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# **Observing atmospheric methane from space**

**Enabling satellite-borne observations of methane  
in the cloudy atmosphere**

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VRIJE UNIVERSITEIT

# Observing atmospheric methane from space

ACADEMISCH PROEFSCHRIFT

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op gezag van de rector magnificus  
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door

Dinand Schepers

geboren te Hardenberg

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*To my family,  
to my friends.*

This thesis was reviewed by members of the reviewing committee:

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prof. dr. M.C. Krol	Wageningen University
prof. dr. T. Röckmann	Utrecht University
dr. P. Stammes	Royal Netherlands Meteorological Institute (KNMI)

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# Summary

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Methane (CH<sub>4</sub>) constitutes the most abundant hydrocarbon trace gas in the Earth's atmosphere. Its present-day globally averaged atmospheric concentration is approximately 1850 parts per billion per volume (ppbv), more than double the estimated abundance of atmospheric methane in pre-industrial times. The role of atmospheric methane in global climate change is a reason for this high concentration and growth rate to be of concern and gaining a quantitative understanding regarding sources and sinks of atmospheric methane is of great importance to develop mitigation policies for climate change and air pollution.

## Space-borne observations of atmospheric methane

Methane is released to the Earth's atmosphere by a wide variety of processes of natural and anthropogenic origin. Around 40% of total emissions stem from natural sources, with 70% of this being emitted from wetlands ecosystems. The 60% anthropogenic methane emissions predominantly originate from agricultural sources and the fossil fuel industry.

Through inverse modelling, the strength of individual methane sources can be inferred from observations of atmospheric methane abundances. By inverting a general circulation model of the Earth atmosphere, fluxes at the surface-atmosphere boundary can be inferred from measured gradients in atmospheric methane concentrations .

Observations of atmospheric methane abundances are provided by the Japanese Greenhouse Gas Observing Satellite (GOSAT), the world's first dedicated satellite mission for observing atmospheric methane and carbon dioxide concentrations from space. The Fourier transform spectrometer (FTS) TANSO-FTS on-board GOSAT measures Earth-radiances in several spectral windows between 0.758 and 2.08  $\mu\text{m}$  with a high spectral resolution. In its default nadir observation mode, the TANSO-FTS instrument continuously sweeps back and forth over a  $\pm 35^\circ$  cross-track range, taking three to five nadir soundings cross-track. When GOSAT is overflying a body of water in a suitable geometry, the instrument operates in solar glint mode i.e. pointing directly into the specular reflection of the solar beam.

Information on methane concentrations in the atmosphere can be retrieved



from a short-wavelength infrared (SWIR) Earth-radiance measurement by exploiting the methane absorption lines around 1600 nm. To extract this information, a forward model is required to simulate the radiance observation as a function of the atmospheric state. Subsequently, the forward model is inverted to infer the atmospheric trace gas concentration that is statistically most likely to underlie the measured radiance.

Following the Beer-Lambert law, the depth of a trace gas absorption line is directly related to the amount of molecules of that particular trace gas that are present along the light path of the measurement. Consequently, correctly estimating the length of the effective light path is crucial for correctly inferring total column atmospheric methane concentrations. Especially since scattering by aerosol and molecules is significant when considering short-wavelength infrared radiation.

To estimate and account for the effective light path, two conceptually different retrieval approaches exist: the physics-based approach and the so-called proxy approach. The proxy approach simultaneously retrieves methane and CO<sub>2</sub> abundances, neglecting all scattering by molecules and aerosols along the light path. The error on the retrieved CO<sub>2</sub> abundance with respect to prior knowledge on CO<sub>2</sub> concentrations provides information on the level of scattering that was neglected. This information is subsequently used to correct the retrieved methane concentration for the effects of atmospheric scattering. In contrast to the proxy method, the physics-based retrieval strategy explicitly accounts for modifications of the light path due to atmospheric scattering. To that end it aims to retrieve the atmospheric scattering conditions from the radiance measurement together with the methane total column.

