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Article

Quantitative Remote Sensing of Sulfur Dioxide Emissions from Industrial Plants Using Passive Fourier Transform Infrared (FTIR) Spectroscopy

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Abstract

The remote monitoring and quantification of industrial gas emissions, such as sulfur dioxide (SO₂), are critical for environmental protection. This research demonstrates the application of passive Fourier Transform Infrared (FTIR) spectroscopy for the remote detection and quantitative analysis of SO₂ emissions from a metallurgical plant chimney. Infrared spectra were acquired at a stand-off distance of 570 m within the 7–14 μm spectral range at a resolution of 4 cm^{−1}. Path-integrated SO₂ concentrations were determined through cross-sectional scanning of the gas plume. To translate these optical measurements into an emission rate, the atmospheric dispersion of the plume was modeled using the Pasquill–Briggs approach, incorporating source parameters and meteorological data. Over two experimental series, the calculated average SO₂ emission rates were 15 kg/s and 22 kg/s, with coefficients of variation of 45.2% and 32.8%, respectively. This work highlights the value of FTIR spectroscopy as a powerful analytical tool for the remote, molecular-specific monitoring of atmospheric pollutants, providing a methodology applicable to the environmental chemistry of industrial emissions.

Keywords: metallurgical plants; emission rate; sulfur dioxide; environmental monitoring; infrared Fourier transform spectroscopy; Pasquill-Briggs; air pollution

1. Introduction

The development of industrial sectors such as energy, metallurgy, chemicals, textiles, and pharmaceuticals is accompanied by an increase in emissions of harmful substances into the atmosphere [1]. These pollutants not only degrade the quality of life but also pose a serious threat to health [2].

The detection and identification of gas pollutants play an important role in environmental monitoring. It allows for tracking their sources, assessing the degree of danger, and developing effective control methods [3]. Currently, infrared (IR) Fourier spectroscopy [4,5], electrochemical sensing [6], differential absorption lidars [7–11], differential optical absorption spectroscopy [12–14], non-dispersive IR spectroscopy [15,16], and others are used to monitor atmospheric pollution.

Modeling the spread of pollutants in the air includes various approaches that can be categorized based on their mathematical frameworks and scales of application. Gaussian models are based on the assumption that the pollutants are normally distributed in the atmosphere; for example, AERMOD (EPA) [17], ADMS (CERC) [18], and ISC3 [19]. Lagrangian models calculate the trajectories of individual pollution particles, accounting for wind and turbulence; for example, HYSPLIT (NOAA) [20], FLEXPART [21], CALPUFF [22]. Eulerian models consider the atmosphere by dividing it into cells, for each of which the transport equations are solved; for example, CMAQ (EPA) [23], CHIMERE (EC) [24], and WRF-Chem [25]. Empirical models describe the scattering of clouds of heavy gas released into the atmosphere near the land surface; for example, B and McQ [26,27], VDI Guidelines [28].

Lagrangian models effectively account for changing weather conditions and complex terrain by utilizing three-dimensional meteorological fields and adaptively calculating particle trajectories. This

makes them suitable for modeling accidental emissions and long-range pollution transport. However, a significant limitation of these models is their poor suitability for accounting for complex chemical reactions in the atmosphere, due to the computational difficulties associated with interacting, independently moving particles. Typically, Lagrangian models implement only the simplest chemical processes, such as radioactive decay or first-order linear reactions, while photochemical cycles, secondary aerosol formation, and other complex transformations remain beyond their scope. To solve this problem, hybrid approaches that combine Lagrangian and Eulerian methods are often used, enabling the modeling of complex atmospheric processes.

In contrast, Eulerian models have several significant disadvantages. Their primary weakness is limited spatial resolution, which leads to the "smearing" of point emission sources (such as chimneys) across the cells of the computational grid. This is especially critical for local-scale tasks where precise spatial detail is important. Furthermore, these models require substantial computing resources to increase detail; for instance, halving the grid spacing leads to an eightfold increase in computational load. They are also highly sensitive to the quality of the meteorological data, as small inaccuracies in wind or temperature fields can result in significant forecast inaccuracies.

