Stratospheric gas variations: Comparing ACE and MKIV balloon

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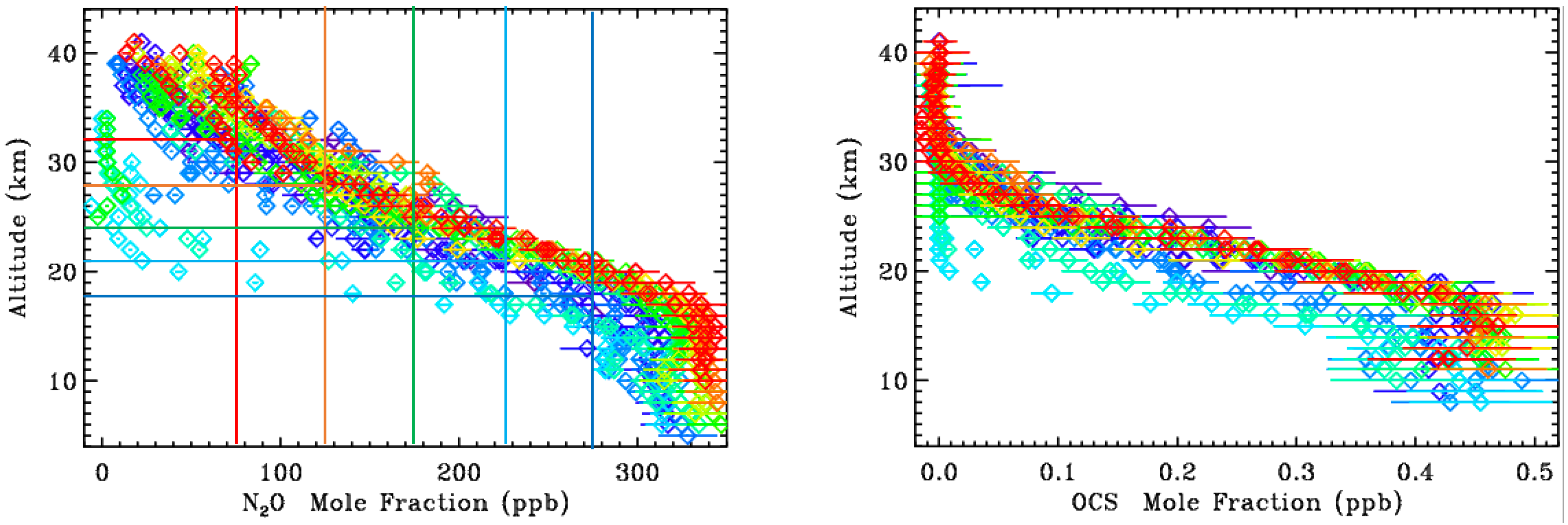
The MKIV interferometer is a solar occultation FTS covering the 650-5650 cm^{-1} range at 0.01 cm^{-1} resolution (56 cm MOPD) for balloon flights. It has performed 28 balloon flights (mostly from 35N), ground-based observations, and has previously flown 3 campaigns on NASA DC-8 aircraft. No close co-incidences with ACE: late Sep at 35N falls in a hole in ACE coverage.

MKIV averages only 1 occultation per year. So using MKIV data for trend assessment is difficult, because even for balloon flights launched from the same site and month, the origin of the probed airmasses varies from flight to flight due to stratospheric transport differences. One year the the MKIV may be sampling sub-tropical airmasses, whereas the next year, from the exact same location and date, it might sample mid-latitude airmasses.

Using a tracer N_2O as a vertical ordinate helps remove some of this dynamical variability, allowing the underlying trends to be more clearly seen.

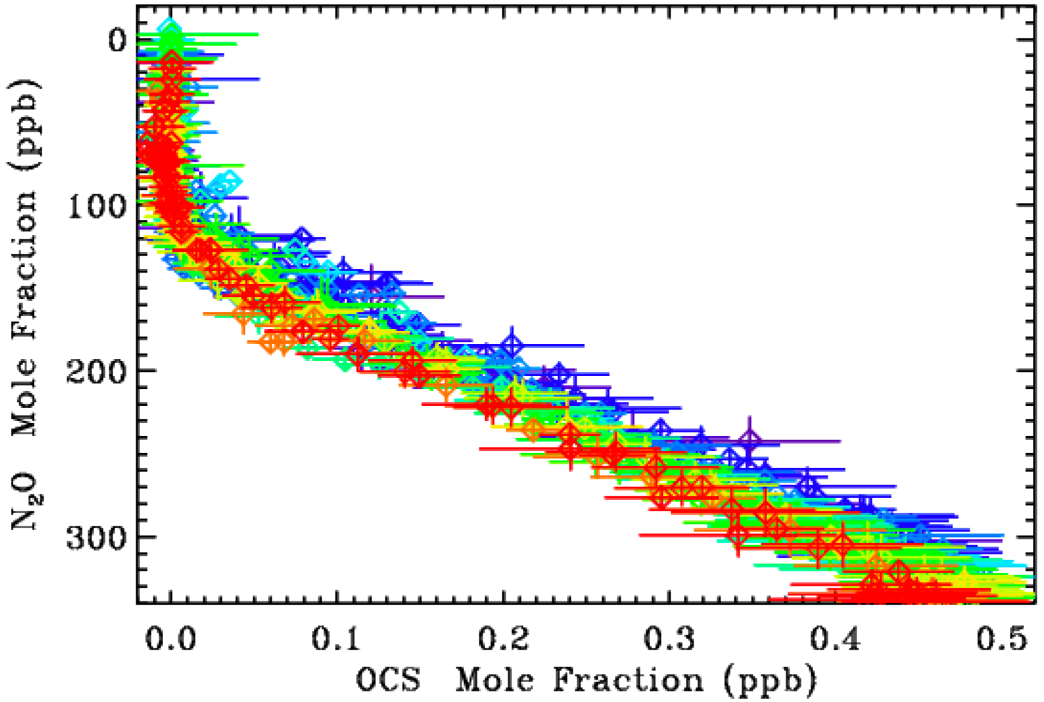
Example: MkIV profiles of N_2O and OCS versus altitude

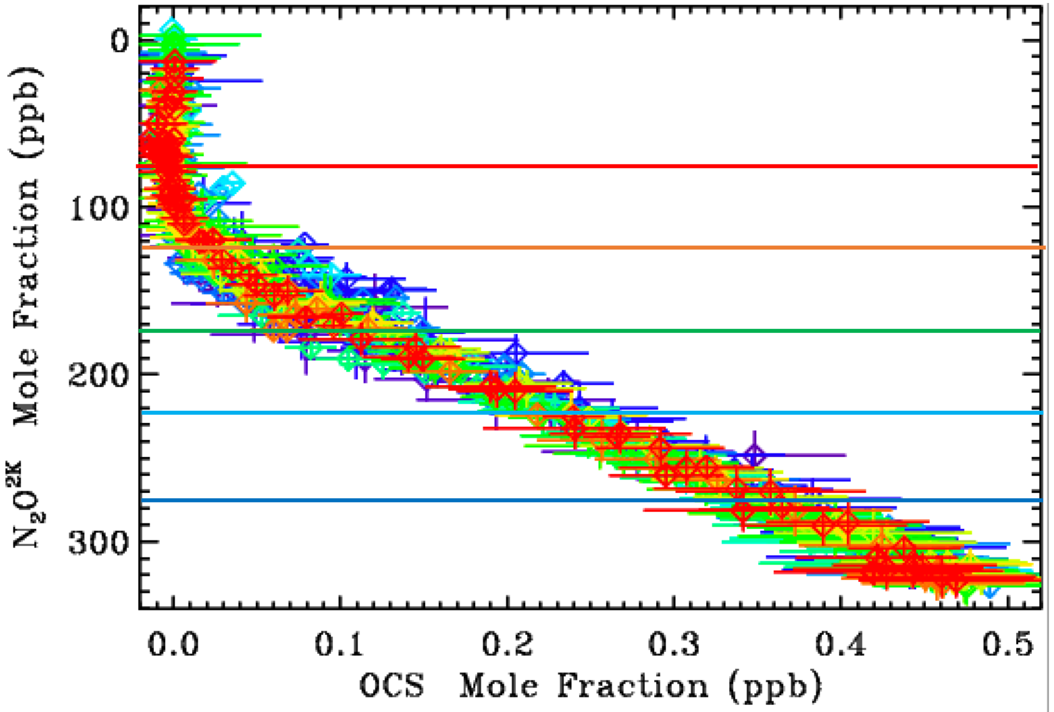


Profiles of N_2O (left) and OCS (right) color-coded by year (blue = 1989; green = 2004; red = 2021, etc.). OCS is shorter lived and therefore decreases more rapidly with altitude, but there is a similarity in the behavior of the N_2O and OCS profiles.

Profiles are variable from flight to flight. High latitude flights (cyan, green) have less gas at a given altitude due to downward transport at high latitudes. But even among the mid-latitude flights, there is still a lot of variation. So, when we try to estimate the trends at altitudes of 19,22, 24, 27, and 32 km, the results look very noisy (next slide)

Plotting versus altitude above the tropopause or potential temperature (not shown) reduces scatter, but not by much.





Compact relationship between N_2O and OCS

Measured OCS and N_2O mole fractions plotted against each other, color-coded by year (purple=1989, blue=1992, green=2004; red=2023).

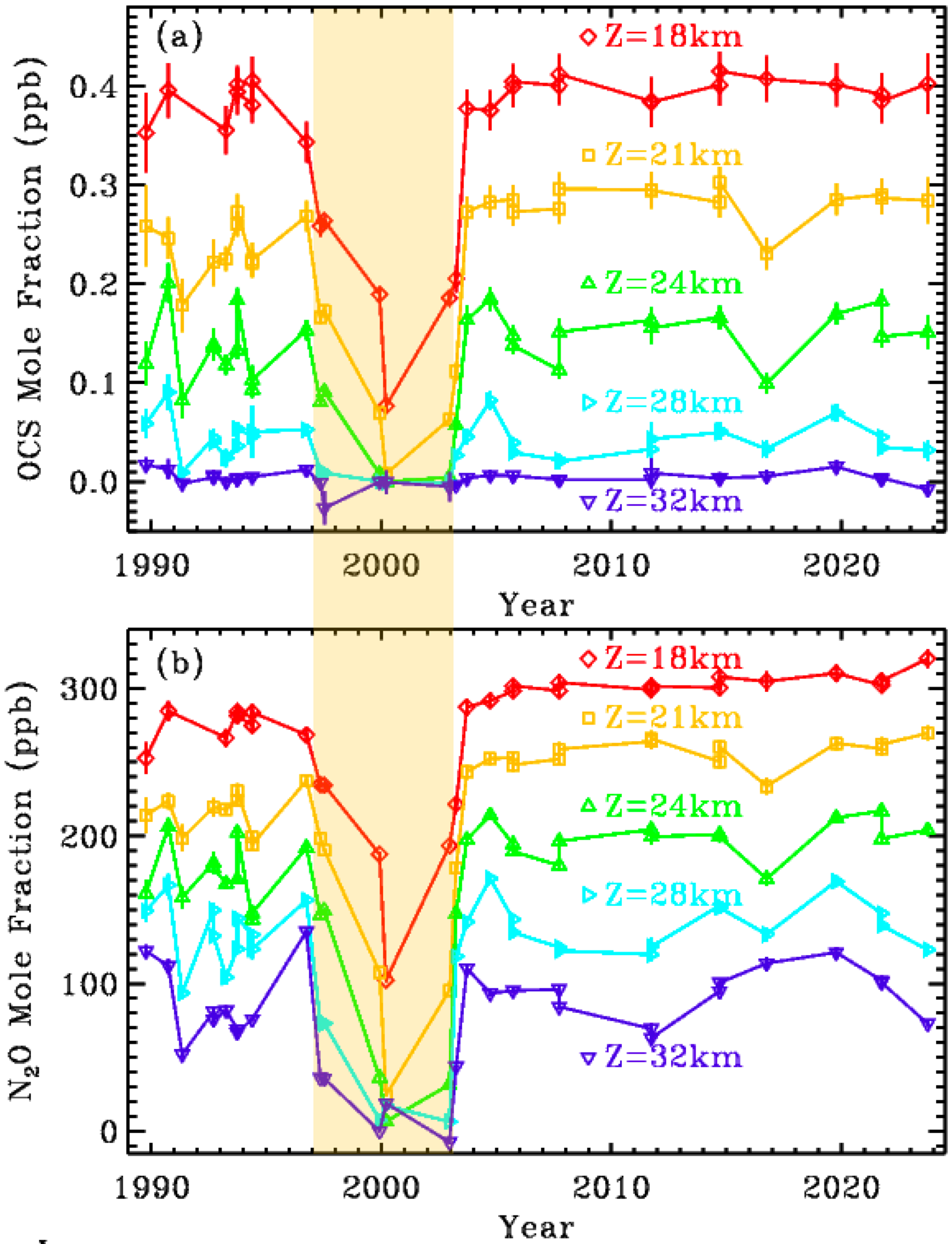
Top panel showing raw data shows a creeping increase of N_2O or decrease of OCS over time. Since we know from other measurements (e.g. in situ and ACE) that N_2O has been increasing by 0.26% per year over the past 30 years, we can correct the measured N_2O values back to the value that they would have been in 2000 using the equation

$$N_2O^{2K} = N_2O/[1+0.0026*(year-2000)]$$

The lower panel shows the OCS plotted versus N_2O^{2K}. There is no obvious creep over time since all the different colors lie on the same compact relationship. This implies little or no OCS trend.

