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The Atmospheric Chemistry Experiment Fourier transform spectrometer (ACE-FTS) version 4.1 retrievals: Trends and seasonal distributions ☆

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ABSTRACT

The Atmospheric Chemistry Experiment (ACE) satellite measures infrared transmission spectra of the atmosphere with a Fourier Transform Spectrometer (FTS) using the Sun as a light source. ACE provides a global view of atmospheric composition from altitude profiles of volume mixing ratios of 44 molecules starting in February 2004. The current version of ACE-FTS processing is 4.1 released in July 2020. Compared to v.4.0, the trends and altitude-latitude distributions have changed only slightly. Quarterly altitude-latitude distributions have been computed to highlight seasonal effects. Generally, the tropospheric volume mixing ratios of v.4.1 agree well with surface measurements made by the NOAA (National Oceanic and Atmospheric Administration) flask network and the AGAGE network. The revised ACE trends provide a quantitative state-of-the-atmosphere report.

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1. Introduction

The ACE-FTS instrument records atmospheric transmission spectra in the infrared using the Sun as a light source (solar occultation) [1]. Processing versions 4.0 and 4.1 provide altitude volume mixing ratio profiles (VMRs) of 44 molecules plus 24 isotopologues [2]. Very recently the global trend values for VMRs of 44 ACE molecules (H_2O, O_3, N_2O, NO, NO_2, HNO_3, N_2O_5, H_2O_2, HO_2NO_2, O_2, N_2, SO_2, HCl, HF, ClO, $ClONO_2$, CFC-11, CFC-12, CFC-113, COF_2, $COCl_2$, COFCl, CF_4, SF_6, CH_3Cl, CCl_4, HCFC-22, HCFC-141b, HCFC-142b, HFC-134a, HFC-23, CO, CH_4, CH_3OH, H_2CO, HCOOH, C_2H_2, C_2H_6, OCS, HCN, $CH_3C(O)CH_3$, CH_3CN, PAN ($CH_3C(O)OONO_2$), and low altitude CO_2) were calculated for 16 years using v.4.0 [3]; in addition, altitude-latitude VMR distributions were plotted for the duration of the mission.

In July 2020, v.4.1 was released which improved on v.4.0, for example, by recovering additional occultations. The most significant change, however, was improvement in the low altitude retrievals, for example, by including the effects of wavenumber variation of aerosol and cloud extinction during analysis of the N_2 continuum [2], which changed the retrieved tangent heights. This had a small effect on the trends but changed the VMR values for many tropospheric molecules. In this note, we have therefore updated the trend values reported previously [3] and made explicit comparisons of v.4.1 ACE VMRs with those published by NOAA and AGAGE [4]. The global altitude-latitude distribution figures are very similar for v.4.1 and v.4.0 so we have not reproduced them but instead have provided the underlying v.4.1 digital data for the altitude-latitude distributions for the overall mission as well as for each quarter to show seasonal effects.

2. Methods

The ACE-FTS data used are the v4.1 Level 2 ACE data products [2] available via signup online (https://databace.scisat.ca/l2signup.php). The same analysis method was used as described by Bernath et al. [3] and will not be repeated here. The new product presented in this paper is the average altitude-latitude distribution calculated for each quarter: December, January, February (DJF); March, April, May (MAM); June, July, August (JJA); and September, October, November (SON). A single figure with 4 quarters is provided for each molecule for the entire mission (March 2004 – November 2019) as well as the underlying digital data.

The ACE orbit concentrates occultation measurements at high latitudes [1]. Thus, trend calculations involving averages over a particular latitude range will implicitly be weighted toward higher latitudes within that range, as a consequence of larger number of data points contributing to the average.

☆ This paper is dedicated in memory of Michael Mishchenko.

