



Effects of Temperature and Humidity on the Absorption Spectrum and Concentration of N₂O Using an Open-Path Sensor System

Jiahong Chen, Yuefeng Zhao *, Zhihao Feng, Nan Zhang, Yanxuan Wang, Zhiqiang Shen, Zongmin Kang and Qingsong Li

Shandong Provincial Engineering and Technical Center of Light Manipulations & Shandong Provincial Key Laboratory of Optics and Photonic Device, School of Physics and Electronics, Shandong Normal University, Jinan 250014, China

* Correspondence: yuefengzhao@sdnu.edu.cn

Abstract: This paper examines the effects of temperature and humidity on the absorption spectrum and concentration of nitrous oxide (N₂O) using a compact, portable open optical-path gas detection sensor system. We obtained the absorption coefficient and widened the linear function of the N₂O absorption spectrum related to temperature by theoretical analysis and the high-resolution transmission molecular absorption database (HITRAN). Afterward, we conducted real-time monitoring of N₂O in both campus and laboratory environments using lasers for a duration of 32 h and 6 h, respectively, and the results were compared and analyzed with the theoretical derivation. The results show that the concentration of N₂O increased with increasing environmental temperature but decreased with increasing humidity. Furthermore, the variations in temperature and humidity significantly affected the peak values of the second-harmonic (2f) and first-harmonic (1f) signals. Finally, the temperature N₂O concentration and humidity N₂O curves were calibrated separately, and temperature changes were positively correlated with the N₂O concentration, while humidity changes were negatively correlated with the N₂O concentration. The experimental results indicate that the concentration of N₂O and its absorption spectra are influenced by humidity and temperature, which has a significant reference value in the absorption and measurement of N₂O in practical applications.

Keywords: N₂O; quantum cascade laser; temperature; humidity; real-time monitoring



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1. Introduction

Since China's proposal of the "carbon peak" and "carbon neutrality" [1,2] goals, the government and society have attached great importance to the problems of air pollution and global warming [3]. Nitrous oxide (N₂O) is among the six greenhouse gases [4] under the Kyoto Protocol. N₂O content is relatively low compared to carbon dioxide (CO₂), but its global warming potential is about 310 times [5] that of CO₂. In addition, it is destructive to ozone (O₃) [6]. There are many reasons for the changes in N₂O concentrations in the atmosphere, which are partly due to anthropogenic activities [7], such as the widespread use of fertilizers in agricultural activities. The concentrations of other gases in the atmosphere, as well as the wind speed and direction, are all correlated with changes in N₂O concentrations [8]. At the macro level, temperature and humidity are also factors affecting the absorption coefficient of N₂O gas. However, relatively few studies have been conducted on the specific effects of temperature and humidity on N₂O gas, and analysis has also been lacking on the influence of temperature and humidity on the absorption spectrum and the concentration of N₂O. Moreover, some uncertainty and variability remain in the observations of the relationship between N₂O gas concentrations and temperature and humidity. The reasons for these discrepancies may be regional differences, differences in observation methods, and imperfections in data, which are all important bases for

measuring the N_2O concentration in atmospheric, medical, combustion, and agricultural processes [9,10]. Thus, further research and exploration, combined with additional field observations and modeling experiments, can uncover the mechanism of temperature and humidity on the N_2O concentration. Consequently, providing a scientific basis for this concentration is essential for reducing N_2O emissions, controlling climate change, and promoting sustainable development and environmental protection.

Several optical detection techniques have been developed to detect the composition of gases in the atmosphere, such as cavity-enhanced absorption spectroscopy (CEAS) [11], cavity ring-down spectroscopy (CRDS) [12], and photo-acoustic spectroscopy (PAS) [13]. However, these techniques are imperfect. For example, for CEAS technology, off-axis cavity-enhanced absorption spectra can effectively avoid the F-P effect [14,15]. Thus, it is easy to carry out pattern matching but difficult to eliminate the F-P effect when using coaxial CEAS. Moreover, CRDS can be immune to laser intensity fluctuations, but has difficulty obtaining cavity ring-down spectra in many wavebands owing to the limited availability of laser light sources and high-reflectance mirrors. Nevertheless, PAS technology, which is more sensitive than the above two technologies, costs more and is rarely used in gas detection [16,17]. Further, PAS technology has some disadvantages; for example, the photo-acoustic signal is proportional to light power, which can create some serious problems with the accuracy and stability of the measurement. Tunable diode laser absorption spectroscopy (TDLAS) [18] is a versatile technique with high sensitivity and high resolution that is also widely used in the study of high-temperature gases. TDLAS mainly includes direct absorption [19] and wavelength modulation [20,21]. This gas detection method, combining wavelength modulation technology and second-harmonic (2f) detection technology, has been the most commonly used method in atmospheric environment detection and gas detection [22,23]. Recently, Li et al. [24] proposed TDLAS technology based on the 2f detection technology based on wavelength modulation spectroscopy (2f-WMS) and long optical-path detection technology to realize the detection of N_2O gas. Its lowest detection limit is 1.98 ppb. Ren et al. [25] developed a gas sensor system based on a quantum cascade laser (QCL) using a compact multi-pass gas cell (MGC). Combined with the 2f detection technology based on wavelength modulation spectroscopy (WMS), the detection limits of methane (CH_4) and N_2O in the atmosphere can reach 5.9 ppb and 2.6 ppb, respectively. Most of these techniques are closed optical paths, although they allow for fast responses and relatively high sensitivity. This restriction severely limits the detection range [26,27], resulting in low use of continuous monitoring and limiting the practicality of large-scale monitoring.

