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Global distributions of acetone in the upper troposphere from MIPAS–E spectra

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Abstract

This study reports the first global measurements of acetone in the upper troposphere. Profiles are obtained between 6 and 15 km from measurements made by the Michelson Interferometer for Passive Atmospheric Sounding onboard Envisat (MIPAS–E) from August 2003. The best accuracy and precision is obtained at lower altitudes in the mid-latitudes and poles. We report a strong hemispheric difference in the acetone volume mixing ratios (vmrs) with the highest average concentrations in the Northern Hemisphere (NH) mid-latitude upper troposphere of between 2000 ppt and 3000 ppt. These high distributions are possibly linked to higher vegetative emissions during the NH summer season. Globally, concentrations range between 750 ppt and 3000 ppt in the middle troposphere (300 to 500 hPa) and between 50 ppt and 2000 ppt in the upper troposphere (120 to 200 hPa). The results show the likely strong influence of mid-latitude and boreal processes on the acetone summertime distribution. Zonal distributions of acetone show that vmrs of the gas decrease rapidly with increasing altitude (decreasing pressure) but there is reversible transport between the upper troposphere and the lowermost stratosphere. Acetone vmrs remain fairly constant poleward of 45° S with vmrs between 400 ppt and 500 ppt. Northern Hemisphere vmrs poleward of 45° N are consistently higher than this on a day-to-day basis by between 200 ppt and 1000 ppt on average.

1 Introduction

Acetone is present ubiquitously in the troposphere with concentrations in the range 0.2 ppb to 3 ppb as determined from sporadic aircraft measurements (Jacob et al., 2002; Arnold et al., 1997; Knop and Arnold, 1987) and is a major source of hydrogen oxide radicals (HO_x) and peroxyacetyl nitrate (PAN) in the upper troposphere (UT) (Singh et al., 1995; Jaegle et al., 1997, 2001; McKeen et al., 1997; Wennberg et al., 1998; Collins et al., 1999; Muller and Brasseur, 1999). Elevated levels of HO_x imply

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a more photochemically active UT, with enhanced rates of ozone formation, and has implications for the mediation of ozone in low water vapour (less than 100 ppmv) conditions (Folkins and Chatfield, 2000).

The presence of acetone in the upper troposphere was originally detected by the MPIK Heidelberg group (MPIK=Max-Planck-Institut für Kernphysik) (Hauck and Arnold, 1984; Knop and Arnold, 1987). Main sources of acetone include atmospheric oxidation of precursor hydrocarbons (propane, isobutane, and isobutene), biomass burning and direct biogenic emissions with a small percentage from primary anthropogenic emission (Singh et al., 1994; Arnold et al., 1997). Importantly, the sources are still poorly understood (Jacob et al., 2002). The main sinks of acetone include photolysis and reaction with OH radicals and this results in an estimated global mean lifetime for acetone of the order of a month (Gierczak et al., 1998). Another sink mechanism that has been proposed is deposition to the oceans (Singh et al., 1994).

The advent of spaceborne limb-viewing Fourier transform spectrometers has allowed the possibility to globally observe many trace gases in the upper troposphere and lower stratosphere (UTLS). One such instrument is the Michelson Interferometer for Passive Atmospheric sounding onboard Envisat (MIPAS-E) (Fischer et al., 2008; Fischer and Oelhaf, 1996). Operationally generated data sets from the MIPAS-E consist of temperature, H₂O, O₃, CH₄, N₂O, HNO₃, and NO₂ profiles although the high-spectral resolution of the data (0.035 cm⁻¹ unapodized) allows the potential retrieval of many other minor gas species (Burgess et al., 2004; Hoffmann et al., 2005; von Clarmann et al., 2006). We present the first global measurements of acetone in the UTLS using MIPAS-E spectral data and explore the zonal and vertical distribution of the compound during August 2003.

2 Measurements

The MIPAS-E instrument onboard the ENVIRONMENTAL SATellite (Envisat) was successfully launched in March 2002 as part of an ambitious and innovative payload. The

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Envisat is in a Polar orbit at an altitude of 800 km, with an orbital period of about 100 min and a reference orbit repeat cycle of 35 days. The MIPAS–E (Fischer and Oelhaf, 1996; Fischer et al., 2008) is a Fourier Transform Spectrometer that provides continual limb emission measurements in the mid infrared over the range 685 to 2410 cm^{-1} (14.6 to 4.15 μm) at an unapodized resolution of 0.035 cm^{-1} during the nearly continuous first two years of flight. This paper concentrates on measurements from the full spectral resolution period of MIPAS–E; the instrument subsequently operated in a reduced spectral mode from August 2004 onwards. The instrument's field of view is approximately $3\times 30\times 400$ km and one complete limb sequence of measurements in nominal mode consists of 17 spectra with tangent altitudes at 68 km, 60 km, 52 km, 47 km, 42 km and continuing downwards to 6 km in 3 km intervals.