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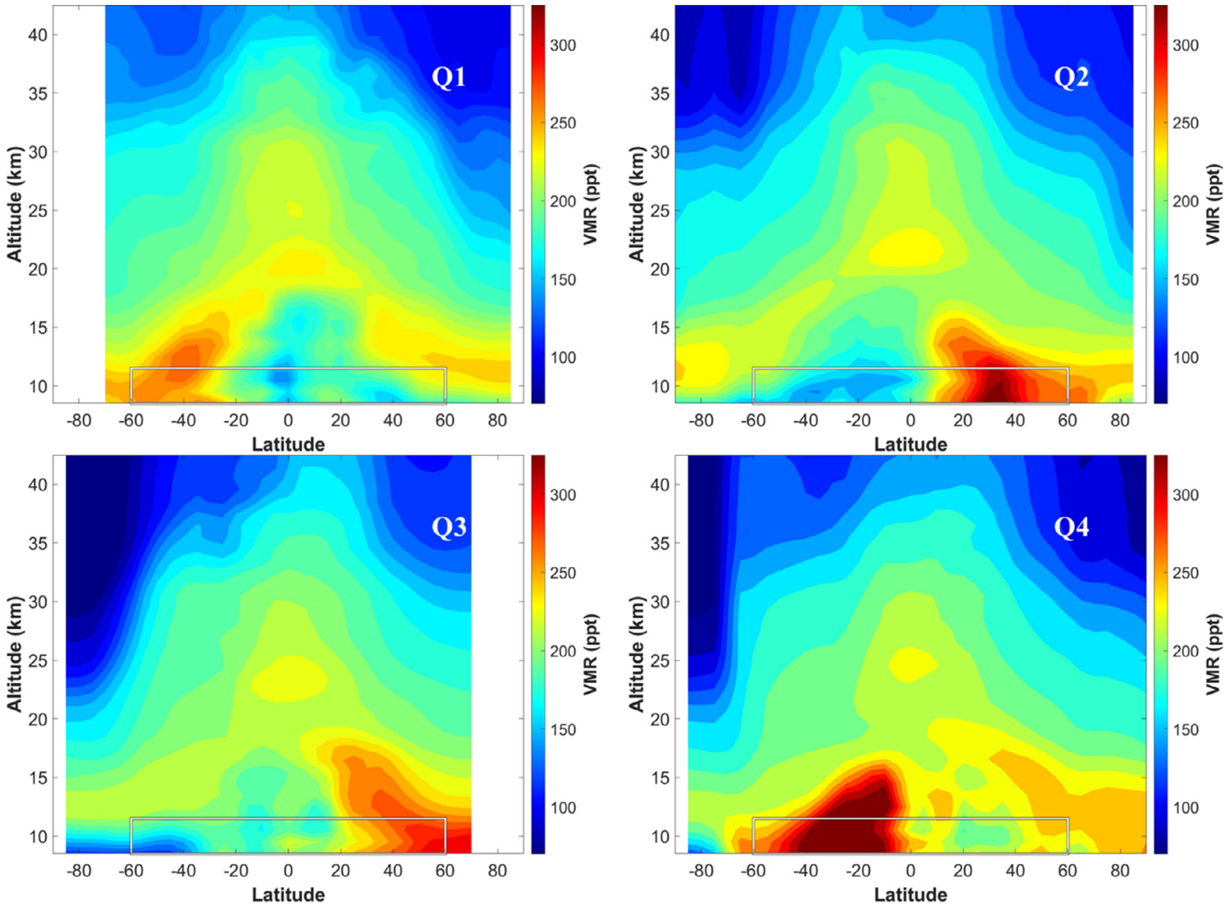

Fig. 1. Mission average quarterly altitude-latitude distributions for HCN for DJF (Q1), MAM (Q2), JJA (Q3) and SON (Q4).

3. Results and discussion

The ACE-FTS trend values for 44 molecules, summarized in Table 1 in Bernath et al. [3], have changed slightly for v.4.1. The revised values for this table are presented in Table 1. The revised trend plots and the underlying digital values for v.4.1 are provided as supplementary data.

Many ACE molecules also display seasonal variations, or more accurately quarterly variations because of the ACE sampling pattern [1]. The mission average altitude-latitude data was divided into 4 quarters: Q1, December, January, February (DJF); Q2, March, April, May (MAM); Q3, June, July, August (JJA); Q4, September, October, November (SON). Fig. 1 shows the quarterly altitude-latitude distribution plot for HCN with the rectangular box indicating the region that was used for the trend analysis. The regions chosen for trend analysis are the same as for v4.0 [1] and the regions selected are provided in the supplementary data plots.

