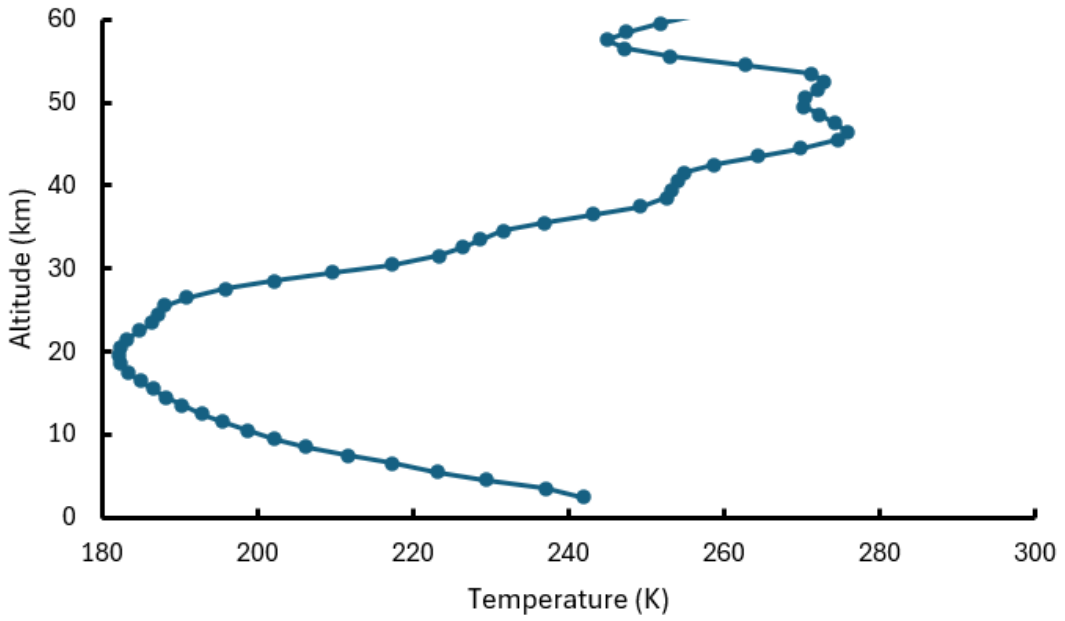
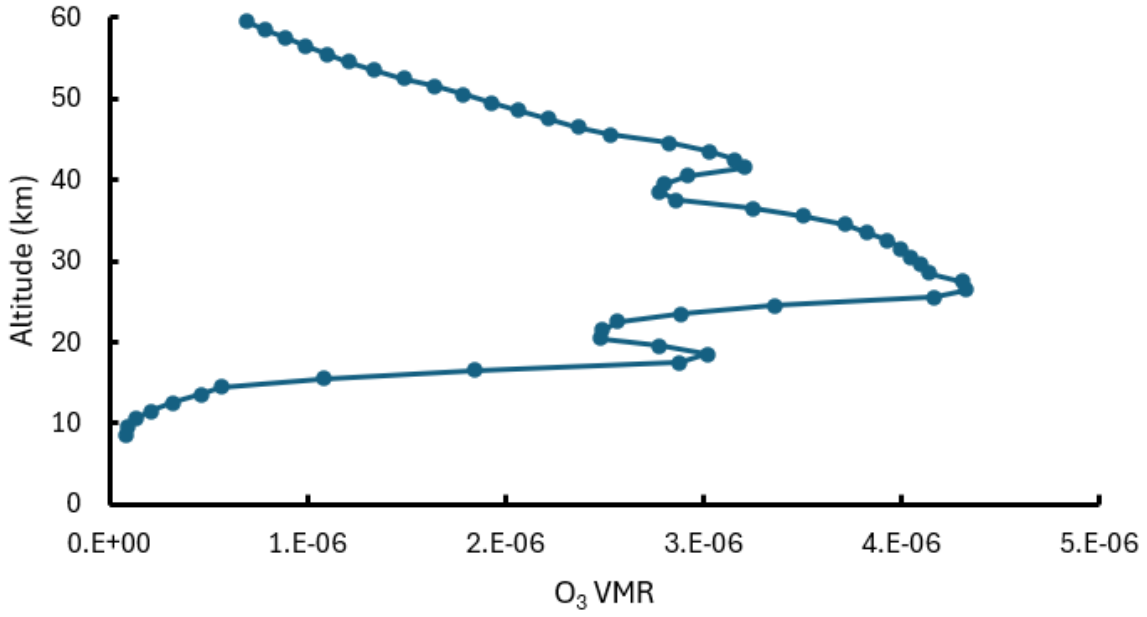
This article investigates average seasonal enhancements in pollutants related to biomass burning using measurements from the MIPAS instrument. Ratios of the VMRs for different molecules are evaluated, relating relative enhancements of different constituents compared to enhancements in HCN to expectations for emission ratios from various types of vegetation, identifying when unexpectedly high levels relative to HCN suggest sources other than biomass burning. In general, it is well written and thorough, a good summary of what MIPAS sees for this class of molecule. However, there were a few issues that I felt should be addressed.

