Comment on “Strategy for high-accuracy-and-precision retrieval of atmospheric methane from the mid-infrared FTIR network“ by R. Sussmann et al.

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Abstract

We acknowledge that the work presented by Sussmann et al is a careful study of the current NDACC infrared working group (IRWG) CH4 retrieval guideline. The authors prove that other schemes can be found which are superior to the current official NDACC guideline, especially for wet sites. However, we have objections with regards to the proposed retrieval scheme to replace the current NDACC retrieval guideline, as the new scheme still might require further optimization. Our main concern is (1) that two of the proposed microwindows (MW 3 and MW 5 in Sussmann et al.) suffer from strong H2O (HDO) interference, (2) that the highly variable interfering species H2O (HDO) is not treated with the required care to minimise the interference error and (3) that a remigration to outdated HITRAN line lists is required. We outline how the retrieval recipe proposed by Sussmann et al. could be further improved (in the following, we refer to the retrieval scheme proposed by Sussmann et al. as MIR-GBM), for this purpose we investigate a refined version of MIR-GBM, outline an alternative retrieval approach and test all three schemes at several FTS sites under different geophysical conditions.

Introduction

The current NDACC IRWG retrieval guideline for CH4 is essentially a result of work done within the EU projects UTFIR (www.nilu.no/uftir) and HYMN (www.knmi.nl/samenw/hymn). Within both projects, extensive retrieval tests have been performed. In addition, within HYMN, laboratory cell measurements of CH4 have been taken at different temperatures and pressures (CH4 in air) to reveal possible problems with the CH4 line parameters provided in HITRAN (www.cfa.harvard.edu/HITRAN). These cell measurements were performed in 2007 within a collaborative effort of the HYMN project partners Univ. of Bremen and KIT, IMK-ASF. It turned out that the spectroscopic data of CH4 in the MIR microwindows under consideration for HYMN work have reached a rather mature shape in recent HITRAN versions - in contrast to the NIR: in this latter spectral domain these cell measurements allowed for a significant improvement of the CH4 line parameters (Frankenberg et al., ACP 8, 5061-5075, 2008) which were adopted in HITRAN 2008. The NDACC recommendation to use the most recent HITRAN spectroscopy (HIT 2008 with official updates) was an overall strategic decision which has been adopted for the reanalysis of all NDACC FTIR species during the NDACC Infrared working group (IRWG) meeting held in Garmisch in 2009.

Recently, it became evident that the current NDACC IRWG CH4 guideline falls short under certain conditions. There are essentially two reasons for the unfavourable reassessment of the current retrieval guideline: firstly, we now aim at a further improved precision of the NDACC CH4 data product (to meet precision levels in NDACC FTIR greenhouse gas data products which are competitive to those claimed by TCCON (www.tccon.caltech.edu)). Secondly, when applying the NDACC IRWG CH4 guideline for the analysis of spectra which were recorded at high H2O total columns (C. Vigouroux, BIRA, priv. comm.) the disadvantageous effects of the H2O/HDO interference in the recommended microwindows became apparent. As obvious remedy it was suggested to remove the MW with strongest H2O/HDO interference from the set of the 5 NDACC MWs (these MWs are listed in Sussmann et al., MW5 shows the strongest interference, see Table 1 and Figure1 therein). This modified scheme (MWs 1-4, HITRAN 2008 line lists) has been applied in several recent studies of CH4 as a workaround, but as Sussmann et. al have proven now, this ad-hoc modification can be outperformed by other combinations of MWs and line lists. Evidently, to achieve success in upcoming projects, the NDACC IRWG community has to redefine a new CH4 analysis guideline asap. On the other hand, since a network-wide reanalysis of CH4 and data dissemination via the NDACC database in due time is a non-negligible effort for the working groups involved, a rashly change to a new (possibly still premature) CH4 retrieval recipe might enforce additional modifications of the guideline and further reanalyses in the near future. The aim of this comment is to encourage a second sight on the CH4 retrieval scheme (MIR-GBM) proposed by Sussmann et al. before this scheme is adopted by the network as is. Because the current NDACC IRWG CH4 guideline is outperformed by the MIR-GBM scheme, we outline an alternative improved scheme in the following section.

Description of an alternative retrieval scheme

Table 1 summarizes the 4 MWs which we applied. To avoid confusion with the numbering of MWs used by Sussmann et al., we identify these windows as A, B, C, D. Note that MWB equals MW1, the other MWs suggested here do not have counterparts. MWB and MWC contain strong CH4 lines and are used to determine the target species CH4, MWA and MWD are auxiliary MWs containing HDO (MWA) and H2O (MWD) lines of appropriate strength to determine the amounts of H2O and HDO in the atmosphere during the measurement.