In this work, we used field monitoring data and simulation experiments to study the effects of temperature and humidity on the absorption spectra and concentration of N_2O . First, the absorption spectral function of N_2O was simulated, which was followed by real-time monitoring of N_2O using an open-path sensor system at different temperatures and different humidity ambient conditions, independently. Then, a comprehensive analysis of the data obtained from the monitoring was used mainly to analyze the relationship between the N_2O concentration and the variation in temperature and humidity. The 2f-WMS signal and first-harmonic detection technology based on wavelength modulation spectroscopy (1f-WMS) signal were extracted and analyzed under different environmental conditions. The results revealed that the concentration of N_2O increased with the increase in temperature but that the concentration of N_2O decreased with the increase in humidity. Thereafter, the curves of temperature and N_2O concentration, humidity, and N_2O concentration were fitted and calibrated, and it was found that the changes in temperature and humidity were closely related to N_2O concentration. Thus, it is important to study the effect of temperature and humidity on the concentration of N_2O .

2. Theory

According to the Beer–Lambert law [28,29], the absorption intensity of a laser through a unit gas sample is given by the following test:

$$I_t = I_0 e^{-\alpha(v)LC} \quad (1)$$

where I_0 is incident light intensity, I_t is transmitted light intensity, L is the effective path of incident light intensity through the gas to be measured, and C is the concentration of the gas sample to be measured. The spectral absorption coefficient is $\alpha(v)$, expressed as

$$\alpha(v) = PS(T)\psi(v) = N\sigma(v) \quad (2)$$

where P , $S(T)$, T , and $\psi(v)$ represent the pressure, absorption line strength, temperature, and linear function, respectively. N is the particle population density, and $\sigma(v)$ is the absorption cross section, where P equals 1 atm. According to Equation (2), the intensity of gas absorption spectra is affected by temperature. Suppose that the environment of the gas is at any T ; its absorption spectral line intensity $S(T)$ can be expressed as

$$S(T) = S(T_0) \frac{Q(T_0)}{Q(T)} \times \frac{\exp[-hcE_i''/kT]}{\exp[-hcE_i''/kT_0]} \times \frac{1 - \exp[-hcv_{0,i}/kT]}{1 - \exp[-hcv_{0,i}/kT_0]} \quad (3)$$

where h is Planck's constant, v_0 is the central absorption frequency, c is the speed of light, E_i'' represents the energy of low-transition states, k is Boltzmann's constant, Q is the total molecular partition function [30], and T_0 is the total molecular partition function. When T_0 is 296 K, combined with the HITRAN database [31,32], the absorption spectral line intensity of gas molecules under this temperature condition can be obtained as $S(T_0) = 7.903 \times 10^{-19} \text{ cm.mol}^{-1}$. N_2O has spectral line widening owing to the simultaneous action of gas pressure and temperature; in this case, the widening function is the combination of the Lorentz curve and a Gaussian function under pressure. The Lorentz linear function can be expressed as

$$f_l(v) = \frac{1}{2\pi} \frac{\Delta v_L}{(v - v_0)^2 + \left(\frac{\Delta v_L}{2}\right)^2} \quad (4)$$

$$\Delta v_L = 2\gamma_{ari}(296/T)^n p \quad (5)$$

where v is the central frequency of a laser, Δv_L is the collision spread, γ_{ari} is the half-width coefficient of N_2O , and n is the collision broadening index. The change in ambient temperature promotes the irregular thermal motion of gas molecules, so when the temperature is dominant, the Gaussian function should be used to represent the absorption spectral line of gas. The Gaussian function is expressed as follows:

$$f_g(v) = \frac{2}{\Delta v_D} \sqrt{\frac{\ln 2}{\pi}} \exp\left[-\frac{4(v - v_0)^2 \ln 2}{\Delta v_D^2}\right] \quad (6)$$

$$\Delta v_D = \frac{v_0}{c} \sqrt{\frac{8KT \ln 2}{M}} = 7.1623 \times 10^{-7} v_0 \sqrt{\frac{T}{M}} \quad (7)$$

where Δv_D is the Doppler widening.

