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UPPER TROPOSPHERE LOWER STRATOSPHERE COMPOSITION CHANGE IN
TROPICAL CYCLONES: ASSESSMENTS FROM 17 YEARS OF SATELLITE
OBSERVATIONS

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UPPER TROPOSPHERE LOWER STRATOSPHERE COMPOSITION CHANGE IN
TROPICAL CYCLONES: ASSESSMENTS FROM 17 YEARS OF SATELLITE
OBSERVATIONS

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BY THE COMMITTEE CONSISTING OF

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Abstract

Tropical cyclones (TCs) are known for a variety of impacts, including threats to life and property, although their effect on the chemical composition of the upper troposphere and lower stratosphere (UTLS) remains understudied. The transport of air between the troposphere and stratosphere, known as stratosphere-troposphere exchange (STE), involves greenhouse gases such as ozone and water vapor whose impact on Earth's radiation budget and climate is most sensitive to changes in UTLS composition. Therefore, we examine observations of STE in TCs and relate these changes to TC and environmental characteristics. We utilize 17 years of trace gas profiles from the Microwave Limb Sounder (MLS) aboard the Aura satellite in conjunction with TC track information and environmental data to relate UTLS ozone and water vapor changes to TC intensity, distance from TC center, and environmental wind shear. We find that accounting for varying tropopause heights and basin-specific background composition is important to accurate assessment of UTLS composition changes within a TC. We also find that TCs are associated with increased water vapor throughout most of the UTLS and dehydration at tropopause level, while ozone is reduced greatly in the upper troposphere. All diagnosed UTLS composition changes demonstrate significant sensitivity to distance from TC center and TC intensity. Additionally, we find that TC-induced UTLS H₂O changes are highly dependent on shear magnitude, where weaker shear is associated with more pronounced tropospheric hydration and tropopause dehydration.

Chapter 1

Introduction

Stratosphere-troposphere exchange (STE) is the critical process by which chemically distinct stratospheric air is irreversibly transported to the troposphere or vice versa (Holton et al. 1995). To diagnose STE, ozone (O_3) can be used as a stratospheric tracer (Rodgers et al. 1990); the concentration of O_3 is high in the stratosphere and uniformly low in the troposphere. Analogously, water vapor (H_2O) commonly serves as a tracer of tropospheric air. Both H_2O and O_3 are critical greenhouse gases; H_2O is a major contributor to global radiative forcing, including in the stratosphere, where decreases in concentration have been linked to slower global surface temperature increases (Kiehl and Trenberth 1997; Solomon et al. 2010). Additionally, the radiative sensitivity to H_2O change is largest within the lower stratosphere (Banerjee et al. 2019), and therefore small composition changes can have large climate impacts. Similarly, the vertical distribution of O_3 plays a critical radiative role, as reductions in upper troposphere and lower stratosphere (UTLS) O_3 concentration are associated with surface cooling (Lacis et al. 1990). Thus, the resulting changes in UTLS chemical composition driven by STE have profound implications for Earth's climate system (Holton et al. 1995). Despite the known importance of UTLS composition, our understanding of STE processes and mixing remain limited (Riese et al. 2012). Therefore, developing a better understanding of STE pathways is critical to improvements in modeling Earth's radiative budget.

Past work has identified numerous processes that affect the chemical composition of the UTLS. It has been observed that tropospheric air enters the stratosphere in the tropics, travels poleward within the stratosphere, and sinks back into the troposphere in the extratropics; this slow transport is known as the Brewer-Dobson circulation (Brewer 1949; Dobson 1956) and is driven by extratropical pumping induced by dissipating planetary-scale

and gravity waves in the stratosphere (Butchart 2014). Of these, breaking Rossby waves dominate (Holton et al. 1995). In addition to their dynamical impact on stratospheric circulation, Rossby wave breaking events can also impact the chemical composition of the UTLS when they occur near the tropopause, especially along isentropic surfaces near the subtropical jet (McIntyre and Palmer 1983; Liu and Barnes 2018). Isentropic exchange also occurs within tropopause folds, where stratospheric and tropospheric air overturn due to ageostrophic circulations in upper tropospheric jets and within frontal zones (Shapiro 1980; Keyser and Shapiro 1986). Although tropopause folding represents a majority of STE in the midlatitudes, more rapid processes such as overshooting deep convection have been observed (Jellis et al. 2023; Homeyer and Bowman 2021); over tropical landmasses, this overshooting convection has been shown to contribute significantly to STE and UTLS composition (Pommereau 2010). These processes demonstrate the necessity of considering numerous dynamical phenomena when analyzing UTLS composition and its evolution.

Tropical cyclones (TCs) are another type of weather phenomena that facilitates STE. TCs have a lifespan of days to weeks and are associated with modifications to large-scale upper tropospheric flow (Merrill 1988), increases in upper tropospheric H_2O (Ray and Rosenlof 2007), and decreases in upper tropospheric O_3 (Zhan and Wang 2012). Although TCs comprise a small fraction of deep tropical convection, they represent a disproportionate amount of tropopause-overshooting convection in this region ($\approx 30\%$; Romps and Kuang 2009) and can induce up to 30% of tropical troposphere-to-stratosphere transport (Zhan and Wang 2012); therefore, given their relatively long lifespan in comparison to other phenomena, evaluating the effects of STE within TCs is critical. Furthermore, TC intensity is projected to increase in a warming climate (Emanuel 1987), and overshooting convection frequency is expected to exponentially increase with increasing SSTs (Griffin 2017). Therefore, it is essential to investigate the relationship between storm strength and STE within TCs, particularly given the potential feedback loop between TCs and the global climate system.

Previous studies have outlined some STE mechanisms within TCs. Danielsen (1993) found evidence of a lifted tropopause within TCs which intersects UTLS isentropes. Resulting isentropic mixing facilitates the transport of tropospheric air into the lower stratosphere. Within the troposphere, downward transport associated with TC outflow has led to the intrusion of stratospheric air near the outflow edge (Leclair De Bellevue et al. 2006), particularly in strong cyclones (Baray et al. 1999). Additionally, modeling work has shown the importance of overshooting tops within TCs. Overshooting tops have been associated with robust two-way exchange, particularly within the outer rainbands, despite their small scale (Allison et al. 2018; Preston et al. 2019). Notably, Leclair De Bellevue et al. (2007) demonstrate a potential relationship between STE and vertical wind shear; given that the deepest convection occurs downshear of the core of TCs (DeHart et al. 2014), a further investigation of the relationship between shear and STE is warranted. Finally, within the TC eye, deep subsidence of stratospheric air into the troposphere has been observed and evidenced by increased upper tropospheric O_3 (Rodgers et al. 1990), although work by Joiner et al. (2006) showed negligible increases in O_3 within the eye of a TC when adjusted for cloud pressure. Decreased tropospheric and total column O_3 has also been associated with TCs (Newell et al. 1996; Zou and Wu 2005), indicating the dominance of upward transport of O_3 -poor lower tropospheric air.

Aura Microwave Limb Sounder (MLS) satellite observations have been utilized to evaluate STE-induced UTLS composition change in TCs. When utilizing observations from the North Indian Ocean basin from a limited time period, Ratnam et al. (2016) found stratospheric H_2O enhancement and increased tropospheric O_3 near TC center. Using MLS observations from 3 storms, Jiang et al. (2020) found that a H_2O was reduced at tropopause level and increased in the stratosphere near TCs. Leveraging 2 years of MLS data, Ray and Rosenlof (2007) also found evidence of strong TC-induced hydration within the upper troposphere; the strength of the hydration was correlated with storm intensity, indicating a potential link between TC intensity and STE. Collectively, these studies reveal some

observed composition changes resulting from STE within TCs, although they have used relatively limited sample sizes. Additionally, previous work has focused on a single ocean basin over a limited time period; therefore, a more complete analysis of STE within TCs is warranted.

Given the limitations of previous efforts to improve understanding of STE within TCs, we aim to address these gaps by leveraging a long, global record of satellite observations from the Aura MLS. Specifically, we examine the influence of TCs on UTLS O_3 and H_2O composition over a 17-year period, from 2005 through 2021, representing the longest analyzed observational record of TC effects on the UTLS. This increased temporal coverage and sample size allow for detailed exploration of the relationship between UTLS composition impacts from TCs and storm characteristics, which we investigate in multiple coordinate systems to evaluate the validity of previously identified processes on a global scale. In particular, we investigate how storm intensity, radial distance from the TC center, and environmental deep-layer wind shear relate to changes in UTLS O_3 and H_2O ; quantifying these impacts, particularly in terms of vertical distance from the tropopause, is critical to improving understanding of how TCs impact Earth's radiation budget and climate.