The proper handling of interfering species is a well-known problem in FTIR remote sensing. An interfering species generates spectral signatures in one or more MWs which are applied in the retrieval of the target species. H2O (and its isotopic variants) is among the most awkward interfering species, because it is highly variable even on short timescales. Both the total column of H2O as well as its vertical VMR profile shape show significant variability. A useful review of the interference problem can be found in a prior publication by Sussmann et al. (ACP, 7, 3537-3557, 2007). In the case under consideration here, Sussmann et al. conclude that the H2O (HDO) interference is the main obstacle in achieving a CH4 measurement precision sufficient to observe the seasonal variability at wet sites. In our feeling, the best strategy to attack this problem on the CH4 column encompasses two measures: (1) select CH4 MWs which minimize the H2O (HDO) impact and (2) include profile retrievals of HDO and H2O. To avoid an impact by those CH4 signatures located in the auxiliary MWs on the CH4 solution, the derivatives wrt CH4 are set to zero in MWA and MWD during the retrieval process. In MWD we deweighted the spectral subset from 2941.88 to 2942.07 cm-1 to minimize the impact of a weak H2O line with inconsistent broadening parameters.

The CH4 windows suggested here (MWB and MWC) suffer significantly less from H2O (HDO) interference than do MW3 and MW5 favoured by Sussmann et al.. Unfortunately, current HITRAN line lists do not describe the CH4 line blend in MWC perfectly well, so we decided to apply an ad-hod correction of HITRAN 2008 line parameters to further improve fit quality. The selection of MW3 and especially MW5 in MIR-GBM seems implausible: while diagnosing that H2O (HDO) interference is the main problem, they retain MWs with significant interference in their MW set - under tropical conditions HDO becomes the leading absorber in MW5!

Figure 1 shows a fit to a spectrum recorded in Karlsruhe (date 100720, UT 084843, average summer H2O column) covering MWA to MWD. Figure 2 shows a fit to the same spectrum using MWs 1, 3, 5 as proposed by Sussmann et al.. Figures 3 and 4 show sample fits for Izana and Kiruna (in winter, early February). The fit quality is satisfying for all stations, the systematic residuals for Kiruna noticeable in the CH4 line centers could be reduced by relaxing the smoothness constraint on the CH4 profile, allowing for an NDOF of about 3 (magenta residuals in Fig. 4) instead of 2. Due to subsidence of air during polar winter, horizontal wind shear and mixing across the vortex boundary the stratospheric part of the CH4 profile can be very variable at a polar site, so the MIR-GBM recommendation of NDOF = 2 (see page 2978 of Sussmann et al.) and an altitude-constant regularization seems to be suboptimal in such conditions. But this would open another point of discussion while our primary concern is the selection of MWs. We therefore stick to the MIR-GBM recommendation (NDOF = 2) for all retrieval setups investigated here.

Finally, the decision to remigrate to outdated HITRAN line lists as recommended by Sussmann et al. is not fully satisfactory. For example, in MW1 (MWB), the fit residuals indicate that HITRAN 2008 is superior (note e.g. line position of CH4 line at 2614.02 cm-1 and strength of H2O signature at 2614.50 cm-1).

Intercomparison of timeseries of different retrieval setups

In this Section we compare the three different CH4 retrieval schemes (MIR-GBM, MIR-GBM refined, our approach) for three FTS sites which cover a considerable range of relevant station parameters (ground pressure, atmospheric temperature, humidity): the polar site Kiruna (northern Sweden), the midlatitude site Karlsruhe (Germany), and the high-altitude observatory Izana (Tenerife). Karlsruhe is located in the Rhine valley, during summer H2O columns similar to those observed in Wollongong (the wettest site used in the Sussmann et al study, see Fig. 6 therein) occur frequently. For the refined MIR-GBM setup: we use the MWs and line lists as in the original MIR-GBM setup, but instead of scaling a climatological H2O profile for each station we scale daily mean VMR profiles for H2O and HDO derived from separate dedicated H2O and HDO profile retrievals.

In Fig. 5, we show ratios of columns for different retrieval setups for the measurements taken in Karlsruhe. A nonzero slope for the ratio MIR-GBM refined / MIR-GBM is found, probably because the H2O (HDO) profile shape varies as function of total column. It is reasonable to assume that the MIR-GBM refined scheme is the superior choice over MIR-GBM, it is in better agreement with our setup at all investigated sites (see Figs. 6 + 7 for Kiruna and Izana). The column ratio “our approach / MIR-GBM refined” is nearly independent of H2O column for Karlsruhe and Kiruna. There is a residual slope for Izana. To gain further insight we correlated short-term variations (applying a high-pass filtering on the time series) of the H2O column with associated changes of the tropospheric CH4 VMR for Izana. The results are shown in Fig. 8 for both retrieval setups. The correlations differ between MIR-GBM refined and our approach – our approach shows a stronger anticorrelation. However, it would be premature to conclude from this result that MIR-GBM refined is the setup with least H2O interference because a certain amount of anticorrelation between CH4 VMR and H2O column is expected in this statistical analysis. (Roughly simplified: higher H2O columns correlate with tropical origin of observed airmass + latitudinal gradient of CH4 VMR on northern hemisphere). Further studies using the in situ night time CH4 VMR measurements (~representative for the free troposphere) performed at Izana could help to establish the correct correlation from independent measurements.