The spectral reference data for acetone came from a set of infrared absorption cross-sections measured at the Molecular Spectroscopy Facility at the Rutherford Appleton Laboratory and described in Waterfall (2004). Here, a set of temperature and pressure dependent absorption cross-sections were derived covering a temperature range between 298 K and 214 K. Measurements were recorded at a spectral resolution of approximately 0.03 cm^{-1} (defined as 0.9/optical pathlength) and Norton-Beer strong apodization. For the ν_7 (C–C stretch) acetone band (between 1195 and 1240 cm^{-1}), these measurements have been estimated to have total errors varying between 4 and 10% depending on the frequency and the temperature. Very recent work by Harrison et al. (2010) determined infrared absorption cross sections for acetone in a similar spectral range to Waterfall (2004) but extending their measurements down to 194 K. Their estimated uncertainty on the cross-sections of 4% give confidence that our error estimate is conservative.

The presence of acetone spectral signatures in MIPAS data was demonstrated by Remedios et al. (2007a). The MIPAS retrievals described in this paper utilise the 1218 cm^{-1} band, with a narrow microwindow selected between 1216.75 cm^{-1} and 1217.5 cm^{-1} . For this spectral range the errors on the individual cross-sections are estimated to range between 5 to 7%. An additional source of error may be introduced

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within the radiative transfer model when extrapolating the data to the appropriate atmospheric conditions, and a more conservative estimate of 10% has been used in the calculation of the retrieval error detailed later in this paper. This estimate also accounts for cases where the temperature of the measurement is less than 214 K (the lowest temperature at which the acetone cross-sections were measured). This is particularly relevant for tropical acetone retrievals where it was found that 32% (1199 out of 3737 cases) had a temperature of less than 214 K in the upper troposphere.

For this study we use the same retrieval approach as described by Moore and Remedios (2010) and the MIPAS Orbital Retrieval using Sequential Estimation (MORSE) scheme (Dudhia, personal communication). Using an average acetone climatology in mid-latitudes (20 to 65 degrees of latitude) based on TOMCAT model output (N. Richards, personal communication), the ν_7 C–C stretch at 1216 cm^{-1} is observable according to radiative transfer simulations at 9 km and 12 km globally above the MIPAS–E band B noise equivalent spectral radiance (NESR). By 15 km, the signal decreases rapidly, so we may only expect to measure acetone (and not reproduce just the a priori) where there is a significant enhancement of acetone compared to the background (i.e. in a biomass burning event). The apodized instrument line-shape (AILS) data are based on pre-flight estimates (H. Nett, personal communication, 2003) with line parameters and absorption cross-sections, apart from acetone, taken from HITRAN 2004 with updates up to the end of 2008 applied (Rothman et al., 2005). The quantities for pressure, temperature, water vapour, methane and nitrous oxide are pre-retrieved for each scan using the same scheme. All other species are taken from the reference atmospheres of Remedios et al. (2007b). We restrict the retrieval altitude range from 6 to 21 km.

Using the MORSE retrieval scheme, we retrieve pressure/temperature (jointly), water vapour, ozone (O_3), nitric acid (HNO_3), methane (CH_4) and nitrous oxide (N_2O) before we retrieve acetone vmrs. The MORSE scheme was designed to be able to replicate and improve upon the parameters retrieved by the European Space agency operational processor (Raspollini et al., 2006). Our retrieval tests show that MORSE is

performing as well as the the operational processor and hence we use some parameter uncertainties based upon validated data from the operational processor. We believe that our error estimates therefore represent an upper limit to the total error.

A detailed error analysis for acetone is shown in Fig. 2 for four separate latitude bands – tropical (20° S to 20° N), mid-latitudes (20° to 65° in each Hemisphere), Polar summer (65° to 90° in the summer Hemisphere) and Polar winter (65° to 90° in the winter Hemisphere). Both the random retrieval noise and systematic errors (model parameter, gain, instrument line shape and spectroscopy) are shown along with the total error.