Firstly, I disagree with setting the tropopause height to 14.7 km for measurements within the polar vortex where the temperature minimum is around 20 km. I agree that the assumptions associated with the WMO definition of tropopause have broken down in that situation, but a tropopause of 14.7 km seems way too high in this latitude region. This can be illustrated by looking at the chemical makeup in the vicinity of 14.7 km within the Antarctic polar vortex.

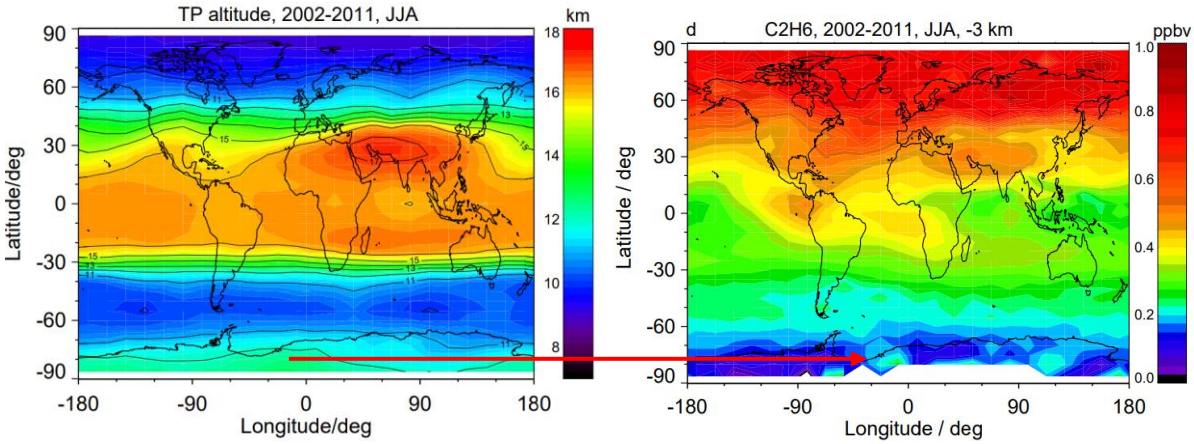


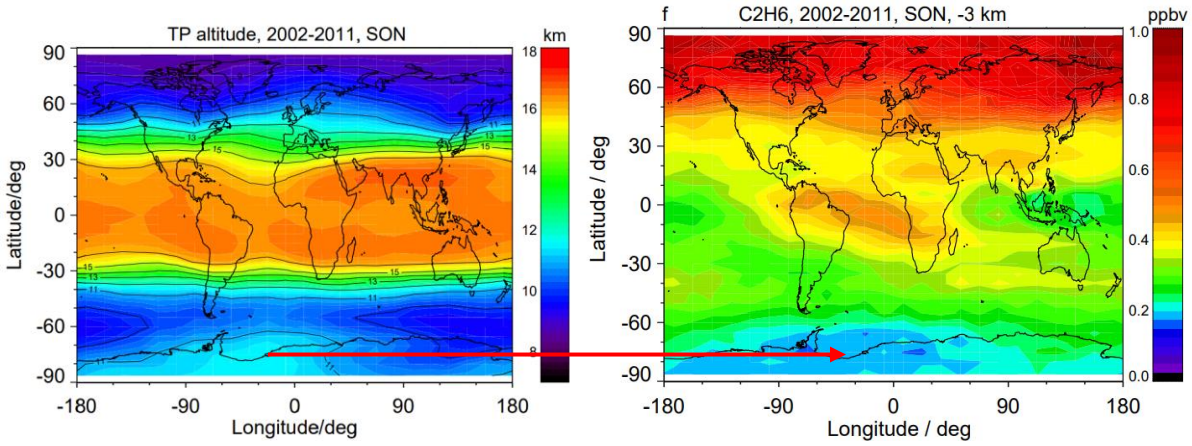
Above: the temperature profile from occultation ss108064 from the Atmospheric Chemistry Experiment Fourier transform spectrometer (ACE-FTS), measured September 1, 2023, near latitude 80 °S. Applying the WMO definition of tropopause would yield a value near 20 km.