HCN is produced almost entirely by fires so the seasonal changes in Fig. 1 are due to the changing locations of biomass burning. In DJF (Q1), fires are typically in Australia, in MAM (Q2) at mid-latitudes in the Northern hemisphere, in JJA (Q3) in the boreal forests and in SON (Q4) in the Amazon and Central Africa. The figures and digital data for these quarterly plots are provided as supplementary data for all 44 molecules.

As described by Boone et al. [2,5], v.4.0/4.1 processing uses temperature and pressure for 5-18 km in altitude from the Canadian weather service model and determines the tangent height of each measurement from the N_2 collision-induced absorption (CIA). In v.4.1, a fitted baseline slope parameter was included in each N_2 CIA microwindow to allow for the wavenumber variation of aerosol/cloud extinction. This (and other changes) altered the tangent heights for v.4.1 relative to v.4.0, which in turn changed the VMRs of tropospheric molecules because air density is an exponential function of altitude. A comparison (Table 2) of a set of ACE v.4.1/v.4.0 VMRs has been made with NOAA and AGAGE global values reported for 2016 in the 2018 Ozone Assessment [4]. The 2016 ACE-FTS values (and one standard deviation statistical errors) were obtained from the linear trend lines, substituting 2016.5 for the year for v.4.1/v.4.0. For two of the molecules (CHF_3 and CF_4), AGAGE (Advanced Global Atmospheric Gases) values are quoted [4] because no NOAA values were listed. For N_2 and O_2, the values are from Gatley et al. [6]. Generally, the agreement is good for v.4.1: within about 5% except for those molecules that display anomalous altitude-latitude distributions (HCFC-141b, HCFC-142b, CHF_3, CF_4) because of spectroscopic interference [3]. In most cases the v.4.1 VMR values are in better agreement than v.4.0 values with NOAA/AGAGE values (Table 2). The atmospheric regions used to determine the ACE trend values are given in the v.4.1 trend plots provided as supplementary data.

The 2018 Ozone Assessment [4] contains annual change values obtained by taking the difference in VMRs from 2015 to 2016. The NOAA annual change values (except for two gases noted above) are also reported in Table 2 along with the ACE v.4.1 multiyear trend values extracted from Table 1. Agreement between NOAA and AGAGE global *in situ* annual change values and ACE remote sensing trends in the upper troposphere is generally good; notable exceptions are HCFC-141b, CH_3Cl and OCS. Notice that in spite of biases, the ACE-FTS v.4.0/4.1 trend values almost always agree well with NOAA and AGAGE annual change values.

Table 1
Linear trend values for the 44 ACE-FTS molecules in %/year and ppt/year for v.4.1. One standard deviation in the last digits is quoted in parentheses. For those molecules for which two linear trends were used, the most recent trend is in the table and the earlier trend is given in a footnote.