Firstly, the systematic model parameter errors were calculated using measured biases in MIPAS–E data. An uncertainty of 20% has been indicated for MIPAS–E water vapour (Lahoz et al., 2004), 10% for ozone (Cortesi et al., 2007), 10% for nitric acid (Wang et al., 2007), 5% for methane (Payan et al., 2009) and 4% for nitrous oxide (Payan et al., 2009). Uncertainties of 1 K for temperature (Ridolfi et al., 2007) and 4% for pressure (Raspollini et al., 2006) were used. The error for the other contaminants was expected to be small; we assumed a 10% uncertainty for each of them. The errors due to uncertainties in these gases were likely to be lower in the real measurements than calculated here. The uncertainty of the instrument gain and instrument offset were taken to be 2% and 2 nW/(cm² sr cm⁻¹), respectively (Spang et al., 2005). Spectroscopic inaccuracies of acetone cross-section data were set conservatively to 10%.

The total error on a single retrieved vmr is calculated to be mainly below 40% at pressures between 250 hPa and 350 hPa, although the water vapour contamination in the tropics limits errors to around 80%. At pressures lower than this the random retrieval error component increases rapidly in all regions and the total error on a single profile varies between 50% and 120%. In this paper we examine the average global behaviour and the random retrieval error in Fig. 4 is in effect much lower than this.

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3 Results

We have used one month of high-resolution MIPAS-E data to investigate the global-average distribution of acetone in the upper troposphere and lower-stratosphere (UTLS) for August 2003. Previous work by (Singh et al., 1994) estimates that 21% of the acetone source is due to direct biogenic emissions and we chose to look at August 2003 due to this period representing the end-point of the Northern Hemisphere growing season. August also encapsulates the Central African biomass burning season (another potential source of acetone).

To detect clouds in the spectra we used a simple ratio approach described by (Spang et al., 2004) by computing a ratio between the mean radiance in the 788.20 to 796.25 cm^{-1} and 832.30 to 834.40 cm^{-1} . Where the ratio is less than 4, the spectra are flagged as cloudy (that is having some cloud fraction in the field of view) and no retrieval performed on these. There were 26 699 quality-flagged scans recorded by the MIPAS-E during the August 2003. At the 9 km nominal altitude level of MIPAS-E, we found that this removed 18213 (68%) of data. Of these remaining data, the retrieval converged for 7175 scans, a success rate of over 85%.

From orbit 07466 on 4 August 2003, we show the quality of residual fit for two different cases (Fig. 3). It can be seen that the residual fit without acetone included in the simulation is up to 50 $\text{nW}/(\text{cm}^2 \text{sr cm}^{-1})$ away from the measured radiance. With retrieved acetone (and continuum) included, the residual is well within the reported MIPAS-E noise of 20 $\text{nW}/(\text{cm}^2 \text{sr cm}^{-1})$ reported by (Kleinert et al., 2007) in both cases. It should be emphasized that although we retrieve acetone from a small microwindow (1216.75 to 1217.5 cm^{-1}), Fig. 3 shows that this still improves the residual fit over the wider 1215 to 1230 cm^{-1} range, particularly where there is an enhancement of acetone (Fig. 3a).

Figure 4 shows the global-mean distribution of acetone at an average pressure of 277 hPa and 185 hPa, respectively, interpolated onto a 20° longitude by 10° latitude grid. What we find at both levels is a distinct hemispheric difference between the higher volume mixing ratios (vmrs) in the Northern Hemisphere (NH) compared to lower vmrs

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in the Southern Hemisphere (SH). Although not shown, we have tested whether the variability may be due to a latitudinal variation in the pointing of the MIPAS–E but we find no significant change in retrieved pressure with latitude. At 277 hPa, the highest average acetone vmrs are found over Greenland and Western Siberia (between 1.6 and 2.0 ppb). As Greenland is unlikely to be a major source of acetone, due to the lack of vegetation, it is likely the acetone has originated from North America. Between Central Africa and South America, we see acetone vmrs ranging between 1.0 and 1.4 ppb. The Southern Hemisphere vmrs are generally below 600 ppt in the monthly average suggesting there are few sources, and suggesting that if the ocean were a likely source of acetone, then the contribution to the global atmospheric budget is small. At 185 hPa, the inter-hemispheric differences are still clear, although the maximum vmrs are below 600 ppt.

