# Global satellite observations of greenhouse gas emissions

# by A.P.H. Goede and co-workers

The EVERGREEN project, funded by the European Commission's 5th Framework Environmental Programme for better exploitation of Earth Observation data, has demonstrated the benefits of new methods for the exploitation of satellite data in climate and air pollution research and application. In particular, the SCIAMACHY instrument on board the European Earth Observation satellite ENVISAT has produced the first retrievals of greenhouse gas emissions from space, generates ozone measurements that improve the weather forecast and delivers an operational service for air pollution monitoring and predictions

In this paper the results of the EVERGREEN project are summarised and focussed on the SCIAMACHY measurements of methane (CH4), carbon dioxide (CO2) and carbon monoxide (CO) in the troposphere. But also the measurements by MIPAS of the vertical distribution of these gases in the upper troposphere and lower stratosphere are analysed. Both SCIAMACHY and MIPAS are spectrometers on board the ESA environmental satellite ENVISAT, which was launched March 1st, 2002 with a scheduled operational life time of 5 years. The measurements by MOPITT, a Canadian instrument on the NASA EOS Terra satellite launched in December 1999, provide additional information on tropospheric carbon monoxide. Global, regional, yearly and seasonal variations of CH4, CO2 and CO over the years 2003-2005 are analysed and compared with theoretical models and ground based measurements. Inverse modelling studies based on satellite data have revealed for the first time a number of significant discrepancies in the CH4 and CO emissions compared with existing emission estimates.

#### **CLIMATE CHANGE**

A useful parameter in climate research is radiative forcing, the change in net irradiance (W/m2) at tropopause height (between 8 and 18 km) exerted by a change in greenhouse gas concentrations. For small changes this provides a measure for the change in temperature at the earth surface. Greenhouse gas radiative forcing can be determined from measurement of their concentration distribution.

The underlying parameters for greenhouse gas concentra-

tions are the sources and sinks. Emissions may be estimated from measured emission factors in combination with statistical data, the so-called bottom-up approach used by countries reporting under the United Framework Convention on Climate Change (UNFCCC) and the Kyoto Protocol. Emissions can also be derived from measurement of the concentration distribution and inverse modelling, the so-called top-down approach. Discrepancies exist between the two approaches whilst uncertainties are considerable. This limits our understanding of climate change and adversely affects verification of greenhouse gas emission inventories.

Inverse modelling has been carried out from ground based observations, but results are limited by the scarce and unbalanced distribution of ground stations [Bergamaschi, 2005]. From their vantage points in space satellite measurements have the benefit of global coverage, but measurement accuracy is a challenge. This challenge was met by EVERGREEN (EnVisat for Environmental Regulation of GREENhouse gases), a European Commission RTD project carried out by a consortium of 12 European partners [Goede, 2006].

#### Methane

Methane is, after carbon dioxide, the second most important anthropogenic greenhouse gas, contributing directly 0.48 Wm-2 to the total anthropogenic radiative forcing of 2.63 Wm-2 by well-mixed greenhouse gases (IPCC, 2007). In addition, there is an indirect radiative forcing of about 0.3 Wm-2 through the formation of other greenhouse gases, notably tropospheric ozone and stratospheric water vapor. Although the global annual source strength of methane ( $550 \pm 50$  Tg yr-1) is relatively well constrained, considerable uncertainties exist in the partitioning of sources and their spatial and temporal distribution.

Vibrations in the methane molecule produce characteristic absorption lines in the near-infrared at around 1.6 and 2.3 µm. This part of the spectrum is covered by the SCIAMACHY channels 6 and 8. From the ratio of the earthshine radiance and the solar irradiance, total column abundances of methane can be retrieved by means of differential optical absorption spectroscopy (DOAS). Since these are the first ever attempts to retrieve methane information from a space-based instrument operating in this part of the spectrum, the EVERGREEN project engaged three independent retrieval algorithms' developments: IUP Bremen (WFM-DOAS), SRON (IMLM) and IUP Heidelberg (IMAP-DOAS). SRON focussed on methane retrieval from channel 8 [Gloudemans, 2005], IUP Heidelberg on channel 6 [Frankenberg, 2006] and IUP Bremen did both [Buchwitz 2005a/b, 2006].

