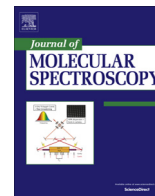




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Seasonal variations of acetone in the upper troposphere–lower stratosphere of the northern midlatitudes as observed by ACE-FTS

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ABSTRACT

This study reports on the climatological acetone distribution and seasonal variations in the upper troposphere and lower stratosphere of the northern midlatitudes, derived from observations by the Atmospheric Chemistry Experiment Fourier Transform Spectrometer (ACE-FTS) onboard SCISAT. The acetone profiles retrieved from 5 to ~20 km cover the period from January 2004 to September 2010. The 1σ statistical fitting errors are typically ~5–20% within the upper troposphere (UT), increasing in the lower stratosphere (LS) with decreasing acetone. The systematic errors range between 15% and 20%. The largest UT acetone mixing ratios (~1200 ppt on average in July over Siberia) are observed in summer in the northern mid- and high latitudes. Mixing ratios are larger over continental regions than over the ocean. Comparisons with airborne measurements available in the literature point toward a possible underestimation in acetone retrieved from ACE-FTS. The largest differences occur primarily in winter and for the background values. This underestimation is attributed to the complexity of the spectral region used for the retrieval. The annual cycle of acetone for the 30–70°N midlatitude band shows a maximum during summer, reflecting the annual cycle of the primary terrestrial biogenic source of acetone. By comparison with ACE-FTS, the LMDz-INCA global climate-chemistry model systematically overestimates acetone mixing ratios lower than 400 ppt. This overestimation is thus generalized for the lower stratosphere, the Tropics and beyond 70°N for the upper troposphere. In contrast, in the upper troposphere of the 30–70°N region, where the acetone levels are the highest (>450 ppt on average), the model–observation differences are in the range of the observation uncertainty. However, in this region, the model fails to capture the annual cycle of acetone, culminating in July. A seasonal cycle can only be obtained by considering high biogenic emissions but this cycle is shifted toward autumn, likely indicating an underestimation of the chemical destruction in the northern high latitudes.

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1. Introduction

Acetone (CH_3COCH_3) is one of the most abundant oxygenated volatile organic compounds (OVOCs) in the troposphere [1,2]. Its impact on the oxidative capacity of the dry extratropical upper troposphere, as a HOx source, was thought to be one of the most important until mid-2000s, e.g. [2–6]. Its major contribution in the upper troposphere (UT) and the lowermost stratosphere (LMS) has been confirmed, especially in summer and autumn. It has been recently quantified based on the CARIBIC experiment

(Civil Aircraft for the Regular Investigation of the atmosphere Based on an Instrument Container) and the model ECHAM5/MESy [7], assuming the recently revised acetone photolysis quantum yield [8,9].

Large uncertainties remain in the acetone budget [10]. The main sources of acetone are primary terrestrial biogenic and biomass burning emissions, secondary chemical production, and a small contribution from the decay of dead plant matter. The ocean is either a source or a sink depending on microbial activity, light and temperature [10–12]. Photolysis, oxidation and dry deposition over land and ocean are the main sinks of acetone [1,10,13]. Many of the acetone budget studies have been constrained using data from aircraft field campaigns compiled by Emmons et al. [14].

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However, given the variability of acetone and the scarcity of campaigns, the dataset is not representative of a complete acetone climatology. More recently, the availability of the CARIBIC aircraft measurements has improved our knowledge of the description of the acetone distribution in the UT/LMS (lowermost stratosphere), especially in the northern midlatitude band between 35°N and 56°N [7,15]. Space-borne instruments are powerful tools to complement in situ measurements since they provide global distributions of the atmospheric composition, especially those of VOCs, e.g. [16–22]. Retrieving acetone from remote-sensing instruments is very challenging because: (i) acetone absorbs in the infrared in a particularly dense spectral region, (ii) the acetone absorption cross-sections are weak and (iii) the concentration of acetone in the atmosphere is rather low (0.2–3 ppb, e.g. [10]). First retrievals from infrared spectrometers were performed from balloon-borne instruments [23] and from the spaceborne ACE-FTS in biomass burning plumes [19]. The first global distribution has been retrieved from MIPAS for one month in August 2003 [21].

In this study, we present a recent acetone ACE-FTS research product based on ACE processing version 3.0 [24]. The principal objective of the study is to evaluate the capability of this new ACE-FTS acetone product to describe the global distribution of acetone in the UTLS as well as the seasonal variations and the transport occurring in this region. Comparisons with measurements available from the literature, and in particular CARIBIC observations in the UTLS [15], are used for this purpose. Comparisons with the state-of-the-art global chemical transport model LMDz-INCA [25] are also discussed. The acetone profile retrievals are described and evaluated in Section 2. The model is described in Section 3. Sections 4 and 5 discuss the global acetone distribution in the UTLS and its annual cycle as observed by ACE-FTS and simulated by LMDz-INCA. Section 6 summarizes the conclusions of the study.

