Optimizing tropospheric retrievals at Eureka

Camille Viatte and Kimberly Strong

Department of Physics, University of Toronto, Toronto, ON, Canada

Context

✓ Several studies have identified pollution transport pathways to the Arctic on the basis of model simulations [1], but our ability to verify these pathways through chemical observations has been limited. Also, our ability to predict trace gas concentrations and variability in models, such as degradation mechanisms of NMVOC's [2], are poorly understood and have yet to be satisfactorily quantified.

✓ We present five years of observations of seven tropospheric species (CO, HCN, C_2H_6, C_2H_2, CH_3OH, HCOOH, and H_2CO) measured using a ground-based FTIR (Bruker IFS 125HR) at the Polar Environment Atmospheric Research Laboratory (PEARL), located at Eureka, Nunavut, Canada (80N, 86W). Total columns of CH_3OH, HCOOH showed here are retrieved for the first time from ground-based FTIR measurements in the High Arctic region.

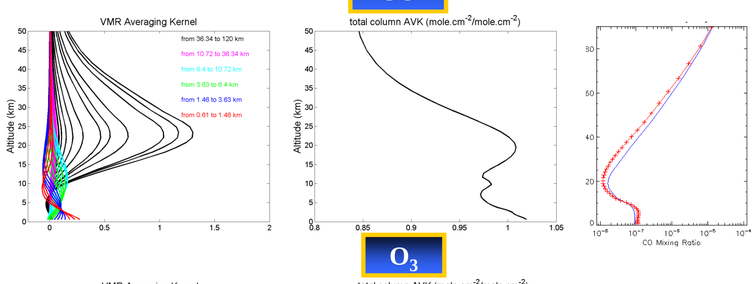
✓ All retrievals have been performed with the SFIT2 [3] v3.94 algorithm (with the HITRAN 2008 spectral linelist), based on the Optimal Estimation Method formalism of Rodgers [4].

Tropospheric species retrievals

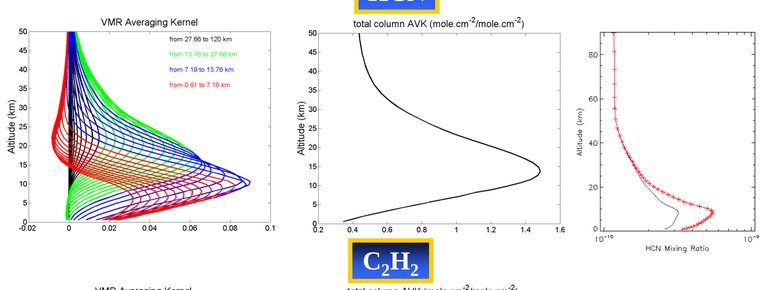
Target species	micro-windows	interfering species	a priori vmr	Sa diagonal value	S:N
CO	2057.684-2058.00, 2069.560-2069.760, 2157.507-2159.144	O_3, CO_2, OCS, H_2O, N_2O	WACCM V6	0.2	200
HCN	3268.000-3268.380, 3287.000-3287.480	H_2O	WACCM V6	0.5	200
C_2H_6	2976.660-2976.950, 2983.200-2983.550, 2986.500-2986.950	H_2O, O_3	WACCM V4V5	0.3	250
O_3	1000.00-1005.00	H_2O, CO_2, O_3^{676}, O_3^{667}, O_3^{686}, O_3^{668}, C_2H_4	WACCM V5	0.3 and larger at O_3layer	100
C_2H_2	3250.430-3250.770, 3255.180-3255.725, 3304.825-3305.350	H_2O, HDO	GC Toon Kiruna991203 Mk4-flight 6-34km, outside spitprim.set divided by 2	0.5	200
CH_3OH	992.000-998.700, 1029.000-1037.000	O_3, O_3^{686}, O_3^{668}, O_3^{676}, O_3^{667}, H_2O, CO_2	WACCM V6	0.2	200
HCOOH	1104.650-1105.600	HDO, O_3, O_3^{668}, O_3^{676}, H_2O, NH_3, CCL_2F_2, CHF_2CL, CH_4	WACCM V6	1	800
H_2CO	2778.120-2778.800, 2780.600-2781.170	CH_4, CO_2, O_3, N_2O	WACCM V6 divided by 2	1	500