Conclusions

We have shown that our retrieval setup generates CH4 columns in good agreement with the MIR-GBM and MIR-GBM refined setup. MIR-GBM and MIR-GBM refined results show different correlations with the H2O column. Our scheme selects CH4 MWs which minimize perturbing H2O (HDO) absorptions and it handles the problematic interference species H2O (HDO) in a rigorous manner. Even when compared to the MIR-GBM refined setup, one might expect that our scheme will prove advantageous for extremely wet sites (e.g. tropical FTS station in Suriname, ship measurements in the tropics, sea-level FTS on La Reunion). In these conditions the HDO absorption in MW5 will essentially cover the target CH4 signature. However, the scheme proposed here is still tentative. It might be e.g. be possible to replace MWC by another choice (avoiding an ad-hoc adjustment of CH4 HITRAN 2008 line parameters) or MWD by another H2O window which allows for a better fit quality. Finally, one could merge certain aspects of our suggestion with other proposals. We hope that the discussion on the NDACC CH4 retrieval recipe will soon converge towards a solution which is positively rated by all NDACC infrared working groups and will satisfy the precision demands for all sites for years to come.

Tables

Table 1: MWs

MW ID	MW bounds	Main absorbers	comments
A	2611.60,2613.35 cm-1	HDO, CO2	Auxiliary MW, CH4 Jacobian zeroed
B	2613.70,2615.40 cm-1	CH4, CO2 weak: HDO	Corresponds to MW1 in Sussmann et al.
C	2914.70,2915.15 cm-1	CH4, NO2 weak: H2O, HDO	slight adjustments of HIT08 CH4 line parms on CH4 line blend performed
D	2941.23,2942.23 cm-1	CH4, H2O weak: O3	Auxiliary MW, CH4 Jacobian zeroed Deweighting of residuals from 2941.88 to 2942.07

Figures

Fig. 1: Fits in MWA to MWD for spectrum 100720/084843SC recorded in Karlsruhe. (Black: measurement, red: calculation, blue: expanded residual, dashed line in MWD: deweighted region). Line lists taken from HITRAN 2008 (update H2O 2009 included, CH4 modified to match the line blend in MWC)