Chapter 2

Data & Methods

2.1 Aura MLS

The Aura satellite follows a sun-synchronous orbital track and MLS has produced nearly continuous profiles of atmospheric composition since its launch in 2004. For analysis, version 5 MLS profiles from 2005 through 2021 are used; data after 2021 are not included to avoid biases resulting from global stratospheric hydration following the 2022 Hunga Tonga-Hunga Ha’apai volcanic eruption Millán et al. (2022). MLS retrieves profiles every 160 km along orbit, with the spatial resolution, accuracy, precision, and retrieval ranges of O₃ and H₂O retrievals recorded in Table 1 Livesey et al. (2022). Notably, the dataset has a relatively coarse spatial resolution; therefore, fine-scale details of TC-associated transport cannot be resolved using these satellite observations. However, the spatial resolution of observations is comparable to the scale of TCs and can be utilized to analyze overall UTLS composition changes in TCs with confidence (owing to robust statistics possible from large sample sizes).

2.2 IBTrACS

The International Best Track Archive for Climate Stewardship (IBTrACS; Knapp et al. 2010) is a comprehensive dataset that synthesizes TC track data from a variety of international reporting agencies. IBTrACS data contains information about the type of system (whether it is tropical, subtropical, or extratropical), as well as an intensity classification (disturbance, tropical wave, tropical depression, tropical storm, or hurricane). For this project, only tropical storms (sustained 10-m winds of 35–63 kts) and hurricanes (sustained

10-m winds of ≥ 64 kts) are considered due to diminishing UTLS impacts at low storm intensities (see Section 3.1). IBTrACS also includes TC location at 3-hour increments at a minimum, as well as data on TC strength and ocean basin of origin. In this analysis, TC profiles from the North Atlantic (NA), South Indian (SI), East Pacific (EP), West Pacific (WP), and South Pacific (SP) basins are utilized. These basins represent nearly all globally observed TCs within the 17-year analysis period, as illustrated in Fig. 2.1.

Using location data from both IBTrACS and MLS profiles, TC characteristics and UTLS composition data are matched, providing an abundant number of samples (summarized in Table 2.2). MLS profiles taken within 1.5 hours and 1000 km of a TC location are flagged as TC profiles, while non-TC profiles are between 1000 and 1500 km removed from the center of a TC (though sampled at the same times). These non-TC profiles provide a reference for the local background environment beyond the main circulation of the TC but are still representative of an environment in which a TC would form, limiting the effects of UTLS seasonal variability.

2.3 MERRA-2 Reanalysis

For environmental conditions, we use 3-hourly assimilations of the global atmosphere from the NASA Modern-Era Retrospective analysis for Research and Applications, Version 2 (MERRA-2). MERRA-2 spans 1979 to the present and has a horizontal resolution of 0.625×0.5 degrees longitude-latitude with 72 vertical levels (corresponding to ≈ 1100 -m grid spacing within the UTLS; Gelaro et al. 2017). MERRA-2 is suitable for this study because its representation of the tropopause and tropospheric winds are comparatively accurate to alternative reanalyses (Xian and Homeyer 2019; Wu et al. 2024), especially relative to the coarse resolution of the MLS profiles. The deep-layer (850–200 hPa) wind shear vector at 3-hourly intervals for each TC is computed following a common approach in TC studies (e.g., see Kaplan and DeMaria 2003; Chen et al. 2006; Hence and Houze 2011): the average value of all MERRA-2 grid points within an annular ring 200–800 km from TC

center, aiming to mitigate influence from the primary circulation of the TC. For tropopause analysis, we apply the World Meteorological Organization (1957) lapse rate tropopause (LRT) definition to MERRA-2 analyses, which has been shown to identify the tropopause height more accurately within than the cold point tropopause definition when considering the tropics (Pan et al. 2018). This reanalysis data is matched with Aura MLS profiles via linear interpolation in space and time.

2.4 Tropopause-Relative Analysis

Because the troposphere and stratosphere are chemically distinct, it is important to evaluate changes in UTLS composition based on whether they occur above or below the tropopause. Thus, in order to accurately assess STE within TCs, MLS profiles are converted into tropopause-relative logarithmic pressure coordinates for both O_3 and H_2O observations, minimizing the effect of variable tropopause heights on the resulting analysis of multiple profiles (see Fig. 2.2). First, tropopause pressure at the MLS profile locations is sourced from MERRA-2 reanalysis data. Next, 12 tropopause-relative bins are created for each profile at a resolution of 0.075 decades of pressure (equivalent to approximately 1 km of vertical distance in the UTLS, as done in Tinney and Homeyer (2021)). Trace gas concentrations are then averaged within tropopause-relative logarithmic pressure bins for comparison of MLS profile samples. Finally, tropopause-relative profiles are transformed back into pressure space for some analyses using the average tropopause pressure for all samples. This allows for reliable comparison of UTLS composition between profiles by avoiding contaminating stratospheric samples with tropospheric observations (and vice versa). This study is the first to employ this technique for the evaluation of UTLS composition change within TCs. Figure 2.3 shows the resulting bulk impacts on assessed UTLS composition change when using pressure coordinates and tropopause-relative coordinates. For O_3 , these changes are most apparent for tropical storms, which seemingly result in increased UTLS

O_3 in standard pressure coordinates (Fig. 2.3a); however, when adjusting to a tropopause-relative framework, tropical storms clearly result in decreased UTLS O_3 (Fig 2.3c). This result removes the effects of mixing tropospheric and stratospheric air and is consistent with past work (e.g. Zou and Wu 2005). UTLS H_2O patterns are similarly altered when converted to a tropopause-relative framework; for major hurricanes, H_2O changes are positive throughout the UTLS in standard pressure coordinates (Fig. 2.3b), while in tropopause-relative coordinates there is a decrease in H_2O near the tropopause (Fig. 2.3d). These UTLS composition changes are only visible in a tropopause-relative framework.

2.5 Basin Relative Analysis

There is great spatial variability in background concentrations of O_3 and H_2O within the UTLS (see Figure 2.4), even within the tropics. Therefore, composition changes are best assessed relative to typical concentrations in the local TC environment. To accomplish this, background profiles are calculated and averaged in tropopause-relative coordinates for reference using only non-TC profiles taken over each basin. As summarized previously (see Section 2.2), these background calculations are restricted to samples from TC times to limit the impacts of seasonal variation. Figure 2.4 demonstrates the large observed variability in UTLS background composition between basins from MLS measurements, especially near and below the tropopause. For instance, H_2O concentrations around the tropopause are on average around 3 ppmv for the SP and SI ocean basins; in contrast, the NA tropopause has a background concentration of ≈ 10 ppmv, more than triple some other basins, due to variability from meteorological and anthropogenic factors. Similar differences are found in the upper troposphere for O_3 . Therefore, it is important to consider which basin a TC forms in to accurately evaluate STE and UTLS composition change. After identifying the ocean basin, the basin-averaged background composition is subtracted from each tropopause-relative TC profile and resulting differences are commensurately averaged to assess composition change. All subsequent analysis is performed using basin-relative considerations.

In Figure 2.5, we demonstrate the necessity of this method by analyzing samples of hurricane-intensity TCs. When considering profiles from the NA basin (relatively high background O_3 and H_2O in the UTLS, as demonstrated in Fig. 2.4) and the SI basin (relatively low background O_3 and H_2O in the UTLS), we find significant absolute concentration differences between basin and TC profiles. The O_3 concentration within NA TCs is significantly higher than the concentration within SI TCs throughout some parts of the upper troposphere (minima ≈ 50 ppbv versus ≈ 30 ppbv). UTLS H_2O shows similar variations; near the tropopause, the average H_2O concentration is ≈ 3 ppmv for SI TCs, but ≈ 10 ppmv for NA TCs (as previously mentioned). Changes relative to average basin-specific background profiles (Figs. 2.5c & 2.5d), however, demonstrate that assessed UTLS composition change becomes more comparable between basins when accounting for their respective backgrounds. There are some complex differences in the profiles of UTLS H_2O changes from TCs, but the vertical structure is similar overall, such as hydration and reduced O_3 in the upper troposphere, a slightly drier tropopause region, and weak stratospheric hydration that is not as readily evident in the absolute concentration plots. Therefore, basin-relative analysis is essential for aggregating TC profiles and assessing UTLS composition change across ocean basins.

2.6 Limb Geometry

An important consideration when leveraging Aura MLS data for studying TCs is the geometry of the limb measurement. Namely, the width of the limb path is approximately 15 km for all MLS observables, but the length of the limb path (along the line of sight) varies by MLS observable and altitude. For UTLS observations, H_2O limb lengths are 185–210 km, while O_3 limb lengths are much coarser at 300–450 km (see Table 2.1). Given such limb geometry, the orientation of the limb measurement relative to a TC determines its utility for assessing TC impacts on UTLS composition as a function of distance from TC center

(radius). Figure 2.6 illustrates this relationship. Namely, Figs. 2.6a & 2.6b show the possible extremes of limb path orientation as that perpendicular to TC center (orientation = 0°) and parallel to TC center (orientation = 90°), respectively. Figs. 2.6c & 2.6d subsequently reveal the resulting radii from TC center limb lengths of 200 km and 400 km, respectively, would span at varying limb orientations and distances from TC center. Near TC center (within one-quarter the limb length), the radii span of a given measurement is generally insensitive to limb orientation as it is roughly collocated with TC center and equivalent to half the limb length. At distances from TC center greater than one-quarter the limb length, a large sensitivity in the radii spanned by a measurement arises due to limb orientation, with orientations $\leq 30^\circ$ resulting in radii spans at or below those near TC center. For these reasons, we determine the limb orientation relative to TC center for all MLS profiles to facilitate analyses partitioned by distance from TC center. Specifically, for further analysis we retain all observations regardless of limb orientation at distances from TC center within one-quarter the limb length and restrict analyses at greater distances to profiles with limb orientations $\leq 30^\circ$ only.