In addition to algorithm challenges, there are a number of instrument challenges to be met. The near-infrared detectors of SCIAMACHY suffer from an increasing amount of dead and bad pixels, high dark currents leading to a low signal to noise ratio, and an ice-layer building up on the cooled detector surfaces (150 K), altering the instrument slit function. These complications are not foreseen in the official ESA data products and had to be resolved by the project.

In the EVERGREEN proposal, total column measurements of O2 were proposed as a proxy for the light path in the conversion of methane columns into column averaged mixing ratios. However, as it turned out, O2 was not very suitable because the photon scattering at 0.76  $\mu$ m, the O2-A absorption band, differed significantly from the scattering at 1.65  $\mu$ m, the methane absorption band. To overcome this problem, CO2 retrievals in a nearby retrieval window at 1.55  $\mu$ m were used as a proxy for the light path of the CH4 retrieval in channel 6 [Frankenberg, 2005]. For channel 8 scattering is less dominant. Here, surface pressure has been employed as a scaling factor.

The methane product developed under the EVERGREEN project has achieved the high precision (1-2%) required to deduce emissions from inverse modelling. Systematic errors (biases) that determine accuracy are still subject to further investigation. Precision and accuracy have been validated by ground based FTIR measurements performed under the Network for the Detection of Atmospheric Composition Change (NDACC).

In some cases, important information over the oceans has been retrieved. This has been made possible by the normalized DOAS retrieval approach, in which the normalization of methane by carbon dioxide allows clouded pixels to be taken into account. Clouded pixels provide for high reflectivity over the ocean, which is needed to obtain sufficient signal strength of the reflected radiation measured by SCIAMACHY. Ocean information proves essential for successful inverse modelling of global methane emissions.

# Higher methane emissions in the tropics

The first SCIAMACHY retrievals of methane obtained with the IMAP-DOAS algorithm over the time period August to November 2003 were presented by Frankenberg (2005). This time period coincides with peak emissions of rice paddies in Asia. The result shows that the North-South gradient is well reproduced. An important new finding is that regions with enhanced methane abundances are observed, most notably over Asia, central Africa, northern South America and eastern USA. The observations have been averaged over the period August to November 2003 on a  $1^{\circ}x1^{\circ}$  horizontal grid [Frankenberg, 2005].

The most likely cause for high abundances observed in Africa and the USA are wetlands and coal mining, respectively. Methane abundances modelled by the TM3 global atmospheric model (forward model) based on current bottom-up emission inventories for the same time period are in general agreement with measurements. However, large differences between SCIA-MACHY measurements and TM3 model results (of the order of 40-90 ppbv) are observed in the tropical belt especially over the broad-leafed forest areas of South and Central America, West Africa, Indonesia and New Guinea [Frankenberg, 2005]. The conclusion is that tropical methane emissions are significantly underestimated in current bottom-up inventories.

The EVERGREEN project has developed inverse modelling tools that have been applied to the methane measurements from SCIAMACHY. Two different inverse modelling approaches have been pursued: four-dimensional variational (4D-Var) assimilation by KNMI [Meirink, 2006] and synthesis inversion by JRC-IES [Bergamaschi, 2007]. Both approaches are complementary; the former allows the optimization of surface fluxes at high spatial resolution, while the latter is more straightforward and robust.

Data assimilation is a statistical method based on Bayes' theorem that adjusts modelled parameters (such as emissions) to fit observations (such as concentration distributions) and thereby improve the model simulations over the time period of interest. It is shown that 4D-var is an efficient method to deal with large quantities of satellite data and to retrieve emissions at high resolution. Observing System Simulation Experiments (OSSEs) have been performed to demonstrate the feasibility of the method and to investigate the usefulness of SCIAMACHY observations for methane source estimation.