3. Experimental Details

3.1. Sensor Setup

Based on WMS technology and an open optical path, an open optical-path detection system for detecting N_2O gas in the atmosphere was built. The schematic diagram is shown in Figure 1. The sensor system is composed of a light-source module, photoelectric

detection module, and data processing module. The light-source module mainly consists of signal generation, a laser drive, QCL, and an indication light source. To effectively realize the tunable characteristics of laser emission wavelength, we designed the signal generator plate to generate a high-frequency sine wave signal with a frequency of 10 kHz to realize the modulation function and to generate a low-frequency sawtooth wave signal with a frequency of 10 Hz to realize the scanning function. The two signals are superimposed on the laser driver (QC750-Touch™, Ningbo HealthyPhoton Technology, Co., Ltd., Ningbo, China). The laser driver controls the temperature and central emission wavelength of QCL and converts it into an injection current acting on the detection light source QCL so that the emission wavelength of QCL is in the tunable range of $2203.7\text{--}2204.1\text{ cm}^{-1}$.

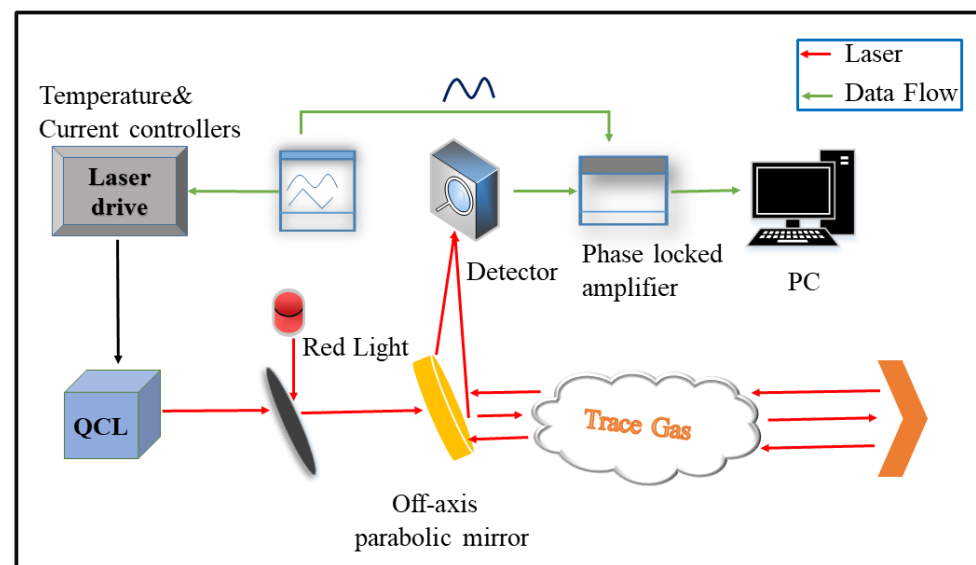


Figure 1. Schematic diagram of N₂O open optical sensor system.

In the photoelectric detection module, two beams of light are sent coaxially through the coaxial collimation system, through the gold-coated off-axis parabolic mirror with a through hole, incident into the atmospheric environment containing N₂O gas molecules. The QCL laser intensity is absorbed by N₂O gas molecules. The signal detected by the detector is transmitted to the lock-in amplifier to filter out the superimposed high-frequency sinusoidal signal from the angle mirror, which is reflected in the direction parallel to the incident light to the parabolic mirror. Thus, we demodulate the required 1f signal and 2f signal. Finally, the data acquisition card (MP4624) records relevant parameters. After data collection by the data acquisition card, the concentration of detected N₂O gas molecules is retrieved through the method of least square fitting and the harmonic ratio. Among them, the demodulated 1f and 2f signals and the real-time concentration change trend are displayed by the PC user interface based on LabVIEW.

3.2. Selection of N₂O Transitions

To achieve effective detection of N₂O gas molecules, we need to select the absorption line intensity and the emission central wavelength of the laser. First, combined with the HITRAN-2016 database, the wave number range of $2000\text{--}2250\text{ cm}^{-1}$ was selected to analyze the region of the absorption spectral line intensity of N₂O, and then carbon monoxide (CO), carbon dioxide (CO₂), and water (H₂O) molecules were simulated and analyzed, as shown in Figure 2. Within this wave number range, the absorption spectra of CO₂ were mainly distributed within the $2000\text{--}2081\text{ cm}^{-1}$ range, and the absorption spectra of CO gas were distributed within the $2025\text{--}2200\text{ cm}^{-1}$ wave number range. The absorption spectra of H₂O gas were distributed before the 2020 cm^{-1} wave number range. The absorption spectra of N₂O gas molecules were mainly distributed in the $2200\text{--}2250\text{ cm}^{-1}$ wave number range,