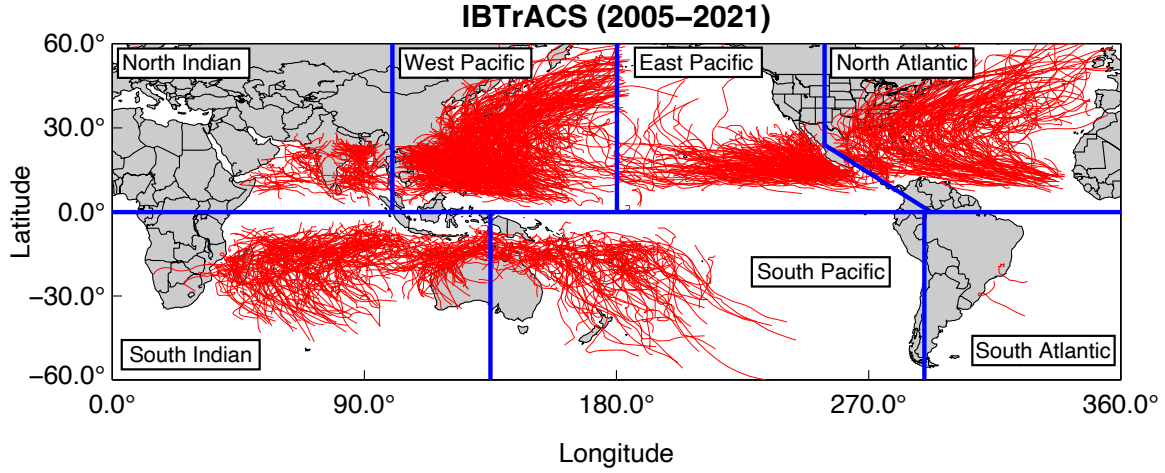


Figure 2.1: All IBTrACS TC tracks (red lines) within the analysis period (2005–2021). Longitude-latitude bounds of IBTrACS basins are indicated by thick blue lines.

Table 2.1: UTLS properties of ozone (O_3) and water vapor (H_2O) MLS observations.

Gas	Δx (km)	Δz (km)	Precision	Accuracy	Retrieval Range
O_3	300–450	2.5–3.5	20–40 ppbv	+7–10%	251–0.03 hPa
H_2O	185–210	1.4–3.1	5–40%	8–25%	316–0.002 hPa

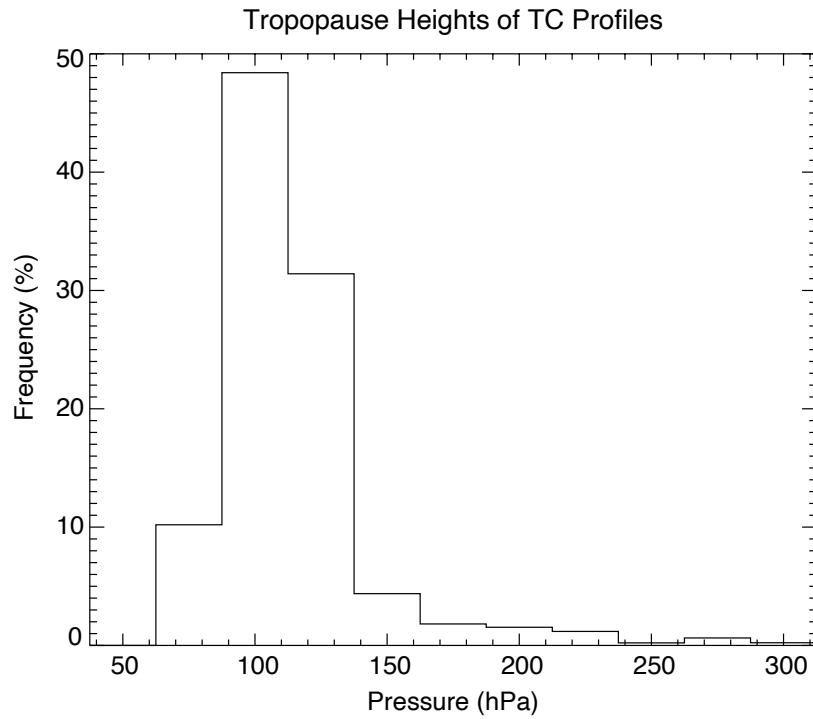


Figure 2.2: A histogram of tropopause heights of all utilized TC profiles.

Table 2.2: Number of TC and Background Profiles by Basin

	WP	EP	SP	NA	SI	Total
Background (non-TC) profiles	36,818	20,579	10,117	26,544	18,692	116,293
Tropical storm profiles	18,316	11,134	5,708	17,500	11,268	63,926
Weak Hurricane profiles	10,142	3,722	2,176	5,467	3,355	24,846
Major Hurricane profiles	1,782	2,013	659	1,879	835	7,168
All TC profiles	30,240	16,869	8,543	24,862	15,458	95,956

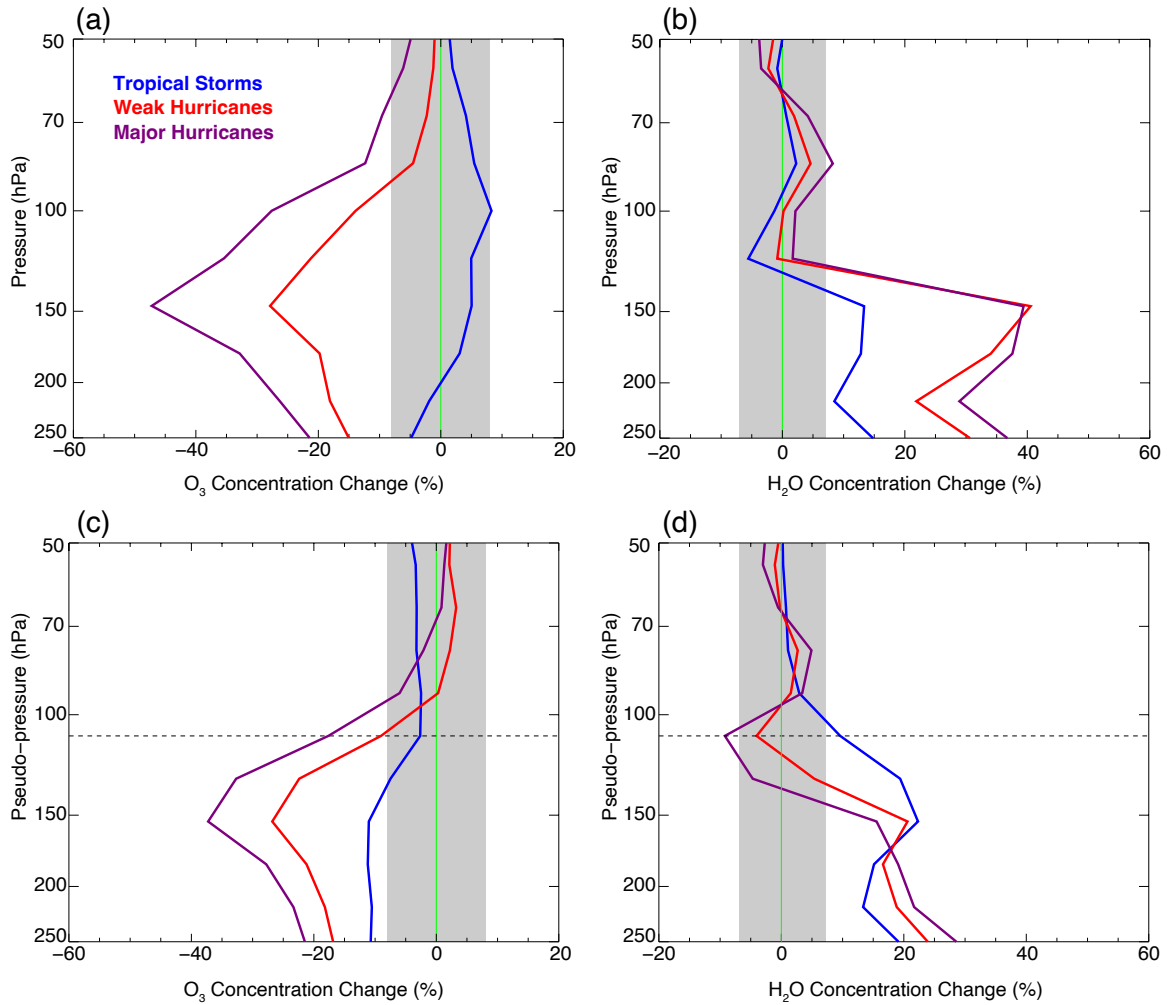


Figure 2.3: Composition change in TCs of tropical storm intensity (blue), hurricane intensity (red), and major hurricane intensity (purple) relative to non-TC profiles (light green reference lines). The dashed line represents the tropopause. Panels (a) and (b) show intensity dependency of ozone and water vapor respectively in raw pressure coordinates, while panels (c) and (d) show intensity dependency of ozone and water vapor in tropopause-relative coordinates. The gray color-filled bars in each panel represent the typical uncertainty of an individual MLS measurement. Analysis includes approximately 63,000 tropical storm profiles, 24,000 weak hurricane profiles, and 7,100 major hurricane profiles.