We can now look for trends on the five isopleths: N_2O^{2K} = 75, 125, 175, 225, 275 ppb, denoted by the colored horizontal lines in lower panel.

The fact that the OCS-N_2O relationship is linear at lower altitudes (N_2O > 150 ppb) means that here these gases both have lifetime greater than transport times. At higher altitudes the relationship becomes curved, since the OCS lifetime is shorter than N_2O's, but remains compact. *Plumb and Ko (1992).*

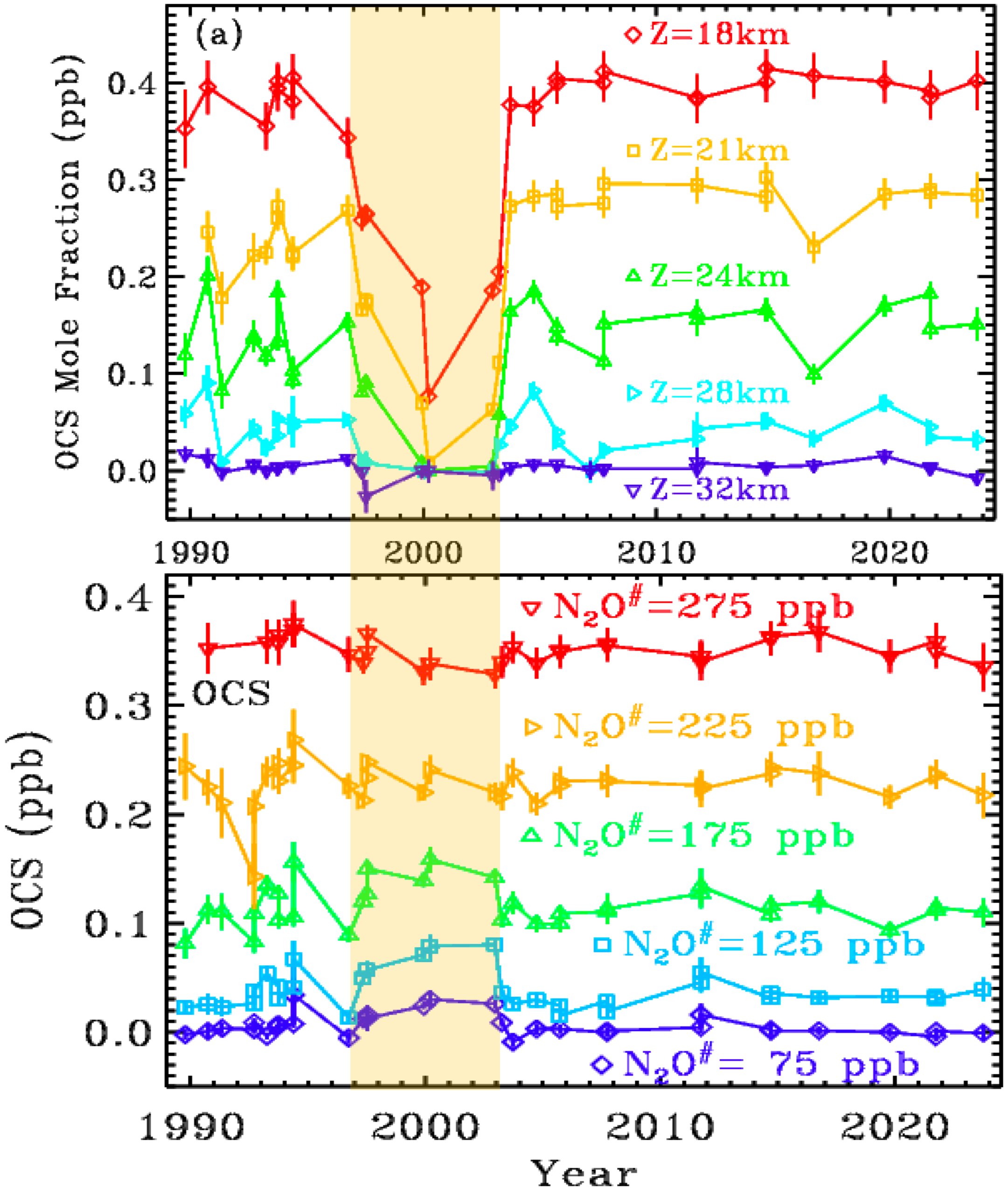


OCS and N2O VMRs at fixed altitudes

Behavior of N2O and OCS is qualitatively very similar. Reduced values for the high-latitude flights (yellow shading) and considerable flight-to-flight variations, even at 35N.

Due to the downward transport in the winter vortex, the N2O and OCS vmrs are down to zero in the late-1999, early 2000, and late 2022 flights at the higher altitudes.

OCS interpolated onto N_2O^{2K} isopleths



Upper panel shows OCS interpolated onto fixed altitude levels, as shown earlier. There is considerable flight-to-flight variability, especially for the six high-latitude flights (yellow shading).

Lower panel shows OCS interpolated onto fixed N_2O isopleths: 75, 125, 175, 225 and 275 ppb. The flight-to-flight variation is reduced in comparison with the upper panel, allowing a more accurate trend determination. The high latitude flights fall into line with the 35N flights, at least at the lower altitudes.

Compared MkIV gas variations with those from ACE as reported by Schmidt et al. (2024)

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Trends in atmospheric composition between 2004–2023 using version 5 ACE-FTS data

Matthew Schmidt [a], Peter Bernath [b] [a], Chris Boone [a], Michael Lecours [a], Johnathan Steffen [a]

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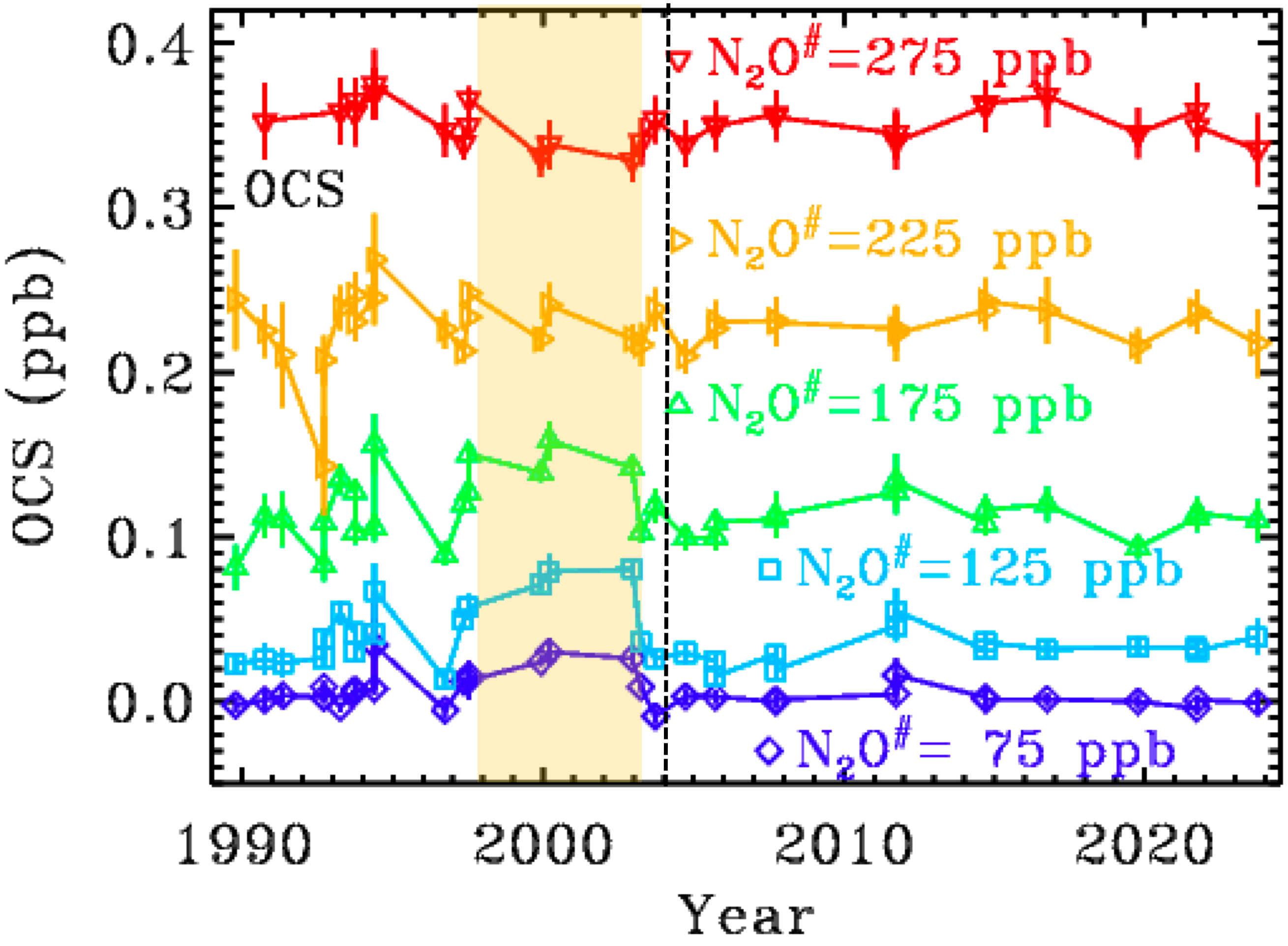
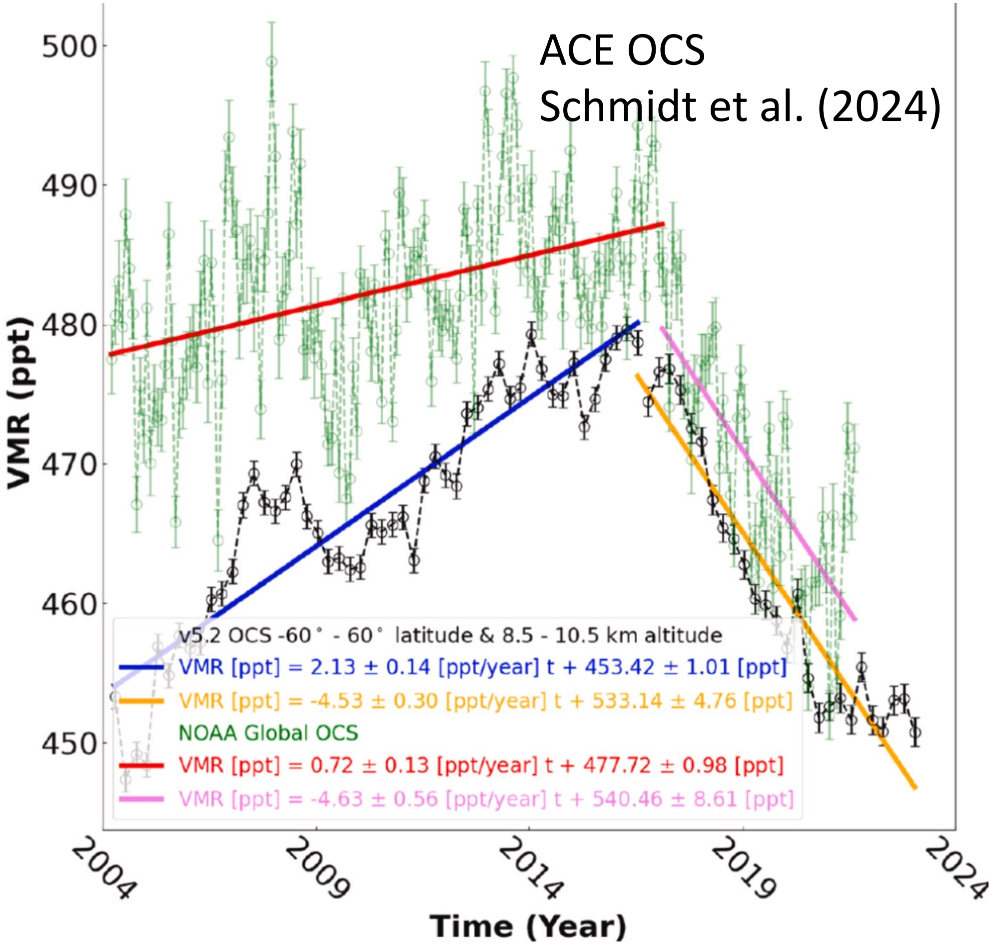
MkIV-ACE: OCS Trend comparison