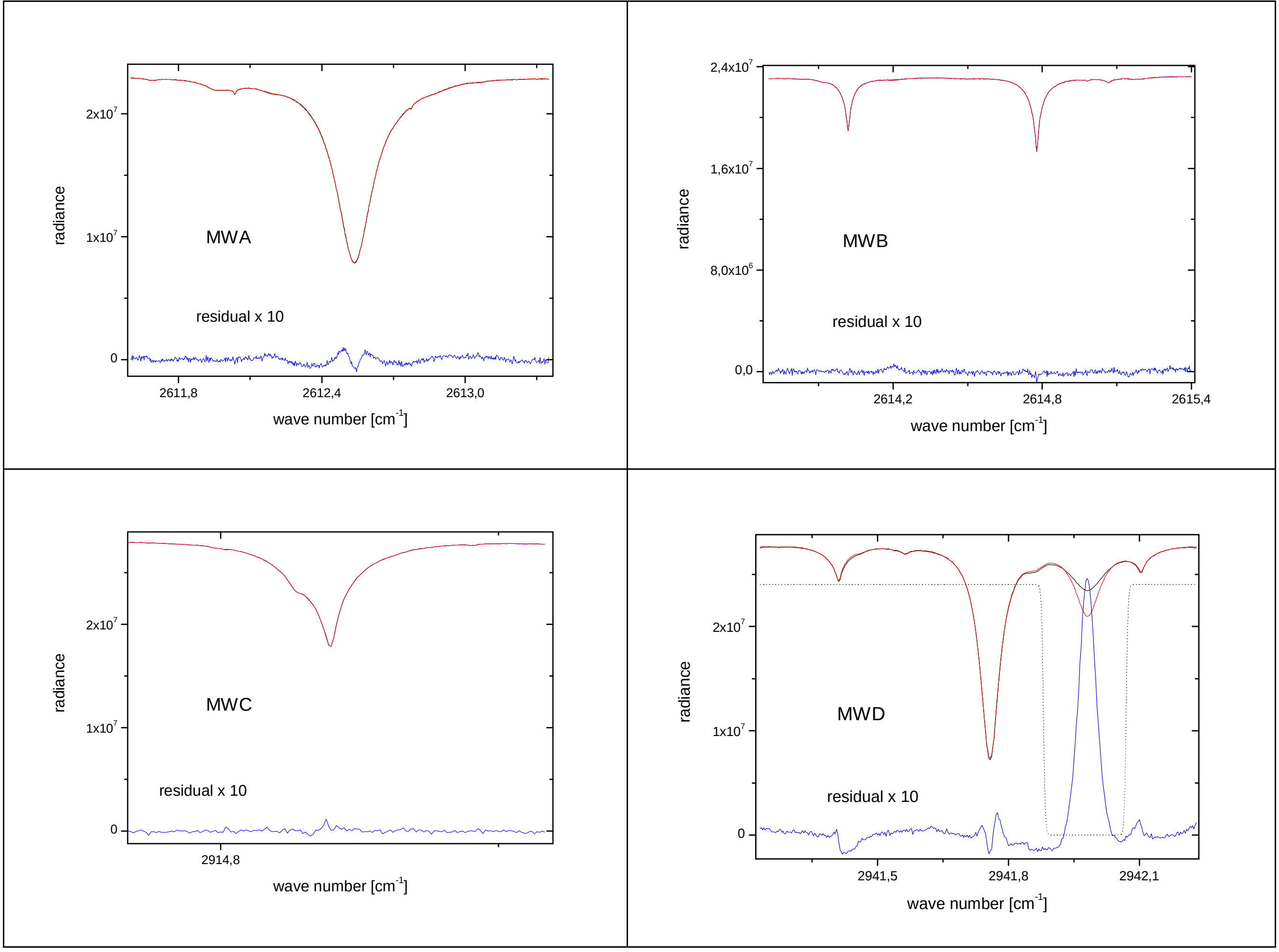


Fig. 2: Fits in MW1, MW3, and MW5 for spectrum 100720/084843SC recorded in Karlsruhe. (Black: measurement, red: calculation, blue: expanded residual). Line lists as recommended in MIR-GBM retrieval setup

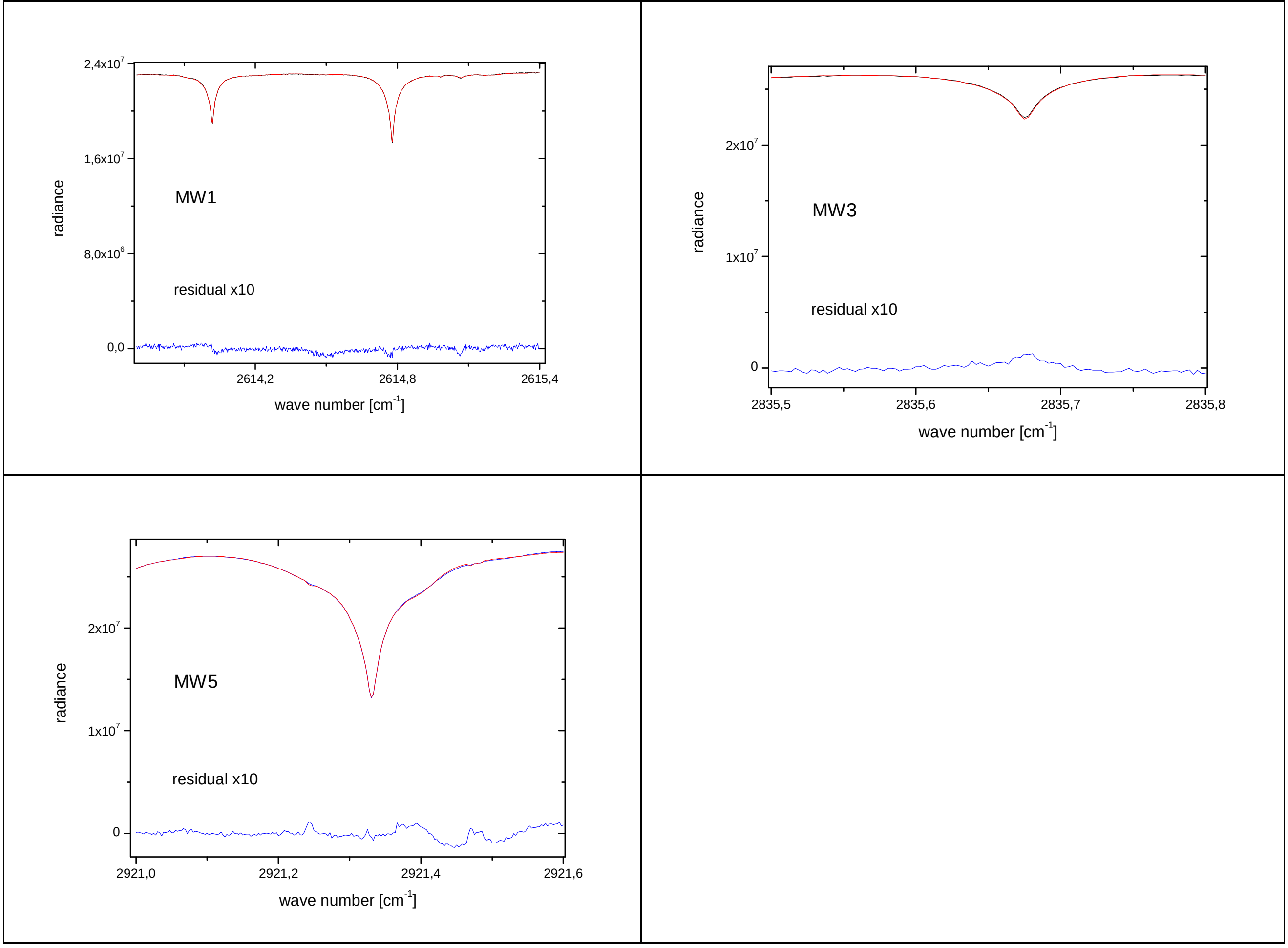


Fig. 3: Fits in MWA to MWD for spectrum 100527/171339SC recorded at the Izana observatory. (Black: measurement, red: calculation, blue: expanded residual, dashed line in MWD: deweighted region). Line lists taken from HITRAN 2008 (update H2O 2009 included, CH4 modified to match the line blend in MWC)