Molecule	Trend, %/year (error)	Trend, ppt/year (error)	Molecule	Trend, %/year (error)	Trend, ppt/year (error)
COClF	-0.85(15)	-0.377(59)	HCN	-0.14(30)	-0.35(70)
H_2O	0.250(30)	13.3(1.5) x10^3	HCOOH	-0.70(28)	-0.32(11)
H_2O_2	0.23(24)	0.47(48)	HF	0.868(45)	17.52(79)
HCFC-22	1.723(56)[a]	4.079(94)[b]	HFC-134a	6.86(22)[c]	5.708(70)[d]
HCFC-141b	0.59(22)[e]	0.169(55)[f]	HNO_3	0.61(20)	50(13)
HCFC-142b	1.06(17)[g]	0.168(21)[h]	HO_2NO_2	0.24(10)	0.54(23)
HCl	-0.297(30)[i]	-7.70(72)[j]	N_2	0.0038(35)	30(28) x10^6
N_2O	0.2852(93)	938(29)	PAN	-13.3(5.4)[k]	-6.06(58)[l]
N_2O_5	0.22(17)	2.3(1.7)	CFC-11	-0.531(25)[m]	-1.225(51)[n]
NO	0.13(16)	10(12)	COF_2	0.35(11)	1.00(28)
NO_2	0.01(11)	0.4(5.2)	$ClONO_2$	-0.55(10)	-4.59(77)
O_2	0.0059(60)	13(13) x10^6	CO_2	0.5484(52)	2206(19) x10^3
O_3	0.095(54)	7.2(4.0) x10^3	C_2H_6	1.34(31)	8.9(1.6)
OCS	-1.44(15)[o]	-6.40(47)[p]	H_2CO	0.19(19)	0.18(17)
$(CH_3)_2CO$	2.31(37)	8.14(94)	C_2H_2	-1.12(38)	-0.81(23)
CCl_4	-1.231(75)	-0.992(50)	CF_4	1.057(20)[q]	0.870(13)[r]
CFC-12	-0.608(18)[s]	-3.156(83)[t]	CFC-113	-1.387(33)	-1.005(19)
CH_3Cl	0.168(40)	1.03(24)	CH_3CN	-0.23(16)	-0.62(40)
CH_3OH	-0.07(19)	-0.4(1.1)	CH_4	0.346(17)[u]	6.26(29)[v] x10^3
CHF_3	3.450(83)[w]	0.840(13)[x]	ClO	0.35(43)	0.82(94)
CO	-0.47(16)	-0.34(11) x10^3	$COCl_2$	-0.63(17)	-0.072(18)
SF_6	3.829(27)[y]	0.3272(23)[z]	SO_2	-0.68(46)	-0.059(36)

[a] 3.854(78) [b] 7.30(11) [c] 10.55(27) [d] 4.520(60) [e] 1.56(26) [f] 0.378(52) [g] 7.18(36) [h] 0.844(26) [i] -0.535(46) [j] -14.3(1.2) [k] -3.1(1.4) [l] -2.06(75) [m] -0.718(28) [n] -1.749(64) [o] 0.505(34) [p] 2.29(15) [q] 0.888(7) [r] 0.682(5) [s] -0.244(25) [t] -1.31(13) [u] 0.331(23) [v] 5.81(39) x10^3 [w] 3.82(18) [x] 0.707(20) [y] 4.437(95) [z] 0.2719(42)

Table 2
Comparison of average ACE-FTS v4.0/v.4.1 tropospheric VMRs and trends for 2016 with NOAA/AGAGE VMRs and annual change values [4].

Molecule	v.4.0 VMRs 2016	v.4.1 VMRs 2016	NOAA & AGAGE 2016	v.4.1 Bias	v.4.1 trends	NOAA & AGAGE 2016
CFC-11	310.4 ± 1.1 ppt	229.9 ± 1.3 ppt	229.8 ppt	-0.04 %	-1.23 ± 0.05 ppt/year	-1.3 ppt/year
CFC-12	498 ± 2 ppt	517 ± 2 ppt	512.2 ppt	-0.94 %	-3.16 ± 0.08 ppt/year	-3.1 ppt/year
CFC-113	64.7 ± 0.4 ppt	67.9 ± 0.4 ppt	71.5 ppt	5.03 %	-1.01 ± 0.02 ppt/year	-0.6 ppt/year
HCFC-22	230 ± 2 ppt	239 ± 2 ppt	237.5 ppt	-0.63 %	4.08 ± 0.09 ppt/year	4.5 ppt/year
HCFC-141b	27 ± 1 ppt	29 ± 1 ppt	24.53 ppt	-18.22 %	0.17 ± 0.06 ppt/year	0.31 ppt/year
HCFC-142b	16 ± 0.4 ppt	16 ± 0.5 ppt	22.01 ppt	27.31 %	0.17 ± 0.02 ppt/year	0.17 ppt/year
CH_3Cl	607 ± 5 ppt	622 ± 5 ppt	559.1 ppt	-11.25 %	1.0 ± 0.2 ppt/year	9.1 ppt/year
CCl_4	100 ± 1 ppt	76 ± 1 ppt	81.2 ppt	6.4 %	-0.99 ± 0.05 ppt/year	-1.0 ppt/year
SF_6	8.43 ± 0.15 ppt	8.71 ± 0.05 ppt	8.9 ppt	2.13 %	0.327 ± 0.002 ppt/year	0.3 ppt/year
N_2O	315.9 ± 0.5 ppb	333.1 ± 0.6 ppb	328.9 ppb	-1.28 %	938 ± 29 ppt/year	800 ppt/year
CH_4	1706 ± 7 ppb	1812 ± 7 ppb	1843 ppb	1.68 %	6.3 ± 0.3 ppb/year	9 ppb/year
CHF_3	23.5 ± 0.3 ppt	24.8 ± 0.2 ppt	28.9 ppt[a]	14.29 %	0.84 ± 0.01 ppt/year	0.8 ppt/year[a]
HFC-134a	84 ± 3 ppt	86 ± 2 ppt	89.6 ppt	4.02 %	5.71 ± 0.07 ppt/year	6.1 ppt/year
CO_2	400.6 ± 0.8 ppm	412.1 ± 0.4 ppm	402.5 ppm	-2.4 %	2.21 ± 0.02 ppm/year	2.24 ppm/year[b]
N_2	78.63 ± 0.09 %	79.48 ± 0.09 %	78.08 %	-1.79 %	-	-
O_2	21.37 ± 0.02 %	21.58 ± 0.02 %	20.94 %	-3.06 %	-	-
CF_4	81.9 ± 0.3 ppt	74.0 ± 0.2 ppt	82.7 ppt[a]	10.52 %	0.87 ± 0.01 ppt/year	0.8 ppt/year[a]
OCS	444 ± 11 ppt	466 ± 13 ppt	505 ppt	7.7%	2.3 ± 0.2 ppt/year	6 ppt/year