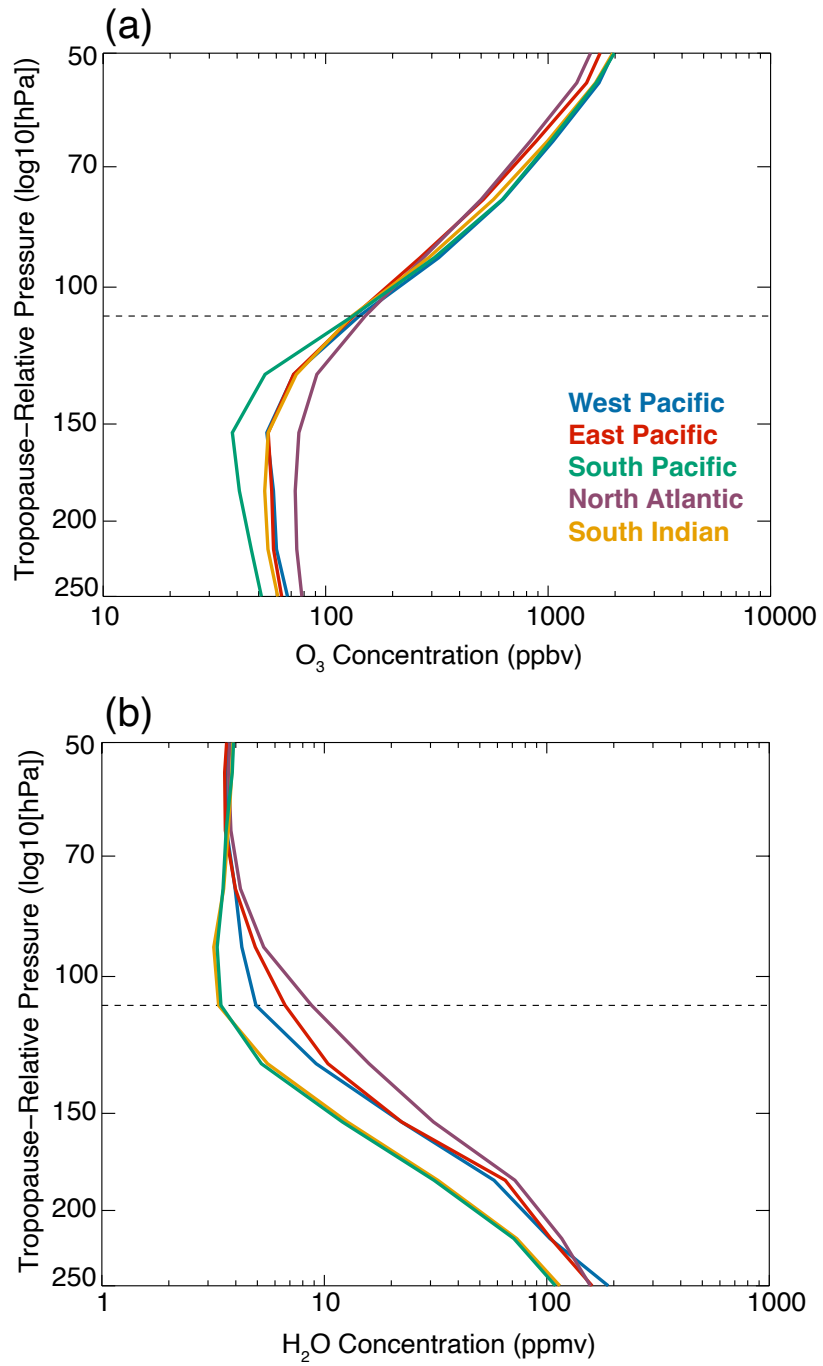


Figure 2.4: Background concentrations of UTLS ozone (a) and water vapor (b) in all analyzed basins plotted in tropopause relative coordinates. Colors corresponding to each basin are defined on the plot.

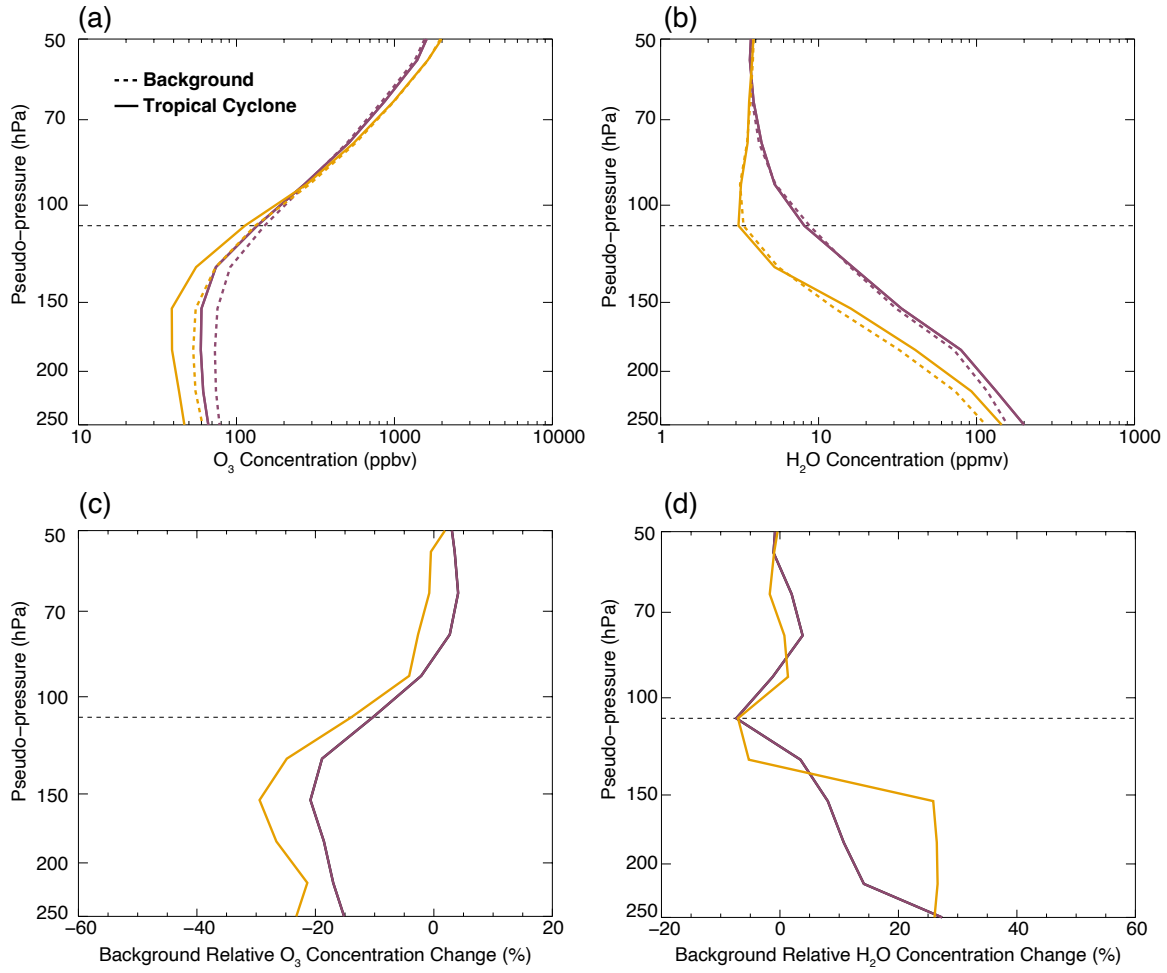


Figure 2.5: Panels (a) and (b) show the tropopause-relative absolute concentrations of ozone and water vapor, respectively, for the North Atlantic (purple) and South Indian (orange) ocean basins, with hurricane profiles represented by solid colored lines and background profiles represented by dashed colored lines. Plots c and d are as above, but with percent change relative to respective basin background conditions. The black dashed line represents the tropopause on each panel.