In conjunction with ECMWF analyses of potential temperature and potential vorticity, we show the monthly average zonal distribution of acetone in the upper troposphere and lowermost stratosphere (Fig. 5). The hemispheric gradient of acetone is pronounced between 200 and 500 hPa, with acetone vmrs between 2.5 and 3 ppb poleward of 50° N. The tropics show a slight enhancement of acetone in the average, with vmrs ranging between 500 and 1500 ppt, but there is no evidence of acetone vmrs exceeding 500 ppt in the tropical upper troposphere between 200 hPa and the 380 K theta surface. The data also suggest some mixing of air between the 2 PVU surface and the 380 K level (as referred to by Holton et al. (1995) as the middleworld), with little penetration of the overworld (theta greater than 380 K).

Analysing the data at the 9 km nominal altitude level (an average pressure of 277 hPa), there is a marked decrease in 5-day average acetone data poleward of 45° N (Fig. 6) with a difference of over 300 ppt between the start and end of August 2003. This may be due to a decrease in biogenic activity over the course of the month. There is a much lower variability in the tropics and poleward of 45° S over the month. It is interesting to note that, in these data, tropical vmrs are consistently lower than NH vmrs by between 300 and 900 ppt in August 2003.

4 Conclusions

Global distributions of acetone (C_3H_6O) in the upper troposphere and lowermost stratosphere (6–15 km altitude range, approximately 500 to 150 hPa pressure range) have been obtained from MIPAS–E spectral measurements during August 2003. The vertical cross-section of acetone has been determined with a strong Northern Hemisphere and Southern Hemisphere contrast with acetone volume mixing ratios (vmrs) of between 2.5 and 3 ppb poleward of $50^\circ N$ and between 300 hPa and 500 hPa. The high vmrs extend to $90^\circ N$ suggesting that there is a high input from mid-latitude emissions (both biogenic and some biomass burning) during the August period. Northern Hemisphere vmrs of acetone in the UT decrease steadily over the month, suggesting that a decrease in biogenic activity coupled with the destruction of acetone is observable in the data. The concentration of acetone in the tropics varies by less than 150 ppt over August 2003, ranging between 700 and 850 ppt on average. Southern Hemisphere vmrs remain steady between 400 and 500 ppt over August.

These new, global, measurements of acetone will likely help in the modelling of chemical processes in the upper troposphere giving a new insight into chemical processes in the upper troposphere. It is likely to enhance knowledge in particular of Arctic chemical processes where in-situ observations are sparse. Although these data are limited by high cloud-occurrence frequency in the tropics, the data show acetone enhancements linked to biomass burning regions over Central Africa and Southern America. A likely progression of this study is to measure the seasonal cycle of acetone in the upper troposphere and compare these data to a chemical transport model.

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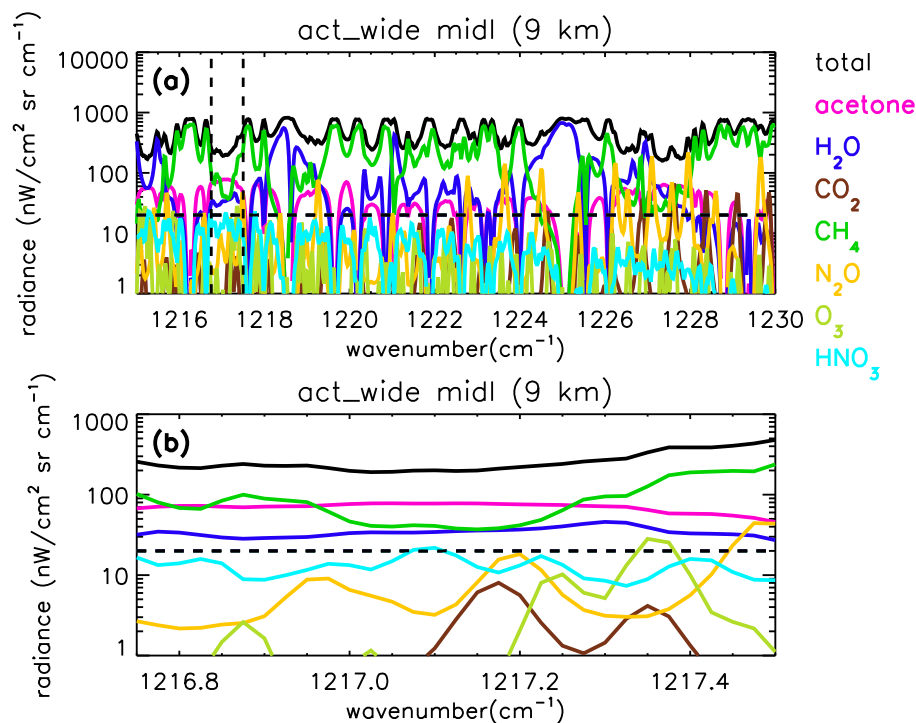


