Observation and simulation of ethane (C_2H_6) at 23 FTIR sites

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BACKGROUND INFORMATION ON ETHANE

-- Ethane is the most abundant non-methane hydrocarbon (NMHC) in the Earth atmosphere. Its main sources are of anthropogenic origin, with globally 62% from leakage during production and transport of natural gas, 20% from biofuel consumption and 18% from biomass burning. In the Southern hemisphere, anthropogenic emissions are lower which makes biomass burning emissions a more significant source. The main removal process is oxidation by the hydroxyl radical (OH), leading to a mean atmospheric lifetime of two months [1].

-- Until recently, a prolonged decrease of its abundance has been documented, at rates of -1 to -2.7%/yr, with global emissions dropping form 14 to 11 Tg/yr over 1984-2010; the decrease was attributed to a reduction in fugitive emissions from the fossil fuel sector [2].

-- However, subsequent investigations have reported an upturn in the ethane trend, characterized by a sharp rise from about 2009 onwards. The ethane increase is attributed to the oil and natural gas production boom in North America [3], although significant changes in OH could also be at play [4, 5].

DEDICATED MODEL SIMULATIONS WITH EMAC

The model simulations used here have been produced with the EMAC 3D Chemistry Climate Model (3D-CCM ECHAM5/MESSy Atmospheric Chemistry), driven by ECMWF analysis data on a ~1.8° x 1.8° horizontal resolution [6].

Four full chemistry simulations were performed, implementing the Master Chemical Mechanism (MCM) scheme and various emission *scenarii*:

-- a first one using the RCP85 emissions as is (Representative Concentration Pathway database v8.5);
-- the **BASE-CASE** simulation, using again the RCP85 emissions, but after scaling by 1.5 the C2-C5 NMHC emissions above 20°N;
-- the **TREND** simulation was obtained by further adding to the BASE-CASE set up a trend in US emissions;
--the **BB-FLAT** simulation repeated the TREND run, but using recurring Biomass Burning (BB) emissions from 2009 onwards;
-- all simulations covered the 2009-2015 time period, but the 2009 data which correspond to the spin-up time period were not used in the present trend investigations;
-- model outputs have been smoothed with FTIR seasonal averaging kernels, but without noticeable impact given the good sensitivity of the retrievals to the whole troposphere and lower stratosphere altitude range.

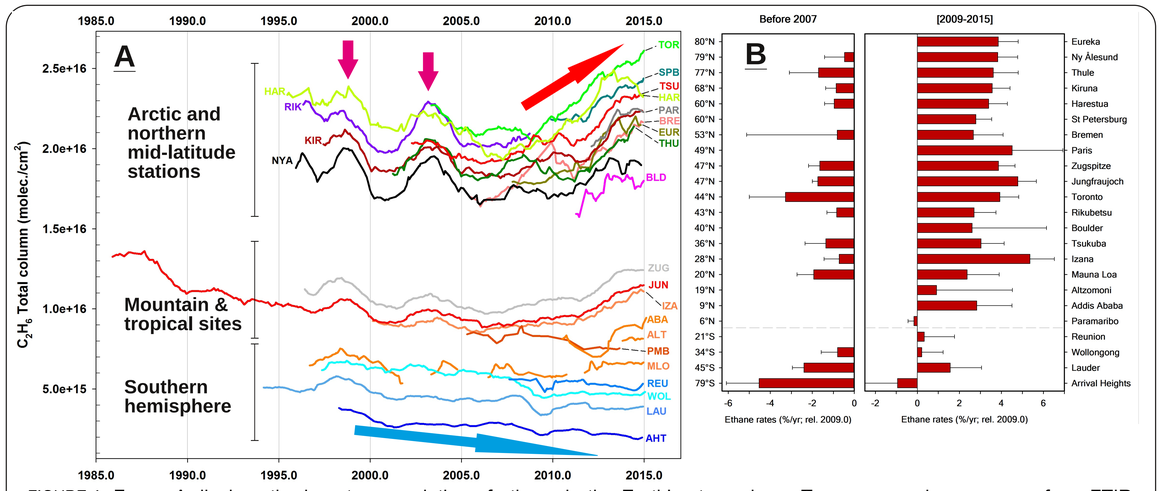

FIGURE 1. Frame A displays the long-term evolution of ethane in the Earth's atmosphere. Two-year running averages from FTIR time series are shown here. The well-known latitudinal gradient is observed [2], with maximum total columns at Arctic and Northern mid-latitude stations, and much less abundance in the Southern hemisphere (SH). Another obvious feature is the C_2H_6 increase in the Northern hemisphere (NH), from about 2006-2009, contrasting with a monotonous decrease in the SH. Two vertical arrows denote local maxima resulting from intense boreal fires (see e.g., [7]). Frame B shows, as a function of latitude, the C_2H_6 relative rates of change derived for two time periods, namely when considering all data available before 2007 (left frame), or between 2009 and 2015 (right frame). The error bars correspond to 2-sigma statistical uncertainty intervals accounting for auto-correlation [8].

