QUANTITATIVE ANALYSIS OF SCIAMACHY CO VARIABILITY AND ITS IMPLICATION FOR CHEMISTRY TRANSPORT MODELS

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ABSTRACT

Carbon Monoxide is an important atmospheric trace gas. It plays a key role in the global OH budget and thus in the cleansing capacity of the atmosphere and is often used as a tracer for pollutant transport. The satellite instrument SCIAMACHY has been measuring CO total columns for several years now, allowing to study its inter and intra-annual variability. We present a quantitative and systematic analysis of SCIAMACHY CO total column measurements for the years 2003 and 2004. SCIAMACHY CO retrievals are hampered by the presence of an ice layer on the detector. However, a detailed correction scheme has been included in the retrieval algorithm, resulting in CO total columns with a precision of 1% for monthly means under ideal circumstances (cloud free pixels, high surface albedo, spatial averaging). For lower surface albedos a precision <10% is obtained. Thus, SCIAMACHY CO total column measurements are of sufficient quality to provide useful new information. Comparisons with a chemistry-transport model simulation show similar spatial patterns for the global distribution of modeled and measured CO. Quantitative comparisons of modeled and measured seasonal variations show a good agreement for very different types of seasonal cycles. Differences do occur but can be attributed to an inaccurate representation of model emissions as is e.g. confirmed by recent updates of biomass burning emission data bases.

INTRODUCTION

Carbon Monoxide (CO) is an important trace gas in the chemistry of the atmosphere. It enables the production of tropospheric ozone and the destruction of the hydroxyl radical, the “cleaning agent” of the atmosphere. It also affects the air quality in the boundary layer. Besides industrial emissions, a substantial but poorly known source of CO is biomass burning. The latter source is very variable throughout seasons, from year to year, and per location. This variability leads to strong temporal and spatial changes in the tropospheric CO concentration. Since the lifetime of CO ranges from several weeks to several months, CO is an excellent tracer of atmospheric transport processes. The SCanning Imaging Absorption spectroMeter for Atmospheric CartograpHY (SCIAMACHY) on board the ENVISAT satellite allows to construct global maps of CO. Global CO maps are also available from thermal infrared measurements by the MOPITT instrument on board the EOS-TERRA satellite (e.g., Drummond and Mand, 1996; Deeter et al., 2003), the Tropospheric Emission Spectrometer (TES) instrument on board EOS-Aura (e.g. Beer et al., 2001), and the Interferometric Monitoring of Greenhouse Gases (IMG) instrument on board the ADEOS satellite (Clerbaux et al., 2003; Barret et al., 2005). All three instruments measure CO in the thermal infrared, with good sensitivity in the free troposphere but in principle no sensitivity in the boundary layer in contrast with SCIAMACHY, which measures in the near-infrared.

Ground-based FTIR measurements provide high quality total column measurements but have very limited spatial coverage [Dils et al., 2005]. Validation with FTIR measurements often consists of comparison with SCIAMACHY CO measurements within a range of several hundreds to thousands of kilometers [Sussman and Buchwitz, 2005; Dils et al., 2005]. As a result, such a validation of SCIAMACHY CO is based on very few truly collocated measurements. In addition, some FTIR stations are located on top of mountains which – due to their small spatial footprint – are also difficult to compare with SCIAMACHY CO total column measurements which have a footprint of 30×120 km^2. Other complications can be low surface albedo, which enhances SCIAMACHY noise errors and clouds, which reduce collocation possibilities [Sussman and Buchwitz, 2005]. Only qualitative comparisons have been done so far using visual identification of spatial patterns correlating SCIAMACHY with MOPITT CO total column measurements or with MODIS fire count maps [Buchwitz et al., 2004, 2005; Frankenberg et al., 2005; de Beek et al., 2006].

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Therefore, de Laat et al. (2006) have compared the retrieved SCIAMACHY CO total columns quantitatively to the atmospheric chemistry model TM4 (Dentener et al., 2003), which provides useful information on global features and seasonality. This paper summarizes the work done by de Laat et al. (2006).