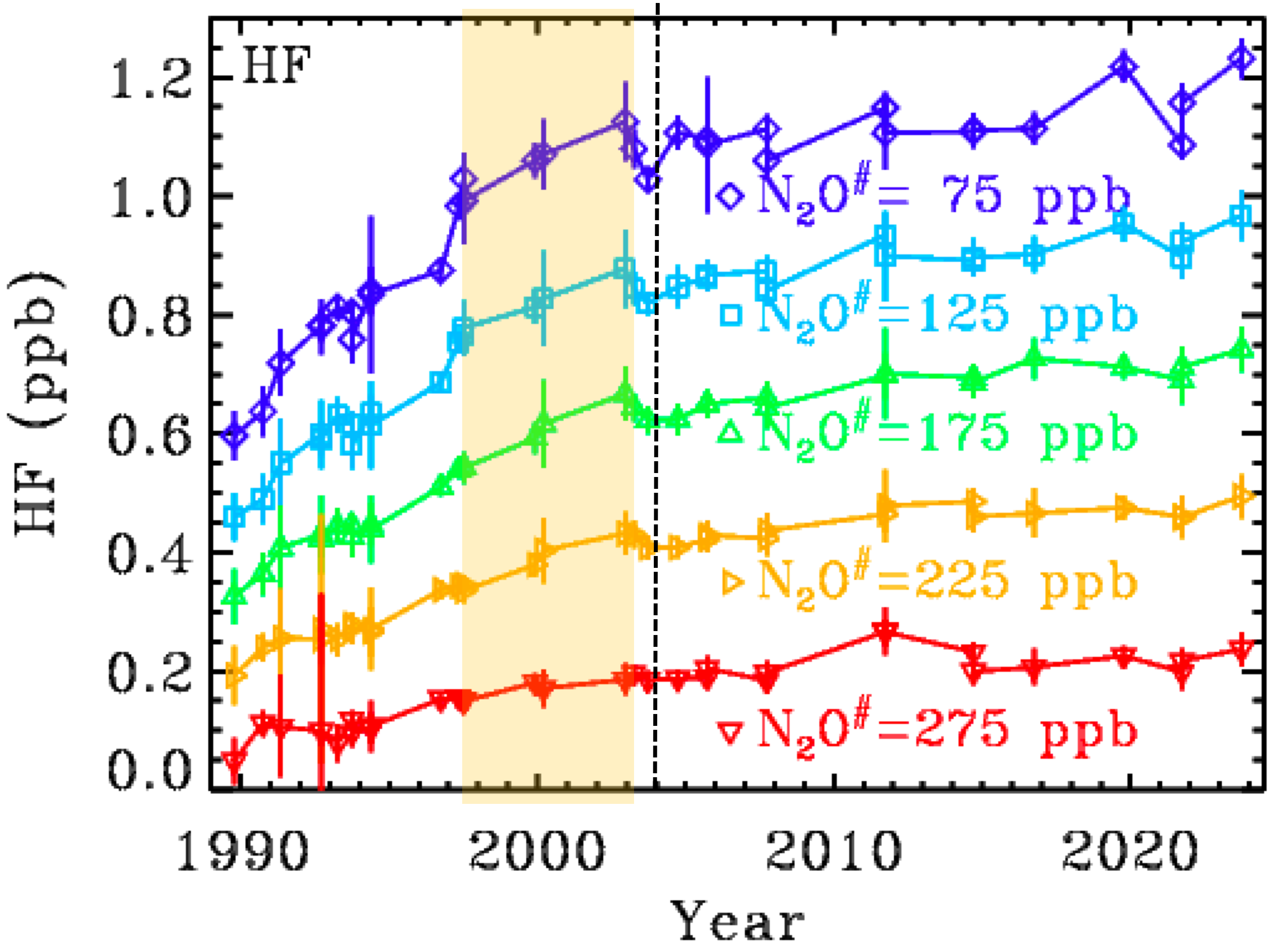


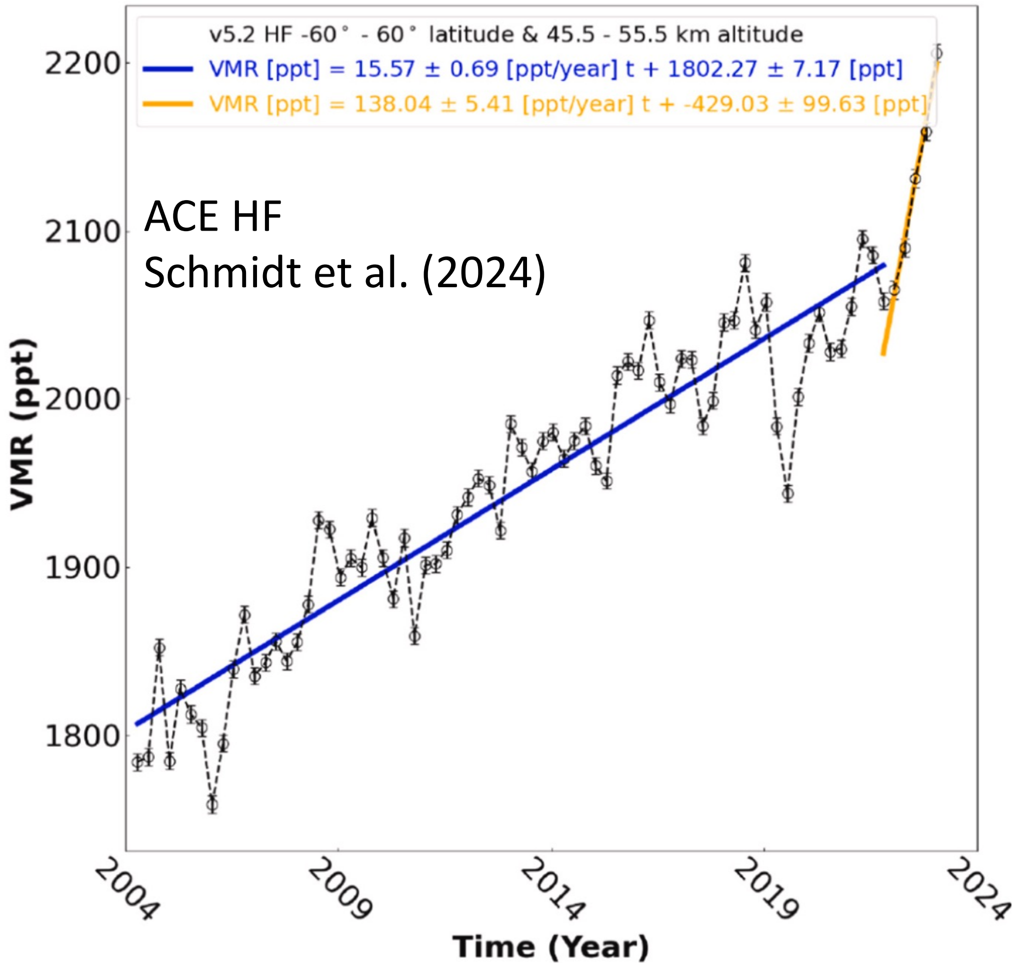
Left: MKIV OCS shows no significant trends in the stratosphere at any of the five N_2O isopleths/altitudes. Vertical dotted black line indicates start of ACE record.

Right: ACE shows a peak in OCS in 2017 over the 8.5 to 10.5 km altitude range, followed by a sharp decrease.

ACE OCS values are 30% larger than those of MKIV (partly due to lower altitude of ACE measurements)

MkIV-ACE HF Trend Comparison

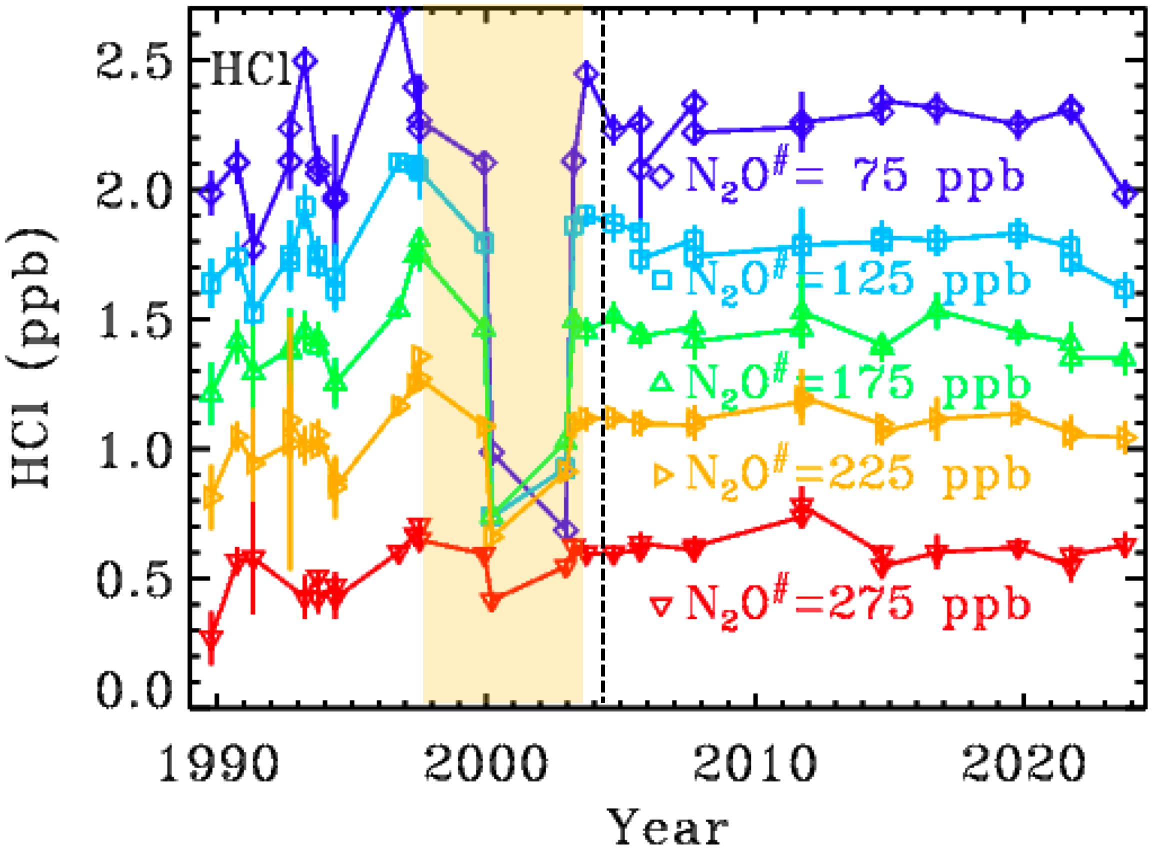


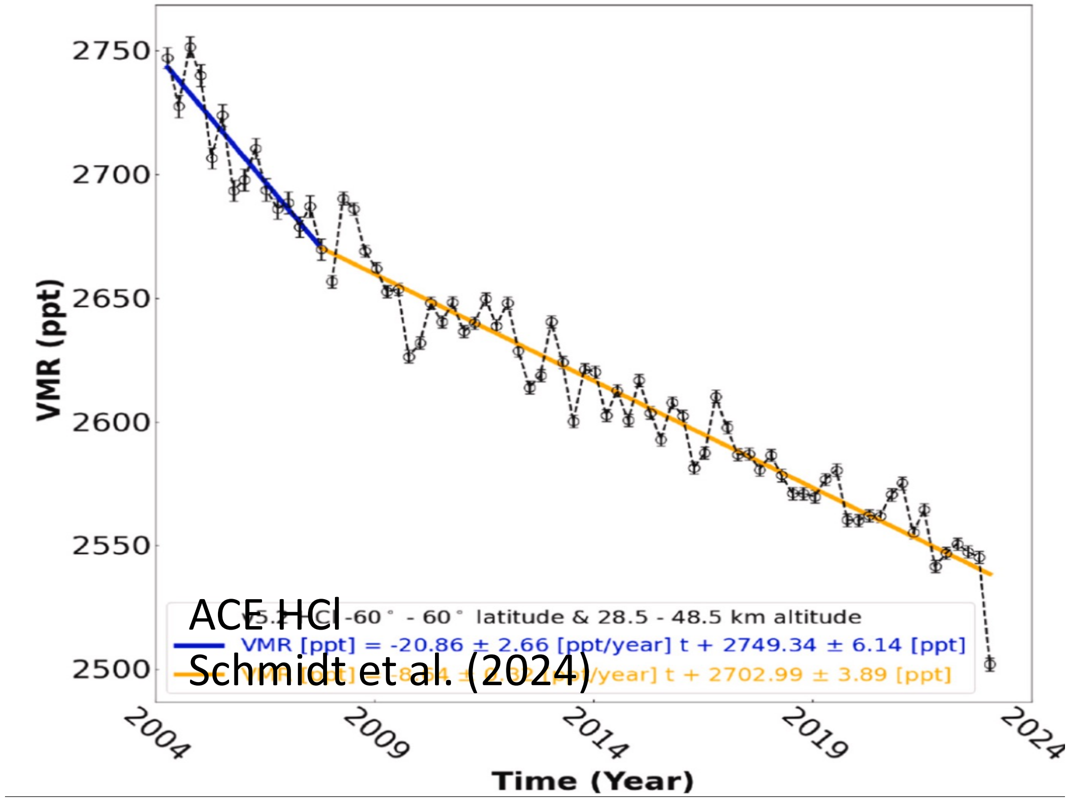


Left: Retrieved MKIV HF vmrs interpolated to various N2O isopleths. Large vmrs from 1997-2003 for high latitudes (Fairbanks and Esrange). The other flights were all from ~35N. HF has increased by more than a factor two 1989-2022, but has now slowed down.

Right: ACE HF vmrs 45-55 km altitude.