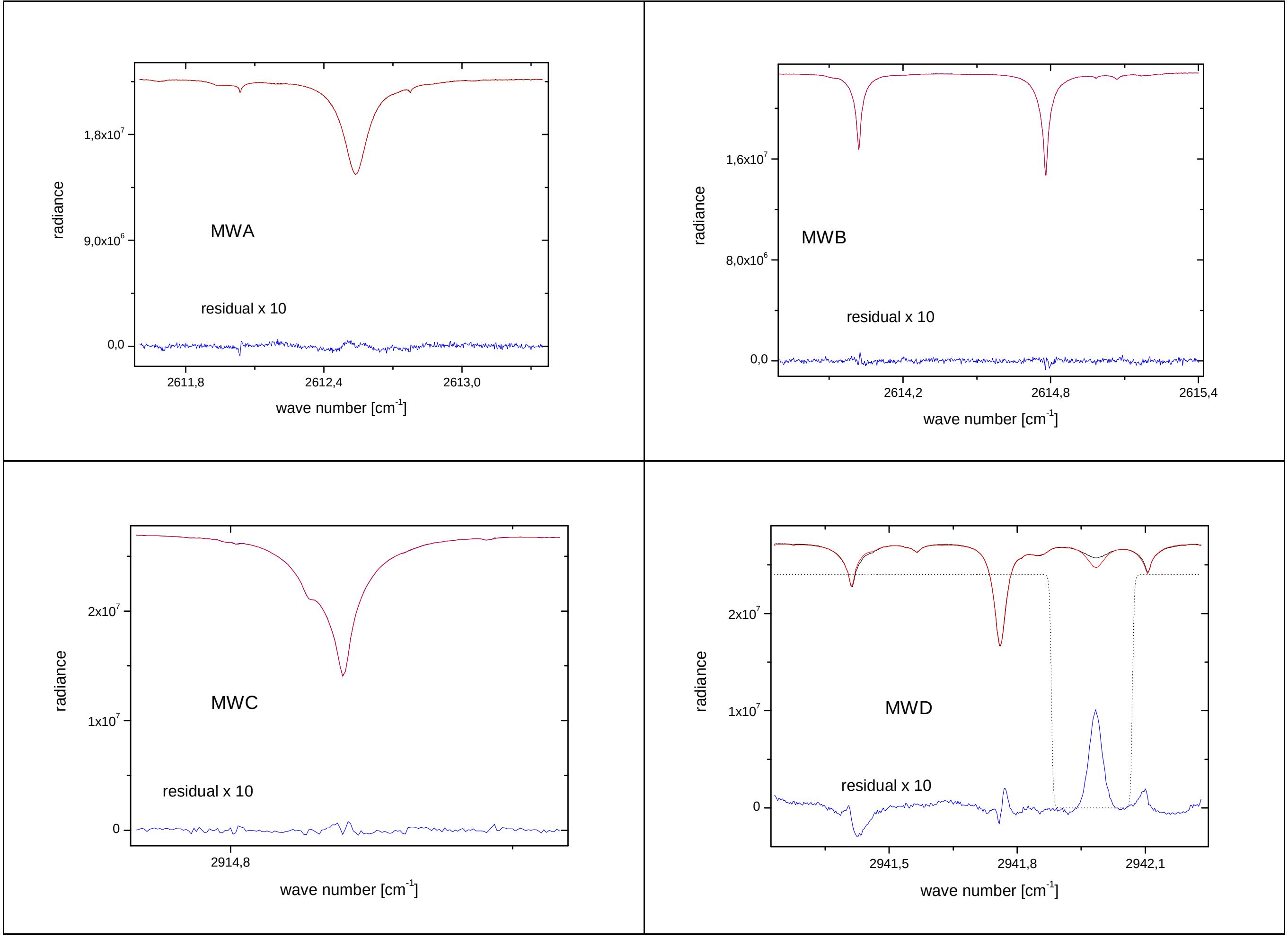


Fig. 4: Fits in MWA to MWD for spectrum 100205/101359SC recorded with the Kiruna FTS. (Black: measurement, red: calculation, blue: expanded residual, dashed line in MWD: deweighted region). Line lists taken from HITRAN 2008 (update H2O 2009 included, CH4 modified to match the line blend in MWC). The magenta fit residuals (vertically shifted for clarity) result if the smoothness constraint on the CH4 profile is reduced.