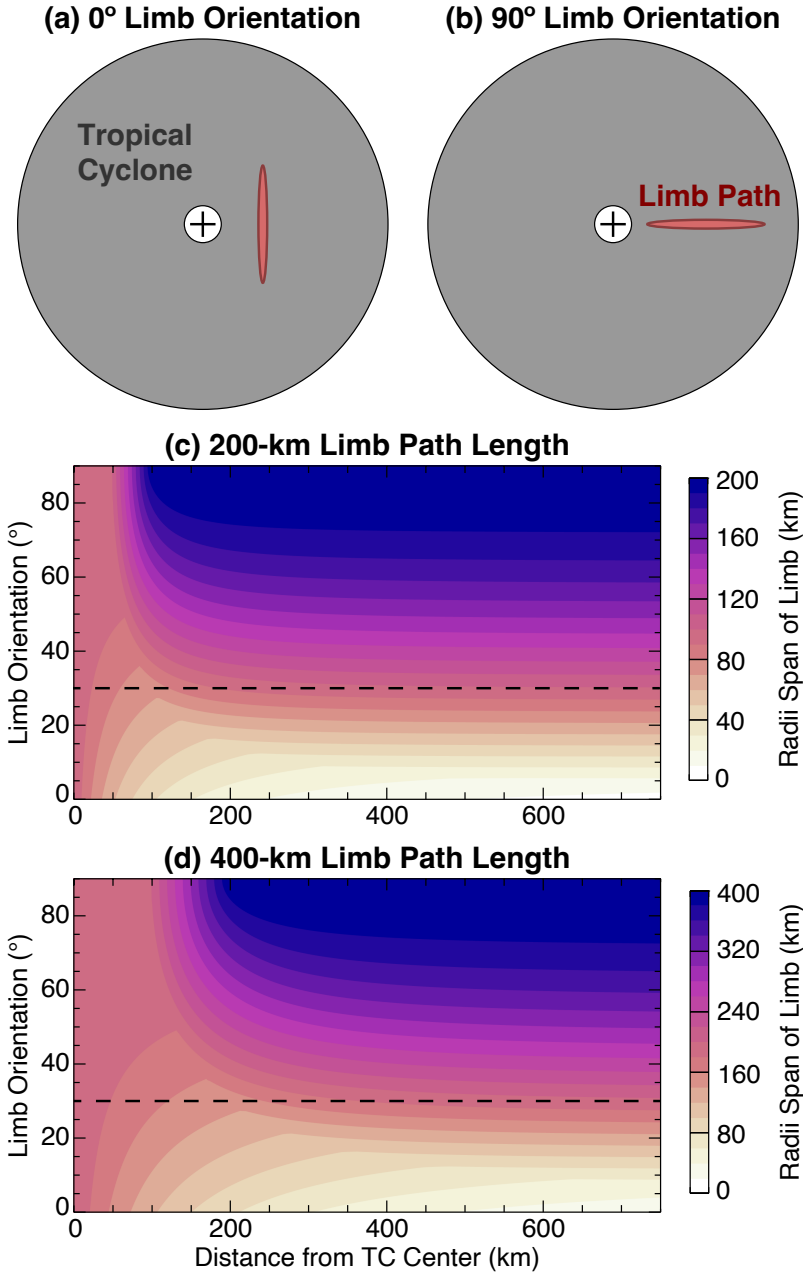


Figure 2.6: An illustration of the impact of MLS limb path geometry on the distances from TC center (radii) spanned by a measurement. Panels (a) and (b) show limb orientations of 0° (perpendicular to TC center) and 90° (parallel to TC center), respectively. Panels (c) and (d) show resulting radii spanned by a limb measurement as a function of distance from TC center and limb orientation for limb lengths of 200 km and 400 km, respectively. The dashed black lines in (c) and (d) lie at a limb orientation of 30°.

Chapter 3

Results

3.1 Intensity dependency

To examine the relationship between TC intensity and UTLS composition change, TC profiles can be grouped based on sustained wind speed. We create three intensity categories: tropical storm intensity (TC profiles with 10-m sustained wind speeds 35–63 kts), weak hurricane intensity (sustained wind speeds of 64–95 kts), and major hurricane intensity (sustained wind speeds ≥ 96 kts). Given the distance dependency of STE processes (see Section 3.2), we require all profiles used for intensity analysis to be within 300 km of TC center (approximately encompassing the inner core of the TC) and employ the limb geometry criteria as defined in Section 2.6. After profiles are filtered and sorted by intensity, average basin-relative composition changes are computed at tropopause-relative altitude.

UTLS O_3 changes are found to be highly sensitive to TC intensity. In Figure 3.3a, there is nearly an 80% average decrease in upper tropospheric O_3 concentration in major hurricanes; this decrease is only $\approx 40\%$ in tropical storms. Additionally, the deficit is more pronounced within major hurricanes near the tropopause and into the lower stratosphere; the interquartile range plot in Figure 3.4a demonstrates a negative shift in the distribution of composition change for hurricane intensity storms, with the greatest decreases occurring for major hurricanes. This effect is diminished with altitude above the tropopause; apparent variations throughout most of the lower stratosphere are small enough to fall below the accuracy of individual MLS O_3 retrievals. Despite this, profiles demonstrate a clear relationship between TC intensity and UTLS O_3 concentration change, where stronger TCs are associated with a more prominent O_3 deficit in the UTLS, particularly near ≈ 2 km below the tropopause (~ 150 hPa). This result is consistent with the results of Zou and Wu (2005), who found that an increase in TC intensity was associated with decreased upper

tropospheric O_3 ; however, our usage of higher sample sizes across multiple ocean basins provides additional evidence of the robustness of this relationship.

H_2O within the UTLS (Fig. 3.3b) also shows high sensitivity to TC intensity, with stronger TCs resulting in larger increases in upper tropospheric H_2O concentration, consistent with that found in Ray and Rosenlof (2007). The biggest differences in this study are found between tropical storms (average enhancements of up to 50% in the upper troposphere) and weak hurricanes (enhancements of nearly 80%). The differences between weak hurricanes and major hurricanes are comparably minor, with relatively overlapping distributions throughout the UTLS (see Fig. 3.4b) except for a significantly drier tropopause within major hurricanes, as found in Jiang et al. (2020) and expected due to previously-diagnosed tropopause cooling and lifting. In particular, the tropopause-level H_2O concentration is around 20% lower than the typical background concentrations within major hurricanes; this feature is not present within tropical storms. Finally, a lower stratospheric hydration signal is strongest for major hurricanes, where H_2O concentrations increase by more than 10% on average ≈ 2 km above tropopause (~ 80 hPa). This can be seen in the shifting of interquartile ranges for major hurricanes in particular (Fig 3.4b), where more subtle increases are found for weak hurricanes and tropical storms. Overall, there is a clear link between TC intensity and UTLS H_2O change.

3.2 Distance dependency

To evaluate the relationship between UTLS composition change and distance from TC center we bin tropopause- and basin-relative profiles every 100 km and employ the limb geometry constraints from Section 2.6 for all bins except that closest to TC center. Due to higher resolution of MLS H_2O retrievals, the innermost 100 km from TC center is divided further into 50-km bins. Additionally, only hurricanes are included in this analysis since UTLS impacts from tropical storms are comparatively weak. These constraints ensure an accurate accumulation of UTLS composition changes.

The resultant analysis for O_3 (Fig. 3.5a) demonstrates an intense and significant upper tropospheric deficit. The intensity of this deficit is maximized near TC center and decreases with distance, with the maximum change in O_3 concentration centered just below the tropopause. Additionally, this O_3 deficit weakly extends into the lower stratosphere, particularly within the inner 200 km of the storm, mirroring O_3 decreases near hurricanes found by Zou and Wu (2005). Although previous studies indicate that subsidence within the eye of the TC could enhance upper tropospheric O_3 , this effect is not evident in this analysis. However, given that the 100-km bin size of O_3 observations is significantly larger than a typical TC eye and the MLS limb retrieval path for O_3 will span up to 200 km from TC center even when coincident, this is not surprising. Namely, previous work has indicated the subsidence feature is most evident within 50 km of TC center and is adjacent to the strongest vertical transport of O_3 -poor air within deep convection of the eyewall (Gordon and Homeyer 2025). The competition between the narrow subsidence and broader eyewall convection likely obscures potential upper tropospheric O_3 increases from observation by MLS. Despite this, these results demonstrate a clear relationship between a change in UTLS O_3 concentration and distance from the center of a TC, as O_3 changes are most robust near TC center; therefore, distance from TC center is an important consideration when evaluating UTLS O_3 change.

UTLS H_2O changes in TCs are also largely significant and dependent on distance from TC center. Figure 3.5b shows that TCs result in strong hydration of the upper troposphere, with average changes in H_2O exceeding 50%. These effects decrease with distance, especially beyond 400 km from TC center. Additionally, weak hydration is observed in the lower stratosphere, particularly within the inner 200 km, where average H_2O increases of 10–15% are present. Finally, there is a clear reduction in H_2O at all distances from TC center evaluated that is coincident with the tropopause. This result is in agreement with Jiang et al. (2020), who found TC-induced adiabatic lift created a cold anomaly above the tropopause. This feature was found to enhance H_2O deposition on TC-lofted cloud ice,

which subsequently falls to lower altitudes, thereby drying the tropopause. We find that this feature is most prominent within 50–100 km from TC center and significant despite smaller sample sizes within this bin, although weak tropopause dehydration is observed ubiquitously beyond the center of the storm. Thus, we conclude that UTLS H₂O impacts are highly dependent on distance, especially at altitudes removed from the tropopause.