On the basis of OSSE's, the impact of a number of parameters on the error in the retrieved methane emission field has been analysed. These parameters include the measurement error, the error introduced by the presence of clouds and the spatial resolution of the emission field. Some important conclusions regarding the SCIAMACHY measurements have been drawn: (i) The observations at their estimated precision of 1.5 to 2% can contribute considerably to uncertainty reduction in monthly, sub continental (~500 km) methane source strengths, (ii) Systematic measurement errors well below 1% have a dramatic impact on the guality of the derived emission fields. Hence, every effort should be made to identify and remove/ correct such systematic errors, (iii) It is essential to incorporate partly clouded pixels in order to sufficiently constrain the inverse modelling, (iv) The uncertainty in measured cloud parameters will at some point become the limiting factor for methane emission retrieval, rather than the uncertainty in measured methane itself.

An initial inversion for the year 2004 of real SCIAMACHY observations has been performed. The 4D-Var system succeeds in producing an analysis that very closely matches the satellite observations over the entire globe. However, particularly at high latitudes, the satellite data suggest much more seasonal variability than can be accommodated by the combination of surface observations and models. This indicates considerable biases in the satellite data and/or in the model.

A work-around strategy is to fit a latitudinal bias directly in the inversion. This strategy has been followed for the synthesis inversion.

The synthesis inversion simultaneously uses the NOAA surface observations of CH4 and the IMAP SCIAMACHY retrievals and includes a polynomial correction to compensate for the systematic bias [Bergamaschi, 2007]. The results suggest significantly larger tropical emissions as compared with both a priori estimates and with inversion based on the surface measurements alone, see Figure 1. The large tropical CH4 emissions derived from the SCIAMACHY observations are attributed to larger emissions mainly from tropical wetlands, but also an increase of CH4 emissions from termites and a decrease of the soil sink. The recent finding of CH4 emissions from plants under aerobic conditions [Keppler, 2006] could help to explain these larger emissions, but this suggestion is subject of the current scientific debate [Dueck, 2007].



Figure 1. A priori emissions (top) and inversion increment for 2 different inversion scenarios: S1 includes surface measurements only; S3 includes both satellite and surface observations. Total emissions for the year 2003 are shown [Bergamaschi, 2007]. Copyright 2007 American Geophysical Union. Reproduced by permission of American Geophysical Union.

### Methane radiative forcing

Results above refer to total column measurements. To assess the effect on the radiation balance and radiative forcing, the vertical distribution of methane must be known. Measurements by MIPAS could provide this information.

In order to assess the sensitivity of radiation budget mod-

elling to stratospheric methane measurements, the average and twice the standard deviation profile measured by MIPAS have been fed into the model. The resulting change in radiation budget is very small (< 0.01 W/m2). A similar conclusion was reached for radiative forcing. First, troposphere mixing ratios were assumed over the entire stratosphere. This increased the forcing by 0.1%. Next, methane was removed from the stratosphere altogether yielding a decrease of 3.4%. It can be concluded that the satellite measurements of stratospheric methane do not lead to a significant improvement of the global radiation budget and radiative forcing [Frieß, 2004].

Therefore, there is a large premium on extending the vertical profile measurement information from the stratosphere down into the troposphere. The altitude range of the operational MIPAS methane product is typically 8 to 50 km with an error budget of the order of 10%. Not included in this figure is the impact of clouds which generally leads to a systematic positive bias in the upper troposphere range. Based on cloudfiltered MIPAS measurements, the increase in radiative forcing since pre-industrial time due to methane has been calculated, assuming a surface mixing ratio of 0.7 ppm in the unperturbed pre-industrial case. The relative spatial methane distribution has been assumed unchanged, but stratospheric adjustment of temperatures has been taken into account. The calculated direct radiative forcing of methane is 0.443 Wm-2, in agreement with IPCC figures. For consistency, the radiative forcing by CO2 and N2O has also been calculated. Results are 1.46 Wm-2 for CO2 and 0.15 Wm-2 for N2O, also in agreement with IPCC figures [Myhre, 2006].