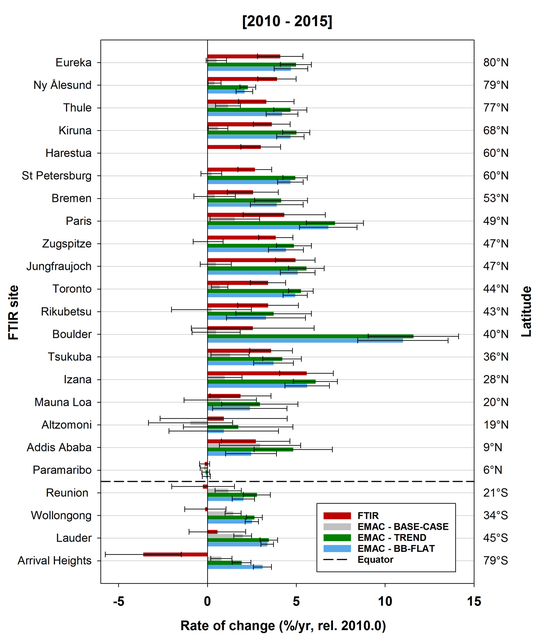

FIGURE 3. OBSERVED AND MODELED TRENDS FOR [2010-2015]

-- Underestimation of the NMHC emissions in the main inventories is confirmed for RCP85. A scaling by 1.5 was needed (>20°N) to match the NH C_2H_6 abundances. The same order of magnitude was determined in previous studies (x2 for HTAP2; see [10]; x1.4 for NEI; see [11]).

-- It is clear from FIGURE 3 that the BASE-CASE EMAC simulation is unable to capture the C_2H_6 trends of 3-5%/yr observed in the NH, while we notice a statistical agreement for the tropical and SH rates of changes (the Arrival Heights Antarctic station being a notable exception).

-- Accounting for additional emissions associated with the oil & gas sector, evaluated at ~7 Tg over [2009-2015] and localized in North America, allows the EMAC-TREND simulation to produce adequate rates of change for the NH.

-- In the latter case however, the SH trends are too large when compared to the observations. An additional run repeating the 2009 BB emissions until 2015 was performed to check that uneven BB emissions are not responsible for the SH trend. But the BB-FLAT simulation provides SH trends commensurate with the EMAC-TREND simulation, confirming the difficulty for the model to represent the situation prevailing in the SH.

FIGURE 2 - NDACC FTIR sites location.

NETWORKS & SITES, INSTRUMENTATION AND TOOLS

-- Very high resolution (up to 0.003 cm^{-1}) and signal-to-noise ratios (>1000) infrared solar spectra are recorded year-round, at a suite of ground-based stations affiliated with the NDACC and/or the TCCON network. See frame B of **Figure 1** for the station names and latitudes and **Figure 2** for a map of the current official NDACC sites.

-- Fourier Transform InfraRed (FTIR) spectrometers are operated at these stations under clear-sky conditions. They are equipped with cooled detectors and cover the infrared region of the electromagnetic spectrum, encompassing the ethane features near 2980 cm^{-1}.

-- The retrievals are performed either with the SFIT4 (v0.9.4.4) or the PROFFIT algorithms which are based on the semi-empirical implementation of the Optimal Estimation Method of Rodgers [9]. Two independent pieces of information are retrieved in most cases, allowing the monitoring of the ethane evolution in both the troposphere and lower stratosphere.

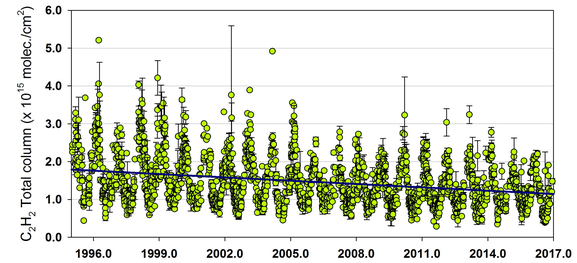

FIGURE 4. Recent studies [4, 5] invoke changes/drops in the OH sink as a possible cause for the observed rise in methane after ~2007, with implications also for C_2H_6 and other alkanes, in particular when evaluating the magnitude of emissions needed to capture the C_2H_6 increase. Within this framework, acetylene (C_2H_2) is another relevant indicator since its main sink is, as for C_2H_6, oxidation by OH, while it has dissimilar sources (biofuel and fossil fuel combustion are the main contributors, before biomass burning). The daily mean time series of C_2H_2 derived from the FTIR monitoring program at the Jungfraujoch reproduced above does not show any trend upturn, with a rate of change after 2009 remaining statistically consistent (at 2-sigma) with the one derived for the whole interval (i.e., -1.6±0.2 %/yr, rel. to 1995.0). Based on this evidence, a drastic change in OH over the recent years is unlikely.

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