## **This research**

The research that is presented in this thesis constitutes an effort to further understand and develop methane retrievals from GOSAT SWIR radiance observations. The first chapter of this thesis is dedicated to an in-depth performance comparison and validation study that compares the proxy and physics-based retrieval methods applied to GOSAT soundings. This study, based on the first 19 months of GOSAT soundings in the immediate vicinity of 12 validation measurements sites of the Total Carbon Column Observing Network (TCCON) identified no significant differences in performance between the two retrieval approaches.

However, a subsequent comparison based on a global data set did show differences in retrieval performance exceeding 1% on regional scales. It was shown that the physics based method tends to overestimate methane abundances un-

der conditions of strong atmospheric scattering over a highly reflective ground surface. Such conditions are typically found in desert-like regions as the Sahara or the Arabian Peninsula. The proxy method on the other hand suffered from retrieval errors introduced by inaccurate a priori CO<sub>2</sub> abundances that propagate directly into the retrieved methane total columns.

While the proxy method is most affected by inaccuracies in a priori data, the physics-based method proved limited in strongly scattering atmospheric conditions. The latter greatly reduces the number of useful physics based retrievals. However, the physics-based method does have the most potential for further development of the retrieval methodology. In the remainder of this thesis the physics-based retrieval was further developed such that it can be applied to GOSAT soundings with enhanced scattering by aerosol and water clouds.

To that end, a new radiative transfer model, called LINTRAN v2.0, was developed to reduce the numerical effort involved in simulating radiance measurements in strongly scattering atmospheres. At its core lies a mathematical framework that allows splitting the modelled radiance in a contribution that originates from light that was scattered  $N$  times or less and a contribution from light that is scattered more than  $N$  times. The contribution of light scattered  $N$  times or less is solved analytically, the high-order scattering contribution is subsequently solved for using a numerical approach. The framework was implemented with  $N=2$ . The numerical performance of this radiative transfer model is significantly better than the previous model version, translating into speed-up of calculations in a cloudy atmosphere with a factor 42, without a loss in model accuracy.

LINTRAN v2.0 was implemented in the forward model of the RemoTeC physics-based retrieval algorithm with the goal of retrieving methane abundances from cloudy GOSAT nadir soundings over the ocean. The proposed approach simulates all light scattering in the sampled atmosphere by a single-layer water cloud characterized by a Gaussian height distribution of cloud droplets. The height and the geometrical thickness of the cloud layer as well as the size and amount of cloud droplets are inferred from the GOSAT measurement together with the column abundances of CO<sub>2</sub> and CH<sub>4</sub>.

The resulting methane and carbon dioxide total column retrievals were validated with ground-based measurements from 8 TCCON sites. All of which are located on an island or near an oceanic coast line. In terms of retrieval accuracy and precision, the proposed physics-based methane retrieval for cloudy ocean soundings performs slightly worse than the RemoTeC physics-based method for cloud-free observations over land. Nonetheless, the retrieved methane columns do contain valuable information on methane abundances over the oceans that beforehand were only sparsely covered.

## Outlook

The TCCON-based validation study of methane retrievals in cloudy GOSAT soundings over oceans represents an important first step toward applying the proposed retrieval to the complete (sub)set of cloudy GOSAT ocean observations. If successful, this would significantly increase geographical coverage of physics-based CH<sub>4</sub> retrievals from GOSAT SWIR observations, especially in the Southern Hemisphere. Moreover, it might be attempted to apply the same retrieval to cloudy soundings over land. To do so, it would be best to target optically thick and unbroken cloud layers to minimise the effect of reflection by the underlying surface.

In light of inverse modelling of methane sources, one should bear in mind that methane retrievals in cloudy atmospheres lack sensitivity below the retrieved cloud height where methane sources are generally located. The retrievals do however contain useful information to constrain methane transport and abundances of in the free troposphere.

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The scientist has a lot of  
experience with ignorance and  
doubt and uncertainty,  
and this experience is of very great  
importance,  
I think.

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Richard P. Feynman