and they were far from the absorption spectra of water vapor and other gases, reducing interference. At around 2203.7 cm^{-1} , the absorption spectra of N_2O gas were the strongest. Therefore, we set the position of the N_2O absorption line to 2203.7333 cm^{-1} , which was used as the wave number of the QCL emission center. The corresponding spectral line intensity was $7.903 \times 10^{-19}\text{ (cm}^{-1}\cdot\text{mol}^{-1})$. The central current and temperature of QCL were set at 330 mA and $36.0\text{ }^\circ\text{C}$, respectively.

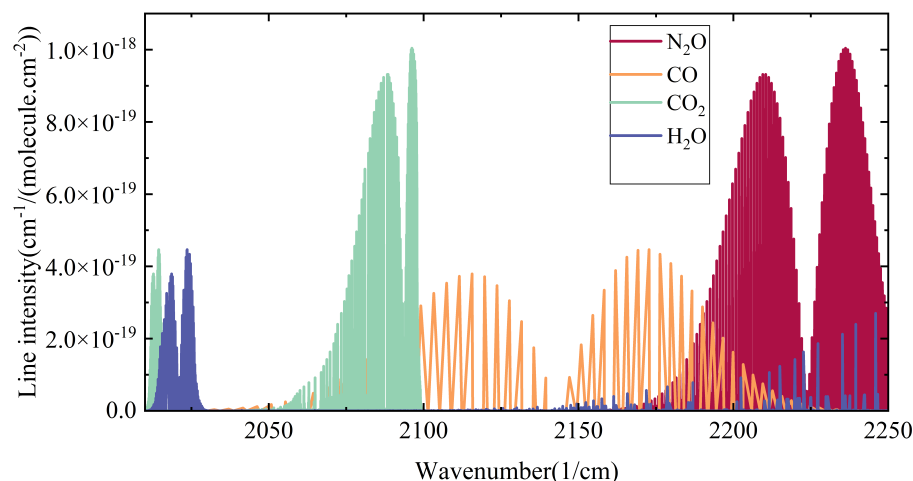


Figure 2. The intensity distribution of absorption lines of N_2O , CO , CO_2 , and H_2O in the range of $2000\text{--}2250\text{ cm}^{-1}$.

3.3. Sensor Calibration

We used a standard mixture of N_2O and N_2 at a concentration of 20 ppm (20 ppm of N_2O in N_2 , from Nanjing Special Gas Co., Ltd., Nanjing, China), which was poured into a standard gas tank with a length of 10 cm to calibrate the instrument. To effectively avoid the interference of N_2O in the atmosphere during the calibration, we put the off-axis parabolic mirror and the angular mirror as close together as possible; after that, real-time monitoring was conducted for 20 min. Moreover, 64 times the signal average could effectively eliminate the random error of the system measurement. By analyzing the N_2O concentration measured in the calibration process, we found that the average value was 19.929 ppm and that the standard deviation was 0.012 ppm, which indicates that the measurement accuracy of the system is high.

4. Results and Discussion

4.1. Effect of Temperature on the Absorption Spectrum and Concentration of N_2O

The concentration of gas molecules is influenced by various environmental parameters, such as temperature and pressure. Temperature is an important factor affecting the measurement of trace gases. Therefore, through a simulation, the change law of the absorption spectral line and the comprehensive widening line function of gas molecules were analyzed when the temperature changed independently, which is an important basis for measuring the concentration of N_2O in the atmospheric environment. The absorption line intensity $S(T)$ of gas molecules is a temperature-dependent function, and it could be obtained that $S(T)$ was $7.903 \times 10^{-19}\text{ (cm}^{-1}\cdot\text{mol}^{-1})$ by searching the HITRAN database. According to Equation (3), the magnitude of the N_2O spectral line intensity at different temperatures could be deduced, and a temperature variation range of 270–320 K was selected to calculate the relative spectral line intensity at different temperatures. As shown in Figure 3a, the relative spectral line intensities gradually weakened with the continuous increase in temperature, and the rate of weakening gradually increased.

The trend of the integrated broadening function value with temperature under different central wave number conditions was further analyzed, taking the wave number variation range of $2203.5\text{--}2204.0\text{ cm}^{-1}$ as an example. Then, the trend of the integrated

broadening line shape function was simulated and analyzed in the temperature range of 0–1000 K, as shown in Figure 3b. As the temperature increased, two trends were observed in the integrated broadening function value over the temperature range 0–1000 K. First, with the increase in temperature, the integrated broadening function value increased continuously, but the growth trend gradually slowed and tended to become stable. Second, with the increase in temperature, the integrated broadening function value first continuously increased to the maximum value and then gradually decreased and tended to become stable. In addition, under the same temperature condition, the comprehensive widening function value corresponding to the wave number closer to the absorption peak was the largest.