Gaussian models are widely used for calculating pollutant dispersion from industrial chimneys due to their optimal combination of accuracy, simplicity, and established validity. This study employs the Pasquill-Briggs model to simulate sulfur dioxide (SO₂) emissions from chimneys. This approach requires a minimal set of input data, making it convenient for operational emission spread calculations. Compared to more complex Eulerian or Lagrangian approaches, the Gaussian model demands an order of magnitude fewer computational resources and less time while maintaining sufficient accuracy for industrial facilities with stationary emission sources.

This paper explores the use of remote sensing measurements via Fourier transform infrared (FTIR) spectroscopy to estimate the SO₂ emission rate from the chimneys of metallurgical plants. The Pasquill-Briggs model is used to calculate the emission rate from the measured path-integrated gas concentrations. This application requires supplementary data on gas outlet conditions (e.g., chimney height, gas exit velocity, and wind speed) as well as atmospheric characteristics (such as stability and the degree of solar insolation).

2. Materials and Methods

In this paper, we applied the Pasquill-Briggs model to estimate the flue gas distribution, and by solving the inverse problem, we were able to estimate the flue gas emission rate. The Pasquill-Briggs model describes the distribution of pollutants in the atmosphere as a Gaussian distribution. This approach is based on the turbulent diffusion equation with the following key assumptions [29,30]:

- dispersion in the horizontal and vertical planes is described by a Gaussian distribution with standard deviations σ_y and σ_z along the y and z axes, respectively;
- the average wind speed acting on the flow remains constant throughout the layer, and the wind direction does not change;
- the gas emission rate is constant;
- the flow can be reflected from the earth surface.

Taking these assumptions into account, the spatial distribution of the SO₂ concentration $c(x, y, z)$ from a point source with a constant wind directed along the x axis can be represented by the following formula within the framework of the Gaussian model (see Figure 1)

$$c(x, y, z) = \frac{q}{2\pi\sigma_y\sigma_z u} \exp \frac{-y^2}{2\sigma_y^2} \left(\exp \frac{-(z-H)^2}{2\sigma_z^2} + \exp \frac{-(z+H)^2}{2\sigma_z^2} \right), \quad (1)$$

where q is the gas emission rate [kg/s]; H is the height of the flare axis above ground level [m]; u is the horizontal wind speed along the torch axis [m/s]. In the Pasquill-Briggs model, the values of the scattering functions σ_y and σ_z are determined by the stability class of the atmosphere at a distance of

100 m to 10 km from the source. The stability class of the atmosphere is determined by wind speed, the degree of solar radiation, and cloud cover [31].

Taking into account the rise of the plume when modeling the dispersion of pollutants is critically important for the correct assessment of the effective height of the emission H , defined as the sum of the physical height of the source h and the vertical displacement of the plume Δh . For hot emissions characterized by significant buoyancy ($T_s \gg T_a$), the rise of the plume is mainly due to thermal convection and is described by semi-empirical Briggs formulas [32]:

$$\Delta h = 1.6 \frac{(F_b x_c^2)^{1/3}}{u} \left[\frac{2}{5} + \frac{16x}{25x_c} + \frac{11}{5} \left(\frac{x}{x_c} \right)^2 \right] \left(1 + \frac{4x}{5x_c} \right)^{-2}, \quad (2)$$

where F_b is a flux parameter [m^4/s^3]: $F_b = g v_s r^2 \left(\frac{T_s - T_a}{T_s} \right)$, $g = 9.81$ – free fall acceleration [m/s^2]; $v_s = 4.36$ – vertical speed [m/s]; $r = 4.36$ – exit radius [m]; T_s, T_a – the exit gas and ambient temperature [K]; $x_c = A F_b^{2/5} h^{3/5}$ – the distance downwind at which environmental turbulence begins to dominate the dispersion process is known as the critical distance [m]; $A = 2.16$ [$s^{6/5}/m^{6/5}$].

