

We are grateful to Boyan Tatarov for reading the manuscript and providing important suggestions. Raman measurements at ppm level are challenging and many factors should be considered.

*“Reproducibility of the results: Over the last decade I have looked for a methane Raman signal at 2914 cm<sup>-1</sup> 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 H<sub>2</sub>O 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.”*

We are not the only ones who see the Raman signal from background methane, for example (Heaps, Wm. S. and Burris 1996). Keeping in mind that cross section of methane is about 8 times higher than that of nitrogen, for 2ppm mixing ratio the methane signal should be  $10^6/16=6*10^4$  weaker than that of N<sub>2</sub>. For counting rate 100 MHz in nitrogen channel, methane signal should be ~1.7 KHz, which is low but definitely detectable.

*“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*

Actually  $6*10^4$

*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-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.”*

Yes, it is important to compare nitrogen and methane Raman signals. However lidar signals in Fig.2 are not calibrated and can't be used for this: only small portion of spitted nitrogen Raman backscatter was used for the measurements. To perform calibration in Fig.1 we show nitrogen Raman signal before system modification (normal operation) and after. Before modification we glued analog and photon counting signals so equivalent counting rate at 1000 m height is about 500 MHz. Comparing the signals we estimate attenuation in modified N<sub>2</sub> channel to be about factor 185. Methane signal is also shown in Fig.1 and at 2000 m it is about  $5*10^4$  lower than

nitrogen signal. Transmission of N<sub>2</sub> interference filter was slightly lower than that in CH<sub>4</sub> channel, so we find the agreement between nitrogen and methane signals to be very reasonable.

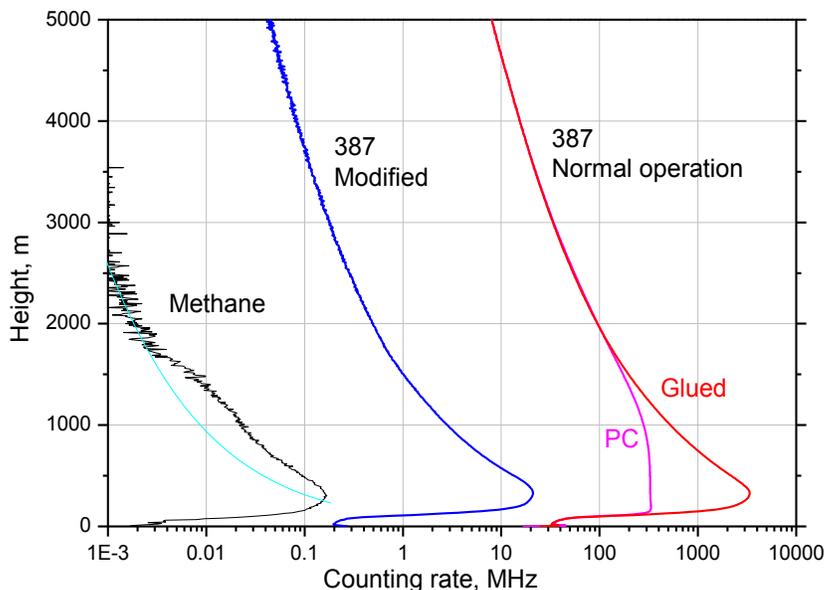


Fig.1.

All these estimations are added to the revised manuscript.

*“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/opticalfilter-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 H<sub>2</sub>O molecules. The optical depth is OD5 for wavelengths larger than 540nm nitrogen, oxygen and H<sub>2</sub>O 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.”*

The filter transmission curve, simulated by Alluxa, is shown in Fig.2.

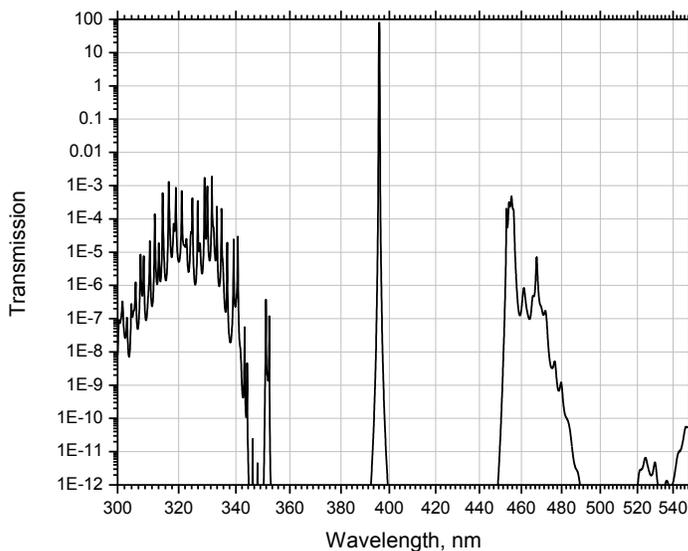


Fig.2. Transmission curve of the methane filter (simulation).

Filter transmission at 395.7 is about 80%, so suppression of rotational lines near 350 nm was more than OD9. Additional notch filter from Edmund ( 355nm TECHSPEC® OD>4 Notch Filter), provided OD4 suppression of rotational anti-Stokes line down to 348 nm. Dichroic mirror at the entrance of the lidar receiving module removed 90% of UV radiation in 340-360 nm range. So total suppression of rotational Raman signal was more than OD13. Suppression of signals in 300-340 nm range by filter is above OD6, transmission of optical path in lidar receiving module for this wavelength range was less than 5%, so total suppression of OD7 should be sufficient to remove anti-stokes Raman scattering of nitrogen and oxygen. To check the influence of 532nm scattering and corresponding Raman components, during test measurements we inserted an additional UV filter with transmission of less than 5% in 500 – 750 nm range. No noticeable changes in CH<sub>4</sub> Raman signals were discovered. Moreover, during filter testing, we inserted the Glan prism in laser beam to remove 532 nm radiation. Again, we didn't see noticeable changes in CH<sub>4</sub> signal. So we don't expect that 532 nm radiation scattering can be an issue. But definitely, in our future measurements we will try to improve transmission characteristics of CH<sub>4</sub> channel by using additional filters. We added the filter transmission curve to revised manuscript.

*“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<sup>-1</sup> (395.3nm) - 0.4nm shift from the author's Interference filter peak transmission, Ethanol 2943cm<sup>-1</sup> (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<sup>-1</sup> (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.”*

In some special cases (extreme industrial emissions?) such interference probably may occur, but we don't expect it in free troposphere. Besides shift of ~0.4 nm significantly reduces interfering signals.

*“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 CH<sub>4</sub> is observed.” is no proof of concept unless supported by independent observations. In the present form, the reliability of the results is highly speculative.”*

We agree with Boyan, that independent reference data are critically important. Unfortunately it is not easy to get vertical distribution of CH<sub>4</sub>. We plan to use Fourier spectrometer measurements in future for validation.

*“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.”*

We discussed in the manuscript, that methane could be generated during the forest fires. On another hand, at a moment we can't completely exclude possibility the interference of fluorescence of aerosol or gases.

*“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 multichannel lidar spectrometer," Opt. Express 20, 20800-20807 (2012)”*

Yes, as mentioned, in future measurements we plan to introduce the control channel at 393 nm to monitor possible fluorescence. The reference is added to revised manuscript.