MkIV-ACE HCl Trend Comparison



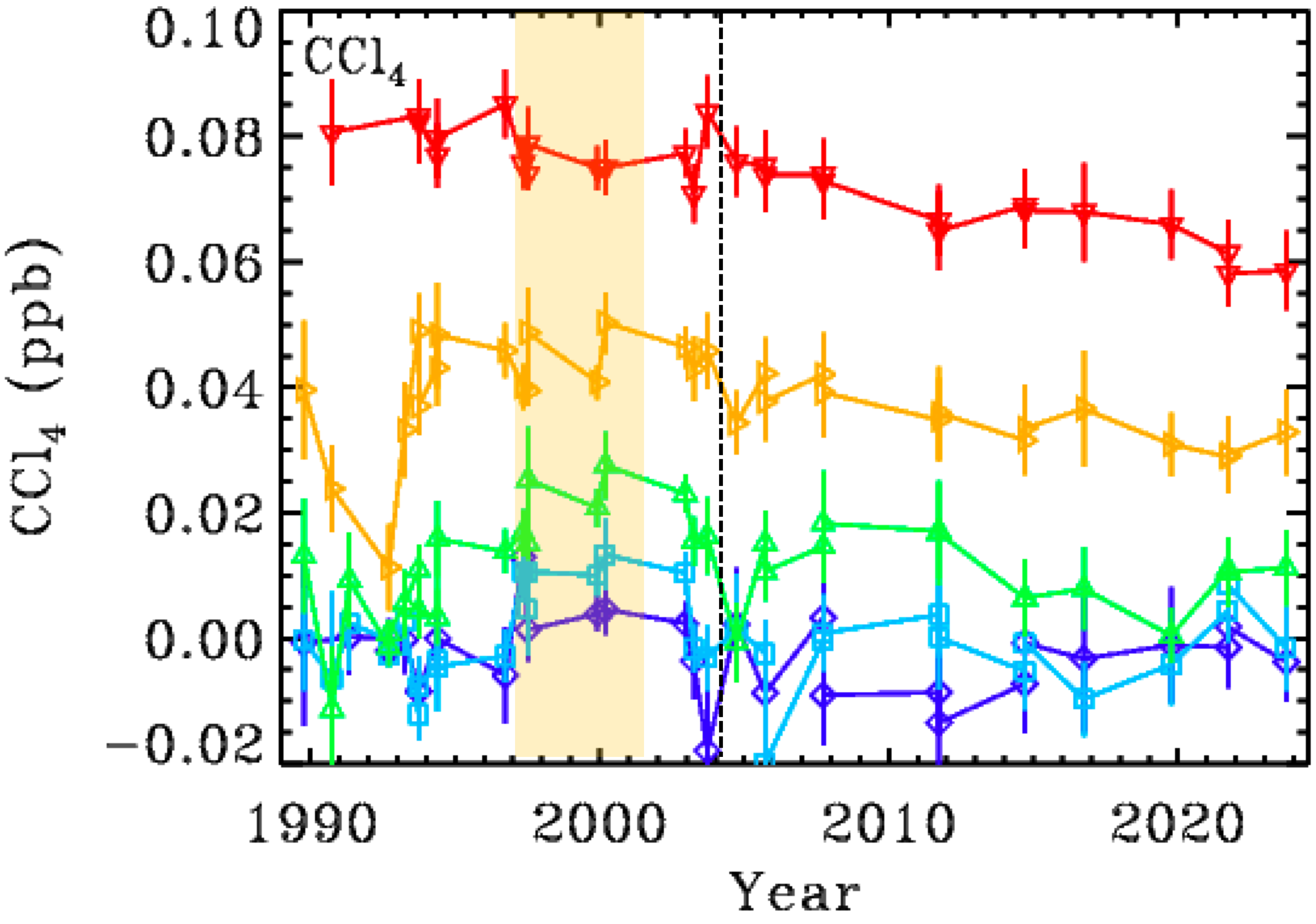


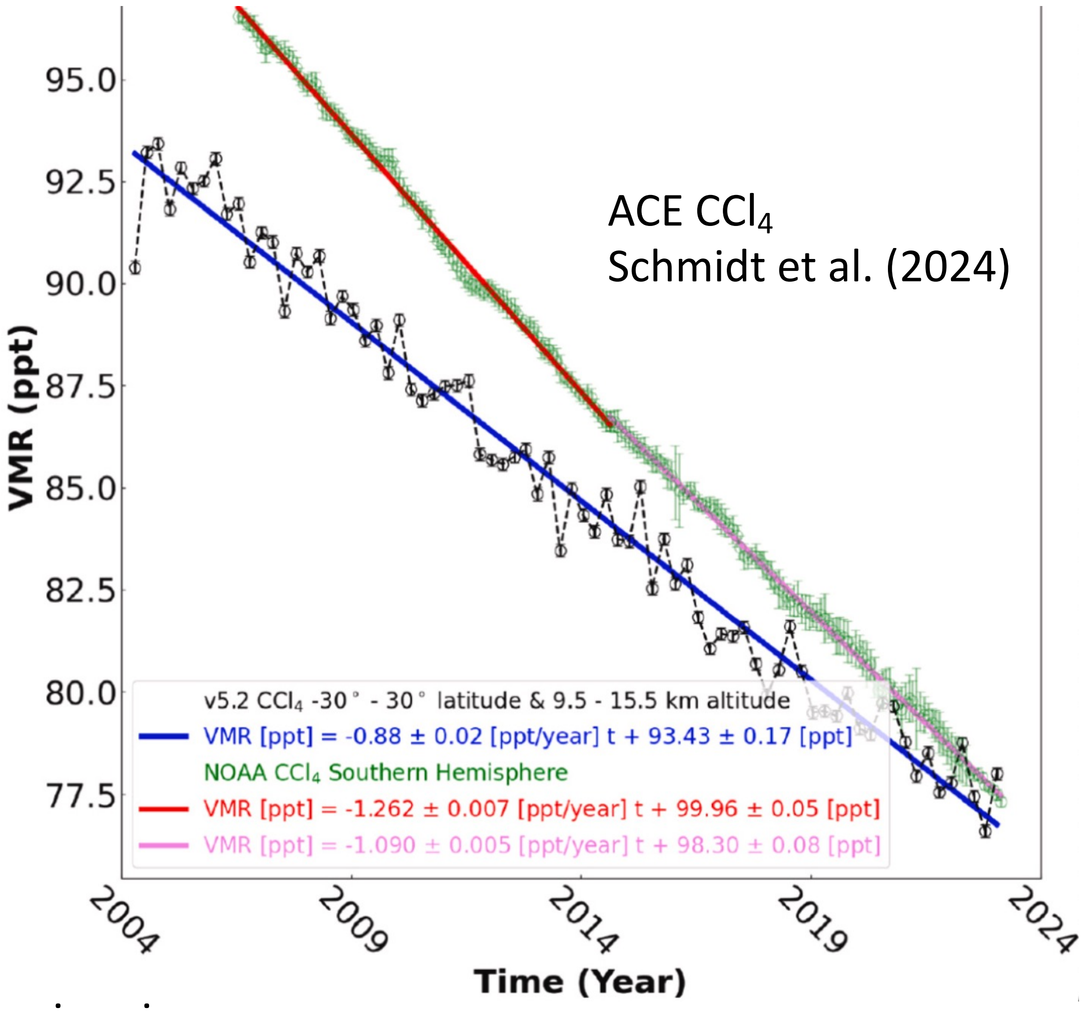
MkIV saw increasing HCl from 1989 to 2000. Two of four high-latitude flights in the winter vortex (yellow shading) show large heterogeneous loss of HCl.

The ACE HCl measurements (right panel) cover the 28.5—48.5 km altitude range, which is above the highest MKIV altitude range (75 ppb of N_2O), and therefore provides slightly larger HCl VMRs.

ACE sees a 6% total decrease (from 2750 to 2585 ppt) over 16 years. Possibly inconsistent with MKIV, although different altitudes are sampled. Ground-based HCl columns saw increasing HCl from 2007-2012 in the NH (Mahieu et al, 2014).

MkIV-ACE CCl_4 Trend Comparison





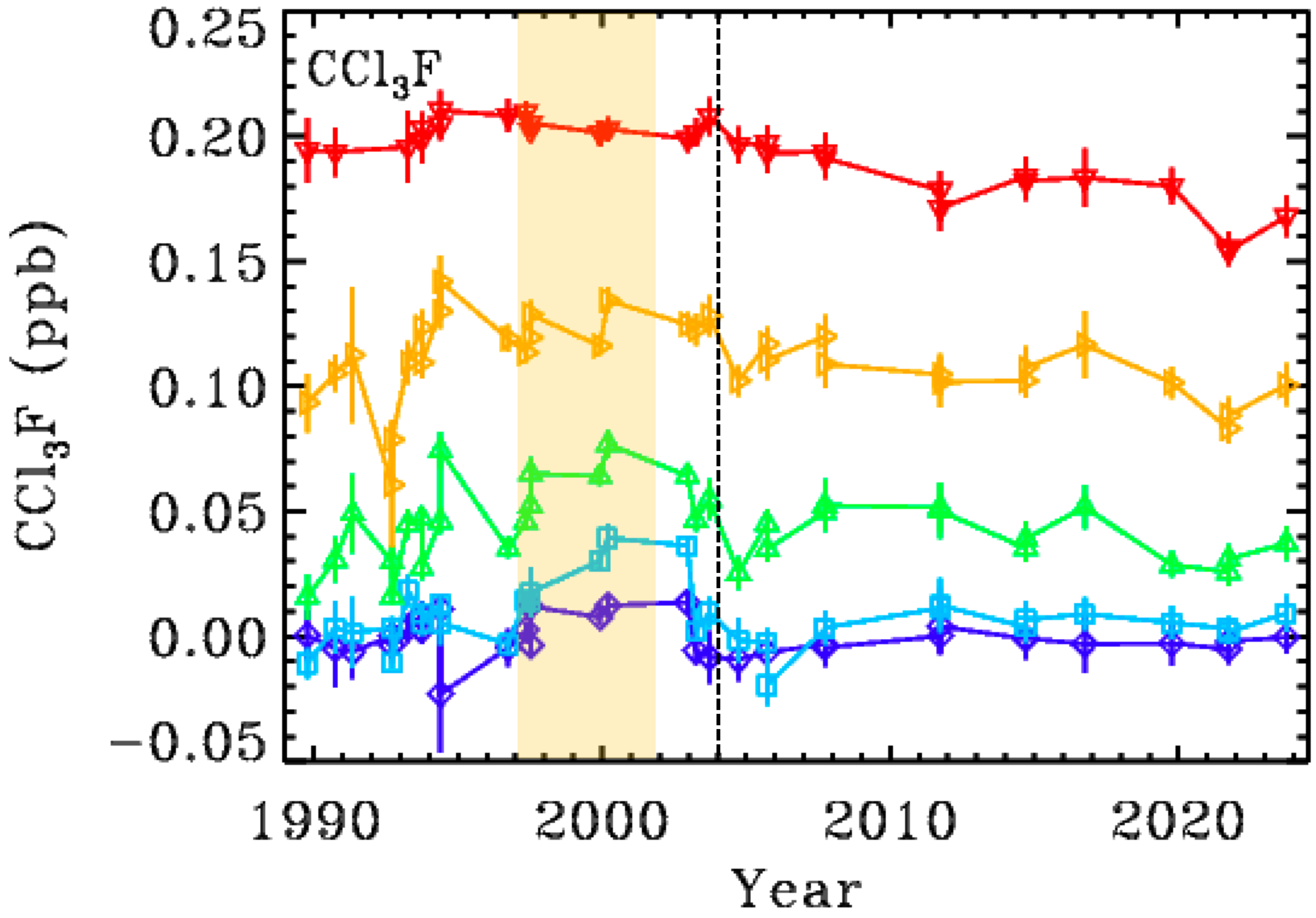
ACE CCl_4
Schmidt et al. (2024)

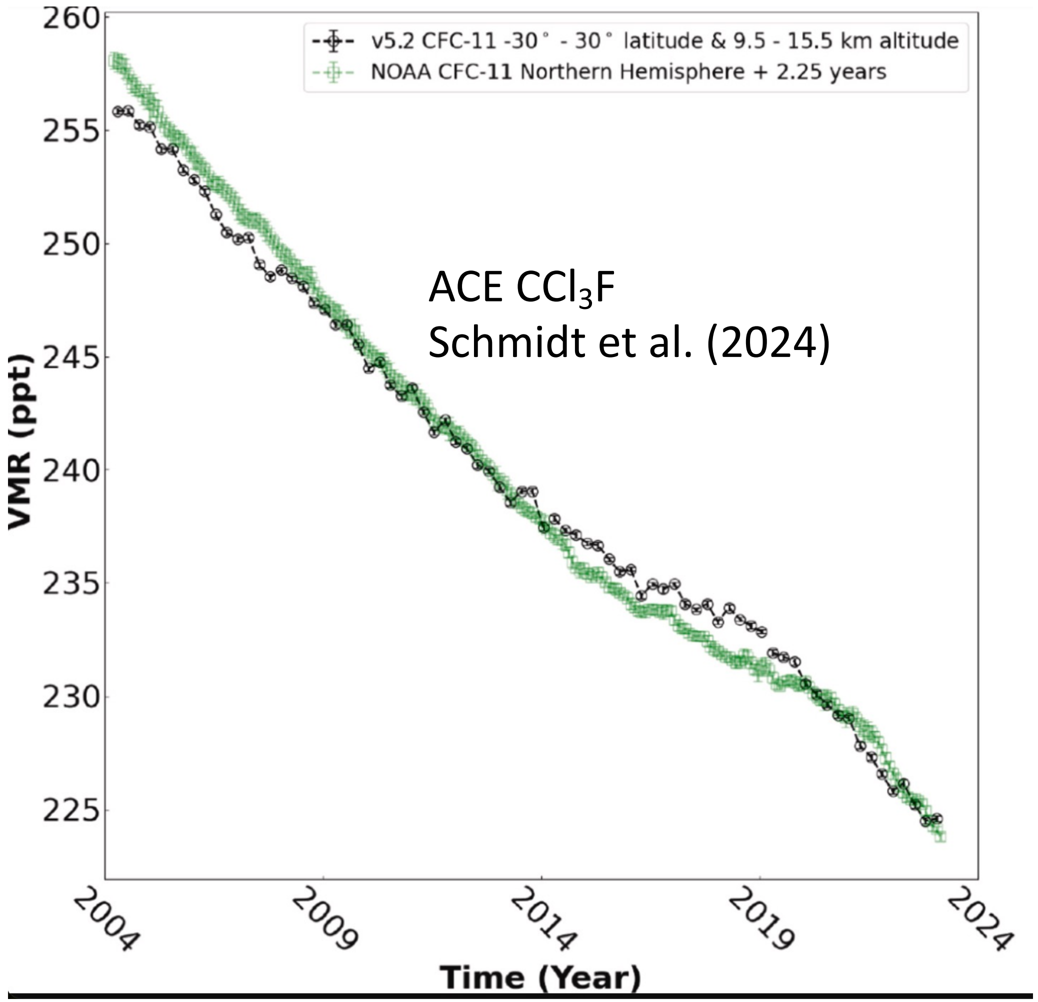
MkIV CCl_4 peaked around 1995 at lowest altitude (red). Has been decreasing since.

