Understanding organic tracers in the troposphere using ACE and GEOS-Chem

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1. Introduction
This poster is comparing data obtained with ACE-FTS (Atmospheric Chemistry Experiment – Fourier Transform Spectrometer) and GEOS-Chem for ethane, ethene and carbon monoxide. Comparisons results in model improvements and developments to better understand the complex behaviour of these molecules in the free troposphere. The ACE-FTS (http://www.ace.uwaterloo.ca) is the main instrument on board the Canadian satellite SCISAT-1 working primarily in solar occultation with a resolution of 0.02 cm⁻¹ in the 2.2 to 13.3 μm (750 – 4400 cm⁻¹) spectral range (Bernath et al., 2005). The instrument provides altitude profile information for volume mixing ratios (VMRs) of trace gases between 85°N and 85°S. The ACE orbit (650 km, 74° inclination) gives near global coverage whilst the solar occultation technique lets us obtain high signal to noise ratios spectra. The retrieval of VMRs is done using a global fit algorithm (Boone et al., 2005). GEOS-Chem (http://acmg.seas.harvard.edu/geos/) is a global 3-D Chemical Transport Model able to simulate trace gases and aerosol distributions in the troposphere (Bey et al., 2001). The model is driven by assimilated meteorological observations from the Goddard Earth Observing System (GEOS). Emissions, sinks, chemistry and photochemistry are all evaluated to produce a realistic simulation of the atmosphere composition.

Figure 1 shows the ACE-FTS spectral region used for the retrieval of ethane and the ethene model emissions corrected after the successful comparison between the model and the satellite data.

Fig. 1. Left panel. Spectral region used in the retrieval of C₂H₆ at 2976.5 cm⁻¹. Looking at the residuals we can see how the inclusion of C₂H₆ in the forward model improves the retrieval.

Right panel shows the 2005 C₂H₆, GEOS-Chem simulated emissions. Following comparison of the simulated concentrations with the ACE data the model emissions mechanism was modified. It was found that the high concentration values obtained for the southern hemisphere were linked to abnormal high emissions in southern tropical latitudes over Africa.

2. Results
Figure 2 shows the comparison between satellite data and the model simulations. Reported ACE VMRs belong to the version 3.0 dataset whilst the GEOS-Chem version used for this study is the v8-02-03 with the dicarbonyl chemical package to account for the ethyne chemistry. The simulated concentrations of carbon monoxide in the northern hemisphere are small in comparison with the retrieved ACE CO VMRs. This has been found in comparisons with other satellite data sets like TES or MOPITT by Kopacz et al. (2010) leading them to the production of a new CO emissions budget that can explain better the observed concentrations. Ethene and ethyne retrieved by ACE have been compared to the GEOS-Chem output. The hemispheric asymmetry linked to the anthropogenic emissions as well as the seasonal variation is well captured by both the model and the satellite data for all three molecules. Global emissions budgets of C₂H₂ and C₂H₆ for the year 2005 have been computed using the satellite data to constrain the emissions. We have obtained a budget of 12 Tg C per year for C₂H₂, and 5 Tg C per year for C₂H₆.

Fig. 2. Top row CO middle row C₂H₂ and lower row C₂H₆ concentrations from ACE (left column), GEOS-Chem (central column) and difference between the ACE data and the model simulation (right column). The global bias between the model and the satellite data is -12% for CO, -7% for C₂H₂ and 1% for C₂H₆. The bias has been defined following the equation:

\[ \text{Bias} = \frac{\text{Model mean} - \text{Satellite mean}}{\text{Satellite mean}} \times 100 \]

Looking at the long term trends for these species using ACE data we have only looked at the period 2004-2007 but ACE data record runs from 2004 till today. We can conclude that the free troposphere concentration of ethane and ethyne is not showing any important interannual trend. We can therefore conclude (given the relatively short lifetime of these molecules in the atmosphere, 2 months for C₂H₂ and 2-4 weeks for C₂H₆) that the anthropogenic amounts injected to the atmosphere each year are remaining fairly constant. Figure 3 illustrates this situation for C₂H₆.

Fig. 3. Long term C₂H₆ concentrations do not show any significant trend. Due to the relative short lifetime of 2 months the model finds difficulties simulating the changes in concentrations produced by random episodes like biomass burning. Despite of this situation the general trend of the simulated data is in good agreement with the satellite.

3. Conclusions
We have developed a new ACE dataset for C₂H₂ and C₂H₆. We have compared these new data sets and the old CO dataset with the GEOS-Chem model to indentify possible issues in the model or the satellite data. The model was over estimating the CO concentration in the northern hemisphere as well as the C₂H₂ in the southern hemisphere. This leads us to the calculation of new global emissions budgets for C₂H₂ (12 Tg C) and C₂H₆ (5 Tg C) for the year 2005.

References