3.3 Shear Dependency

To evaluate environmental influence on STE within TCs, we examine the effects of deep-layer vertical wind shear on UTLS composition change. For hurricanes only, we group MLS profiles by shear magnitude into 3 classes: weak shear ($< 5 \text{ m s}^{-1}$), moderate shear ($5\text{--}10 \text{ m s}^{-1}$), and strong shear ($> 10 \text{ m s}^{-1}$). Given the demonstrated relationship between TC convection and deep-layer shear, we further classify profiles as located upshear or downshear of TC center and apply the limb geometry constraint. Finally, as also done for the TC intensity analysis, we include only profiles within 300 km from TC center.

Our results show a minor shear dependency for upper tropospheric O₃ and a substantial shear dependency for UTLS H₂O. When examining UTLS O₃, upper tropospheric deficits are stronger on the downshear side of the storm (Fig. 3.1a). These differences are most apparent in strongly sheared environments, where average upper tropospheric O₃ decreases upshear are around 50% compared to 70% downshear. However, closer to the tropopause and into the stratosphere, these differences become negligible. There are limited differences between weak, moderate, and strong shear profiles otherwise, apart from a tendency for a reduced depth of O₃ deficits with increasing shear (especially on the upshear side); there is significant overlap between the interquartile ranges of each shear category (see Fig. 3.2). In contrast, UTLS H₂O shows substantial sensitivity to deep-layer shear (Fig. 3.1b). In the upper troposphere, upshear profiles have substantially higher H₂O than downshear profiles when shear is weak (around a 60% average increase in H₂O for downshear profiles, while

upshear profiles have a nearly 100% average increase in upper tropospheric H₂O). In moderate shear environments, upshear and downshear profiles are nearly identical; however, in strong shear environments, upper tropospheric H₂O is generally higher for downshear profiles closer to the tropopause. Given that the downshear side of the storm generally has the strongest convection in a sheared environment, while upshear locations have weaker updrafts (DeHart et al. 2014), these differences make sense. Notably, tropopause dehydration is the most prominent in low shear environments, where an average H₂O decrease of ~20% is observed; this dehydration feature is completely absent in strongly sheared environments. Finally, UTLS H₂O shows much stronger sensitivity to shear magnitude on the upshear side of the storm than downshear; there is much more variability in the interquartile ranges of upshear profiles than downshear profiles within the UTLS (See Fig. 3.2) Thus, we find that upper tropospheric O₃ changes are somewhat dependent on deep-layer wind shear, while UTLS H₂O changes are highly affected by shear.

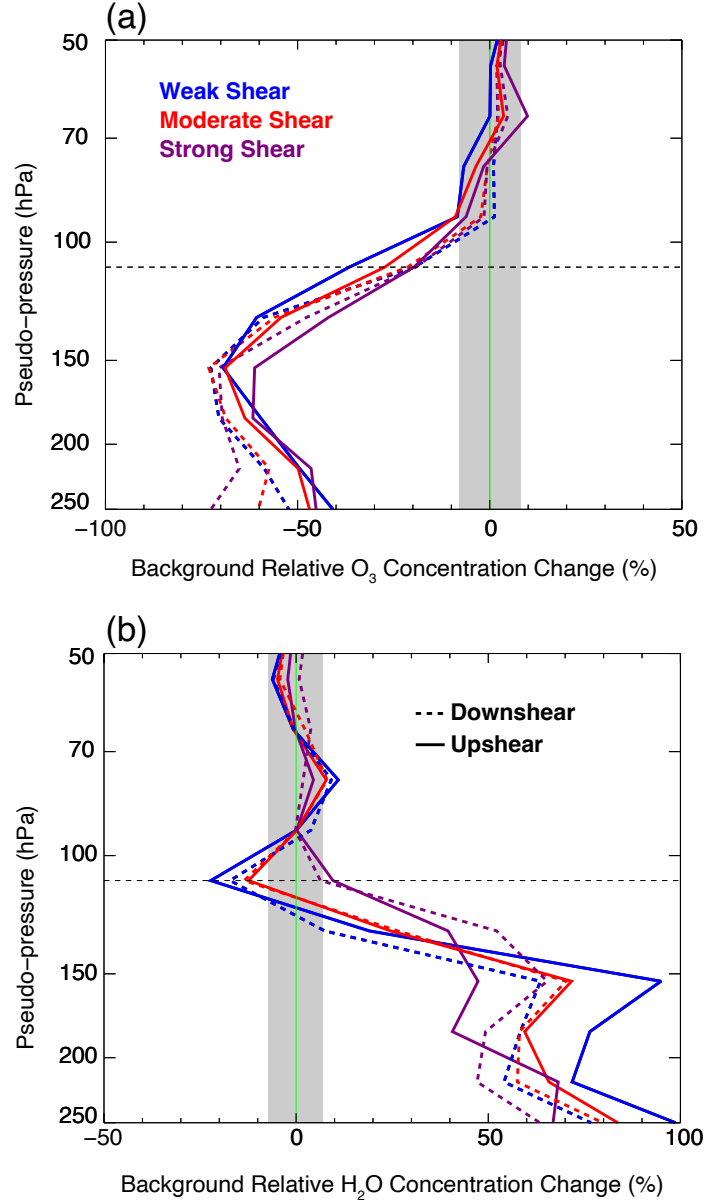


Figure 3.1: Basin and tropopause-relative plot showing the percent change of (a) UTLS ozone and (b) UTLS water vapor for hurricanes in environments with weak shear in blue, moderate shear in red, and strong shear in purple. The solid colored lines represent upshear profiles, while the dotted colored lines represent downshear profiles. The dotted black line represents the tropopause, the green line marks a 0% change, and the gray color-fill represents the typical uncertainty of an MLS measurement. Analysis includes approximately 150 hurricane profiles for weak shear cases, 500 profiles for moderate shear cases, and 130 profiles for strong shear cases for upshear and downshear analysis.

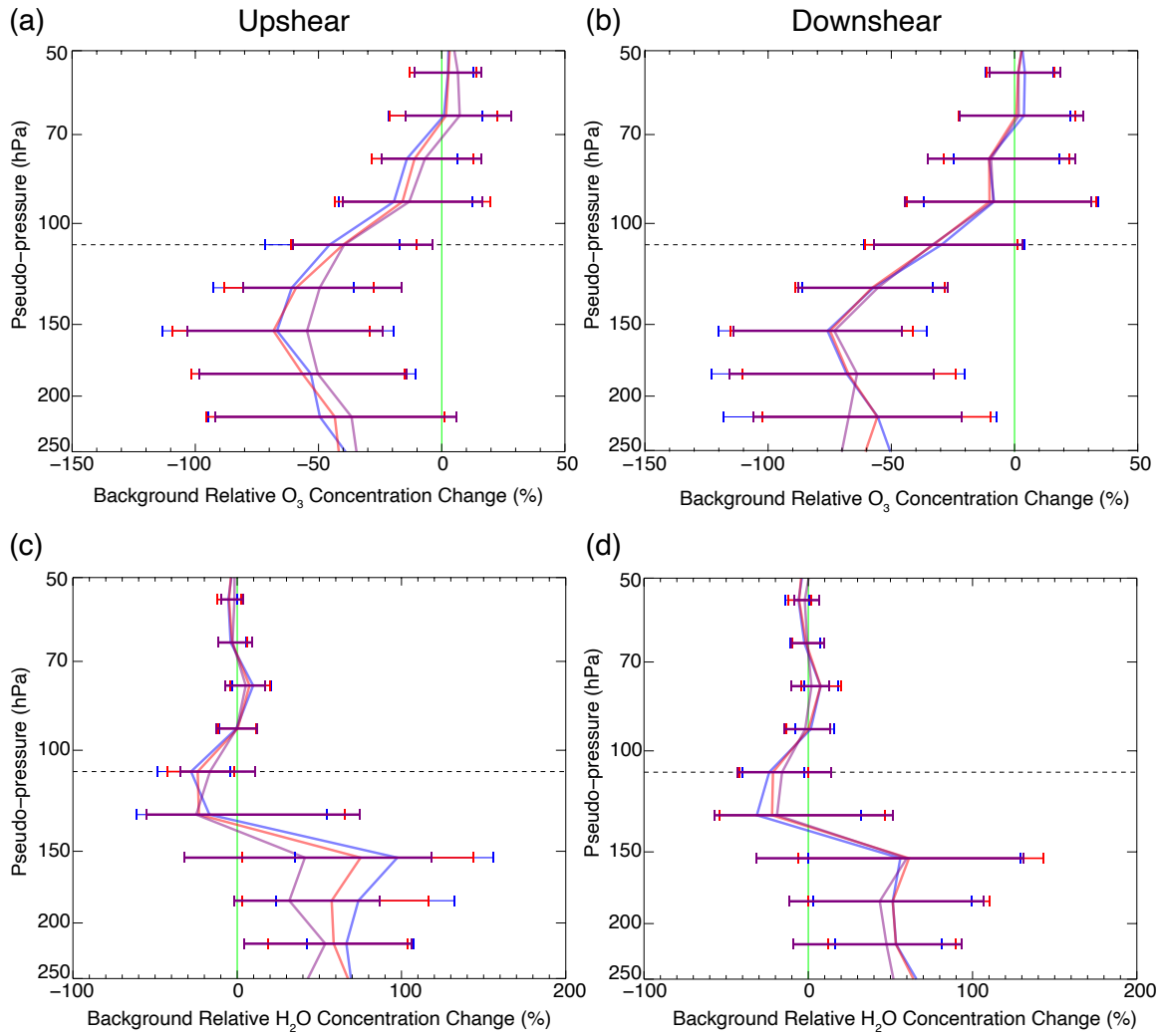


Figure 3.2: Basin and tropopause-relative plot showing the percent change of UTLS ozone (panels a and b) and UTLS water vapor (panels c and d) for hurricanes in environments with weak shear in blue, moderate shear in red, and strong shear in purple. Analysis is divided into upshear profiles (panels a and c) and downshear profiles (panels b and d). The solid colored lines represent median percent changes, while the bars represent the interquartile range for each shear intensity. The dotted black line represents the tropopause and the green line marks a 0% change. Analysis includes approximately 150 hurricane profiles for weak shear cases, 500 profiles for moderate shear cases, and 130 profiles for strong shear cases for upshear and downshear analysis.

