

We thank the referee for their effort and time spent in revising our manuscript. In the following we respond to their comments and hope to be able to convince the referee of the quality of our work (red color indicates the referee's comments and black our responses).

General comment

The paper addresses the problem of defining a tropospheric CH₄ averaged column for NDACC FTIR CH₄ retrieved profiles. Although the paper addresses an interesting problem, it does not provide a clear outline of the scientific methods and assumptions. I was not able to check the validity of equations (5), (6) and (7), nor could I find a mathematical motivation for equations (10), (11) and (12). Perhaps the a posteriori method is introduced elsewhere in the literature, but then the necessary references should be made.

RESPONSE:

The referee misses a clear scientific outline of our manuscript. The main reason for their discontent seems to be problems in checking the validity of Eqs. (5) – (7) and a missing motivation for Eqs. (10) – (12). We are sorry about this discontent and in the following we would like to briefly depict our manuscript's outline and in the context of the specific comments explain and motivate the equations. In our opinion, the outline should be captured from the titles of the different sections. In the following we propose some minor modifications of these titles and we hope that this satisfactorily addresses the referee's complains.

The INTRODUCTION describes the problem of CH₄ remote sensing. There are different CH₄ signals (depicted in Fig. 1) or if the term signal is not favored by the referee we can also say that there are CH₄ variations that take part in different regions of the atmosphere: a very local signal in the boundary layer (variation that we index with BL), a regional scale signal in the lower /middle troposphere (variation that we index with TRO), and a signal due to large scale variations in the tropopause altitude, i.e., in the UTLS (variations that we index with UTLS). We are interested in the "TRO" signal/variation.

SECTION 2 ("Ground-based NDACC FTIR: experiment, tropospheric CH₄ retrieval setup, and error estimation") presents the experiments and the retrieval. The error estimation suggests that the CH₄ variations in the UTLS are a leading error source for the "TRO" product. This result of the error estimation is the motivation for the a posteriori correction method which is described in Subsection "2.3.3: A posteriori correction for optimal independence of retrieved tropospheric and stratospheric CH₄". We propose changing this subsection title to "A posteriori correction for an improved estimation of the lower/middle tropospheric CH₄ signals/variations". We think that this title makes clearer what we are aiming at and it avoids the term "optimal". In addition the equation of this subsection will be set in context to the work of Rodgers and Connor (2003),

thereby we hope being able to better convince the referee about the usefulness of our approach.

SECTION 3: The remote sensing product is compared to GAW data and we need to make sure that we pair remote sensing data (which are very weakly affected by local signals/variations) with GAW data (which can contain very local signals/variations) that are comparable. The approach for achieving this comparability is explained in Section 3. In order to make the content of Section 3 even clearer we will change from “Pairing the ground-based FTIR and surface in-situ datasets” to “Pairing the ground-based FTIR data with comparable surface in-situ data”.

Section 4 shows the comparison and Section 5 the conclusion.

In the following we address the specific comments of the referee. There we will also address their concerns and questions in the context of Eqs. (5)-(7) and (10)-(12).

Specific Comments

(1) The paper contains statements that are not entirely clear to me:

- p637,l1: ‘. . .the tropospheric signal is much smaller than the boundary layer or the UTLS signal’: what does signal mean?

RESPONSE:

Here we talk about the atmospheric CH₄ variations that are actually present in the atmosphere.

We think that the total atmospheric CH₄ variations can be classified into three different groups: 1) a very local and surface near variations (we call it boundary layer signal, BL), 2) a regional scale signal in the lower /middle troposphere (we call it tropospheric signal, TRO), and 3) a signal due to large scale variations in the tropopause altitude, i.e., in the UTLS (we call it UTLS signal).

The local scale signals which we call BL can be rather large (CH₄ concentrations can vary between 1800 ppb and 2200 ppb on a day-to-day time scale). On the other hand, the regional scale tropospheric CH₄ day-to-day variation, which we call TRO, is much smaller (within 50 ppb).

- p642,l2: ‘. . .we calculate the tropospheric column-averaged CH₄ mole fraction directly from the measured spectrum’: do you mean from a retrieved CH₄ profile (‘directly from the measured spectrum’ is confusing)?

RESPONSE:

We directly retrieve profiles from the spectra. Our method is different from a method typically used for near infrared retrievals, where total columns are retrieved from the spectra and then subsequently the tropospheric columns are

obtained after the retrieval process by a correction using HF or N₂O total columns (e.g., Washenfelder et al., 2003).

We will change from “calculate [...] directly from the measured spectra” to “retrieve [...] directly from the measured spectra”.

- p645,l9: Can you describe the temperature uncertainty correlations between the different layers?

RESPONSE:

We assume no temperature error correlations between the different layers. This will be clarified in the AMT version.

- p648,l3: ‘. . .that ensures an optimal separation between the retrieved tropospheric and stratospheric amounts’: what does optimal mean? Did you compare to other methods?