Fig. 1. Plot of modelled radiance contributions for the radiatively active gases in **(a)** the 1215 to 1230 cm^{-1} range and **(b)** 1216.75 to 1217.5 cm^{-1} in the mid-latitudes (20 to 65°). The dotted line represents the noise equivalent spectral radiance (NESR) of $20 \text{ nW}/(\text{cm}^2 \text{sr cm}^{-1})$ as reported in MIPAS–E band–B by Kleinert et al. (2007).

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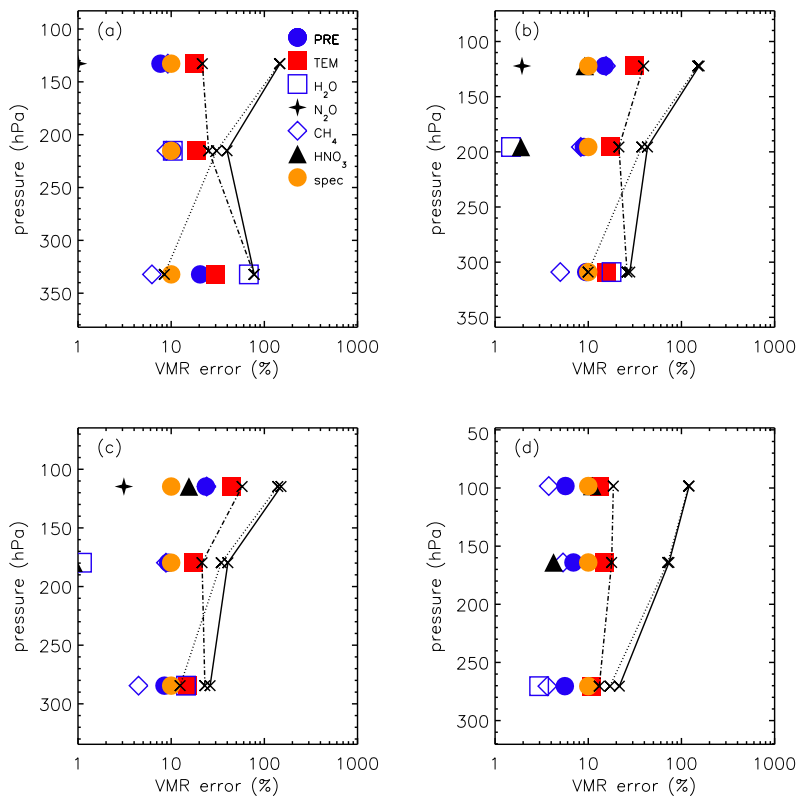


Fig. 2. Error budget for acetone vmr retrievals. The data have been split into regimes, **(a)** 20° S to 20° N – tropical, **(b)** 20 to 65° – mid-latitudes, **(c)** 65 to 90° in the summer hemisphere – Polar summer, **(d)** 65 to 90° in the winter hemisphere – Polar winter. The solid black line represents the total error on a single retrieval. The random (dotted) and systematic (dashed) component of the error are also shown. Systematic errors are dominated by pressure (“PRE”) and temperature (“TEM”), acetone spectroscopy (“SPEC”), and gain uncertainties.