The geographical distribution of the radiative forcing is shown in Figure 2 [Myhre, 2006]. Generally, a large latitudinal gradient in the forcing is observed, with the highest values in the tropics, reflecting the large temperature gradient between the surface and the upper troposphere. The imprint of clouds can clearly be seen, with reduced radiative forcing in regions with clouds, which also absorb infrared radiation, leaving less energy to be absorbed by methane. The cloud effects can clearly be seen in the zones of convection in the tropics as well as in the belts of low pressure systems at mid latitudes in both hemispheres.



Figure 3. Annual mean CO total columns for the period September 2003 to August 2004 are retrieved from SCIAMACHY by the IMLM retrieval algorithm [Laat, 2006]. Copyright 2006 American Geophysical Union. Reproduced by permission of American Geophysical Union.

# Carbon monoxide emissions

CO emissions have been retrieved by Bayesian inverse modelling, determining the optimum between a priori emissions, chemical observations and the predictions of a global chemical-transport model [Muller, 2005]. Surface measurements and space based monthly averaged CO columns of the MOPITT instrument serve as observations, whilst the model is IMAGES. The bottom-up emission inventory of the EC 4th framework RTD project POET is used as a priori data.

Compared with methane, the relation between emissions and abundances is more non-linear, as CO is a more reactive gas. Therefore, inverse modelling was performed with an adjoint model taking into account the chemical feedbacks. The adjoint technique allows performing grid-based inversions, where the emissions of every model pixel, month and category are optimised. The adjoint model of IMAGES (transport and chemistry, 59 chemical compounds) has been built and verified under the EVERGREEN project. The cost function and its derivatives are calculated in the forward and adjoint models respectively, and are used as input to the minimisation subroutine. This provides new estimates of the emissions, until the cost function minimum is found. The number of iterations needed ranges typically between 30 and 50.

The results show an increase in global anthropogenic and biogenic emissions and a decrease in biomass burning emissions over Africa. Chinese anthropogenic emissions show an increase. Top-down anthropogenic emissions are now estimated to be 760 Tg/yr, i.e. about 100 Tg/yr more than bottom-up estimates.

## **Carbon dioxide**

Carbon dioxide is the most important anthropogenic greenhouse gas. Information on CO2 sources and sinks on the global scale is currently derived from a highly precise but rather sparse network of about 100 ground stations (e.g., NOAA/ ESRL). Satellite measurements provide the global coverage needed to better constrain the inverse models in the derivation of CO2 sources and sinks.

The retrieval of a long-lived and therefore well-mixed gas such as CO2 is extremely challenging, because only small variations in the concentration distribution contain the necessary information on surface sources. Due to its near-infrared nadir observation capability SCIAMACHY is the first satellite instrument that is highly sensitive to CO2 in the boundary layer where most variation occurs [Buchwitz, 2005a]. Therefore, SCIAMACHY CO2 retrievals attempting to retrieve the small concentration differences play a pioneering role which requires the development of a dedicated retrieval algorithm, Weighting Function Modified Differential Optical Absorption Spectroscopy (WFM-DOAS) [Buchwitz, 2000]. WFM-DOAS is a linear leastsquare method based on scaling (or shifting) pre-selected vertical CO2 profiles.

The CO2 columns are retrieved from the spectral fitting window 1.558-1.594 µm of SCIAMACHY channel 6 [Buchwitz, 2005a/b, 2006, 2007b]. Dry air column averaged mixing ratios XCO2 are determined by normalisation with the simultaneously measured oxygen columns retrieved from the O2 A-band. Because of the large spectral distance between the CO2 band and the O2 band, the light path error does not fully cancel in the CO2 to O2 column ratio XCO2 calculated. The resulting XCO2 error contains both random and systematic components. Under high and variable aerosol load this error can become as large as several percent. This problem is overcome by identifying the aerosol contaminated scenes. A similar situation occurs with cloud contaminated scenes. Cloud contaminated ground pixels are identified by a threshold algorithm based on the subpixel information contained in the SCIAMACHY Polarization Measurement Devices.

Figure 4a shows bi-monthly averages of XCO2 retrieved over the northern hemisphere by WFM-DOAS version 0.4 [Buchwitz, 2005b]. Figure 4b shows TM3 model simulations. Both model and measurements have been (re-)sampled on the same grid allowing meaningful comparison. Qualitatively, there is a reasonable agreement between the SCIAMACHY measurements and the model simulations. Quantitatively, there are significant differences with respect to the spatial details and the amplitude of the CO2 spatial and temporal variability.