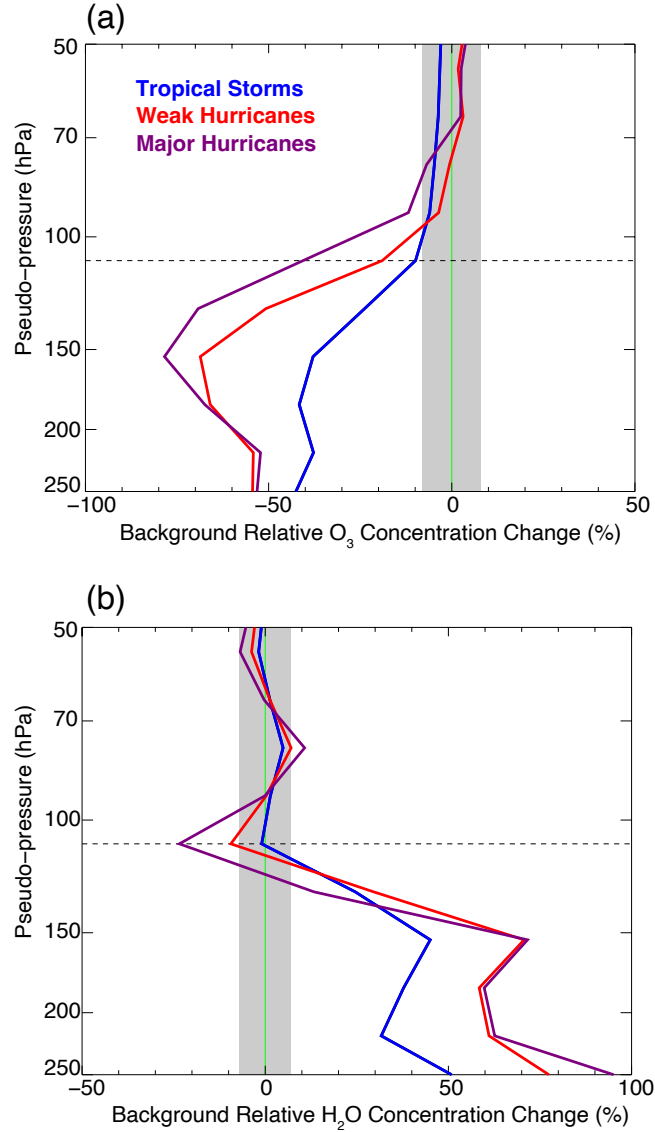


Figure 3.3: Basin-relative plot showing the percent change of (a) UTLS ozone and (b) UTLS water vapor partitioned by TC intensity and in tropopause relative coordinates. The purple line represents major hurricane intensity, the red line is weak hurricane intensity, and the blue line is tropical storm intensity. The horizontal black dashed line represents the tropopause, while the gray color-fill represents the typical uncertainty of an MLS measurement. A green line has been added at 0% concentration change (i.e., indicating the background). Analysis includes approximately 1900 TC profiles for tropical storms, 750 profiles for weak hurricanes, and 240 profiles for major hurricanes.

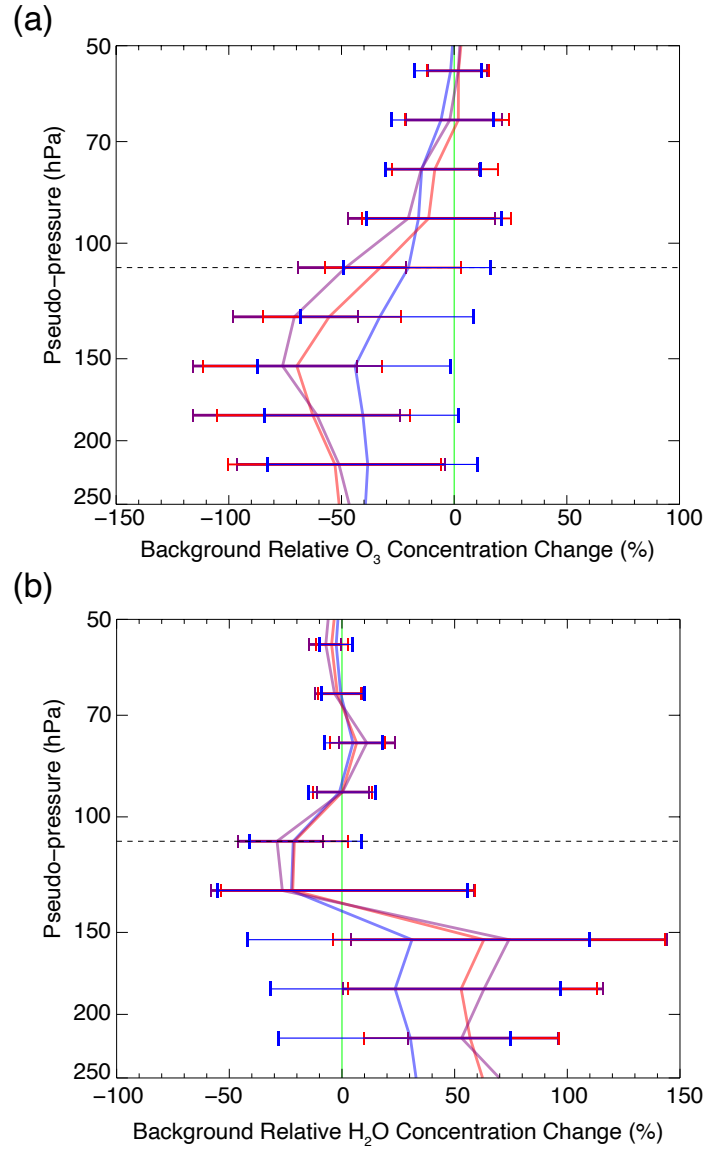


Figure 3.4: Basin-relative plot showing the percent change of (a) UTLS ozone and (b) UTLS water vapor partitioned by TC intensity and in tropopause relative coordinates. The purple line represents major hurricane intensity, the red line is weak hurricane intensity, and the blue line is tropical storm intensity. The solid colored lines represent median percent changes, while bars represent the interquartile range for each intensity category. A green line has been added at 0% concentration change (i.e., indicating the background). Analysis includes approximately 1900 TC profiles for tropical storms, 750 profiles for weak hurricanes, and 240 profiles for major hurricanes.