Error budget (%)	CO	HCN	C_2H_6	O_3tot	O_3trop	C_2H_2	CH_3OH	HCOOH	H_2CO
measurement error	**0.2**	**4.3**	**1.2**	**0.2**	**5.6**	**7**	**4.6**	**16.3**	**10**
smoothing error	**0.1**	**5.9**	**12.2**	**0.4**	**23.3**	**16.7**	**3**	**1.5**	**5.7**
uncertainty on temperature	0.7	0.4	0.3	0.9	3.1	2.9	4.4	2.5	5.5
uncertainty on lineintensity	1.2	4.9	6.4	8.8	21.3	5.7	10	3.6	2.4
uncertainty on linewidth	0.2	3.7	2.4	1.6	13.8	7.7	1.1	0.9	2.1
uncertainty on retrieval parameters	0.1	4.3	2.6	0	0.1	8.2	0	0	1.1
uncertainty on interf. species	0	0.2	0	0	0.2	0.1	0	0.3	24.1
uncertainty on SZA	0.3	0.3	0.3	0.2	0.5	0.7	0.2	0	0.2
total random error	**0.8**	**6.1**	**2.9**	**1**	**6.7**	**11.6**	**6.3**	**16.5**	**26.7**
total random and spectral error	1.5	8.7	7.4	9	28	15.1	11.9	17	26.9
total error	**1.5**	**10.5**	**14.3**	**9**	**37.5**	**22.5**	**12.3**	**17**	**27.5**
N spectra total	3895	1815	1819	1984		1269	1095	1973	1242
DOFS	**2.6**	**1.2**	**1.9**	**6.2**	**2**	**1**	**1.1**	**0.8**	**0.7**