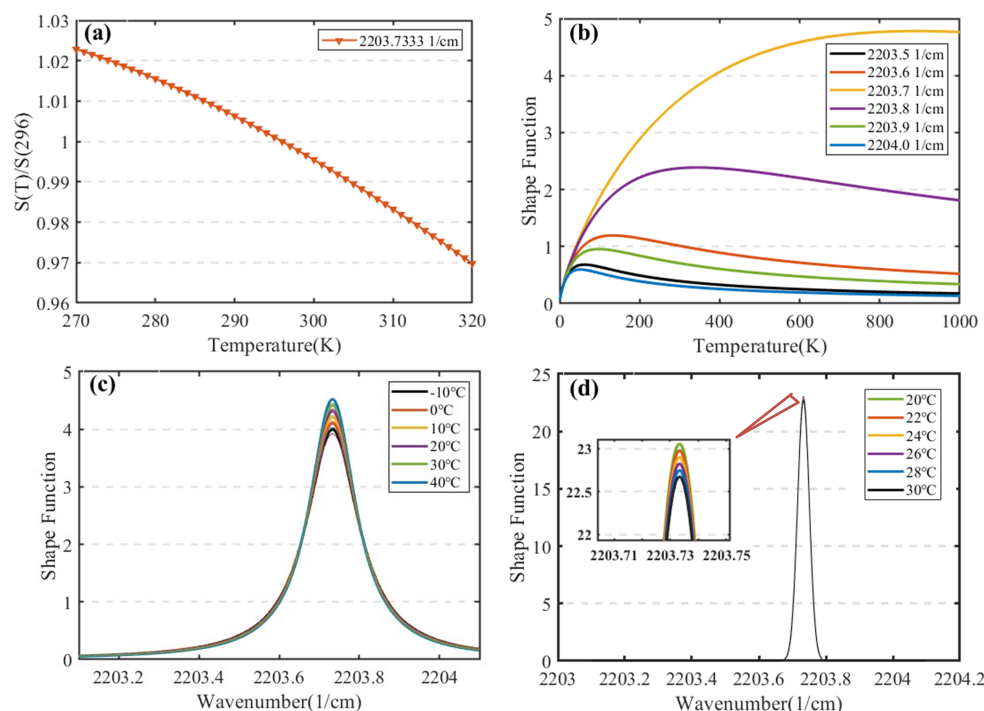


Figure 3. (a) The relationship between relative spectral line intensity and temperature. (b) The trend of its linear function when different wave numbers changed with temperature. (c) The relationship between the comprehensive broadening type function and wave number under different temperature conditions. (d) The peak value of the Gaussian function under different temperature conditions.

To further analyze the effect of temperature change on the integrated broadening line function of gas molecules, we set the gas pressure to 1 atm, the laser center wave number to 2203.73 cm^{-1} , and the tunable range to $2203.3\text{--}2204.1 \text{ cm}^{-1}$, as shown in Figure 3c. A certain correlation existed between the change in the function value and the choice of wave number with the increasing temperature. At the central wave number, the value of the function increased with the increase in temperature. When it deviated from the central wave number, however, the value of the function corresponding to different wave numbers changed with the temperature. When the temperature changed independently, the Gaussian function was selected for analysis. Finally, the Gaussian function was simulated when the ambient temperature was between 20°C and 30°C . As shown in Figure 3d, the higher the temperature, the lower the peak value of the Gaussian function.

To investigate the effect of temperature on N_2O concentration, we placed the sensor in the open area below the campus teaching building of Shandong Normal University ($36^\circ32'49.55''\text{N}$, $116^\circ49'48.47''\text{E}$) for real-time monitoring, and the distance between the angular reflector and the off-axis parabolic mirror was adjusted to set an effective light path of 20 m. Real-time monitoring of N_2O was conducted for 32 h, from 12:00 on 7 September to 20:00 on 8 September 2022. Given the difference between the temperature in the campus and the temperature in the atmosphere measured by the real gas network, a high-precision

temperature and humidity measuring instrument (RS-WS-ETH-6) was used to monitor the temperature of the campus in real time during the experiment, and the measured N_2O concentration was analyzed.