ACE CCl_4 covers the 10—15 km altitude. They show a steady decrease since 2004 .

ACE CCl_4 VMRS are 40% larger than those of the lowest (250 ppb N2O) MKIV .

MkIV-ACE CCl_3F Comparison





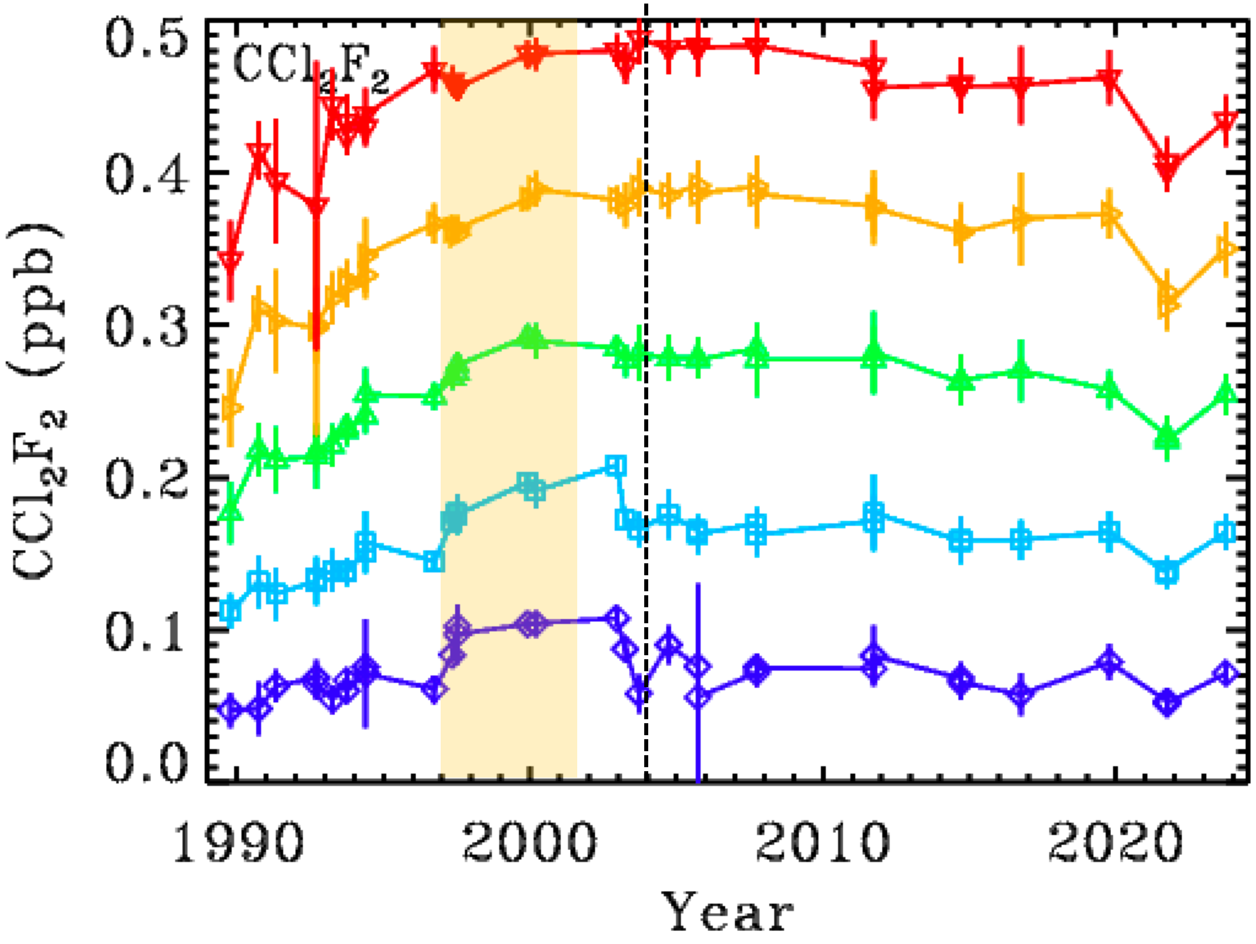
ACE CCl_3F
Schmidt et al. (2024)

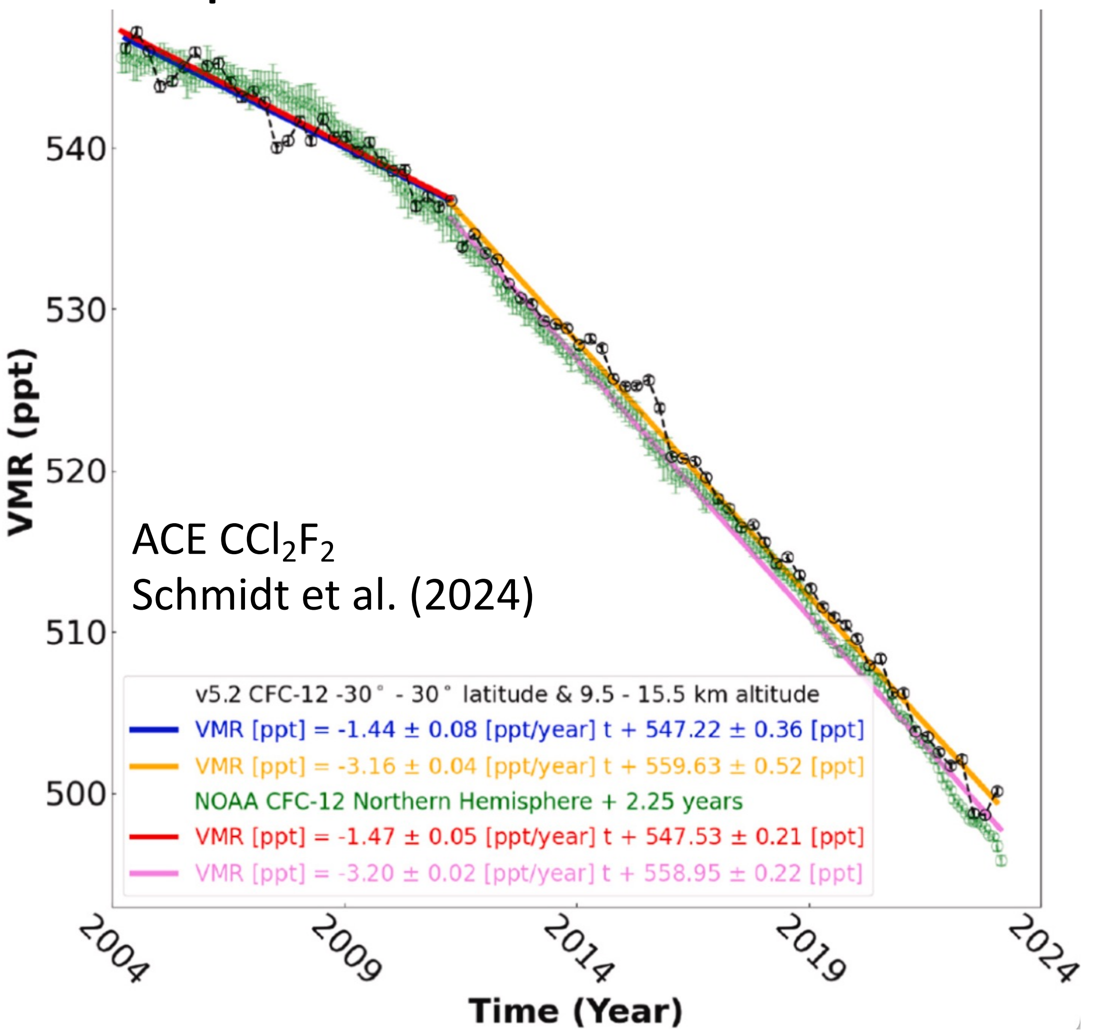
MkIV CCl_3F peaked around 1994 at lowest altitude. Has been decreasing since.

ACE CCl_3F covers the 10—15 km altitude. They show a steady decrease since 2004 .

ACE CCl_3F VMRS are larger than those of MKIV because they represent lower altitudes.