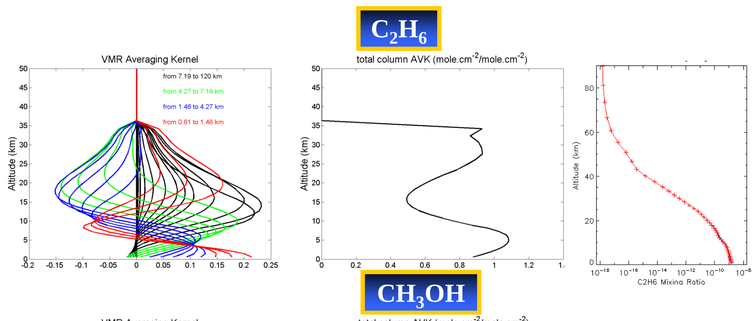
CO



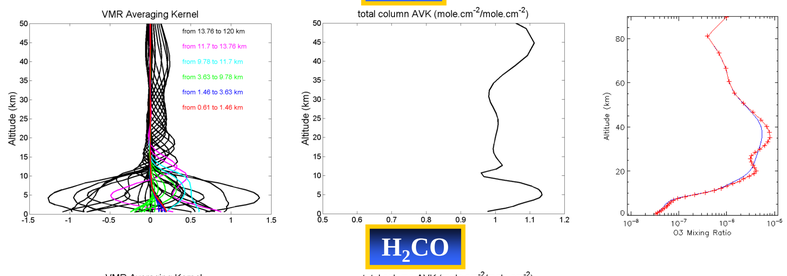
HCN



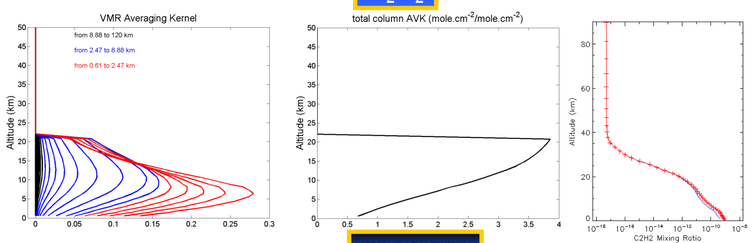
C_2H_6



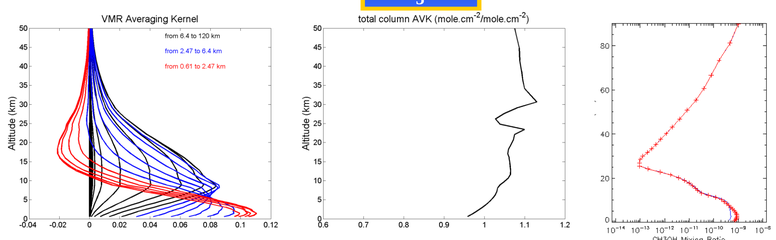
O_3



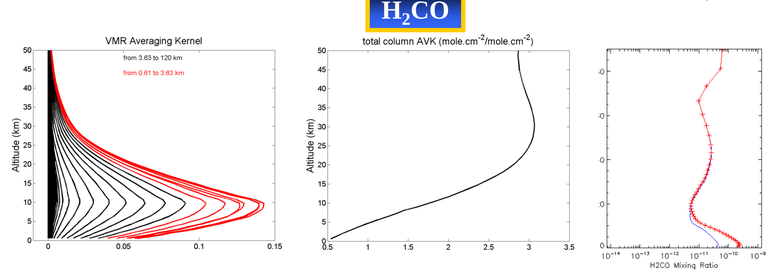
C_2H_2



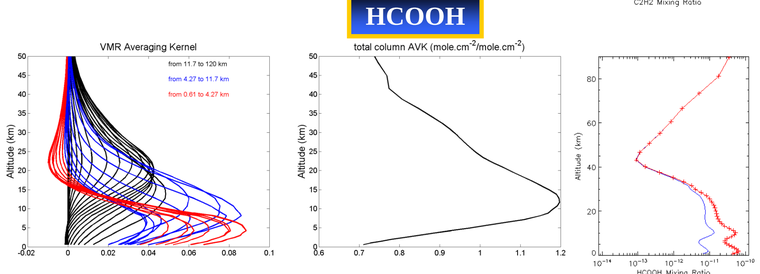
CH_3OH



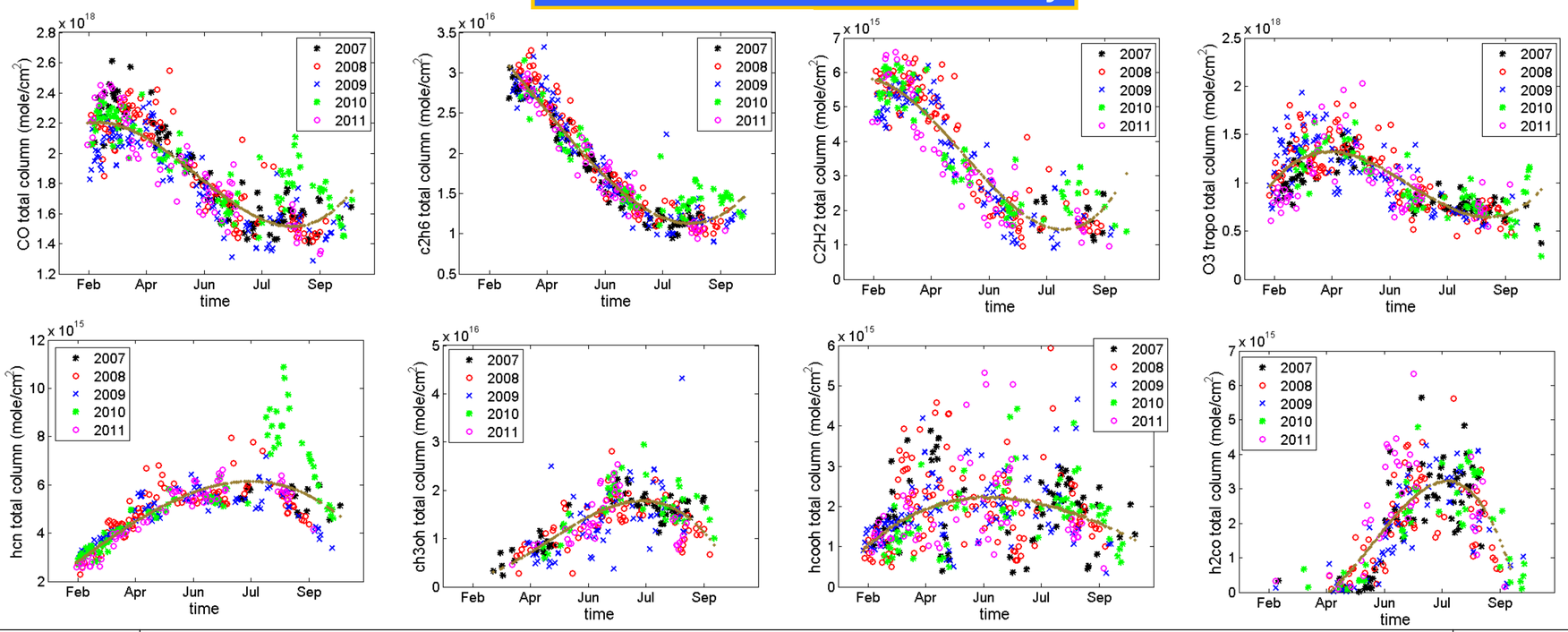
H_2CO



HCOOH



Results : seasonal variability



Monthly mean total columns	February	March	April	May	June	July	August	September	October	Mean	STD
CO x10^{18}	1.83	2.20 (0.14)	2.17(0.14)	1.97(0.14)	1.69(0.10)	1.59(0.17)	1.63(0.17)	1.56(0.14)	1.56(0.08)	1.85	0.31
HCN x10^{15}	2.78(0.15)	3.37(0.48)	4.58(0.59)	5.39(0.54)	5.58(0.34)	5.76(0.72)	6.63(1.60)	5.28(1.10)	4.69(0.58)	4.97	1.29
C_2H_6 x10^{16}		2.85(0.14)	2.66(0.28)	2.07(0.21)	1.55(0.15)	1.29(0.24)	1.20(0.17)	1.23(0.18)	1.24(0.03)	1.77	0.65
C_2H_2 x10^{15}	4.57	5.50(0.53)	5.00(0.79)	3.31(0.74)	2.12(0.58)	1.87(0.71)	1.94(0.59)	1.61(0.04)	1.38	3.95	1.69
CH_3OH x10^{16}		0.42(0.20)	0.74(0.19)	1.08(0.45)	1.57(0.80)	1.82(0.39)	1.53(0.49)	1.36(0.33)		1.49	0.59
HCOOH x10^{15}	1.05(0.25)	1.43(0.64)	2.23(0.93)	1.99(1.05)	2.20(0.86)	1.87(1.05)	2.22(0.88)	1.58(0.72)	1.14(0.47)	1.94	0.90
H_2CO x10^{15}		0.36(0.22)	0.27(0.20)	0.59(0.63)	2.42(1.00)	3.14(1.02)	2.81(0.95)	1.13(0.77)	0.55(0.38)	2.30	1.28

Conclusions

✓ Our retrievals constitute a new dataset of tropospheric species that can be used to constrain and improve model simulations of chemical and dynamical processes in the high Arctic.

✓ Seasonal variabilities in the Arctic of CO, C_2H_6, and C_2H_2 are driven by the OH reaction and long-range transport.

