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Tropical methane emissions: A revised view from SCIAMACHY onboard ENVISAT

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Spectroscopic interferences

We can consider erroneous spectroscopic parameters as systematic errors in our forward model that is used for the retrieval. Given the new spectroscopic dataset by Jenouvrier et al. (2007), we can now quantify this systematic error and show how it leads to an overestimation of methane abundances. For this purpose, we use a simplified retrieval version, scaling only total columns in a least-squares fit without constraints. The forward model ignores scattering and simply computes $\log(I/I_0)$ as a function of viewing geometry and trace gas abundances.

We consider a tropical atmosphere at a solar zenith angle of 30° and run the forward model twice, once with the old HITRAN water spectroscopy and once with the modified updates by Jenouvrier et al. (2007). The total water column is $1.39 \cdot 10^{23}$ molec/cm². The difference of $F = \log(I/I_0)$ between the two runs can be considered as forward model error ΔF , giving rise to a systematic error in the retrieved state vector elements, in our case the methane, carbon dioxide and water total column amount. In an unconstrained least squares approach, this error reads

$$\hat{x} - x = (K^T K)^{-1} K^T \cdot \Delta F,$$

where

$$\begin{aligned} \hat{x} &= \text{retrieved state vector,} \\ x &= \text{true state vector,} \\ K &= \text{Jacobian of the forward model,} \\ \Delta F &= \text{Systematic error in the forward model.} \end{aligned}$$

Fig. S1 illustrates how the systematic error in the forward model propagates into an overestimation of methane. The upper two panels depict the forward model runs and their difference. Panel c shows the entries of the Jacobian for methane, carbon dioxide and water vapor, using a simple scaling factor for the entire atmospheric column as state vector elements. Panel d depicts how the least squares fit

Figure 1: Systematic spectral structures in the forward model and its impact on SCIAMACHY methane measurements. Panel a depicts the forward model runs for HITRAN and the modified Bxl-Reims spectroscopic databases assuming a tropical atmosphere with high water vapor abundances ($1.39 \cdot 10^{23}$ molec/cm²) and a gaussian instrumental lineshape with 1.33 nm FWHM. Panel b shows the resulting forward model error ΔF , assuming the new spectroscopic dataset to represent the truth. Panels a and b are drawn with high spectral sampling while the following panels use SCIAMACHY sampling (about 2 pixels per FWHM) In panel c, entries of the Jacobian $K = dF/dx$ for methane, carbon dioxide and water vapor are displayed. Panel d shows how the least squares approach accomodates the systematic error in the forward model by incorrectly fitting individual trace gas contributions, shown as thin lines in the same color as in panel c. ΔF is shown as black line, $K(\hat{x} - x)$ as red line

accommodates the systematic structures of ΔF by fitting CH₄, CO₂ and H₂O. ΔF is shown as black line, $K(\hat{x} - x)$ as red line and the individual contributions of the trace gases as thin lines (same color as panel c). The resulting errors in the total column of the individual trace gases in this case are as follows:

$$\begin{aligned} \text{CH}_4 &= 3.3\%, \\ \text{CO}_2 &= -3.8\%, \\ \text{H}_2\text{O} &= -6.7\%. \end{aligned}$$

Methane is overestimated by about 3.3%, consistent with our findings using real SCIAMACHY data. However, one should notice that the error in CH₄ also depends on the choice of the state vector (e.g. including temperature derivative or total column scaling vs profile retrieval) and the magnitude of the overestimation should not be interpreted as universal for all SCIAMACHY retrievals. However, it is obvious that especially the dip in ΔF in the vicinity of the methane $2\nu_3$ Q-branch causes a systematic overestimation of methane abundances, depending on the total water vapor column.

Modifications to water spectroscopic parameters

For some transitions, the Bxl-Reims database (Jenouvrier et al. (2007)) either does not provide a pressure shift or air-broadening parameter or they obviously don't fit the atmospheric measurements (some air-broadening coefficients are as low as $0.007 \text{ cm}^{-1}/\text{atm}$, an extreme value that could not be confirmed by the FTIR spectra). For missing values, we attributed a universal pressure shift of $-0.01 \text{ cm}^{-1}/\text{atm}$ and an air-broadening parameter of $0.066 \text{ cm}^{-1}/\text{atm}$. For erroneous broadening coefficients or pressure shifts, we manually adjusted the line parameters to yield reasonable residuals in the FTIR fits. For the few strong water lines within the Q-branch range of the $2\nu_3$ methane band, we also slightly adjusted line strengths to minimize residuals in a range where spectroscopic interferences are most likely. Table S1 lists all changes (numbers in bold font indicate changed parameters).

Table 1: Additional modifications of water spectroscopic parameters.

line position [cm ⁻¹]	line strength at 296K [cm ⁻¹ /(molec cm ⁻²)]	air-broadening [cm ⁻¹ /atm]	pressure shift [cm ⁻¹ /atm]
5975.8916	1.513E-25	0.0680	-0.0230
5975.9562	4.505E-26	0.0550	-0.0190
5976.7235	8.359E-26	0.0600	-0.0240
5981.7928	4.613E-25	0.0570	-0.0150
5983.5389	1.117E-25	0.0460	-0.0030
5985.2589	1.101E-26	0.0565	0.0020
5985.7346	4.722E-25	0.0472	-0.0070
5987.2006	4.811E-26	0.0820	-0.0010
5990.1259	2.593E-25	0.0369	-0.0060
5990.2160	1.621E-25	0.0668	-0.0260
5990.9150	1.625E-25	0.0470	-0.0057
5992.3952	2.367E-25	0.0600	-0.0200
5992.4251	5.906E-25	0.0480	-0.0200
5997.8040	4.308E-26	0.0720	0.0100
6001.3640	6.650E-25	0.0760	0.0030
6001.5430	2.793E-25	0.0870	-0.0070
6003.1479	6.685E-26	0.0590	-0.0210
6003.2566	2.292E-25	0.0590	-0.0080
6037.9375	1.654E-25	0.0520	-0.0200
6039.2624	4.100E-25	0.0680	-0.0140
6063.8059	1.304E-25	0.0420	-0.0215
6069.3211	2.759E-26	0.0320	-0.0230
6069.8212	5.517e-28	0.0500	-0.0100
6087.5067	2.138E-26	0.0430	-0.0250
6087.5288	5.476E-26	0.0450	-0.0250
6099.2651	2.911E-26	0.0660	-0.0080
6099.3000	5.950E-25	0.1060	-0.0060
6099.5803	1.143E-25	0.0830	0.0000
6115.3808	4.668E-26	0.1036	-0.0100
6116.2706	1.110E-25	0.0650	-0.0080