Retrieval and calibration

The CO total columns are retrieved from near-infrared spectra measured by SCIAMACHY between 2324.5 and 2337.9 nm. The near-infrared retrievals have proven more complex due to many instrument/calibration issues as described in Gloudemans et al. (2005). The most important problems are the presence of an unexpected ice layer on the detectors, which varies in time and the increasing number of dead detector pixels (Kleipool et al., 2005). Their effects have been reduced by applying dedicated in-flight decontamination procedures and additional in-flight calibration measurements, as well as improvements to the calibration (Lichtenberg et al., 2005). In addition, in-house knowledge of the instrument and calibration has allowed to derive accurate corrections for the ice layer which are included in the retrieval algorithm. The results presented here are derived with the Iterative Maximum Likelihood Method (IMLM) developed at SRON (Schrijver, 1999; Gloudemans et al., 2005). Results from earlier versions of the IMLM retrieval algorithm have been presented in Straume et al. (2005) and Gloudemans et al. (2005). The current version 6.3 is presented in de Laat et al. (2006) and contains two major improvements as well as some smaller ones: the 2004 version of the HITRAN spectroscopic database has been incorporated in the retrievals (Rothman et al., 2005), and ECMWF data have been used. The quality of the current CO product has been assessed quantitatively in de Laat et al. (2006) through comparisons with chemistry transport simulations.

Model description

The global chemistry-transport model TM4 [Meirink et al., 2005] used for this study is a follow-up of TM3 [Dentener et al., 2003, and references therein]. Anthropogenic CO emissions are based on Van Aardenne et al. [2001], while natural emissions are as in Houweling et al. [1998]. The seasonality of CO biomass emissions is based on climatological emission estimates (Hao and Liu, 1994).

De Laat et al. (2006) show that in general, average CO concentrations as well as long term (monthly) and short term (deseasonalized) variability seen in in-situ measurements are adequately reproduced by the model. The model results can thus be used as a powerful tool to evaluate SCIAMACHY CO total columns.

Filtering of the data and calculation of monthly averages

The SCIAMACHY CO total column errors due to random instrument noise for single measurements are quite large - typically 10-100 %. They depend on the signal level of the SCIAMACHY spectra and are thus related to variations in surface albedo and solar zenith angle. The instrument noise error is calculated using an instrument model which includes pre- and in-flight measurements to deal with contributions of Johnson noise, photoelectron noise, and detector read-out noise.

Due to the large noise errors of the individual SCIAMACHY CO total columns, multiple single column measurements need to be averaged to obtain information on CO with a useful precision. A common method to average measurements with different errors is to use weighted means. The weight of each measurement is taken inversely proportional to the square of the errors:

$$X_{wav} = \frac{\sum w_i x_i}{\sum w_i} \quad in \quad which \quad w_i = \frac{1}{\sigma_i^{2}}$$

With X_{wav} the weighted average, x_i a single column measurement, σ_i the error corresponding to measurement x_i and w_i the weight of this measurement. The error of the weighted mean (X_{wav}) is expressed as:

$$\sigma_{wav} = \frac{1}{\sqrt{\sum w_i}}$$

This method is applied to the retrieved SCIAMACHY CO columns - as well as to collocated TM4 model values – for both spatial and temporal averaging using the instrument-noise errors.

The measurements need to be filtered to remove a number of biases. Only pixels with a cloud cover of less than 20% are used. This cloud cover has been determined from the number of cloud-free polarization measurements (7×30 km) within one SCIAMACHY observation (60-240×30 km) using the SPICI algorithm [Krijger et al., 2005]. CO column measurements for very low surface albedos (< 0.05; some locations over land and all above sea) – and thus very low signal-to-noise - turn out to be often unrealistically high with large noise errors (> 1.5 10^{18} molecules/cm^2) and are removed.

The retrieved CO columns are combined on a monthly 3°×2° longitude-latitude grid, which corresponds to the TM4 model resolution. Because of low land surface albedos, frequent high cloud cover, possible errors at large solar zenith angles at high latitudes, the analysis is restricted to the latitude range of 60°S to 60°N.