Above: the O_3 volume mixing ratio (VMR) profile for ss108064. In the troposphere, O_3 levels should be relatively low. Judging from the above profile, the tropopause at the location of this occultation is somewhere around 10 km. 14.7 km is well into the stratosphere. Unfortunately, the altitude resolution of O_3 measurements from MIPAS presumably means O_3 cannot be used to estimate tropopause height.

The impact of this issue is probably best seen in the distribution plots for C_2H_6. For seasons where high average tropopause heights are seen in the data set, low levels of C_2H_6 are indicated in the distribution plots. This is not a chemical (i.e., low C_2H_6 in the troposphere) effect. Instead, it is a consequence of including stratospheric air (with low levels of C_2H_6) in the average.





The obvious solution would be to use a dynamical tropopause in the analysis rather than the WMO definition. When dealing with measurements inside the polar vortex, it is possible that using a single boundary on potential vorticity (available from MERRA-2) might not suffice, and you would need to use a more sophisticated approach (doi:10.1029/2010JD014343, "Dynamical tropopause based on isentropic potential vorticity gradients" by Kunz et al., JGR Atmospheres, 2011). The alternative would be to simply exclude Antarctic measurement where the WMO tropopause is above some threshold (15 km?). Some distribution plots would likely end up with blank regions, but that would be preferable to reporting mostly stratospheric results as tropospheric. Regions with no data would presumably all be in the Antarctic, which is the least interesting region anyway (scientifically speaking), with no significant sources of these molecules.

The second issue I had related to internal consistency for the different molecules. Enhancement ratios are considered for each molecule separately, which admittedly already makes for a large amount of information to digest, but no synergy between the different molecules was considered. For example, unexpectedly low enhancement ratios were observed for C_2H_2 and C_2H_6 in the tropics/subtropics in October. The quite logical suggestion was made that this indicated aged emissions, with C_2H_2 and C_2H_6 levels having decreased more than HCN because of their shorter lifetimes. However, the following observation was then made for HCOOH in October (page 20, line 26):

Thus both the high correlations as well as the ERs indicate that biomass burning is a major source of the enhanced HCOOH observed in the southern tropics and subtropics during this period.

The conclusion from C_2H_2 and C_2H_6 suggested that on average in October, the fires contributing to their enhancement in this region occurred a long time previously, long enough for the shorter lifetimes relative to HCN to have a significant effect. Then the conclusion for HCOOH in October was the opposite, that there were "fresh" fires contributing to the HCOOH enhancement. For me, it would have been more logical to conclude that the contributions to HCOOH enhancement from biomass burning

should have been relatively low because we generally have aged emissions in October (as suggested by the C_2H_2 and C_2H_6 observations), so the higher enhancement ratios for HCOOH (compared to those observed for C_2H_2 and C_2H_6) indicate biogenic sources are also likely contributing. This would be consistent with the conclusions in the paper suggesting biogenic sources for HCOOH during other time periods. Why would there be biogenic sources of HCOOH during other time periods but not this one?

As the authors indicate, the different vertical resolutions HCN and the other molecules will impact the accuracy of the ratio. They report that degrading C_2H_2 to HCN resolution yields "on average" 10% lower C_2H_2. It was not clear what conditions were being averaged over. Over the global set? The vertical resolution changes with latitude, and so does the tropopause height, which could compound the systematic error. Since the vertical resolution for C_2H_2 is larger at 14 km than it is at 8 km, presumably the impact would be smaller in the tropics than near the poles because there would be less of a resolution mismatch.

Regardless, an error of 10% is probably low compared to the uncertainty from the unknown "emission age" of the air, due to the different lifetimes of the molecules being ratioed. That could perhaps be stressed more when comparing observations of enhancement ratios to emission ratios for various types of vegetation. It is difficult to make definitive statements of agreement/disagreement with the expectations for a particular type of vegetation without knowing how long ago the fire occurred.

Minor comments:

Page 7, line 6: but ca also be caused

ca -> can

Page 7, line 16: caused by confinement of pollution mostly from South- and South-East Asia (see, e.g., Vogel et al., 2015) inside the Asian Monsoon Anticyclone (AMA)

Not just confinement; vertical transport within the monsoon region carries pollution toward the upper troposphere, increasing the measured enhancement for a point 2 km below the tropopause.

Sometimes "Table" is written out fully and sometimes it is abbreviated as "Tab."

Figure 8: North Africa is labelled as NAFR but is referred to as NAF in the text

Page 17, line 26: Like for HCH versus CO

HCH -> HCN

Page 19: no reference to Figure 13 in the text