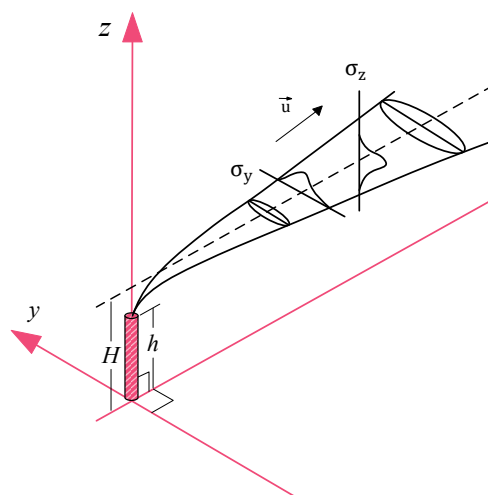


Figure 1. Visualization of flux propagation in the Pasquill-Briggs model.

The most important step in constructing the plume distribution is to obtain the initial parameters of the model. The basic meteorological parameters (wind speed, atmospheric stability class) required for calculations are set directly from the weather forecast at the time of the measurements. The path-integrated gas concentrations in the flux are calculated according to the direction of data recorded on the Fourier spectrometer. The emission rate q is calculated by comparing experimentally obtained path-integrated gas concentrations of pollutants $C_{int}^E(\theta, \phi)$ in the exhaust gas plume with the simulated values $C_{int}^T(\theta, \phi)$, where θ, ϕ – the angle of location and azimuth relative to the observation point. The simulation of path-integrated gas concentrations was carried out as follows:

$$C_{int}^T(\theta, \phi) = \int_0^\infty c_{sp}(r, \theta, \phi) dr, \quad (3)$$

where $c_{sp}(r, \theta, \phi)$ is calculated using the distribution $c(x, y, z)$, by switching to a spherical coordinate system with rotation and shifting the origin to the observation point. For the described transformations, additional parameters are introduced: the azimuth on the tube θ_s and the distance to the tube from the observation point r_s . By varying the direction of wind speed and emission rate q , the minimum discrepancy between the theoretical and experimentally measured series of values is determined:

$$\|C_{int}^T(\theta, \phi) - C_{int}^E(\theta, \phi)\| \xrightarrow{\vec{u}, q} \min \quad (4)$$

The desired value of the ejection power is determined from condition 4. The experimental concentrations were determined by remote sensing using an infrared Fourier spectrometer. Obtaining experimental values $C_{int}^E(\theta, \phi)$ is presented in detail in [1]. The efficiency of SO₂ detection in the infrared range of the spectrum has been shown in [1,33].

3. Experimental Setup

Figure 2 shows a diagram of an experiment on the remote recording of infrared chimney exhaust gas spectra. Table 1 shows the main technical characteristics of the infrared Fourier spectrometer. The Fourier spectrometer was located on an automated pan tilt positioner controlled via the ethernet protocol, with the ability to automatically transmit azimuth and elevation for each measurement, along with a time reference, which makes it possible to use the Pasquill-Briggs model to calculate emissions. The positioning accuracy of the automatic pan tilt positioner is at least 1 mrad.



Figure 2. The experimental setup during the measurements. The red arrow shows infrared radiation in the from the chimney exhaust gases.

Table 1. Main characteristics of the experimental setups.

Parameter	Value
Optical scheme	Michelson Interferometer
Spectral range, μm	7-14
Spectral resolution, cm^{-1}	4
Measurement frequency, Hz	1
FOV, deg	2×2
Detector type	MCT, cooled up to 80 K
Operating mode	Passive (without IR source)

4. Results and Discussion

Measurements of path-integrated gas concentrations were carried out in two series. The series were held over two days, with each episode lasting 30 minutes. The measurement conditions, as

well as the simulation parameters, are shown in Table 2. Figure 3 shows the side view of the plume from the spectrometer. The middle of the crosshair shows the axis of the infrared channel. The red square is shown conditionally. Figure 4 shows an analysis of thermograms of exhaust plumes with an assessment of the temperature contrast.

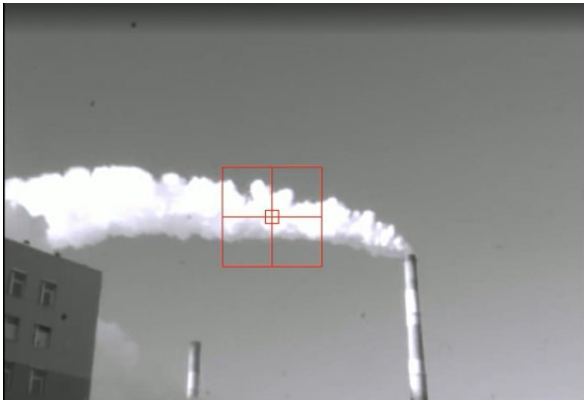


Figure 3. View of the exhaust gas plume from the spectrometer side during measurements.

