

Interactive comment on “Profiling of CH₄ background mixing ratio in the lower troposphere with Raman lidar: a feasibility experiment” by Igor Veselovskii et al.

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Comment on the manuscript “Profiling of CH₄ background mixing ratio in the lower troposphere with Raman lidar: a feasibility experiment” by Igor Veselovskii, Philippe Goloub, Qiaoyun Hu, Thierry Podvin, David N. Whiteman, Michael Korenskiy, and Eduardo Landulfo.

The manuscript presents a Raman lidar for remote measurements of methane. The approach of the authors is generally valid. The authors are renowned experts in the development of Raman lidar systems, as well as in the simulation of the performance of such systems and the analysis of the collected data.

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Nevertheless, there are several points need to be addressed in more detail:

Reproducibility of the results: Over the last decade I have looked for a methane Raman signal at 2914 cm⁻¹ with three multi-channel spectroscopic lidars: at Tsukuba, Japan (Sugimoto et al. 2012), Gwangju, Korea, and now at Hatfield, United Kingdom. During the work with all those instruments I have never managed to detect methane background signals as shown in the manuscript when using a laser power for the emitted light comparable to the one available to the authors. The multi-channel lidars I have worked with are all based on spectrometric and long-pass edge filter isolation of Raman lines rather than single bandpass interference and notch filters as used by the authors. The system in Japan used 100mJ@355nm at 30Hz repetition rate and a 100 cm telescope. At Gwangju we used about 200mJ at 10Hz and a 40 cm telescope. We are not able to observe the background methane signal even with a laser energy of about 300mJ at 10Hz (40 cm telescope) in the spectrometric lidar system at Hatfield. With all these systems we can observe nitrogen and H₂O Raman signals with counting rates of tens or even hundreds MHz when using emission energy below 200mJ, but nothing above the noise levels in the 396nm channel.

Signal isolation: The filter the authors use to isolate the methane line should be described in more details. In fact, the Alluxa interference filter (395.7-0.3 OD12 Ultra Narrow Bandpass Filter) has a rejection ratio (optical depth, OD) of 12 only for some wavelengths. According to the manufacturer's web page (<https://www.alluxa.com/optical-filter-catalog/ultra-narrow-bandpass/395-7-0-3-od12-ultra-narrow-bandpass.html>) this filter has “Blocking Range(s) OD12 (By Design): 353 to 389 nm, 403 to 443 nm, 485 to 540 nm; OD5: 300 to 353 nm, 443 to 485 nm, 540 to 1100 nm”. The filter has OD5 for some of the pure-rotational anti-Stokes lines around 352nm. Using an additional notch filter can provide a good suppression of the pure rotational Raman signal. However, the optical depth is OD5 for almost all anti-Stokes Raman spectra (351 nm to 309 nm) including anti-Stokes scattering by nitrogen, oxygen, and H₂O molecules. The optical depth is OD5 for wavelengths larger than 540nm which includes the Raman signals by

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nitrogen, oxygen and H₂O when using a laser at 532nm. The authors should provide the curves of ATR (Attenuation-Transmission and Reflection) of the particular filter and discuss the suppression/rejection ratio for pumping of the anti-Stokes band by 355nm as well as excitation of the Stokes band of Raman spectra pumped at 532nm.

Interference with other signals: Although the methane Raman line appears to be well isolated there are other Raman lines of atmospheric compounds very close to it. Namely: Propane line at 2890cm⁻¹ (395.3nm) - 0.4nm shift from the author's Interference filter peak transmission, Ethanol 2943cm⁻¹ (396.1nm) - 0.4nm shift from the author's Interference filter peak transmission with Raman cross section of the same magnitude as methane, Methanol 2v6 line at 2955cm⁻¹ (396.3nm) - 0.6nm shift from the Interference filter peak transmission which can be connected to the methane cycle and its interaction with water vapour.

Lack of reference data: As the study assesses the feasibility for profiling of methane with a modified multiwavelength Raman lidar, the authors have to compare their findings to independent data (especially regarding the background concentration) from in-situ measurements or DIAL observations. The conclusion made on page 1, line 20 "The measured methane profiles do not correlate with aerosol backscattering, which corroborates the hypothesis that, in the PBL, not aerosol fluorescence but CH₄ is observed." is no proof of concept unless supported by independent observations. In the present form, the reliability of the results is highly speculative.

Measurement setup: Additional information regarding detector sensitivity and parameters (high voltage and discriminator levels) should be provided. If we take a background concentration of 2ppm and the ratio of Raman backscatter cross sections (methane to nitrogen) of 8, the nitrogen signal should be approximately 1.6×10^5 (160000) higher than the methane signal. However, Fig. 2 shows a nitrogen signal (378nm) that is only approximately 100 times higher than the methane (396nm) signal. If one accounts for the 10% beam splitter applied for the 378nm channel, then the nitrogen signal seems to be 1000 times higher than the methane signal. Hence, the counting rates in the 396-

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nm channel appear to be about two orders of magnitude higher than expected. The authors need to provide information on how they managed to obtain that high count rate.

Data consistency: The authors assume that the background methane concentration in the free troposphere is 2ppm and can differ in the PBL "inside the planetary boundary layer (PBL)" (Page 2, line 47). Yet they present enhanced methane mixing ratios at altitudes far above the PBL. In Fig. 6, the methane mixing ratio is significantly higher from 3000m to 5000m. The authors should discuss possible mechanisms that could lead to the formation of methane plumes in these height ranges and/ or persist in the free troposphere.

Fluorescence: Sugimoto et al. (2012) show that fluorescence can be observed in case of pumping at 355nm. Though the fluorescence maximum has been observed at about 460-470nm, fluorescence interference should be considered as an interfering factor in measurements for which optical pumping at 355nm is used.

Reference: Nobuo Sugimoto, Zhongwei Huang, Tomoaki Nishizawa, Ichiro Matsui, and Boyan Tatarov, "Fluorescence from atmospheric aerosols observed with a multi-channel lidar spectrometer," *Opt. Express* 20, 20800-20807 (2012)

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