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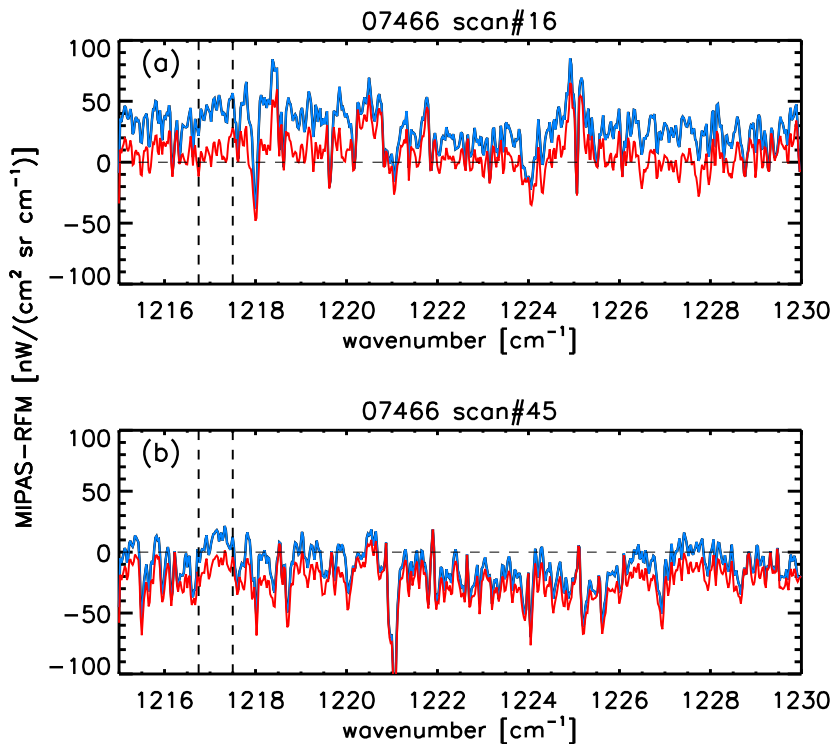


Fig. 3. Examples of MIPAS–E radiance – RFM modelled radiance residuals for orbit 07466 on 4 August 2003 at the nominal 12 km altitude level. Scenes were chosen which represent the typical behaviour of **(a)** enhanced acetone vmr (743 ppt in this scene). Case **(b)** represents acetone levels around the median vmr for August 2003 of 240 ppt (this scene has a vmr of 259 ppt). The blue line is the fit in both cases without considering retrieved acetone and the red line is the fit including retrieved acetone (and continuum). The vertical black dashed lines show the acetone microwindow used. The co-ordinates of the MIPAS–E measurements shown are (a) 9° S, 98° W, (b) 47° N, 93° E.

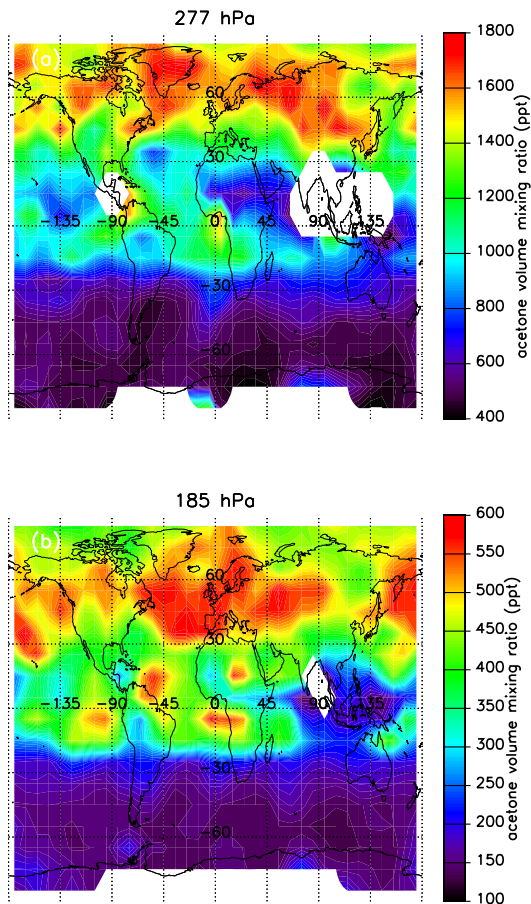


Fig. 4. Global acetone distributions (in units of ppt) at average pressure levels of **(a)** 277 hPa and **(b)** 185 hPa for the month of August 2003. White areas are regions where the data are wholly cloud-contaminated for the month.

