

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

Article of Veselovsky et al. "Profiling of the CH₄ background mixing ratio in the lower troposphere with Raman lidar: a feasibility experiment" is devoted to the problem of remote control of small gas components of the atmosphere. At the same time, the problem of monitoring the total content and spatial distribution of the concentration of greenhouse gases is of particular practical importance in terms of predicting climate change. Methane is one of the most important climate-forming components of the atmosphere, and therefore there is an obvious need for the development of new methods and means of monitoring methane content in the atmosphere. The authors provide a fairly comprehensive analytical overview of the achievements in the field of remote monitoring of methane content in the atmosphere and substantiate the prospects for applying the Raman effect to create a lidar technology for remote monitoring of the vertical distribution of methane concentration.

Taking into account that we are talking about background concentrations of methane (about 2 ppm), experimenters understand that the determination of such low concentrations using the Raman effect is a very difficult task. However, as we see, the difficulties do not stop the authors and as convincingly shown in the publication, this problem can be successfully solved.

Obviously, the authors of the publication are experienced experimenters, lidar developers, who understand well the problems of applying the Raman effect and are able to find the right technical solutions. This is evident from the fact how carefully the authors relate to solving the problem of spectral suppression of possible interfering factors, such as unshifted scattering laser line, an overtone of oxygen, which the experimenters do not always remember, Raman on liquid water in the clouds. At the same time, the samples of interference filters made by the newest technology and having unique characteristics are correctly selected.

It also seems appropriate to carry out preliminary mathematical modeling in order to estimate the expected magnitude of lidar responses, and determine the accumulation time of signals to achieve a given accuracy of measuring the mixing ratio. And the fact that in real experiments the magnitude of lidar responses in the methane channel corresponds to the calculated values is an indirect confirmation of the correctness of the chosen technical solutions.

It should be noted that the most difficult task of lidar measurements of background concentrations of greenhouse gases using the Raman effect in the case of methane is somewhat simplified due to the extremely large scattering cross section of the methane molecule ($1.9 \cdot 10^{29} \text{ cm}^2/\text{sr}$), which is more than 8 times larger than the cross section of the nitrogen molecule. However, even in this case, experimenters are required to display not a small amount of mastery in constructing equipment and taking measurements.

As shown in the publication, the results of lidar measurements of the vertical course of the methane mixing ratio do not contradict generally accepted ideas about the spatial distribution of methane in the atmospheric boundary layer. The absence of a direct correlation between the aerosol backscatter coefficient and the methane mixing ratio indicates a sufficient level of suppression of the unshifted scattering noise and possible liquid water Raman signal.

However, as the authors rightly point out in conclusion, it is impossible to completely exclude the fluorescence factor of an aerosol, given the high sensitivity of the lidar at the level of ppm units. The authors rightly say that the Raman lidar for sensing of water vapor mixing ratio can be easily converted into a lidar for methane sensing. However, the question of the

contribution of aerosol fluorescence to the Raman signal of methane remains open. All the same, to ensure correct measurements of the methane mixing ratio, it is necessary to build a special lidar, equipped with a more powerful laser and a special channel to control the level of aerosol fluorescence. The authors understand this and plan to carry out such work in the future. As for my opinion, I consider it more reliable to carry out research using a multichannel spectrometer, which makes it possible to see the spectral image of the lidar response and to interpret various spectral components of the signal.

Nevertheless, the presented publication convincingly proves the possibility of lidar measurements of the vertical distribution of the methane mixing ratio using lidar. It is obvious that, in the main, the presented results reflect the real vertical distribution of methane concentration. However, as long as there is no control over the level of aerosol fluorescence, it is difficult to trust the measurements of the methane mixing ratio inside the aerosol layers. The magnificent hypothesis of methane brought in the composition of the products of combustion of forest fires (Fig. 6) can also be explained as a result of the fluorescence of smoke aerosol and products of combustion or sublimation of the organic components of wood.

Summarizing, it should be stated:

1. The paper addresses relevant scientific questions within the scope of AMT.
2. The paper presents novel concept of Raman lidar technique, new tool for monitoring of methane mixing ratio, data of methane vertical distribution.
3. Substantial conclusions are mostly reached.
4. The scientific methods and assumptions are valid and clearly outlined.
5. The results are sufficient to support the interpretations and conclusions.
6. The description of experiments and calculations is sufficiently complete and precise to allow their reproduction by fellow scientists.
7. The authors give proper credit to related work and clearly indicate their own original contribution.
8. The title clearly reflects the contents of the paper.
9. The abstract provide a concise and complete summary.
10. The overall presentation is well structured and clear.
11. The language is fluent and precise.
12. Mathematical formulae, symbols, abbreviations, and units are defined and used correctly.
13. Text, formulae, figures and tables are satisfied demands for scientific publications.
14. The number and quality of references are quite appropriate.
15. The amount and quality of supplementary material is appropriate.

I believe that the article under discussion can be published in AMT without significant modifications and changes.

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