Figure 4(a) shows the thermogram of the exhaust plume obtained using the NEC 2640 high-resolution thermal camera from the same position as the spectrometer. Figure 4(b) shows the temperature distribution along the plume from the chimney exit (L is the distance from the chimney exit, m). The thermogram shows that on the day of the measurement, the effective temperature of the underlying surface (sky) is -38.1°C or less, which corresponds to a temperature contrast between the exhaust plume and the underlying surface of the order of 20° or more.

Table 2. Measurement conditions.

Parameter	Value	Series 1	Series 2
Time duration	min	30	30
Chimney height	M	250	250
Wind speed ($H_v = 10\text{ m}$)	m/s	3-4	5-6
Atmospheric stability class	-	D	C
Distance to the chimney R	m	570	590
Azimuth to the chimney α	deg	55	50
Chimney exit radius r	m	4.36	4.36
Ambient air temperature T_a	deg C	-5	-20
Discharge temperature T_s	deg C	55	50
Gas flow speed v_s	m/s	4.5	4.5
Seat angle θ	deg	25-30	25-30
Azimuth ϕ	deg	220-280	220-280

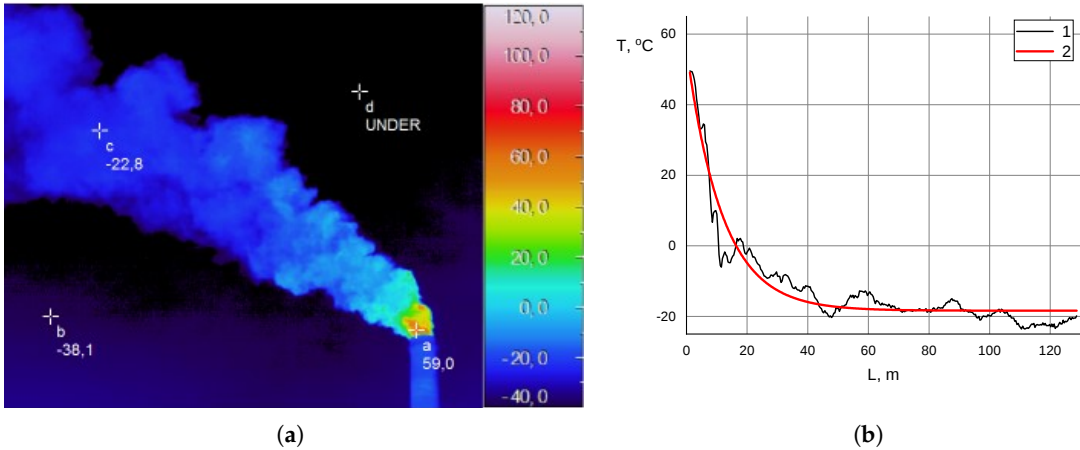


Figure 4. (a) Thermogram of the exhaust plume during experiment No. 2. (b) the temperature distribution by the movement along the plume from the chimney exit, L is the distance from the chimney exit, m.s.

The measurement conditions given in Tables 2 and 3 show the main parameters of the Pasquill-Briggs model. The results of the calculations of the point source plume propagation are shown in Figures 5–7.

Table 3. Parameters of the Pasquill-Briggs model.

Parameter	Value	Series 1	Series 2
Average source emission	kg/s	15.0	22.0
Wind direction α_u	deg	121	143
Buoyant flow F_b	m^4/s^3	90	112
Critical distance x_c	m	359	392
The plume shifting Δh	m	146.6	105.3

The graph shown (Figure 5) illustrates the vertical profile of the SO_2 distribution for various distances from the emission source, calculated using the data from the corresponding experiment No. 1. Near the source (no more than 1000 m), in the zone of active plume rise, the concentration distribution shows a symmetrical Gaussian character (Figures 6 and 7) with a maximum located at an altitude determined by Formula (2).