Results

Figure 1 shows the global distribution of SCIAMACHY CO averaged over one year of data from September 2003 through August 2004. The corresponding TM4 model results, sampled in the same way as the SCIAMACHY CO data, are also shown.

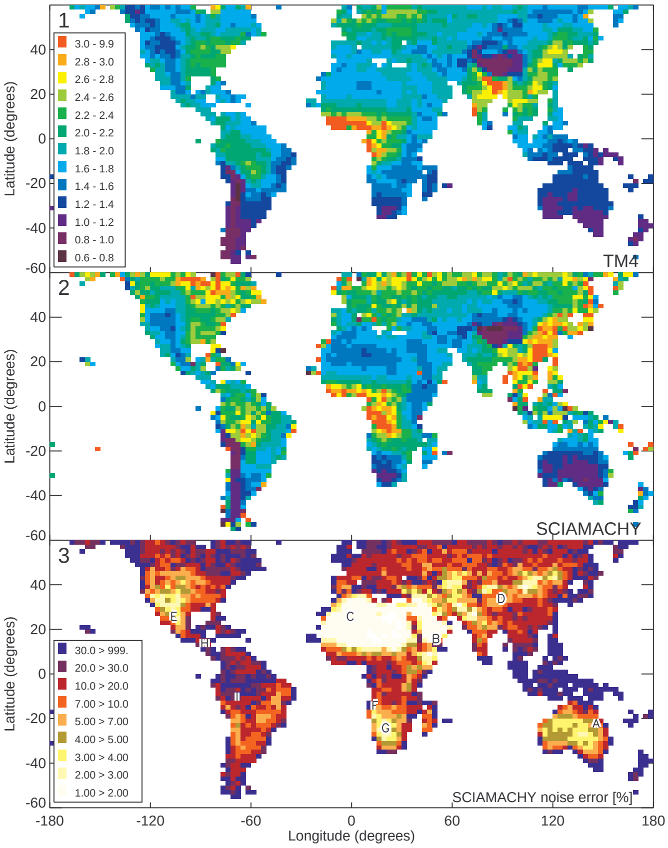

Fig. 1: Annual mean CO total columns (10^{18} molecules/cm^2; September 2003 – August 2004) on 2°×3° resolution for TM4 (panel 1) and SCIAMACHY (panel 2). Panel 3 shows the instrument-noise related SCIAMACHY errors (as % of the annual mean TM4 CO total column value). Indicated in panel 3 are also the locations used in Figure 2. From: de Laat et al. (2006).

A number of similarities between the SCIAMACHY CO total columns and the model can be seen. CO columns are lower over southern South America, southern Africa and over Australia compared to northern mid-latitudes. Emission regions like Amazonia, equatorial Africa (biomass burning), South and Southeast Asia, Europe and North America (anthropogenic) can clearly be discerned, as expected. CO columns are lower over the mountainous regions.

Discrepancies are also visible: SCIAMACHY CO columns are higher than the TM4 model results at northern mid-latitudes, over East Asia, Amazonia, Africa southeast of the equator and over Indonesia. On the other hand, SCIAMACHY CO is lower over India and also over northern Africa (10°-30°N) compared to the model. Furthermore, the spatial variability in CO columns over low surface albedo regions (high latitudes, tropics) is higher in the SCIAMACHY CO total columns than in the model results.

The bottom panel of Fig. 1 shows the corresponding random instrument noise error of the annual mean SCIAMACHY CO. Clearly small errors are obtained for arid regions like deserts, which have a high surface albedo and low cloud cover. Larger errors are found over vegetated regions (tropical rainforests, higher latitudes) and partial sea pixels, reflecting lower surface albedos and more cloudy scenes. Most annual mean CO measurements have noise errors smaller than 10 %.