✓ Seasonal variabilities in the Arctic of HCN, CH_3OH, HCOOH and H_2CO are driven by biogenic emission and short-range transport.

Future work

✓ Study tropospheric O_3 production (or destruction) in extreme events. Because VOC's are important tropospheric O_3 precursors, they can affect the regional and global tropospheric O_3 budget.

✓ Compare our data with the GEOS-Chem model to quantify the possible underestimations (or overestimations) in order to improve our understanding of the chemical and dynamical processes in the Arctic region.

✓ Study biomass burning events, since all of these molecules are released by fires, in order to derive emission ratios that are key parameters used in chemical transport models for emissions.

References

[1] Shindell et al.: A multi-model assessment of pollution transport to the Arctic, Atmos. Chem. Phys., 8, 5353–5372, 2008

[2] Stavrakou et al.: Global emissions of non-methane hydrocarbons deduced from SCIAMACHY formaldehyde columns through 2003-2006, Atmos. Chem. Phys., 9, 8431–8446, 2009

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[4] Rodgers: Inverse Methods for Atmospheric Sounding : Theory and Practice, 200 pp., World Sci., Hackensack, N. J., 2000

Acknowledgements

PEARL/CANDAC: ARIF, AIF/NSRIT, CFCAS, CFI, CSA, EC, GOC-IPY, INAC, NSERC, NSTP, OIT, ORF, PCSP, SEARCH

ACE Arctic Campaigns: CSA, EC, NSERC, NSTP

Logistical and operational support at Eureka:

The wonderful team at the EC Weather Station, CANDAC/PEARL PI James R Drummond, PEARL site manager Pierre Fogal, The CANDAC operators, and Rebecca Batchelor, and Rodica Lindenmaier.