Figure 4a. Carbon dioxide column averaged mixing ratios over the northern hemisphere for the year 2003 as retrieved from SCIAMACHY near-infrared nadir spectra using the initial version 0.4 of the WFM-DOAS retrieval algorithm [Buchwitz, 2005b].



Figure 4b. Carbon dioxide column averaged mixing ratios over the northern hemisphere for year 2003 obtained from TM3 model of MPG-BGC-Jena [Buchwitz, 2005b].

Recently, three years of SCIAMACHY data (2003-2005) have been reprocessed using the improved WFM-DOAS version 1.0 with improved spectral calibration and updated spectroscopic line parameters. This data is currently being analysed and first results are published [Buchwitz, 2007b]. The focus is on large scale Northern Hemispheric features such as the seasonal cycle of CO2 and the year-to-year increase, mainly caused by the burning of fossil fuel. Results show excellent synchronisation of the seasonal cycle, but the amplitude is larger compared with models. In July/August during the growing season the CO2 amplitude measured is 10% to 20% lower, whilst in November/December when rotting sets in, the CO2 amplitude is 10% to 20% higher. These results may be explained by a larger land surface exchange flux of CO2 than

is currently derived in modelled vertical transport (see model comparison). However, it could also be due to problems with the accuracy of the satellite data. Clearly, these interesting results warrant further investigation.

#### VALIDATION

The EVERGREEN data products for CH4, CO and CO2 retrieved from the SCIAMACHY near infrared channels have been validated by an independent team by comparing these data with data from the ground-based FTIR spectrometer network of the NDACC (http://www.ndacc.org).

Due to inherent limitations of the FTIR and SCIAMACHY measurements, validation is not straightforward and several issues had to be resolved. These issues are (i) how to deal with varying ground station altitude (ii) sparse data availability and (iii) difference in observed air masses. The first item was dealt with by normalising all data to ECMWF pressure data and introducing an altitude correction factor. The second item is addressed by temporal interpolation among FTIR data. However, spatial representation remains limited by the high northern latitude locations of some FTIR stations, where SCIAMACHY data quality is known to be poor. The third issue represents an inherent limitation in comparing data above a single ground station with data covering a finite ground pixel size and requires a case by case inspection [Dils, 2007].

The results of the validation exercise with FTIR stations for methane, carbon monoxide, and carbon dioxide are summarised in Table 1. A large number of data have been compared (order 104 for all individual data products). Seasonality is captured relatively well as indicated by the correlation coefficients, except for CO2. Part of this CO2 error is reduced in the new WFM-DOAS version 1.0, not considered in this Table. Scatter for XCH4 has reduced to acceptable level for inverse modelling of emissions to be carried out, whilst scatter for SCIAMACHY CO and for CO2 remains on the high side. However, results are unfavourably biased by the high Northern latitude locations of FTIR stations where CO and CO2 data quality is known to be low.

	-	CO			CH4		COz
	WFMD	IMLM	IMAP	WFMD	IMLM	IMAP	WFMD
	CO	CO	XCO	XCH <sub>4</sub>	CH4	XCH <sub>4</sub>	XCO <sub>2</sub>
	v0.5	v6.3	v0.9	v0.5cur	v6.3	v1.1	v0.4
dias	1.00 +0 \$7	-14.7 +0.90	-259+063	-3.28 + 0.15	-2.33 + 0.10	-0.620.14	-693+0.14
N	22362	12092	14218	42072	5323	22954	7704
Cud	25.1	22.4	23.5	1./3	3.14	19	3.7±
R	0 85	0.83	0.53	0.20	0.7.	0.70	0.45
Cr.s. TP		<u>(_)</u> ۴۴۸			115		10 1

Table 1. SCIAMACHY validation results based on FTIR of the NDSC[Dils, 2006].

