in particles, much of it is released to the gas phase (e.g., Robrecht et al., 2019). Consequently, gaseous HNO<sub>3</sub> remained abundant throughout this period in 2020 (Figure 3h), allowing ClONO<sub>2</sub> regeneration. Again, chemical modeling will be needed to confirm this scenario.

An important question is whether the sustained chlorine activation caused substantial chemical ozone destruction in the SH midlatitudes. (An initial transient reduction in ozone within the confined ANY plume is largely consistent with displacement of stratospheric air by uplifted ozone-poor tropospheric air [Khaykin et al., 2020; Schwartz et al., 2020].) Under the assumption that the organic particles became coated or mixed with sulfuric acid as they aged, Yu et al. (2021) employed heterogeneous reaction rates equal to those for sulfate aerosol at the same temperature in their simulations, predicting chemical loss of 5–7 DU in total column ozone in the SH midlatitudes. Ozone values were lower than climatology after January and modestly redefined the MLS range from mid-March through July (Figure 3i), as also noted by Rieger et al. (2021). Largely on the basis of a lack of corresponding anomalies in temperature (as a proxy for circulation effects) and using a short data record (2015–2020), Rieger et al. (2021) inferred a chemical cause for the observed low ozone in 2020. To distinguish transport- and chemistry-related changes in ozone in the 38°S–54°S EqL band, we performed an MLS “Match” analysis, similar to that done by Livesey et al. (2015) to estimate chemical ozone loss in the winter polar vortices. Conclusive evidence of significant chemical destruction in 2020 was not found (Figure S7 in Supporting Information S1). This is not surprising, given that the 2020 enhancement in reactive chlorine, albeit unprecedented in the midlatitudes, was still an order of magnitude or more weaker than that in a typical winter polar vortex. Moreover, a midlatitude Match analysis is inherently noisier, as far fewer matched measurements are obtained than in the polar regions, where the MLS orbit tracks converge. Like HCl, O<sub>3</sub> is generally anticorrelated with N<sub>2</sub>O in the midlatitude lower stratosphere; if any chemical ozone loss occurred in 2020, it was too small to noticeably alter the transport-dominated climatological O<sub>3</sub>/N<sub>2</sub>O correlation (Figure S8 in Supporting Information S1). These results further suggest that, as for HCl, the low O<sub>3</sub> in late 2020/early 2021 likely arose from transport effects. Finally, we also examined anomalies in MLS partial stratospheric column ozone, which generally track the lower stratospheric mixing ratio anomalies (Figure S8 in Supporting Information S1); the stratospheric column estimates show that changes in ozone of the magnitude (a few percent) simulated by Yu et al. (2021) are well within typical interannual variations. We conclude that the prolonged chlorine repartitioning observed in 2020 induced at most minor changes in ozone.

### 3.2. The Northern Hemisphere Midlatitudes in 2019

The June 2019 eruption of Raikoke (48°N) strongly enhanced stratospheric sulfate aerosol throughout the NH (Kloss et al., 2021b; Peterson et al., 2021). Since Raikoke and ANY caused stratospheric injection of roughly comparable amounts of aerosol at corresponding latitudes during the equivalent season (mid-summer) in successive years, we compare the subsequent evolution of chlorine partitioning in the respective hemispheres. Following the Mt. Pinatubo eruption, much greater sulfate aerosol loading led to enhanced ClO at low and middle latitudes as increased heterogeneous hydrolysis of N<sub>2</sub>O<sub>5</sub> shifted chlorine partitioning in favor of ClO by slowing production of ClONO<sub>2</sub> via  $\text{ClO} + \text{NO}_2$  (Avallone et al., 1993; Dessler et al., 1993; Fahey et al., 1993). In contrast, the net effect of enhanced N<sub>2</sub>O<sub>5</sub> hydrolysis on HCl is minimal, and little post-Pinatubo change in HCl was observed (Dessler et al., 1997).