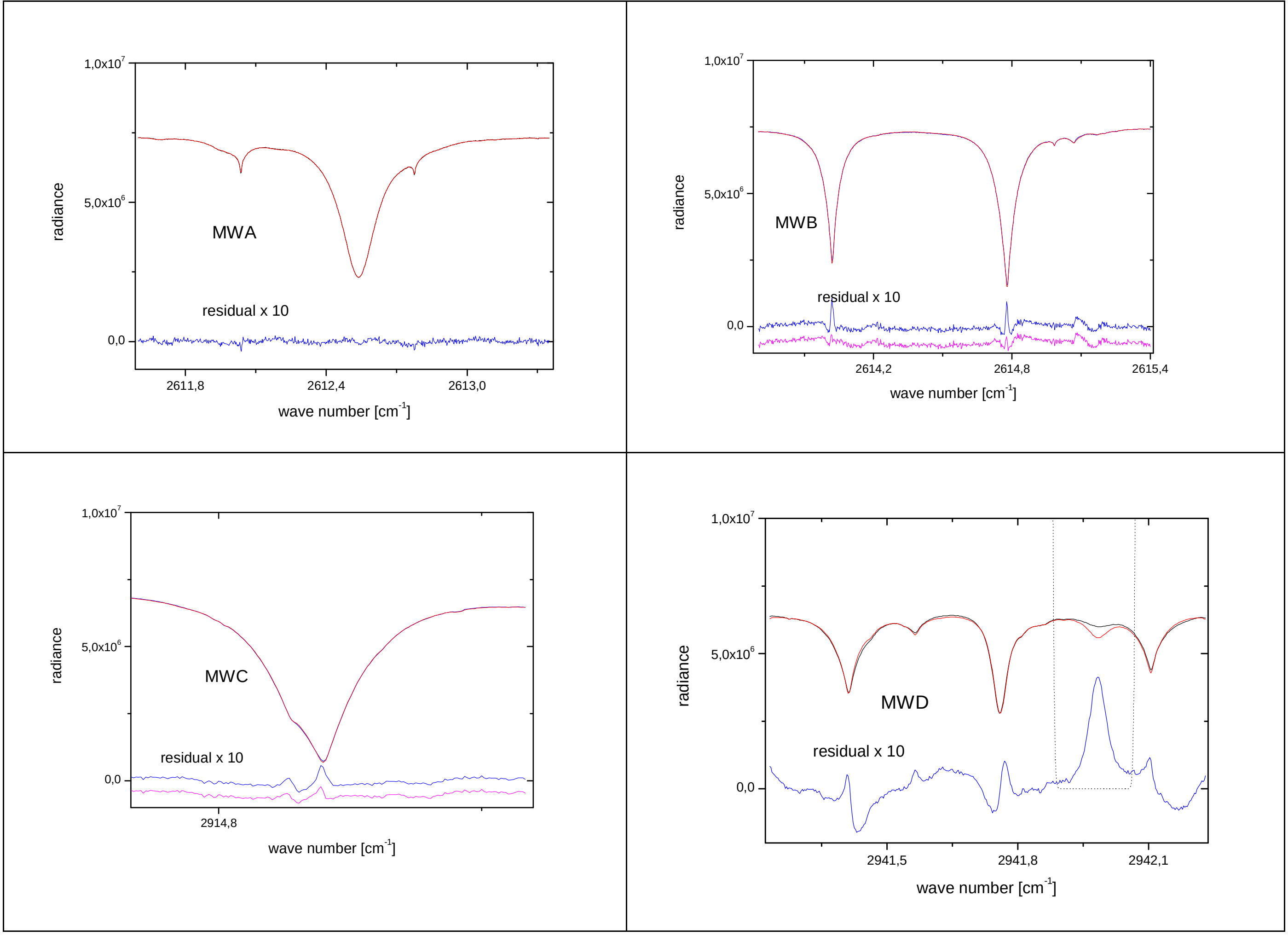


Figure 5: different ratios of retrieved CH4 columns as function of H2O column for Karlsruhe (to be compared with Fig. 6 in Sussmann et al.)