MkIV-ACE CCl_2F_2 Trend Comparison



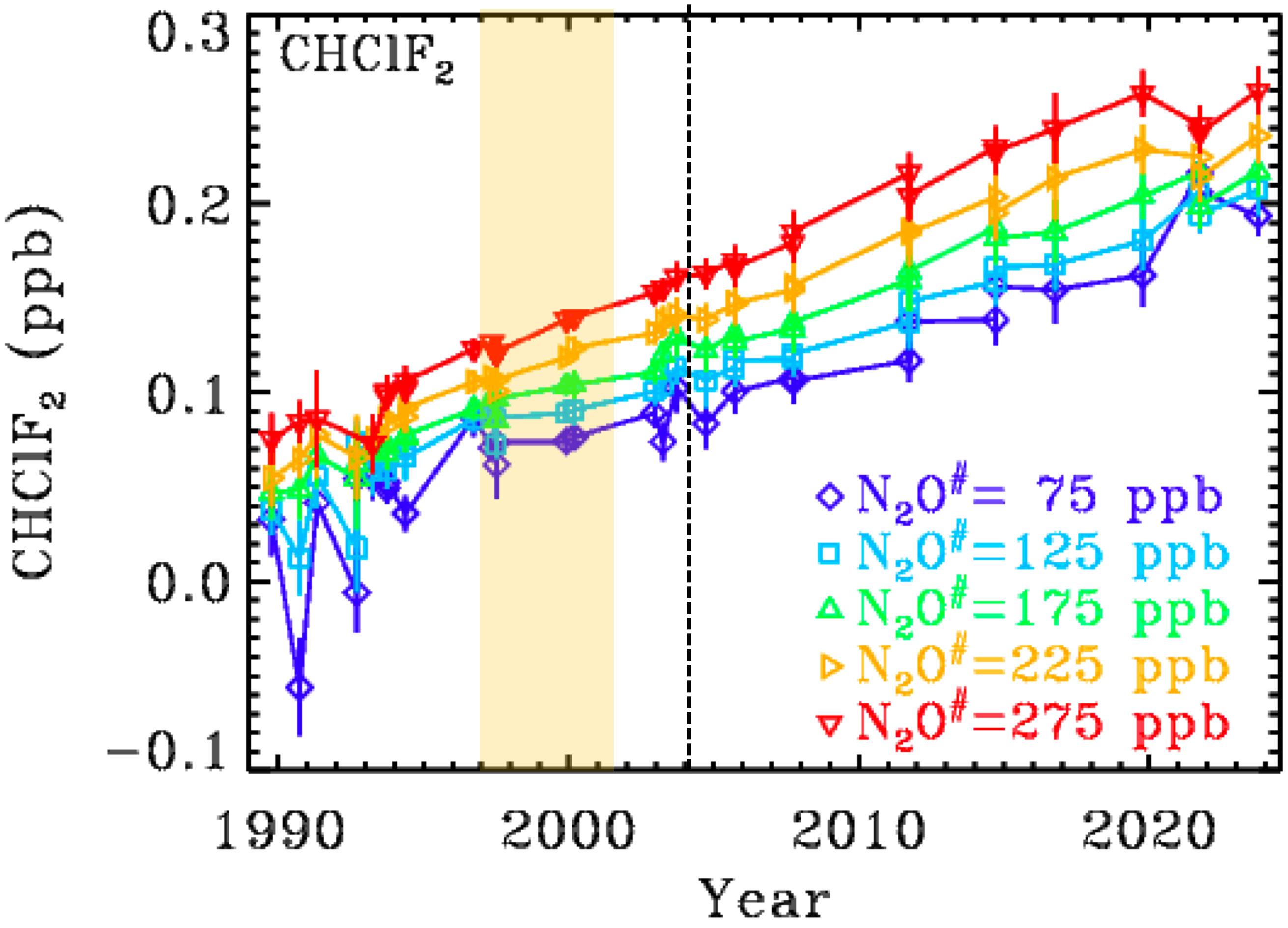


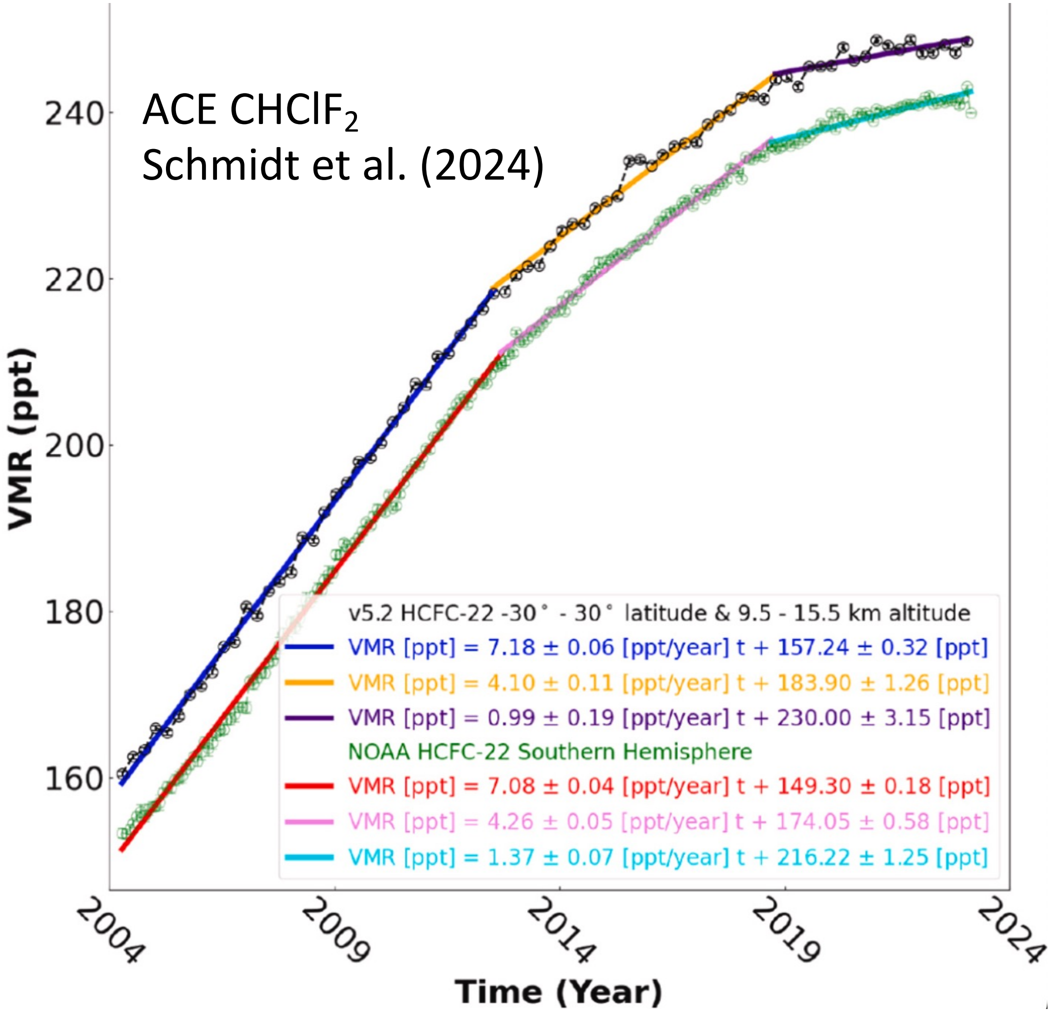
MkIV CCl_2F_2 increased in the 1990s, peaked around 2004. Has been decreasing since.

ACE CCl_2F_2 covers the 5—10 km altitude, which is lower than 275 ppb N2O. They show a linear decrease since 2011.

ACE CCl_2F_2 VMRS are larger than those of MKIV because they represent lower altitudes.

MkIV-ACE $CHClF_2$ Trends



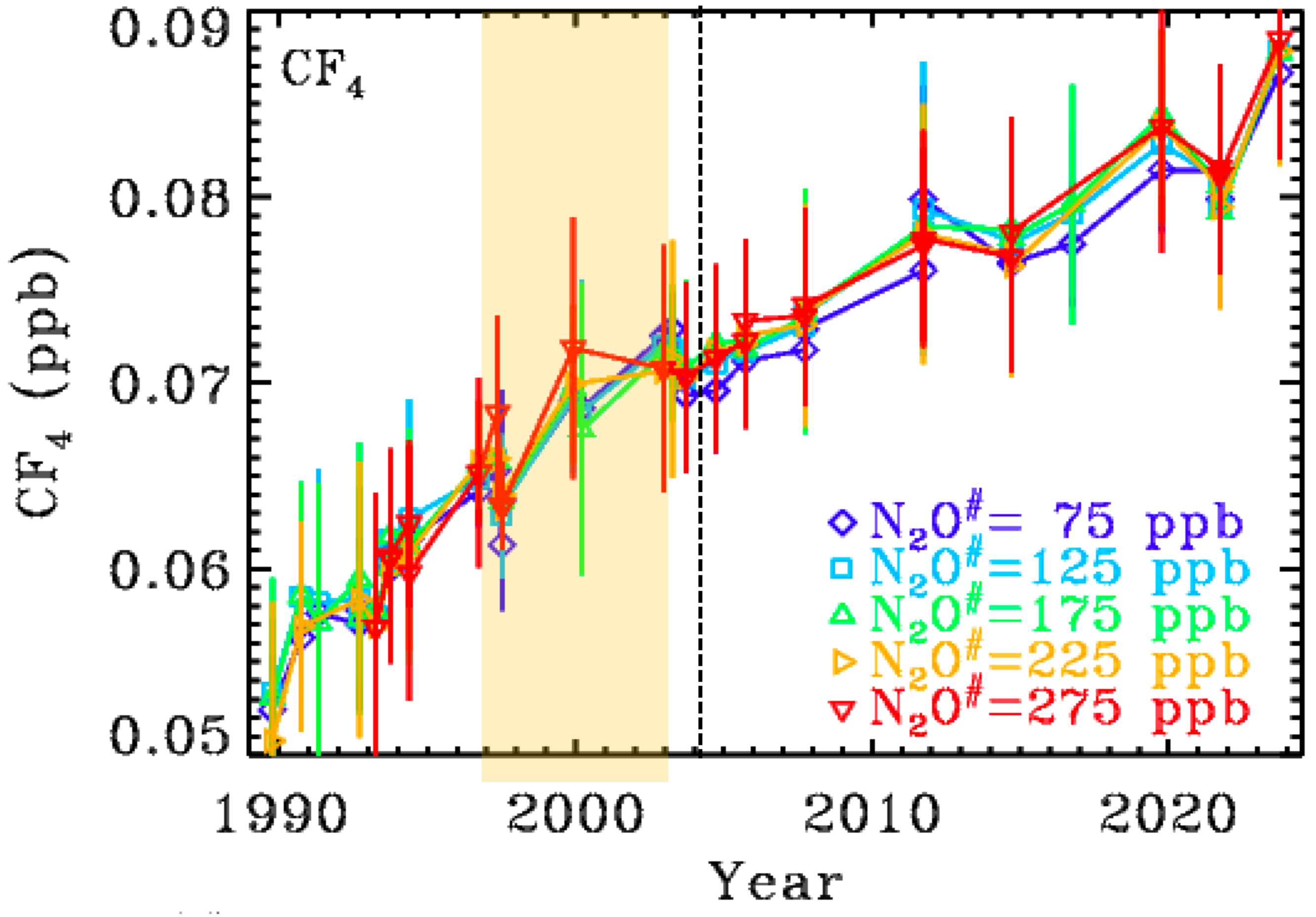


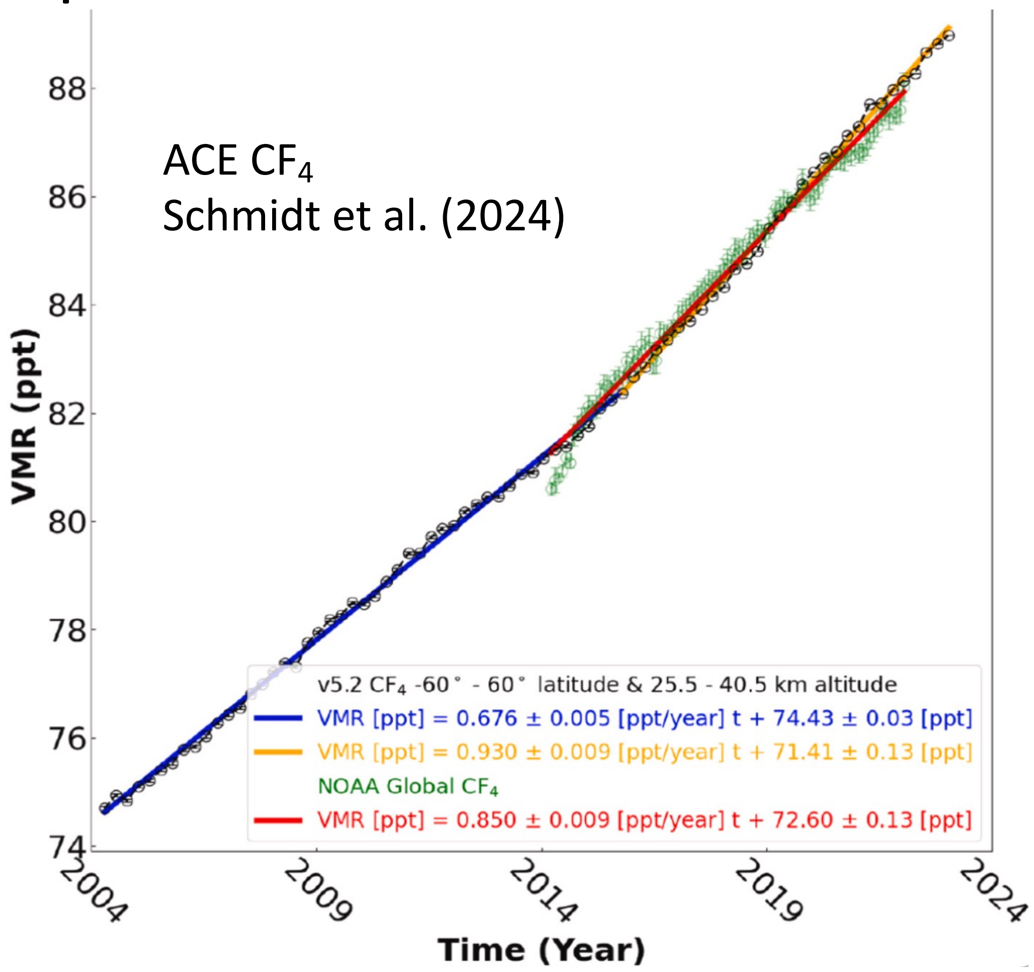
MkIV $CHClF_2$ has tripled since 1989 with an almost linear growth.

ACE $CHClF_2$ show a slowing rate of increase since 2004.

ACE $CHClF_2$ VMRS are similar to those of MKIV at the N_2O=250 ppb level (red), despite representing lower altitudes (5-10 km).