2. ACE-FTS observations

2.1. Retrieval of acetone from ACE-FTS spectra

The ACE-FTS instrument, which covers the spectral region 750–4400 cm^{-1} with a ± 25 cm maximum optical path difference, uses the sun as a light source to record limb transmission through the

Earth's atmosphere during sunrise and sunset ('solar occultation'). Transmittance spectra are obtained by ratioing against exoatmospheric 'high sun' spectra measured each orbit. These spectra, with high signal-to-noise ratios, are recorded through long atmospheric limb paths (~ 300 km effective length), thus providing a low detection threshold for trace species. ACE has an excellent vertical resolution of about 2–3 km in the troposphere and can measure up to 30 occultations per day, with each occultation sampling the atmosphere from 150 km down to the cloud tops (or 5 km in the absence of clouds). The locations of ACE occultations are dictated by the low Earth circular orbit of the SCISAT satellite and the relative position of the sun. Over the course of a year, the ACE-FTS records atmospheric spectra over a large portion of the globe [26].

Version 3.0 of the ACE-FTS retrieval software was used for the acetone volume mixing ratio (VMR) retrievals. Vertical profiles of trace gases (along with temperature and pressure) are derived from the recorded transmittance spectra via a nonlinear least squares global fit to the selected spectral region(s) for all measurements within the altitude range of interest. The microwindow set consists of 16 microwindow 'slices' across the acetone spectral feature, with a common set of fitted baseline parameters (a baseline scaling factor and a baseline slope); all microwindows are listed in Table 1 and an example of fit is given in Fig. 1. An additional four microwindows are utilized to improve the retrieval of various interferers; see the footnotes to Table 1. The 16 'slices' lie between CH_4 and H_2O lines and were chosen to avoid CH_4 line mixing effects. The atmospheric pressure and temperature profiles, and the tangent heights of the measurements were taken from the v3.0 processing of the ACE-FTS data. The abundances of most molecules with absorption features in the microwindow (see Table 2) were adjusted simultaneously with the acetone VMR, except for CFC-12, CFC-114, CFC-115, HCFC-22, HFC-125, CBrF₃, CFC-13 and HFC-143a. Contributions from CFC-12 and HCFC-22 were calculated directly using the v3.0 VMRs, with assumed constant VMR profiles that did not vary with time for the others. Spectroscopic line parameters and absorption cross sections for most molecules were taken from the HITRAN 2004 database [27], with the exception of CHF₃ [28] and CBrF₃ (PNNL; <http://nwir.pnl.gov>), water vapor and ozone [29]. The acetone cross-section dataset, recently included in HITRAN 2012 [30], was obtained from Harrison et al.

Table 1
Summary of the microwindows used in the acetone retrieval.

Center frequency (cm^{-1})	Microwindow width (cm^{-1})	Lower altitude (km)	Upper altitude (km)
1184.65	0.22	$7 - 2\sin^2(\text{latitude}^\circ)$	22
1189.38	0.28	$7 - 2\sin^2(\text{latitude}^\circ)$	22
1191.72	0.24	$7 - 2\sin^2(\text{latitude}^\circ)$	22
1194.50	0.50	$7 - 2\sin^2(\text{latitude}^\circ)$	22
1204.59	0.70	$7 - 2\sin^2(\text{latitude}^\circ)$	22
1206.85	0.50	$7 - 2\sin^2(\text{latitude}^\circ)$	22
1208.25	0.30	$7 - 2\sin^2(\text{latitude}^\circ)$	22
1214.00	0.40	$7 - 2\sin^2(\text{latitude}^\circ)$	22
1217.00	0.35	$7 - 2\sin^2(\text{latitude}^\circ)$	22
1227.85	0.30	$7 - 2\sin^2(\text{latitude}^\circ)$	22
1231.25	0.70	$7 - 2\sin^2(\text{latitude}^\circ)$	22
1232.10	0.40	$7 - 2\sin^2(\text{latitude}^\circ)$	22
1200.40	0.40	$8 - 2\sin^2(\text{latitude}^\circ)$	22
1222.25	0.30	$10 - 2\sin^2(\text{latitude}^\circ)$	22
1233.95	0.30	$9 - 2\sin^2(\text{latitude}^\circ)$	22
1241.53	0.26	$9 - 2\sin^2(\text{latitude}^\circ)$	22
1937.15 ^a	0.70	8	21
1950.10 ^b	0.35	$10 - 2\sin^2(\text{latitude}^\circ)$	22
2566.22 ^c	0.26	9	22
2623.87 ^d	0.90	$8 - 2\sin^2(\text{latitude}^\circ)$	22

^a Included to improve results for interferers H_2^{18}O and H_2^{17}O .

^b Included to improve results for interferer H_2O .

^c Included to improve results for interferers N^{15}NO .

^d Included to improve results for interferers HDO , OC^{18}O , and CH_4 .

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