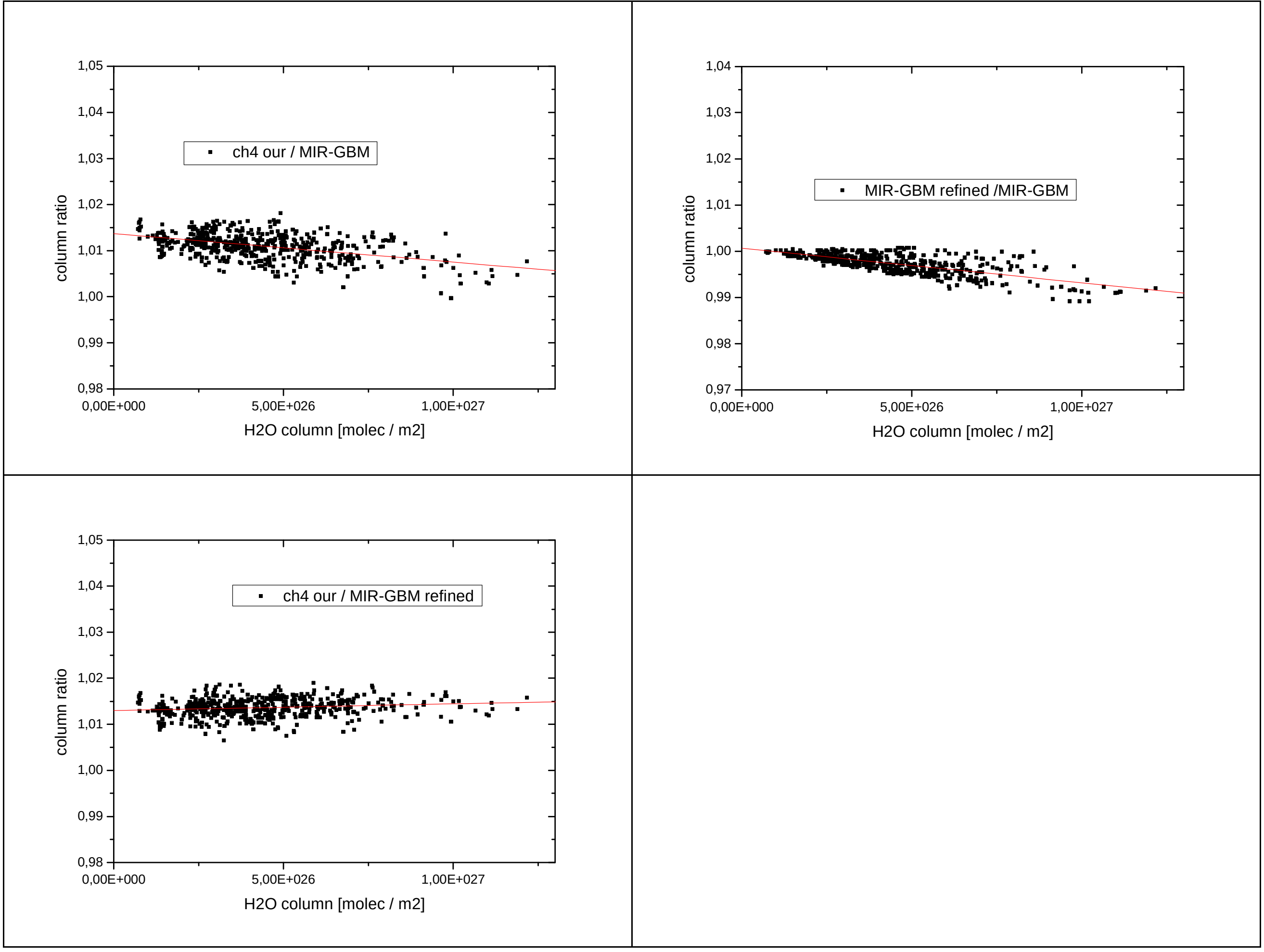
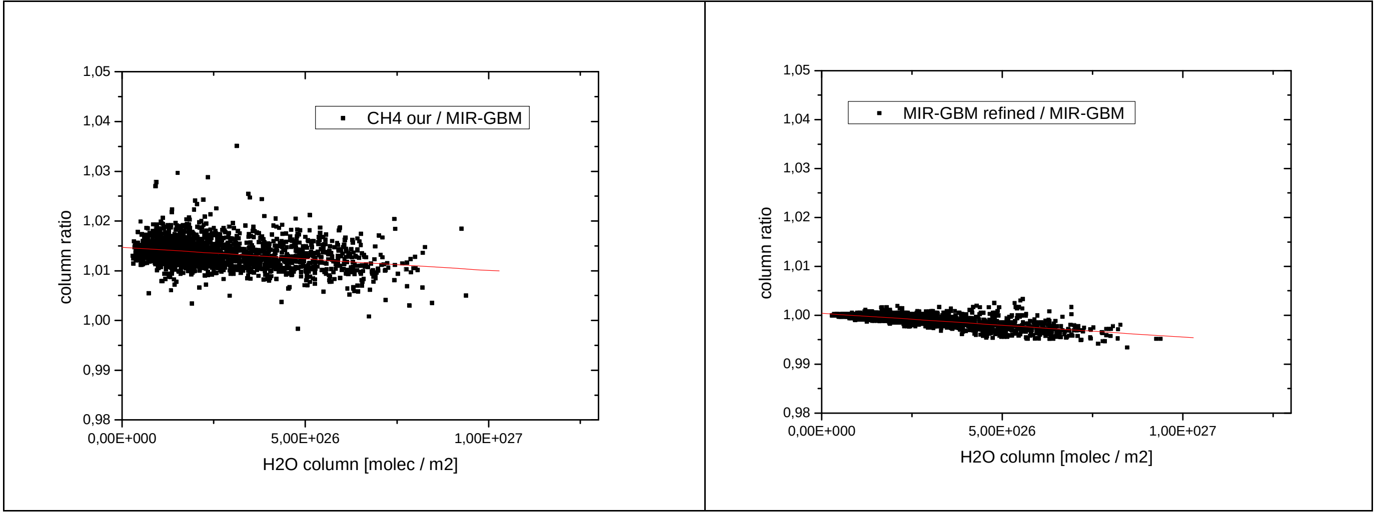


Figure 6: different ratios of retrieved CH4 columns as function of H2O column for Kiruna (to be compared with Fig. 6 in Sussmann et al.)



