Comparing long-term gas trends from ACE and MKIV balloon

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Bernath et al. (2020) recently published the paper "Sixteen-year trends in atmospheric trace gases from orbit" in JQSRT. I was curious to see how well the ACE trends compare with MKIV balloon trends of the same gases.

The MKIV instrument has performed 25 balloon flight over the past 30 years, averaging 1 occultation per year. Most of these were launched from Ft Sumner, New Mexico (35N), with 7 flights from high latitude (Alaska & Sweden). But the MKIV occultations are very sparse. We don't have the luxury of averaging thousands of occultations per year like ACE.

Using MKIV data for trend assessment is difficult, because even for balloon flights launched from the same site and season, the origin of the probed airmasses varies from year to year (and even from day to day) due to wave activity in the stratosphere. So one year the the MKIV may be sampling sub-tropical airmasses transported from 25N, whereas the next year it samples mid-latitude airmasses transported from 45N with lower vmrs of N₂O, CH₄ etc.

So even just considering the Ft. Sumner flights from New Mexico, a range of effective latitudes are sampled, which adds geophysical noise to the measurement, that dwarves the long-term changes of many gases.

How best to extract trends from these accurate but diverse set of observations?

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Example: using a passive tracer (N_2O) to reduce effects of atmospheric transport



▲Z=26km

▼Z=30km

2015

2020

2010

N₂0 Mole

00

50

1990

1995

2000

2005

Year

Instead of considering the vmr profiles as a function of altitude, or pressure, we instead consider them as a function of N₂O. We interpolate the gas vmr altitude profiles onto fixed N₂O isopleths: 100, 150, 200 and 250 ppb.

Panel (a) shows OCS VMRs at 4 different altitudes from 21 to 30 km. Big dip from 1997-2003 (yellow shading) was because these flights were from high latitudes, some inside the polar winter vortex. Also, a small dip in 2016 even at 35N for the lower altitudes (blue and green).

Panel (b) shows the N_2O at the same altitudes, showing very similar behavior.

Panel (c) shows OCS trends seen on fixed N₂O isopleths. The flight to flight variation is greatly reduced allowing a more accurate trend determination

MKIV-ACE: OCS comparison



Upper-Left: MKIV OCS shows no significant trends in the stratosphere (although small trends are seen in tropospheric OCS via ground-based $\bigcirc_{\mathcal{O}}$ column measurements). This figure updates fig.3c in Toon et al. (2018). Hint of a maximum in MkIV OCS in 2016 at the lower levels (N₂O>200) $\bigcirc_{\mathcal{O}}$

Upper-Right: ACE shows an 5% increase from 2004 to 2016, followed by a 4% decrease over the 8.5-10.5 km range (troposphere). ACE vmrs are much higher than MKIV balloon due to the altitude differences.

Lower-Right: MKIV ground-based measurements of tropospheric OCS trends from Toon et al. (2018) show 5% increase from 2004 to 2016.



Another Example: HF

Top panel (a) shows HF vmrs at various altitudes. The large vmrs from 1997-2003 were because these flights were from high latitude, some inside the polar winter vortex.

The 2016 flight from 35N is now a peak in HF relative to neighboring flights at the lower altitudes (blue & green). It seems HF has decreased in recent year

Lower panel (b) panel shows HF vmrs interpolated to fixed N_2O isopleths. The flight to flight variation is greatly reduced. The curves show that HF has increased by more than a factor two 1989-2019, but the rate of increase is slowing. No decrease in recent years.

There is still a residual effect of the high latitudes, especially at the higher altitude (red), probably because the HF-N₂O relationship is latitude dependent. The 2016 flight is now in line with its neighbors.

Isn't this method affected by the trends in N₂O? Yes, but these are well known (0.3%/year) and have been fairly constant over the past 30 years, allowing a simple correction was performed to the MKIV vmrs to account for the N₂O trend.



MKIV-ACE HF Comparison



MkIV HF (upper left) has doubled over the 30-year observation period. But has only increased by about 10% since 2004.

The ACE HF measurements (right panel) cover the 45-55 km altitude range, which is well above the MKIV altitude range, and provides much large HF VMRs. But the rate of increase should be consistent over altitude, in the absence of circulation changes.

ACE sees a 17.516 ppt/year increase, or 280 ppt over 16 years, about a 14% increase.

MKIV-ACE HCI Comparison



MkIV saw increasing HCl from 1989 to 1997. The vortex flights show large heterogeneous loss of HCl. No significant change of HCl since, at any level.

The ACE HCl measurements (right panel) cover the 28.5—48.5 km altitude range, which is well above the MKIV altitude range, and provides much large 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 CCl₂F₂ Comparison



MkIV CCl_2F_2 increased in the 1990s, peaked around 2004, then decreasing by about 5% by 2016, after which it leveled off. ACE CCl_2F_2 covers the 5.5—10.5 km altitude, and show an an accelerating decrease since 2004 reaching 7% by 2020. ACE CCl_2F_2 VMRS are higher than those of MKIV because they represent lower altitudes.

MKIV-ACE CHClF₂ Comparison



MkIV CHClF₂ has tripled since 1989 with an almost linear growth.

ACE CHClF₂ show a slowing rate of increase since 2004, reaching 7% by 2020.

ACE CHClF₂ VMRS are similar to those of MKIV at the N₂O=250 ppb level, despite representing lower altitudes (5-10 km).

MKIV-ACE CHF₃ Comparison



MkIV CHF₃ has more than tripled since 1989 with an almost linear growth. VMRs are noisy at N₂O=100 ppb (30 km).

ACE CHF₃ show an almost constant rate of increase, nearly doubling since 2004, reaching 7% by 2020.

ACE CHF₃ VMRS are were similar to those of MKIV at the N₂O=250 ppb level in 2004, but MkIV's have since grown more

MKIV-ACE N₂O (Fake) Comparison



Since MkIV has used N₂O to define the vertical reference frame, we cannot claim to also measure the N₂O trend. The figure in the upper left is simply a self-consistency check: that when were use N₂O in the same way as the other gases, the assumed N₂O vmrs are returned. Note that in the reference year, 2000, the N₂O vmrs cross their nominal isopleth values. The assumed 0.26%/year rate of increase is apparent.

ACE N₂O vmrs show 0.799 ppm/year increase, about 0.26%/year, which is consistent with that assumed in MKIV analysis.

Summary and Conclusions

ACE measurements of the long-term trends Bernath et al (2020) 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. Averaged altitude range varies from gas to gas.

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 affects 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 this is usually smaller than the dynamical effects that are partially removed.

Comparison of absolute VMR values between MkIV and ACE is difficult due to the different altitudes presented, and the fact that the MKIV data are not fixed in altitude, but fixed in N₂O. But comparison of trends is still valid.

Compared trends between MkIV and ACE seem reasonable so far – no obvious discrepancies seen yet.

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

MKIV balloon profiles available from: https://mark4sun.jpl.nasa.gov/data/mkiv/m420191007__all_balloon.ames

More MKIV trend information available from: <u>http://mark4sun.jpl.nasa.gov/report/MkIV_Balloon_VMR_Trends_2020.pdf</u>

Current MkIV analysis assumes a compact Gas-N₂O relationship that does not vary with latitude. In future, attempt a further (second-order) correction to account for the latitude variation.

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