Figure 4a shows the real-time concentration of N_2O in the campus environment as measured by the sensor. The concentration of N_2O fluctuated in the range of 0.16–0.24 ppm from 12:00 on 7 September to 20:00 on 8 September 2022, owing to environmental differences. Moreover, the concentration of N_2O on campus was slightly lower than that in the atmosphere. We averaged the concentration of N_2O every hour, and Figure 4b shows the mean value and standard deviation of the N_2O concentration and temperature change. It is evident that the temperature increased from 12:00 to 13:00 on 7 September 2022, and that the concentration of N_2O also increased during this hour. From 13:00 to 15:00, the temperature was maintained at 30 °C to 31 °C, a relatively stable state, at which time there were essentially no significant fluctuations in N_2O concentrations, until 6:00 am on 8 September, when the temperature dropped from 31 °C to 18 °C, and the N_2O concentration decreased significantly, from 0.26 ppm to 0.15 ppm. On 8 September 2022, the temperature gradually rose from 6:00 am to 29 °C at 12:00 noon, and the concentration of N_2O also rose to 0.21 ppm.

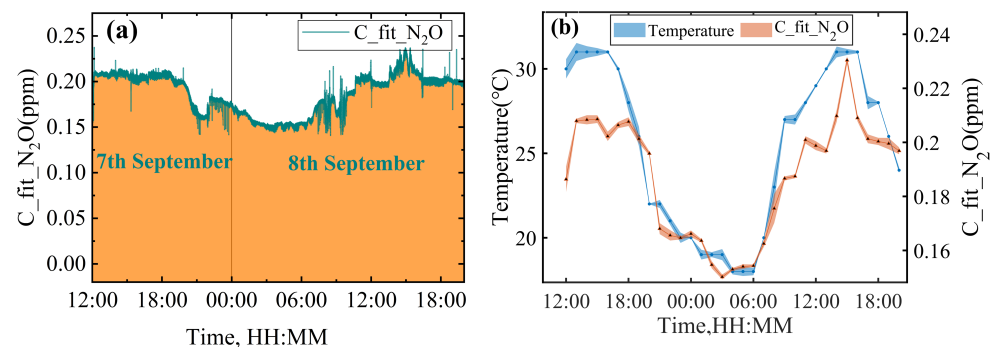


Figure 4. (a) Real-time atmospheric N_2O concentration from 7 September 2022 to 8 September 2022. (b) The dependence of N_2O concentration on temperature.

The maximum standard deviation of the N_2O concentration in this process was 0.54 ppb, and the minimum standard deviation was 0.05 ppb. It can be seen that the concentration of N_2O increased with the increase in ambient temperature. The reason for this result may be that the temperature directly or indirectly affects the emission of N_2O from soil. When the temperature of soil in the environment increases, the denitrification effect enhances the emission of N_2O .

In addition, we extracted the 2f signals and 1f signals at different temperatures during the measurement for analysis. The harmonic signals were mainly extracted at 30 °C, 28 °C, 26 °C, 22 °C, and 20 °C. Figure 5a shows the 2f signal at different temperature moments. We found that the peak of the 2f signal corresponding to the higher temperature was smaller, and that the lower the temperature, the larger the peak of the 2f signal. This result aligns with the Gaussian function peak distribution. Figure 5b shows the primary harmonic signal. The analysis shows that the higher the temperature, the smaller the absolute value of the peak of the corresponding primary harmonic signal. The lower the temperature, the larger the absolute value of the peak of the 1f signal.

Then, a linear fit was performed for N_2O concentrations at different temperatures, and as shown in Figure 6a, the N_2O concentration and temperature were positively correlated, with higher temperatures and higher N_2O concentrations without considering other factors, where $R^2 = 0.87$. The N_2O concentration and 2f/1f signal value were then compared and analyzed, as shown in Figure 6b. When the concentration increased, 2f/1f also increased, which we found by calculating $R^2 = 0.71$.

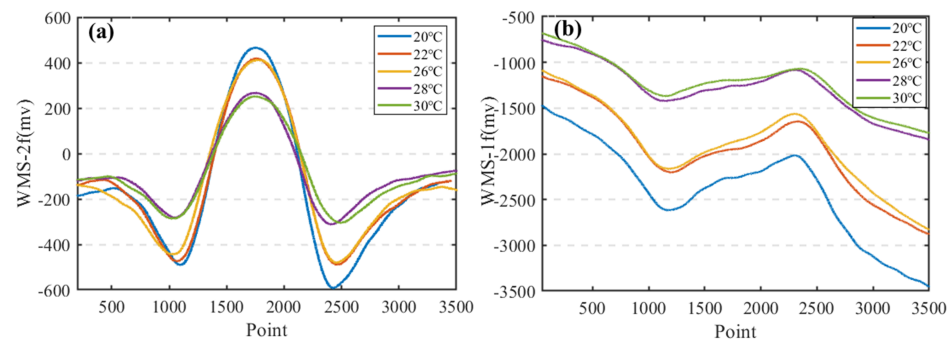


Figure 5. (a) 2f signals at different temperature conditions. (b) 1f signals at different temperature conditions.