MkIV-ACE CF_4 Trend Comparison



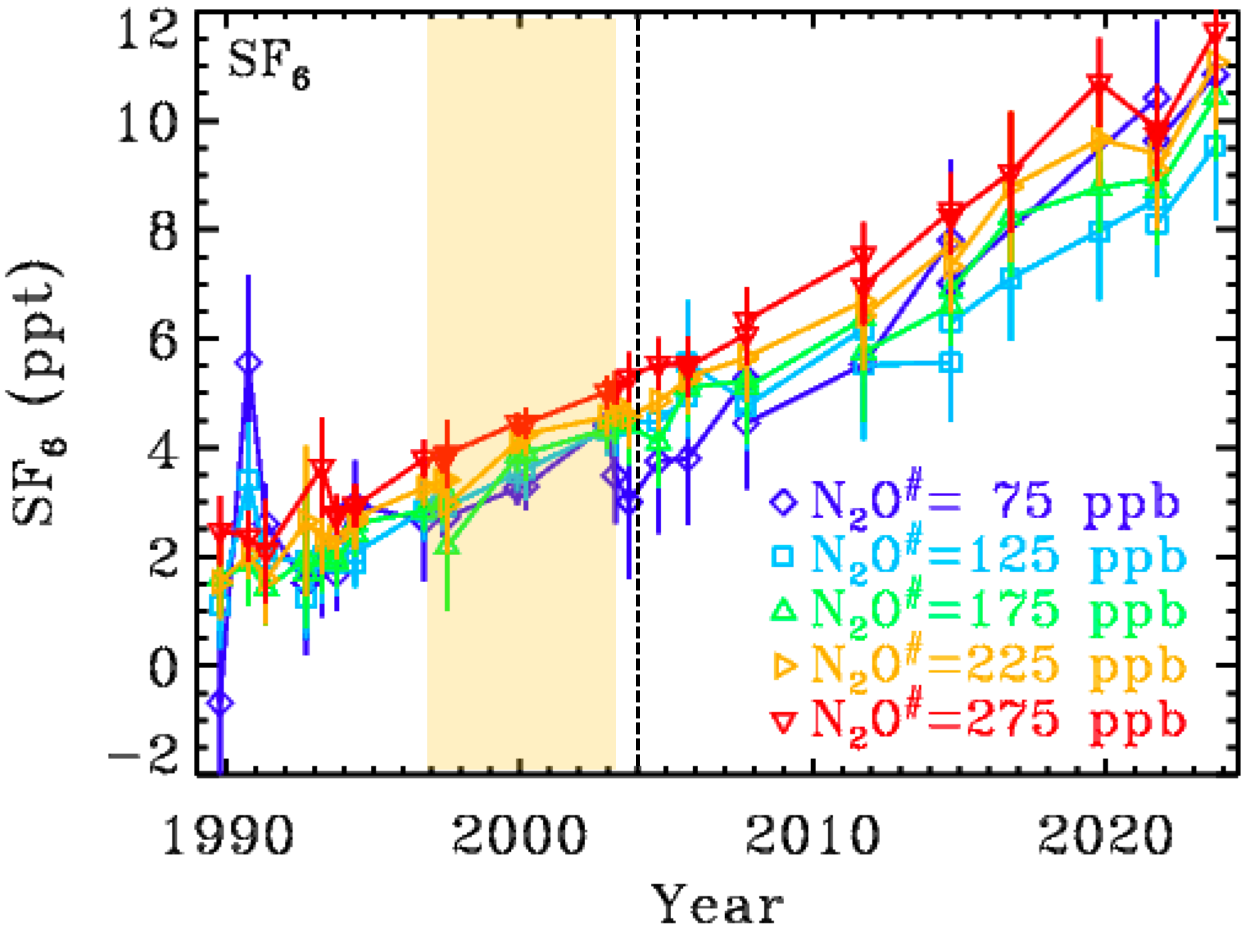


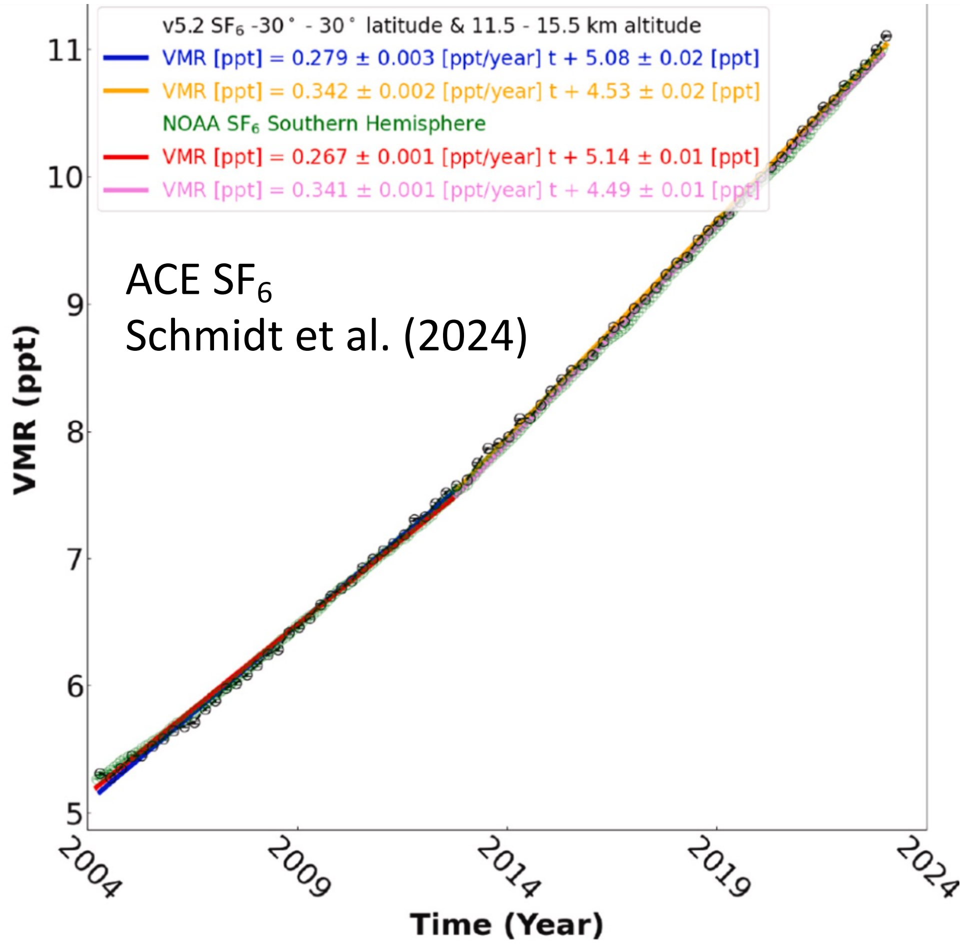
MkIV CF_4 has nearly doubled since 1989 with an almost linear growth. VMRs are noisy at the higher altitudes N_2O=100 ppb (~30km), especially in the early years.

ACE CF_4 25-40 km show an almost constant rate of increase 2004-2015, then a slightly faster increase since 2015.

ACE CF_4 VMRS are are similar to those of MKIV at the N_2O=275 ppb level in 2004

MkIV-ACE SF_6 Trend Comparison

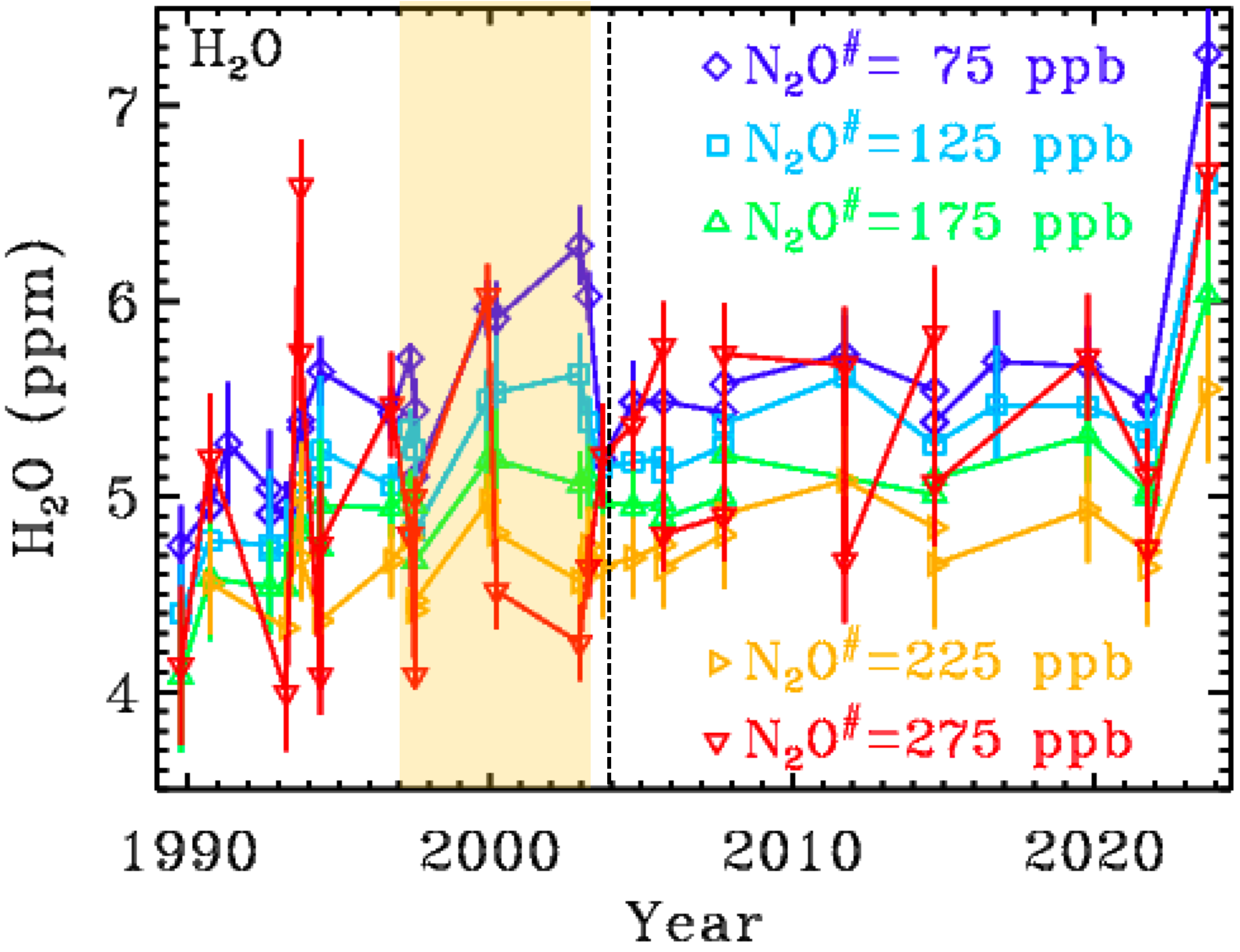


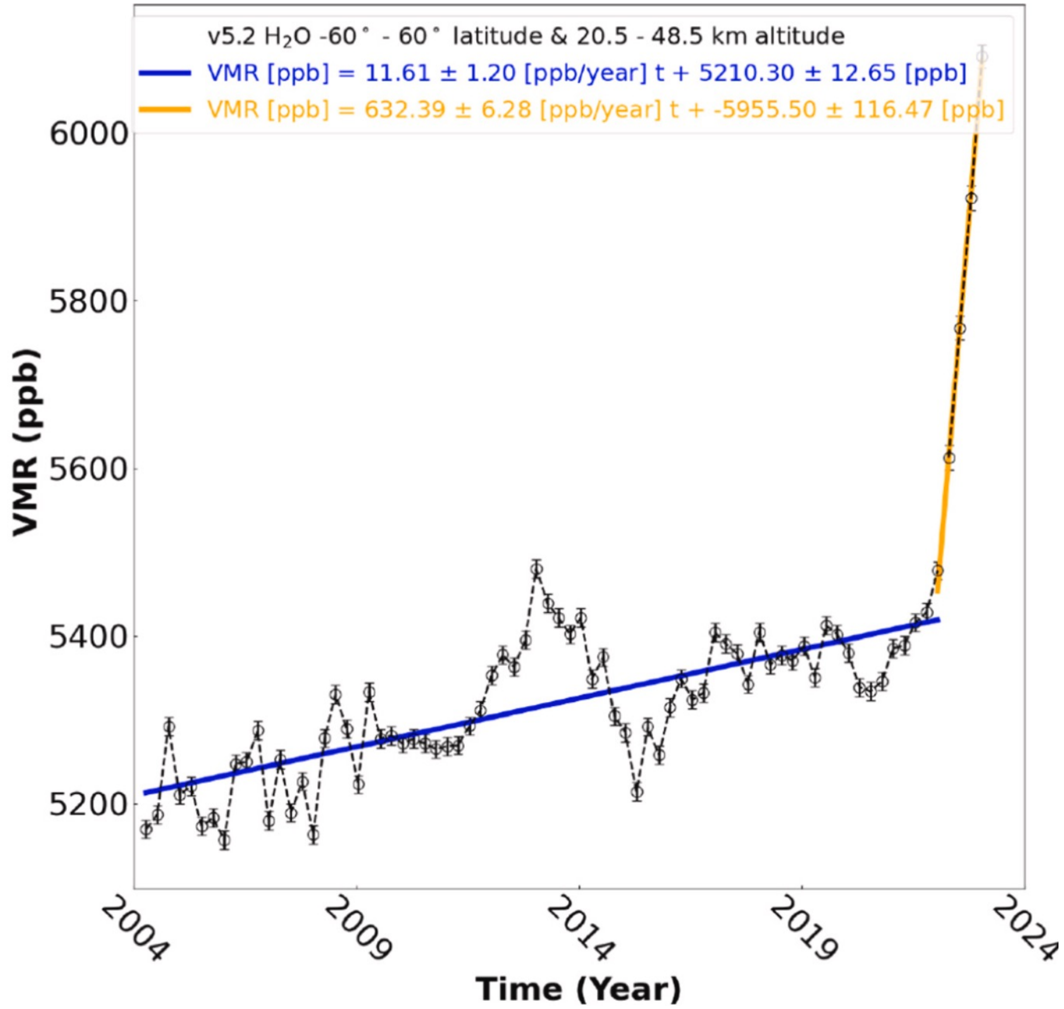


MkIV SF_6 has quintupled since 1989. from 2 ppt to 10 ppt, with a slightly increasing rate of growth. VMRs are noisy at the higher altitudes N_2O=75 ppb (~30km), especially in the early years.

ACE SF_6 averaged over 11.5 to 15.5 km show similar values to the red (275 ppb N2O) MkIV curve.