The errors represent the weighted standard errors (3\*std/ sqrt(N)) of the ensemble of individual weighted biases. Bias is the calculated weighted bias (in %) of the SCIAMACHY data relative to the 3rd order polynomial fit through the ground based FTIR data using the large grid ( $LG = \pm 2.5^{\circ} LAT, \pm 10^{\circ} LON$ ) spatial collocation criteria (the weight w = 1/(err)2, in which err is the error on the individual measurement as given by the data providers). The bias is calculated for all stations. For individual stations bias can differ significantly. N is the number of correlative individual SCIAMACHY data. sigmascat is the percentage 1sigma weighted standard deviation of the daily averaged SCIAMACHY measurements with respect to the polynomial interpolation of the daily FTIR data, corrected for the daily bias. R is the correlation coefficient between the weighted monthly mean SCIAMACHY and FTIR data. Also given are the scatter of the daily averaged FTIR data points relative to their corresponding polynomial fit values (sigmaFTIR) and the target precisions (TP) needed for inverse modelling on a regional scale.

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#### **MODEL COMPARISON**

A comprehensive comparison of 5 atmospheric chemistry transport models TM5, TM4, TM3, IMAGES, and LMDZ has been carried out by the EVERGREEN project [Bergamaschi, 2006] with the objective to analyze differences in model transport, in particular vertical mixing in the boundary layer and in convective transport, synoptic variations, and large scale global circulation, including inter-hemispheric exchange and stratosphere troposphere exchange (STE). Simulations employ tracers with different atmospheric lifetimes: 222Rn (3.8 days), CH4 (~9 years), SF6 (~3000 years). Furthermore, the chemistry has been tested through OH fields derived from full chemistry model simulations.

222Rn simulations show significant differences in vertical transport between models, leading to differences in simulated 222Rn concentrations near the surface of up to a factor 3. The TM4/5 models show the highest 222Rn concentrations near the surface, while the other models display stronger vertical mixing. Simulations with SF6 show significant differences in inter-hemispheric transport between the models, ranging from 6 to 12 months. STE is weaker and probably more realistic (15–16 months) in TM4/5 and LMDZ than in TM3 and IMAGES (7-8 months). CH4 tracer simulations with prescribed OH fields are consistent with the 222Rn and SF6 simulations. Results are also consistent with previous model comparisons such as TransCom2.

Simulated OH fields show significant differences between models near the earth surface, probably due to different emission inventories of CO, NMHC and NOx. In the free troposphere the spatial OH distribution is similar for all models. Also, the seasonal OH variation is consistent for all model runs.

#### CONCLUSION AND FUTURE OUTLOOK

The EVERGREEN project has produced:

• Improved global data products for CH4, CO and CO2 based on SCIAMACHY satellite measurements.

• Validation of these data against the ground-based NDACC FTIR measurements showing good precision of CH4 and adequate precisions of CO and CO2

• Inverse modelling of these data showing higher CH4 emission in the tropics and higher anthropogenic CO emission in South East Asia compared with bottom-up estimates.

• Total column distributions of CO2 over the Northern Hemisphere showing a larger amplitude of the CO2 seasonal cycle measured by SCIAMACHY compared with models.

• Radiation budget and forcing calculations, including MI-PAS satellite measurements for CH4, that are consistent with IPCC figures.

• Data assimilation tools and Observing System Simulation Experiments for inverse modelling of sources and sinks defining criteria for successful inversion.

• Model comparisons identifying weaknesses in modelled vertical transport.

EVERGREEN results have found application in the EC-ESA GMES operational service for atmosphere PROMOTE. Data users may consult the ESA PROMOTE web-site for access to the data [Goede, 2004].

EVERGREEN has provided information for the specification of future space-based climate gas sensors [Barrie, 2004]. Crucial is the understanding and reduction of systematic errors encountered in instrument calibration and data retrieval. Higher detector sensitivity in combination with higher spectral resolution, feasible with today's technology, marks an obvious road to improved data accuracy. Improved accuracy in underlying spectroscopic data remains an important priority, as errors propagate directly into volume mixing ratios. Future satellite sensors should aim at higher temporal and spatial resolution, in particular vertical profile information in the troposphere is necessary in order to understand errors in vertical transport modelled.

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