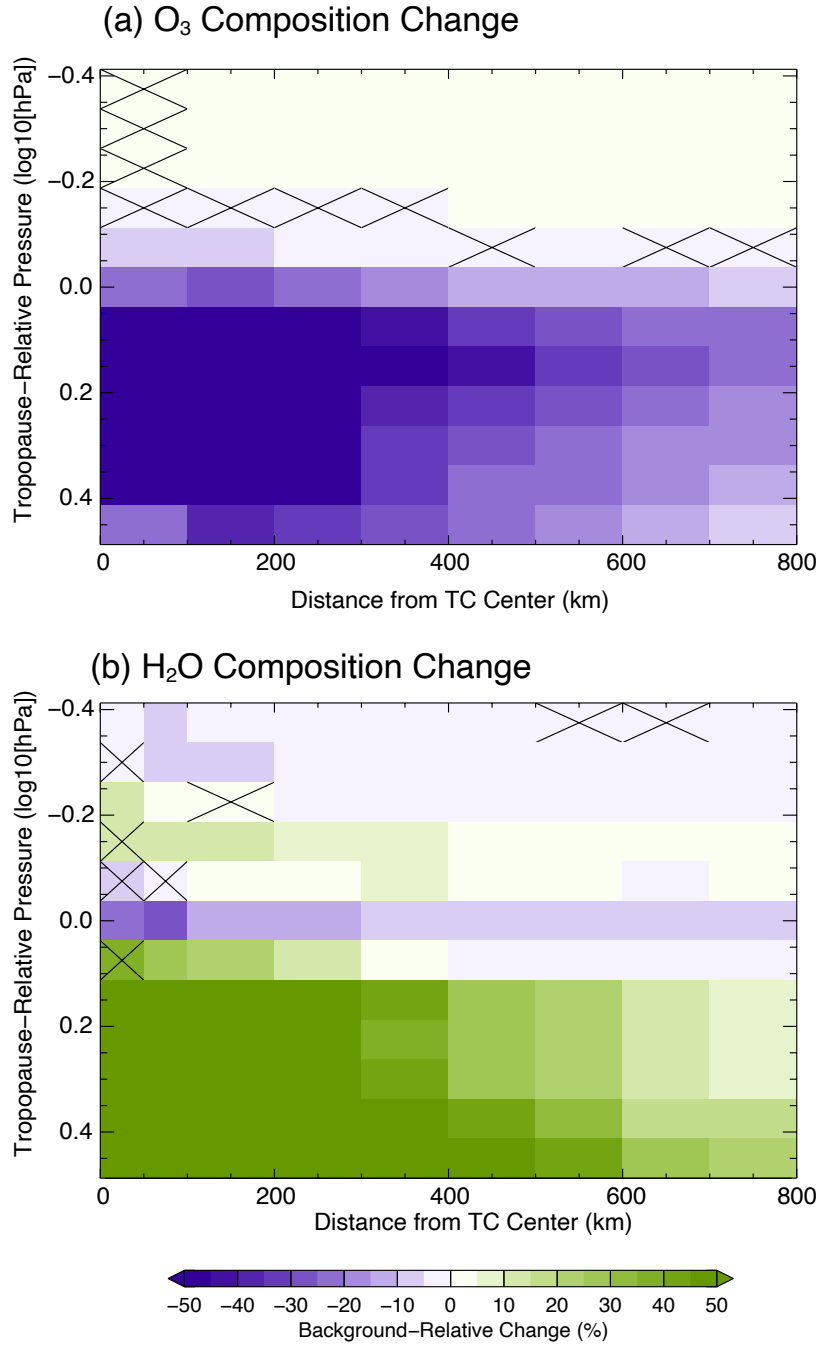


Figure 3.5: Basin-relative plot showing the percent change of (a) UTLS ozone and (b) UTLS water vapor as a function of distance from TC center and in relative pressure to the tropopause. An x is drawn over bins with a p-value greater than 0.01.

Chapter 4

Summary and Conclusions

This study leveraged 17 years of satellite observations to examine the influence of TCs on UTLS composition. In particular, we examined the environmental wind shear, distance and TC intensity dependency of TC-induced changes in UTLS O_3 and H_2O . To do this, we matched Aura MLS trace gas profiles with TC location data from the IBTrACS dataset and environmental conditions from MERRA-2 reanalysis. Given variation in tropopause height across profiles, we employed a tropopause-relative framework to allow for robust assessment of TC-induced composition change (see Fig. 2.3), a first for such studies. Additionally, we found that UTLS background O_3 and H_2O concentrations vary significantly by ocean basin, sometimes by a factor of three times or more (see Figs. 2.4 & 2.5); therefore, TC-induced composition changes must also be calculated relative to the average background to be reliable when considering global samples.

Ultimately, we found that UTLS composition changes from TCs are dependent on both TC intensity and distance from TC center, while shear dependency was most evident for UTLS H_2O . We found that more intense TCs were associated with greater changes in UTLS composition (Fig. 3.3). In particular, we found that within major hurricanes, upper tropospheric O_3 decreased by nearly 80% on average. This strong upper tropospheric deficit is in agreement with the results of Zou and Wu (2005), who found decreased O_3 in hurricanes, especially in more intense storms. We uniquely demonstrate that this result is robust when assessed globally and for a much longer record, and further reveal the variation in O_3 composition impacts with altitude and for weaker TCs. For UTLS H_2O , we found strong upper tropospheric hydration of $\approx 70\%$ on average within hurricanes, also in agreement with past work (e.g., Ray and Rosenlof 2007). We also robustly diagnosed hydration in

the stratosphere, where increasing intensity is related to increasing hydration up to a maximum of $\approx 10\%$ in major hurricanes at altitudes ≈ 2 km above tropopause (see Figs. 3.3 & 3.5), supporting similar results for select basins and limited samples in prior work (Ratnam et al. 2016; Jiang et al. 2020). Again, the detailed variation in altitude and robust global assessment in this study provide greater confidence in and assessment of these outcomes.

We also found that effects on UTLS O_3 and H_2O generally decrease with distance from TC center, particularly beyond around 400 km (Fig. 3.5). In this analysis, the strongest stratospheric hydration signal was found to be within 100 km from TC center and at altitudes ≈ 1.5 – 3.5 km above tropopause, mirroring a similar result from a recent idealized modeling study (Gordon and Homeyer 2025). Additionally, we found a dehydrated tropopause within TCs, especially so within 50–100 km from TC center; this dehydrated tropopause agrees with findings from Jiang et al. (2020). The tropopause-level dehydration was found to be around 20% within major hurricanes (Fig. 3.3b) and only revealed when assessing composition change in tropopause-relative coordinates.

For UTLS composition change sensitivity to deep-layer wind shear, we found that upper tropospheric O_3 deficits are highest on the downshear side of TCs (Fig. 3.1a). Additionally, we found that UTLS H_2O demonstrated a substantial sensitivity to shear, with weak shear environments having stronger UTLS composition changes (Fig. 3.1b). Upper tropospheric H_2O change varied significantly between upshear and downshear profiles in both strong and weak shear environments, with limited differences when shear was moderate. A cautionary note to these shear sensitivities is that TC intensity is also affected by deep-layer wind shear (stronger shear typically weakens TCs), so the diagnosed sensitivities of UTLS composition change to TC intensity and deep-layer wind shear are not entirely independent.

Overall, it is clear from combination of prior studies and our work that STE within TCs significantly modifies O_3 and H_2O composition throughout the UTLS, with impacts strongly affected by storm intensity and distance from TC center; a weaker TC or distances further away from a storm will generally have reduced UTLS composition impacts. The

sensitivity of UTLS composition impacts to TC intensity warrants greater attention to the projected increase in TC intensity in the future as the climate warms (Emanuel 1987). Increased STE and UTLS O_3 and H_2O change will amplify the radiative feedbacks driving climate warming, though which effect will dominate in TCs remains unknown. For example, decreased UTLS O_3 in TCs would reduce or slow warming, while increased UTLS H_2O would enhance or accelerate warming. Thus, the prevailing climate impacts from STE in TCs are in competition and require further detailed study.

While our results provide robust insights into the impact of TCs on the UTLS, several limitations remain. Most notably, given the coarse resolution of MLS observations, this analysis does not allow for evaluation of distinct STE processes within TCs but rather the overall STE budget. Further, due to limb length restrictions, we cannot resolve distance-dependent composition change in bins smaller than 100 km for O_3 and 50 km for H_2O . Thus, although previous work has found subsidence within the eye of TCs (Rodgers et al. 1990; Leclair De Bellevue et al. 2006; Baray et al. 1999; Gordon and Homeyer 2025), it is not possible to diagnose the effect of this process using MLS observations; ultimately, we find significant upper tropospheric O_3 decreases within the inner 100 km of a TC (Fig. 3.3a), implying this region is dominated by lofted lower troposphere low- O_3 air. Additionally, given TC track data does not include information about the size and location of specific TC features (such as the eye, eyewall convection, inner core, rainbands, and extent of upper tropospheric outflow), we cannot relate observed composition change to such TC structure with confidence. Finally, although overall TC sample sizes are large in this study, sample sizes remain prohibitively limited for distance-dependent analysis of major hurricanes only, despite our results showing that major TCs have the largest impacts on UTLS composition within the inner core (Fig. 3.3). Even so, this analysis shows that both weak and major hurricanes have similar overall composition change patterns; therefore, our results using all hurricane samples are likely representative of the patterns seen within major hurricanes but biased toward relatively low amplitudes of composition change.

Given the relationship between UTLS composition change and Earth's radiation budget, as well as the disproportionate influence of TCs on STE (Zhan and Wang 2012), deepening our understanding of STE within TCs remains an important focus for future work. Specifically, further research is needed to determine the impact of individual transport processes such as stratospheric subsidence near TC center on UTLS composition change, given conflicting past work regarding the significance of such transport (e.g. Rodgers et al. 1990; Joiner et al. 2006), as well as overshooting. Additionally, analysis of a longer record of major hurricanes should be leveraged to further explore their impacts on UTLS composition. This is particularly relevant because although overall TC frequency is expected to decrease as the climate warms, the frequency of intense storms is expected to increase Knutson et al. (2010). Because our results show the most intense UTLS composition changes result from major hurricanes, examining the effects of projected TC frequency and intensity change on future UTLS composition modifications is of critical importance. Additionally, although we find that TCs significantly alter the UTLS, the persistence of these changes remains unclear and requires further investigation. Finally, this study does not consider UTLS changes resulting from TCs undergoing extratropical transition; further research is still needed to quantify these impacts. Despite this, our findings contribute to a growing body of evidence regarding the importance of evaluating TC-induced STE and its effects on UTLS composition.

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