Raikoke aerosol did not penetrate as deeply into the stratosphere as that from ANY; thus we focus on 420 K ( $\sim 16$  km). Substantial aerosol arrives at  $38^{\circ}\text{N}$ – $54^{\circ}\text{N}$  EqL in late July (Figure 3n). Temperatures are near their seasonal maximum ( $\sim 220$  K, Figure 3o) at this time, typically too high for heterogeneous processing. No enhancement in ClO is discernible (Figure 3k). As noted earlier, in 2019 HCl was anomalously low throughout the NH. To remove the signature of transport-related anomalies and quantify changes in HCl, differences from the values on the eruption date are shown in Figure S9 in Supporting Information S1 for 2019 abundances and climatology at several potential temperatures. In the days immediately after the eruption, MLS observed a handful of profiles with strongly enhanced HCl (up to 6–8 ppbv, not shown) at the lowest retrieval levels, indicative of direct volcanic injection of HCl into the stratosphere (e.g., Carn et al., 2016), but these few highly localized transient enhancements should not significantly affect EqL-band averages. In general, the evolution of HCl in the months following Raikoke was unremarkable, although small positive deviations from climatology are seen after August at the higher levels. Examination of other NH EqL bands yields similar results (not shown). Thus, as expected given the temperature and humidity of the NH midlatitude lower stratosphere at the time of the eruption, and consistent with the behavior observed after Pinatubo, Raikoke induced no HCl depletion.

#### 4. Conclusions

MLS and ACE-FTS provide unique measurements of the primary components of the stratospheric chlorine budget, along with transport tracers. In the months following ANY, well after distinct plumes dispersed, these data show unprecedented prolonged and pervasive depletion in HCl (50%–60% below climatology) and enhancements in both ClO and ClONO<sub>2</sub> in the SH midlatitude lower stratosphere that did not arise from transport effects. Maximum anomalies were attained in mid-2020. In contrast, the 2019 Raikoke eruption, which injected roughly comparable amounts of aerosol at corresponding latitudes during the equivalent season as ANY, had no significant impact on NH midlatitude chlorine partitioning. Under normal aerosol loading conditions, heterogeneous reaction rates are negligible at the temperatures and water vapor abundances prevailing in the SH midlatitude lower stratosphere when the anomalous chlorine repartitioning was observed in 2020; however, abundant and widespread aged smoke particles from ANY clearly provided ample surfaces on which efficient heterogeneous chlorine activation could take place over a large fraction of the hemisphere for several months. Although such sustained midlatitude enhancement in reactive chlorine is unmatched in the record, it was still far weaker than that in typical winter polar vortices, inducing at most minor changes in ozone.

ANY demonstrated that extreme pyroCbs can have profound impacts on the composition of the lower stratosphere that persist for months. Detailed investigation, including chemical modeling, is needed to identify the specific characteristics of the ANY smoke particles that facilitated such dramatic midlatitude chlorine repartitioning. Deeper understanding of the processes involved is critical to assessing the likelihood of significant changes in midlatitude composition in a future climate in which the stratosphere is expected to be colder, wetter, and more frequently affected by pyroCb events.

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#### Data Availability Statement

The data sets used here are publicly available, as follows. Aura MLS Level 2 data: <https://disc.gsfc.nasa.gov/datasets?page=1&keywords=AURA%20MLS>; Aura MLS Derived Meteorological Products (DMPs): <https://mls.jpl.nasa.gov/eos-aura-mls/dmp> (registration required); ACE-FTS 4.1 data: <http://www.ace.uwaterloo.ca> (registration required at <https://database.scisat.ca/l2signup.php>); OMPS-LP L2 Aerosol Extinction Vertical Profile swath multi-wavelength daily 3slit Collection 2 V2.0 data: <https://doi.org/10.5067/CX2B9NW6FI27> (Taha, 2020); MER-RA-2: (<https://disc.sci.gsfc.nasa.gov/ui/datasets?keywords=%22MERRA-2%22>).

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