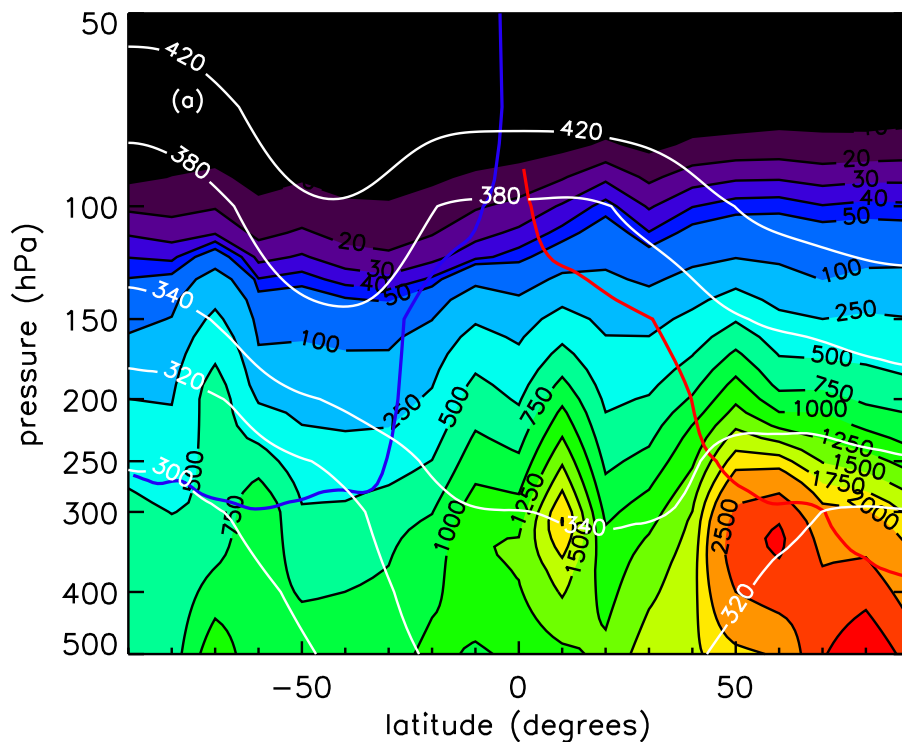


Fig. 5. The zonal acetone distribution of acetone (units of ppt) as a function of pressure for August 2003. The white lines indicate potential temperature (θ) levels in Kelvin and the blue and red lines indicate the 2 PVU potential vorticity levels which are a method of representing the monthly-average tropopause level in the data.

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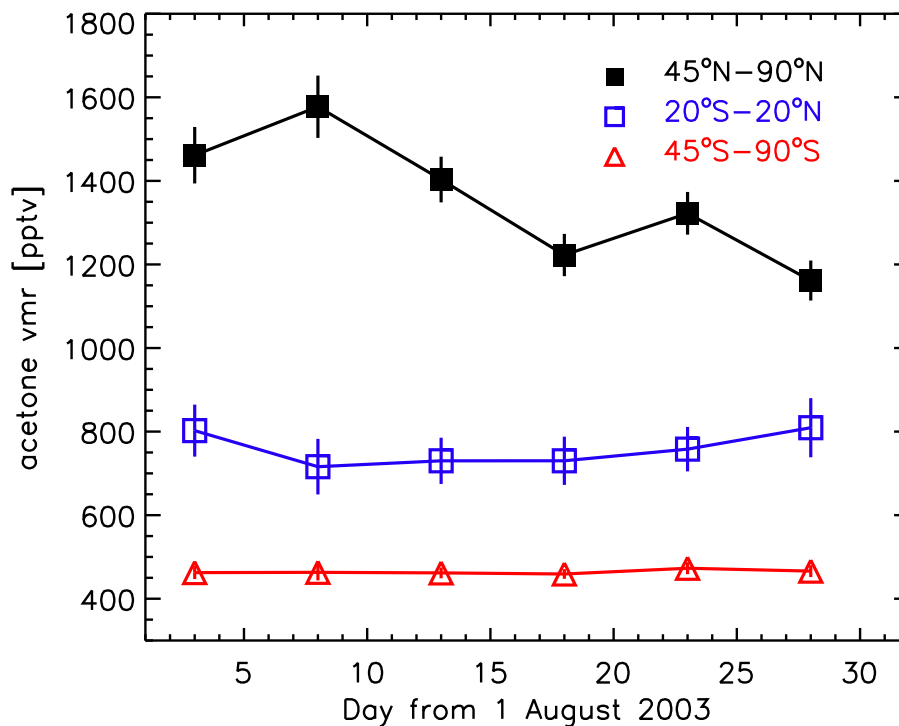


Fig. 6. Daily-average acetone volume mixing ratios for three regions at an average pressure level of 277 hPa; **(a)** 45° N to 90° N, **(b)** 20° S to 20° N and **(c)** 45° S to 90° S.

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