MkIV-ACE H_2O Trend Comparison

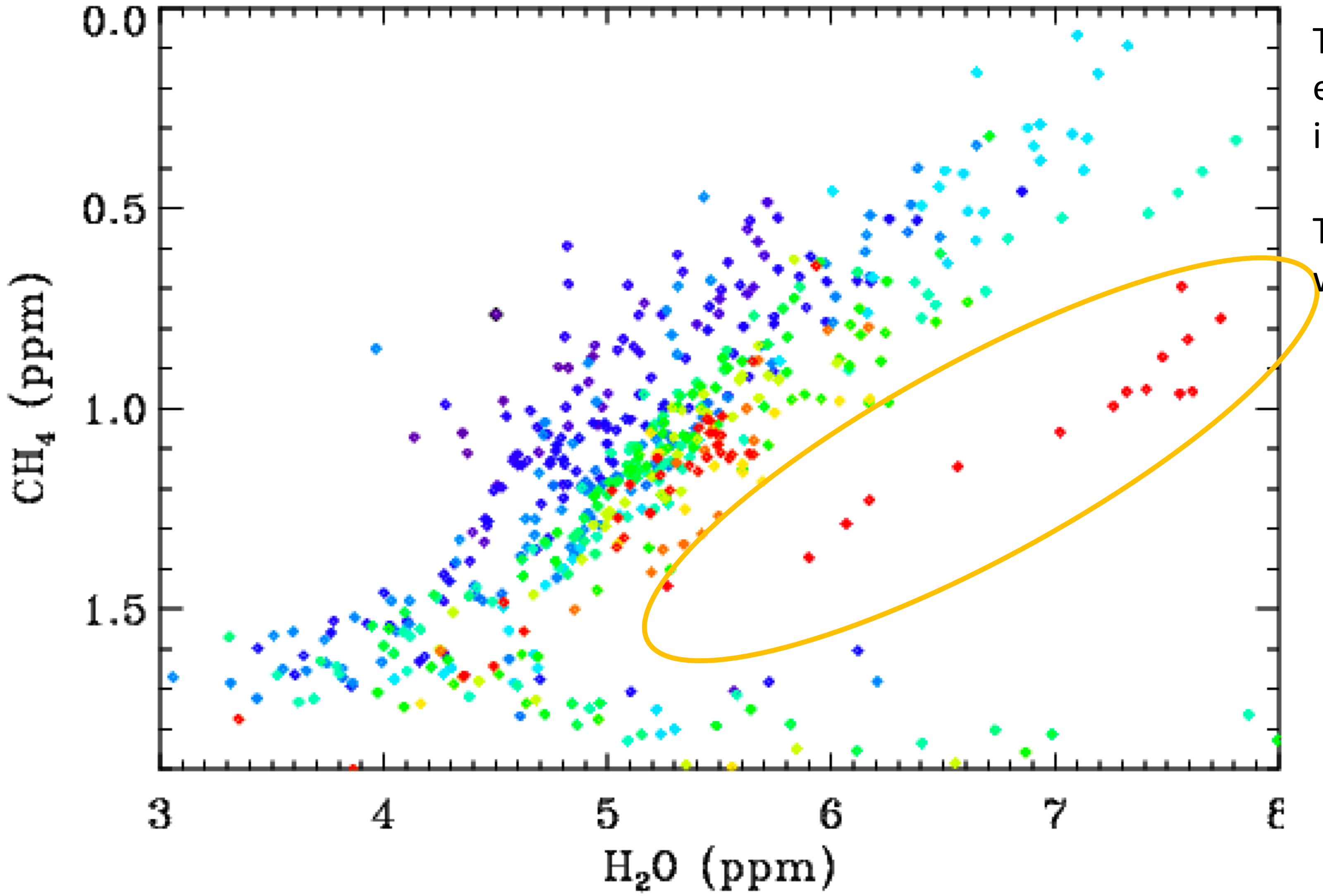




Left: MkIV stratospheric H_2O increased from 1989 to 2000. Then, fairly constant until the Hunga Tunga eruption in Jan 2023.

Right: ACE H_2O show a significant increase in the 20-48 km altitude range between 2004 and 2022. Then a spike in 2023 due to Hunga Tunga.

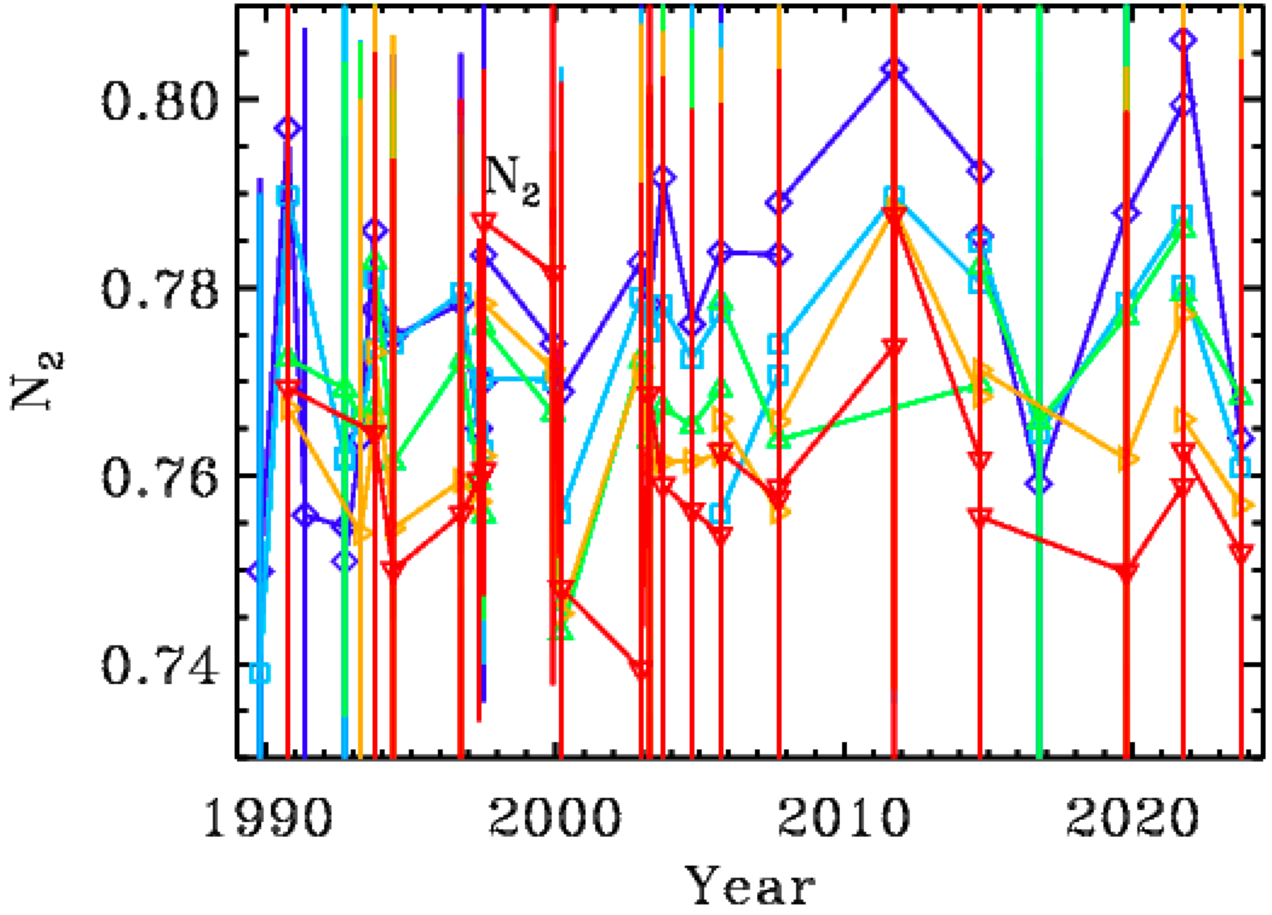
H_2O / CH_4 correlation plot color coded by year (2023 is red)

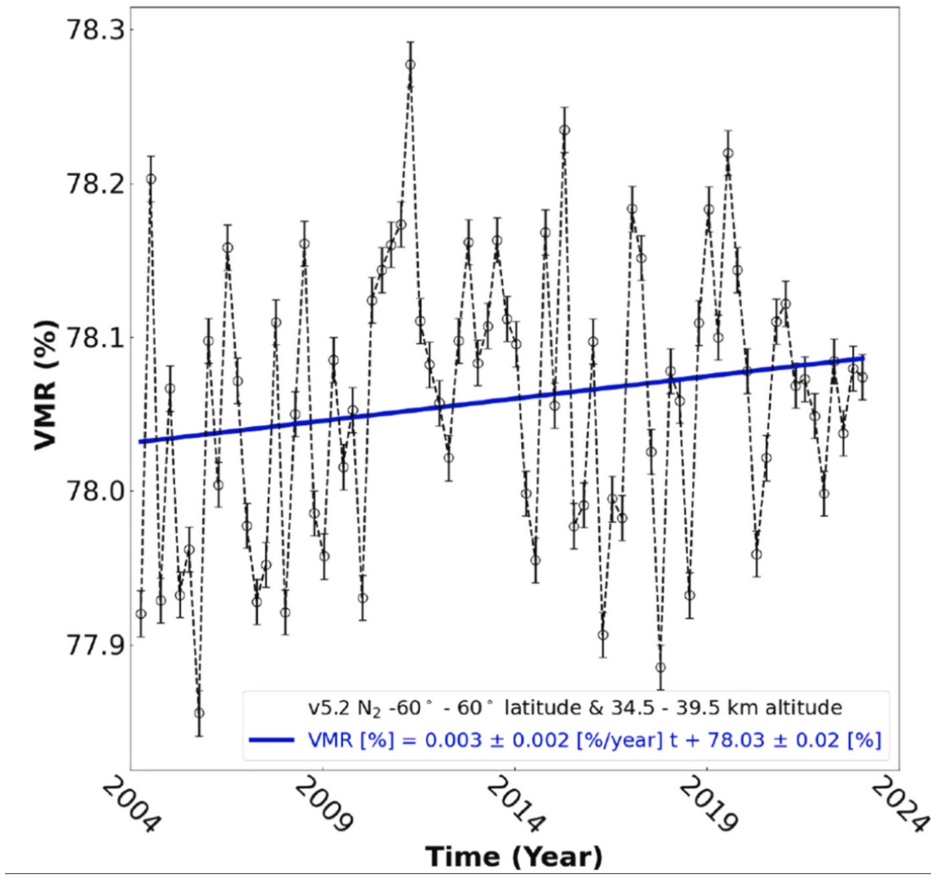


The 2023 points inside the oval show a H2O enhancement of 1 ppm in the lower strat, increasing to 1.5 ppm in the upper strat.

This proves that the 2023 H2O enhancement was not due to transport.

MkIV-ACE N_2 Trend Comparison





Left: MkIV N_2 mole fraction is constant at about 0.77. No trend over time

Right: ACE N_2 averaged over 35-40 km altitude, is constant at 0.780 over the 28-32 km altitude range.

Summary and Conclusions

ACE measurements of the long-term trends Schmidt et al. (2024) are highly precise due to the large number of occultations and in some cases the broad vertical averaging. Currently, no ACE trend information on different altitude ranges.

MkIV has only ~1 occultation per year with large flight-to-flight differences that exceed error bars. Need to “sacrifice” N_2O profiles to remove effects of differences of airmass origin. Can’t average them away like ACE. Using N_2O adds some noise (from N_2O) to the gas trends, but for long-lived gases this is usually smaller than the dynamical effects that are removed.

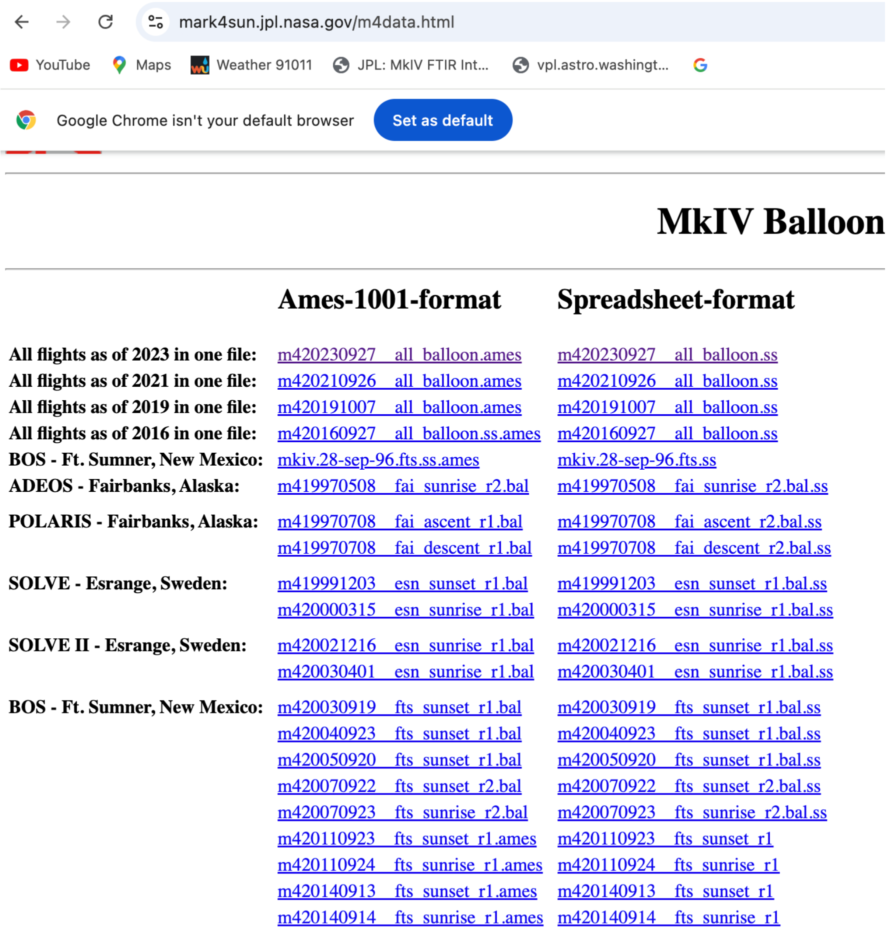
Comparison of absolute VMR values between MkIV and ACE is difficult due to the different altitudes presented (Need to interpolate ACE data onto same N2O isopleths).

Compared trends between MkIV and ACE seem reasonable so far.

All MKIV flights were analyzed with the same software version and linelist.

The MKIV balloon profiles used in this presentation are available from:
https://mark4sun.jpl.nasa.gov/data/mkiv/m420230927__all_balloon.ames

MkIV analysis assumes a compact Gas-N_2O relationship not varying with latitude.



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