Some discrepancies may also be explained by the inaccurate model emissions. Large forest fires occurred at high latitudes in 2003 (Siberia; Yurganov et al. [2005]) and 2004 (Alaska) and their emissions are not included in the model simulation. Similarly, the climatological biomass burning emissions used in the model over equatorial regions may not be representative for this time period. Current estimates of east Asian CO emissions may be too low [Petron et al., 2004]. Higher hydrocarbon emission estimates – a strong indirect source of CO, notably isoprene - are also quite uncertain. In addition, model emissions over India may be overestimated [J. van Aardenne, personal communication]. Recently, preliminary model simulations with the new GFEDv2 biomass burning emission data base (van der Werf et al. 2006) have been performed and show a much better comparison of SCIAMACHY CO and the model simulations.

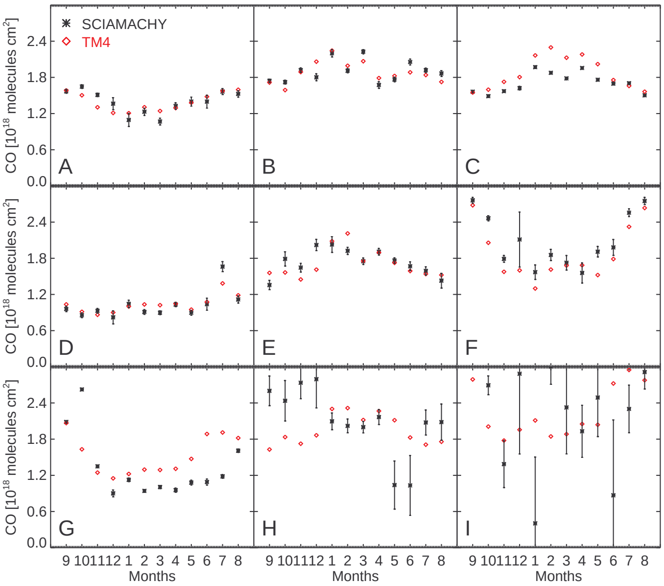

Fig. 2: Comparison of monthly mean CO total columns from SCIAMACHY (black) and TM4 (red) for nine specific locations (2°×3° grid boxes). Error bars denote the SCIAMACHY noise error. From: de Laat et al. (2006).

A quantitative analysis of the SCIAMACHY monthly mean CO total columns is presented in Figure 2. This figure shows the seasonal variation of the SCIAMACHY CO at the nine locations indicated in the bottom panel of Figure 1. For locations A to F in Figure 2 the SCIAMACHY CO total columns and the modeled seasonal variations agree well, with average absolute differences in monthly means well below 10 %. Differences also occur, e.g. over Algeria [C] during January to May, where modeled CO is higher. This pattern is consistently found over northwestern Africa as suggested by Figure 1 and most likely related to inaccurately modeled biomass burning emission in north-equatorial Africa. Simulations including the GFEDv2 data base (van der Werf et al. 2006) compare much better with SCIAMACHY CO.

Note that locations A-F have small noise errors (generally less than 6 % for monthly means) due to a high surface albedo and sufficient ‘cloud-free’ SCIA measurements per month and grid box. Locations [H] and [I] have lower surface albedos and fewer “cloud-free” observations and thus larger noise errors. These large errors hamper a comparison of seasonal variations since the measured month-to-month variations are much smaller than individual monthly mean noise errors. For these locations useful information can only be obtained by averaging over larger regions or time periods.

A remarkable difference is found over South Africa [G] where the measurements have small noise errors. A distinct maximum in the SCIAMACHY CO total columns is seen in September and October 2003 and August 2004. The model results do not show a peak in CO in October 2003 while CO increases after May 2004. Also in this case, simulations including the GFEDv2 data base (van der Werf et al. 2006) compare much better with SCIAMACHY CO, both in absolute values of the maximum and the timing of the maximum, than the simulations using climatological emissions.

Thus, the GFEDv2 biomass burning emission data base clearly is a major improvement over simulations using climatological emissions as used in de Laat et al. (2006). It also highlights the potential use of SCIAMACHY CO measurements to optimize surface emissions

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