[a] Values from AGAGE network.
[b] From ref. [3].

4. Conclusions

The ACE 2004-2020 trends in atmospheric composition highlight changes and provide a quantitative "state-of-the-atmosphere" report (Table 1). The v.4.0 and v.4.1 trend values are similar and improve substantially on the v.3.5/3.6 values (which are not reliable). The VMRs of tropospheric gases changed from v.4.0 to v.4.1 and are now generally in good agreement with independent measurements. ACE quarterly altitude-latitude distributions show seasonal variations as illustrated by HCN.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

CRediT authorship contribution statement

P.F. Bernath: Methodology, Supervision, Writing - original draft. **J. Crouse:** Formal analysis, Visualization. **R.C. Hughes:** Formal analysis. **C.D. Boone:** Writing - review & editing.

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Supplementary materials

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.jqsrt.2020.107409.

References

[1] Bernath PF. Atmospheric Chemistry Experiment (ACE). J Quant Spectrosc Rad Transf 2017;186:3–16.
[2] Boone CD, Bernath PF, Cok D, Steffen J, Jones SC. Version 4 retrievals for Atmospheric Chemistry Experiment Fourier transform spectrometer (ACE-FTS) and imagers. J Quant Spectrosc Rad Transf 2020;247:106939.
[3] Bernath PF, Steffen J, Crouse J, Boone CD. Sixteen-year trends in atmospheric trace gases from orbit. J Quant Spectrosc Rad Transf 2020;253:107178.
[4] Engel A, Rigby M, Burkholder JB, Fernandez RP, Froidevaux L, Hall BD, Hossaini R, Saito T, Vollmer MK, Yao B. Update on ozone-depleting substances (ODSs) and other gases of interest to the Montreal Protocol, Chapter 1. In: Montzka SA, Velders GJM, Krummel PB, Mühle J, Orkin VL, Park S, Shah N, Walter-Terrinoni H, editors. Hydrofluorocarbons (HFCs), Chapter 2 in WMO (World Meteorological Organization), Scientific Assessment of Ozone Depletion: 2018. Global Ozone Research and Monitoring; 2018. p. 588. Project-Report No. 58.
[5] Boone CD, Bernath PF. Tangent height determination from the N_2-continuum for Atmospheric Chemistry Experiment Fourier transform spectrometer. J Quant Spectrosc Rad Transf 2019;238:106481.
[6] Gatley DP, Herrmann S, Kretzschmar H-J. A twenty-first century molar mass for dry air. HVAC&R Res 2008;14:655–62.