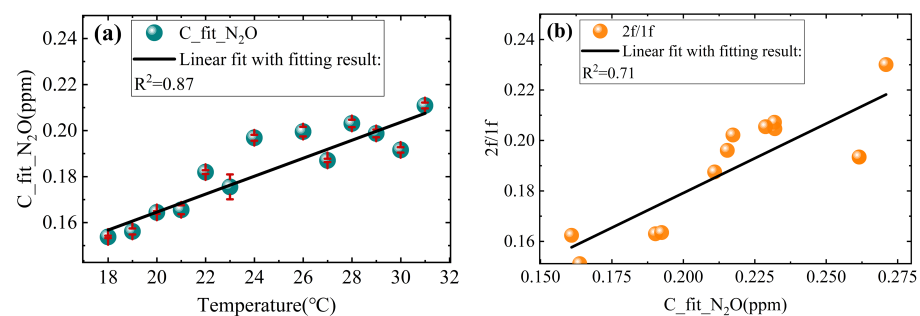


Figure 6. (a) Fitting results for temperature and N₂O concentration. (b) Results of fitting N₂O concentration to the 2f/1f signal.

4.2. Effect of Humidity on the Absorption Spectrum and Concentration of N₂O

In addition to pressure and temperature, humidity may have a certain influence on the concentration of N₂O. We chose a temperature-controlled laboratory environment for an experiment. First, we opened the windows of the laboratory for ventilation for a period of time, to maintain a relative balance between the laboratory environment and the external environment. Here, N₂O is in a natural concentration state, without any changes to the sources of N₂O emissions in the experimental environment. We then closed the doors and windows for a few hours for real-time monitoring, during which time the temperature in the laboratory was controlled at 25 °C using air conditioning. The test began at 12:00 on 6 September 2022, and a high-precision humidity detector was used to determine the humidity level in the laboratory at that time to be 33%. The humidity in the test chamber was then increased, and N₂O was measured continuously. Figure 7a shows the real-time concentration of N₂O in the laboratory. The humidity in the laboratory measured at 11:00 was 56% and reached a maximum of 64% at 12:00. Within 2 h, the concentration decreased from 0.22 ppm at the beginning to 0.19 ppm, with a moderate and continuous increase in humidity. Starting at 12:00, the indoor humidity no longer increased, so the indoor humidity was naturally reduced.

At 14:00, the indoor humidity was recorded as 46%, and the N₂O concentration was significantly higher than that at 12:00. At 16:00, the indoor humidity was reduced to 30%, and the N₂O concentration basically returned to the initial laboratory concentration of 0.21 ppm. Figure 7b shows that the indoor humidity and N₂O concentration were negatively correlated, and that the higher the humidity, the lower the N₂O concentration. One reason for this relationship between the N₂O concentration and humidity is that increased humidity promotes secondary aerosol formation. In addition, there is a certain interaction between N₂O and other gases in the air, the increase in water molecules can disrupt the dynamic balance of gas molecules in the air, causing the concentration of other gases to change [33], which indirectly affects the concentration of N₂O. Changes in humidity can also affect the decomposition of N₂O [34] and its diffusion rate. The 2f signals

and 1f signals at different humidity levels were extracted for analysis, and the 2f signals at the 30%, 46%, 56%, and 64% humidity levels were mainly extracted. As shown in Figure 8a, the lower the humidity, the larger the 2f signal peak; the higher the humidity, the smaller the 2f signal peak. Figure 8b shows the 1f signal at different humidity levels, and it can be seen that the lower the humidity, the larger the 1f signal peak; the higher the humidity, the smaller the corresponding 1f signal peak. Furthermore, the N_2O concentration under different humidity levels was fitted. As shown in Figure 9, when the humidity was higher, the corresponding N_2O concentration was smaller, and the calculated $R^2 = 0.88$.

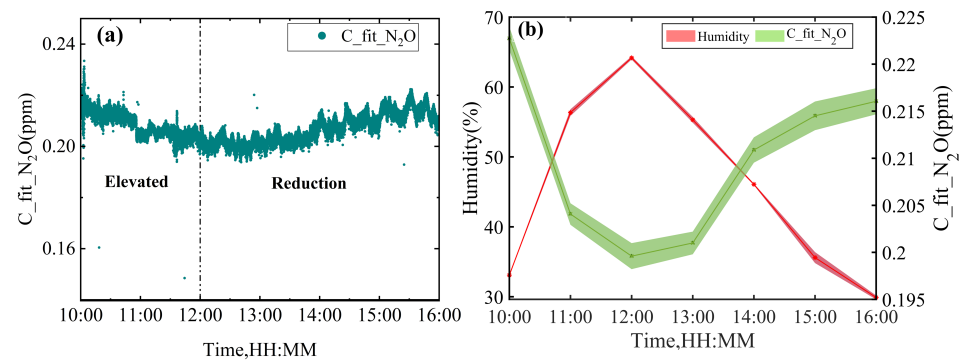


Figure 7. (a) Real-time indoor N_2O concentration on 6 September 2022. (b) N_2O concentration versus humidity curve.