RESPONSE:

Yes, we agree with the referee. We cannot prove that our method is optimal. We can theoretically show that it provides improvement (see estimations of section 2.3.3). We will change from “optimal separation” to “improved separation”.

- p648,l19: ‘The a posteriori correction means an a posteriori optimisation of the retrieval constraints. The constraints are modified in order to get a tropospheric product that is optimally independent of the UTLS’: quantify the meaning of optimization and make this statement more precise: show how equation (12) changes the retrieval constraint (which is Tikhonov-Philips matrix).

RESPONSE:

Eq. (12) modifies the averaging kernel (according to Eq. 11). Since the measurement is not changed the modification of the kernel has to be achieved by changing the constraints of the inverse problem. The statement here is just made in order to briefly describe the working principle of the method. Some mathematical background to such a posteriori modifications (or optimisations) is given in Section 4 of Rodgers and Connor (2003) and we will refer to the Rodgers and Connor (2003) paper in our manuscript in order to better motivate our approach.

- p649,l4: ‘. . . but it provides the best tropospheric CH₄ data quality’: specify the meaning of best. Did you compare your product with other products?

RESPONSE:

We agree and will relativise (see also above). We will change from “best” to “improved”.

(2) p644: Equations (5), (6), (7): I do not understand how these equations have been derived. The smoothing error is obtained from $(A-I)S_a(A-I)^T$ where S_a is the (estimated) covariance of $(x-x_a)$. Indicate how $S_{a,bl}$, $S_{a,tro}$ and $S_{a,url}$ relate to S_a . This should explain why you treat the bl and utls contributions differently in equations (5), (6) and (7).

RESPONSE:

We assume $S_a = S_{a,BL} + S_{a,TRO} + S_{a,UTLS}$. We are interested in the “TRO” signals/variations. We are not interested in the signals/variations called “BL” and “UTLS”, which are thus error sources that propagate into our retrieved state according to $AS_{a,BL}(A)^T$ and $AS_{a,UTLS}(A)^T$. These errors are often called “interference errors”, e.g., Eq. (8) of Rodgers and Connor (2003).

The uncertainty in retrieving the variations called “TRO” are $(A-I)S_{a,TRO}(A-I)^T$. This error is often called the “smoothing error”, e.g., Eq. (7) of Rodgers and Connor (2003).

For the revised manuscript we will explicitly explain that Eqs. (5) and (7) calculates so-called “interference errors” and Eq. (6) so-called “smoothing errors”.

(3) p647: Motivate equations (10), (11) (12). How does this C guarantee an ‘optimal separation’ between tropospheric and stratospheric amounts? On p648 you mention an optimization of the retrieval constraint. Do you mean that the matrix C optimizes the Tikhonov matrix? This seems to be a strong statement. Provide a more mathematical motivation for this statement.

RESPONSE:

We will motivate the approach by referring to respective formulae of Rodgers and Connor (2003). Furthermore, we will replace “optimal” by “improved”.

Rodgers and Connor (2003) provide some mathematical background to such a posteriori modifications (or optimisations). The relevant part of Rodgers and Connors (2003) is Section 4, where “best estimates of a function of the retrieved state vector” are discussed. This is what we are dealing with in our paper, since the tropospheric column-averaged amount is a function of the retrieved state vector:

In case of a retrieval that is optimal in the sense of the Bayes’ theorem, one can simply apply the function to the retrieved state and automatically get the best estimate (see Sect. 4.1 of Rodgers and Connor, 2003). However, we do not completely know the a priori state and the details of its covariance. Instead we use a modeled mean state (WACCM model) and an ad-hoc constraint (Tikhonov-Philips). With the constraint we take care that we do not overinterpret the spectra and thereby tend to over-constrain the problem. We can then calculate the tropospheric column-averaged amount from this retrieved state however, there might be a better estimator of the tropospheric column-averaged amount (see Sect. 4.2 of Rodgers and Connor, 2003). Equation (18) of Rodgers and Connor (2003) shows how this better estimator can be calculated from the retrieved state. Their Eq. (18) is of the same algebraic form as Eq. (12) in our manuscript.

In the revised manuscript we will discuss the mathematical similarity of Eq. (18) of Rodgers and Connor (2003) and our Eq. (12).

ADDITIONAL REFERENCES:

Rodgers and Connor (2003):

Rodgers, C. D. and B. J. Connor, "Intercomparison of remote sounding instruments", J. Geophys. Res., 108, D3, 4116, doi:10.1029/2002JD002299, 2003.

Washenfelder et al. (2003):

Washenfelder, R. A., Wennberg, P. O., and Toon, G. C, "Tropospheric methane retrieved from ground-based near-IR solar absorption spectra", Geophys. Res. Lett., 30, L017969, doi:10.1029/2003GL017969, 2003.