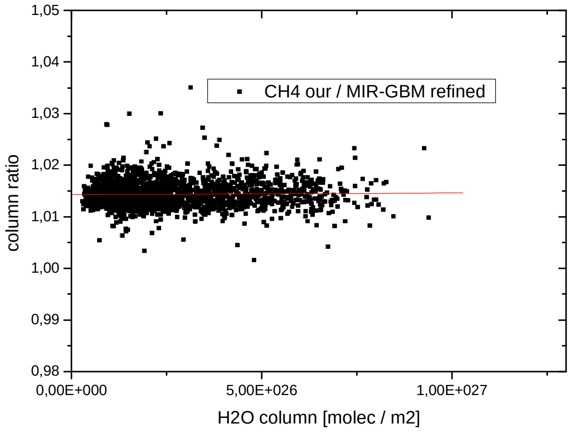


Figure 7: different ratios of retrieved CH4 columns as function of H2O column for Izana (to be compared with Fig. 6 in Sussmann et al.)

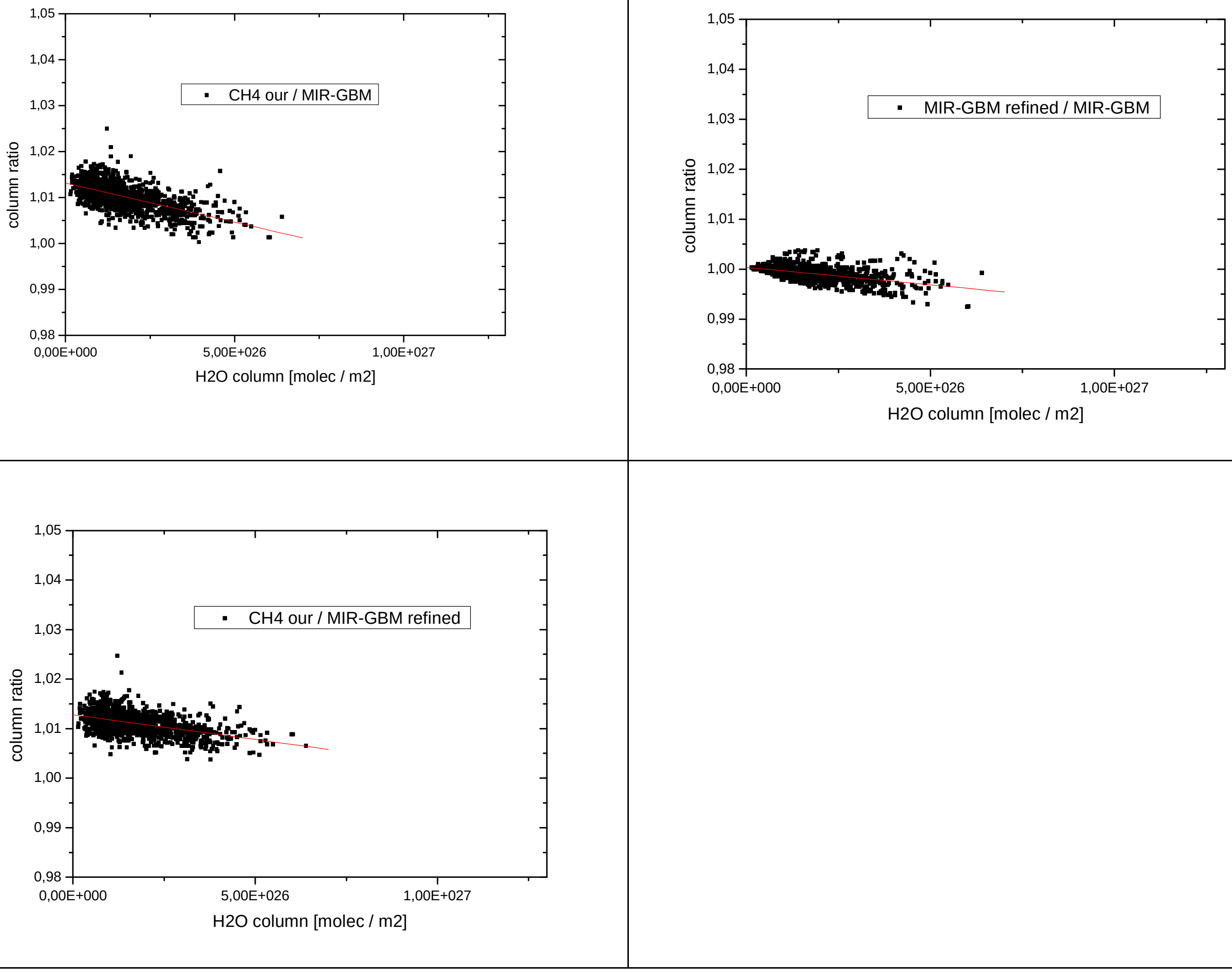


Figure 8: Correlations of CH4 VMR changes with H2O column changes at Izana