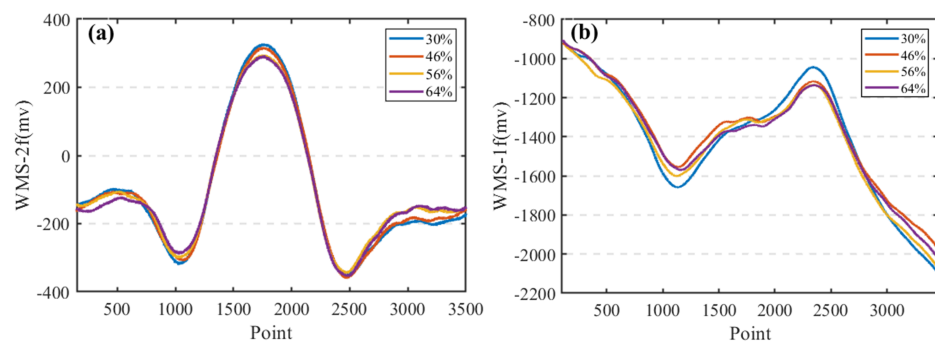


Figure 8. (a) 2f signal under different humidity conditions. (b) 1f signal under different humidity conditions.

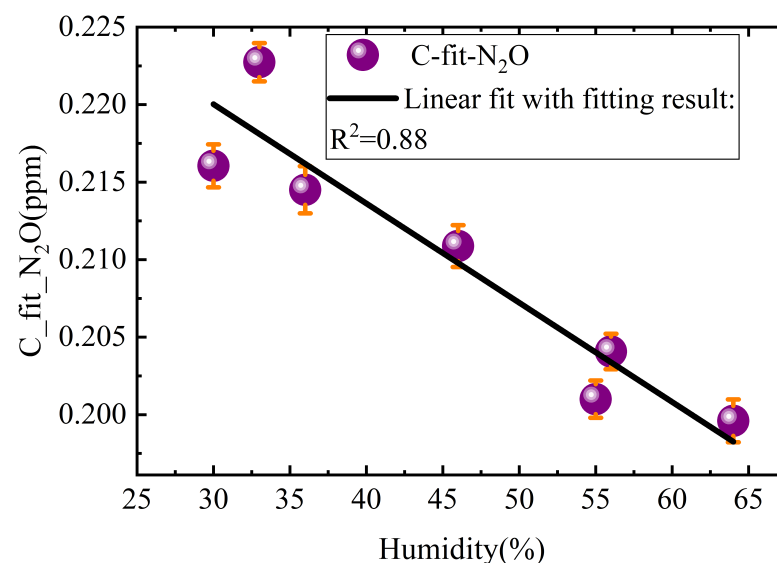


Figure 9. Results of the fit between humidity and N_2O concentration.

5. Conclusions

In this study, we investigated the effects of temperature and humidity on the concentration of N_2O and its absorption spectra using an open-path sensor system. By combining theoretical analysis and field monitoring, we first conducted monitoring of N_2O in a campus environment, analyzing the effects of temperature on its concentration and absorption spectra. We discovered that the concentration of N_2O would increase correspondingly with the increase in temperature. The influence of humidity on N_2O concentration was monitored under the condition that the ambient temperature of the laboratory remained unchanged. The concentration of N_2O was negatively correlated with humidity. The 2f and 1f signals under different temperature and humidity levels were extracted for analysis. We found that the higher the temperature, the smaller the peak value of the 2f and the 1f signals, which accords with the trend of the Gaussian function changing with temperature. Under different humidity conditions, the lower the humidity, the larger the 2f signal peak; the higher the humidity, the smaller the 2f signal. This study is of great significance for analyzing the relationship between N_2O and environmental parameters such as temperature and humidity. We hope that our research findings can assist environmental agencies in formulating more effective environmental policies for different environments. In the future, we can use QCL to analyze the relationship between N_2O and other environmental and gas parameters.

Author Contributions: Conceptualization, J.C.; methodology, J.C.; software(Matlab2020) , Z.F.; validation, N.Z., Z.K. and Q.L.; formal analysis, J.C.; investigation, Y.W.; resources, Y.Z.; data curation, Z.S.; writing—original draft preparation, J.C.; writing—review and editing, J.C. and Y.Z. All authors have read and agreed to the published version of the manuscript.

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Data Availability Statement: The data presented in this study are available upon request from the corresponding author.

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