

This paper presents a modeling approach that integrates least-squares curve fitting to improve the accuracy of gas concentration estimation using tunable diode laser absorption spectroscopy (TDLAS). A mathematical model accounts for absorption, scattering, and reflectance effects. The Levenberg–Marquardt algorithm is applied to extract the fundamental and even harmonic components, enabling precise determination of gas concentration. Experimental results demonstrate a strong correlation between the absorption pulse amplitude and methane concentration with the coefficient of determination R^2 value of 0.989. The proposed method proves efficient, requiring only a few signal cycles for gas concentration estimation, making it suitable for real-time monitoring systems.

Keywords: gas concentration, least-squares curve fitting, even harmonic, harmonic signal.

Introduction. Gas concentration measurement is critical for a wide range of applications, from early fire detection to monitoring air pollution and detecting hazardous gas leaks in pipelines. The ability to accurately detect and measure gases is essential for ensuring safety, environmental protection, and regulatory compliance across various industries. These technologies are also instrumental in monitoring landfills, agricultural sites, and identifying toxic gases, all of which have significant implications for public health and the environment. Furthermore, advancements in mobile gas detectors and measurement systems continue to enhance accessibility and precision, enabling real-time monitoring in diverse settings.

One of the most promising approaches for remote methane detection, even at extremely low concentrations, is optical methods. Tunable diode laser absorption spectroscopy (TDLAS) is among the most effective optical techniques for this purpose. There are three primary variations of TDLAS: direct absorption spectroscopy (DAS) [1], wavelength modulation spectroscopy (WMS) [2], and wavelength modulation-direct absorption spectroscopy (WM-DAS) [3]. The WM-DAS method combines the simplicity and calibration-free benefits of direct absorption spectroscopy with the high sensitivity and noise rejection capabilities of harmonic detection, making it a powerful and reliable technique for methane concentration estimation.

Our method integrates a modeling approach, harmonic analysis and least-squares curve fitting to enhance the accuracy of gas concentration estimation using TDLAS with WM-DAS method. Our models of transmitted laser intensity and the absorption pulse signal are proposed, which are capable of achieving excellent performance with a low computational requirement.

Methodology. To accurately describe and measure gas concentration using tunable diode laser absorption spectroscopy (TDLAS) technology in a complex environment, an additive environmental model of the transmitted laser intensity signal is proposed, taking into account absorption, scattering, and reflectance processes, as described below:

$$I_{tr}(t, K_{env}) = (I_{DC}(t) + I_{RAW}^{f_1}(t) + I_{abs}(t)) \cdot K_{env} + N_{noise}(t) \quad (1)$$

where $I_{RAW}^{f_1}(t) = A_1 \cos(2\pi f_1 t) + B_1 \sin(2\pi f_1 t)$ is the fundamental harmonic related to direct absorption spectroscopy-wavelength modulation (DAS-WM) technology [3],

$I_{abs}(t) = \sum_{k=1}^{K_H/2} (A_{2k} \sin(2\pi \cdot 2kf_1 \cdot t) + B_{2k} \cos(2\pi \cdot 2kf_1 \cdot t))$ is the absorption pulse signal

caused by absorption gas, $\{A_1, B_1\}$ are the linear weight coefficients of the fundamental harmonic, $\{A_{2k}, B_{2k}\}$ are the linear weight coefficients of the even harmonics, K_{env} is the environmental attenuation coefficient due to atmosphere scattering and surface reflection, $K_H = f_s / (2 \cdot f_1)$ is the number of the significant harmonics, $N_{noise}(t)$ is the additive random environment noise, $I_{DC}(t)$ is the DC component, $I_{abs}(t)$ is the absorption pulse signal, f_s is a sampling frequency, f_1 is the fundamental frequency, t is the sample index.

The absorption pulse signal, fundamental harmonic signal, DC component and the attenuation coefficient can be extracted using the Levenberg–Marquardt algorithm [4] is in the least-squares curve fitting problem: given a set of m empirical samples $(t, I_{tr}(t, K_{env}))$ of independent and dependent variables, find the optimal parameters $\beta_{opt} = \{A_1, B_1, I_{DC}, K_{env}\} \cup \{A_{2k}, B_{2k}\}_{k=1, \dots, K_H}$ of the model curve so that the sum of the squares of the deviations is minimized:

$$\beta_{opt} = \underset{\beta}{\operatorname{argmin}} \sum_{t=1}^m \left(I_{tr}(t, K_{env}) - (I_{DC}(t) + I_{RAW}^{f_1}(t) + I_{abs}(t)) \cdot K_{env} \right)^2 \quad (2)$$

Experiment. The laser dataset provided by the department of remote control systems is consist of the digital measured absorption signal with the following parameters: a sampling frequency of $3.2 \cdot 10^5$ Hz, a base frequency of 10 kHz, a resolution of 24 bits per sample and signal lengths $m=32768$ samples.

The experiment was performed with laser output power of 9 nW passing through a methane volume with different concentrations: 100 ppm, 250 ppm, 540 ppm, 1000 ppm and 2000 ppm. For each of five concentration value, 5 measured signals are used.

The absorption pulse amplitude is defined as the sum of the harmonic components' amplitudes divided by the amplitude of the fundamental component. A curve-fitting method [4] is applied to determine the parameters A_{2k} and B_{2k} with the selected maximum order of harmonic component $K_H = 7$. Relationship between amplitude of absorption pulse and methane concentration is shown in **Figure**.

Experimental results show that the correlation coefficient $R^2=0.989$ is close to 1, which indicates a good fit between our observed and expected data. The absorption signal is concentrated in the lower harmonics.

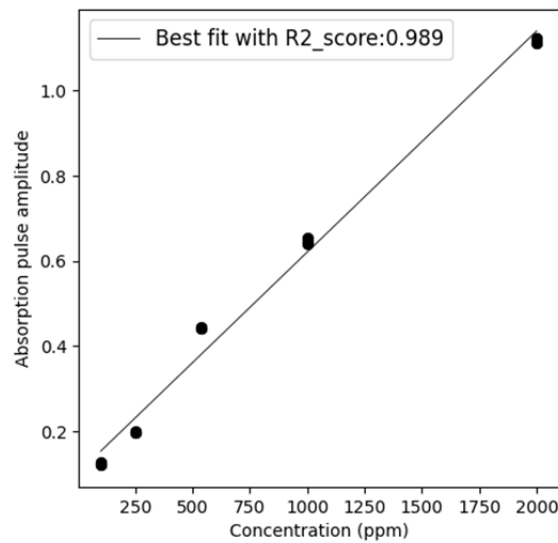


Figure – Relationship between amplitude of absorption pulse and methane concentration

This method is efficient, only 2–3 cycles can be determining the absorption pulse amplitude and, consequently, the gas concentration, making it suitable for real-time monitoring systems.

Conclusion. This paper presents a modeling approach that integrates with least-squares curve fitting for gas concentration measurement using TDLAS. The proposed model considers absorption, scattering, and reflectance effects, enabling accurate extraction of fundamental and even harmonic components using the Levenberg–Marquardt algorithm. Experimental results show a strong correlation ($R^2=0.989$) between absorption pulse amplitude and methane concentration.

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