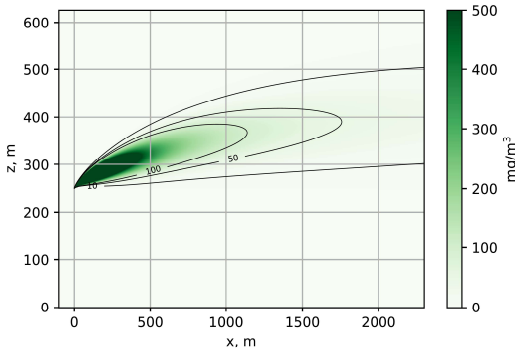


Figure 5. Vertical profile of the model plume, with the origin anchored to the base of the chimney (0, 0, 0), $y = 0$ m.

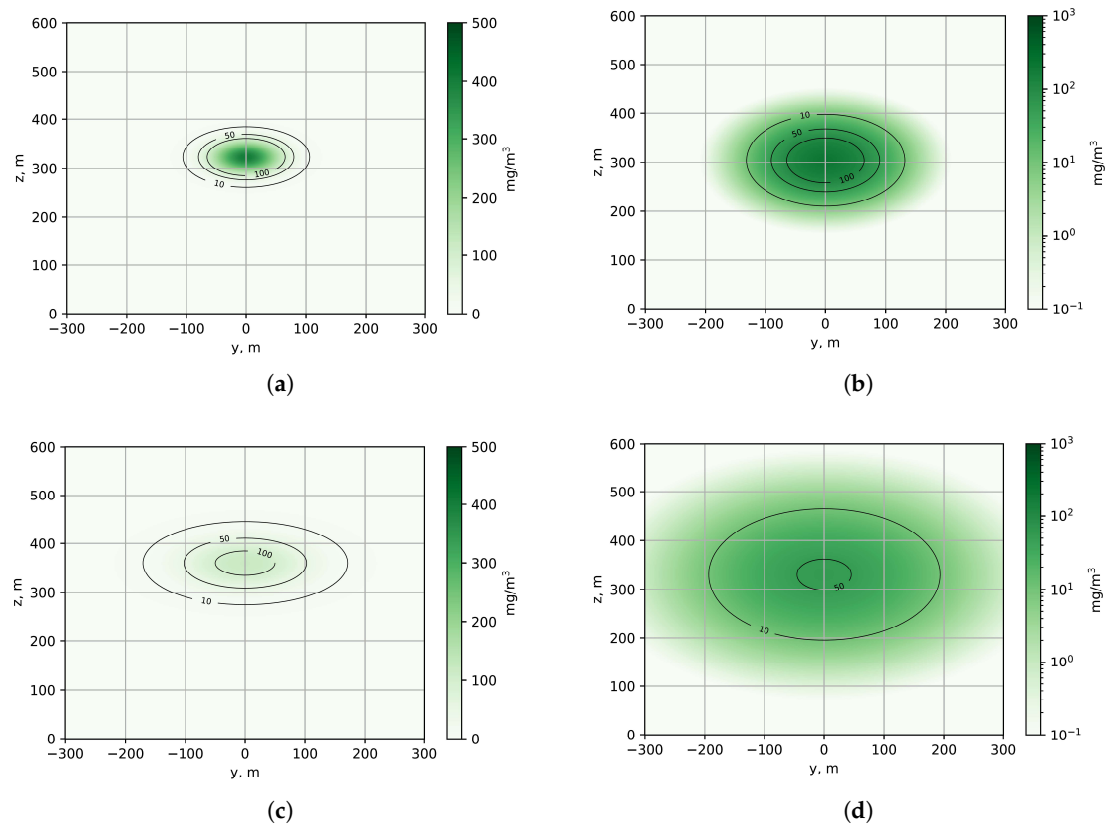


Figure 6. The cross-section of the model plume, the source anchored to the chimney exit (0, 0, 250). $x = 500$ m where a logarithmic scale: (a) – Experiment No. 1; (b) – Experiment No. 2. $x = 1000$ m where a logarithmic scale: (c) – Experiment No. 1; (d) – Experiment No. 2.

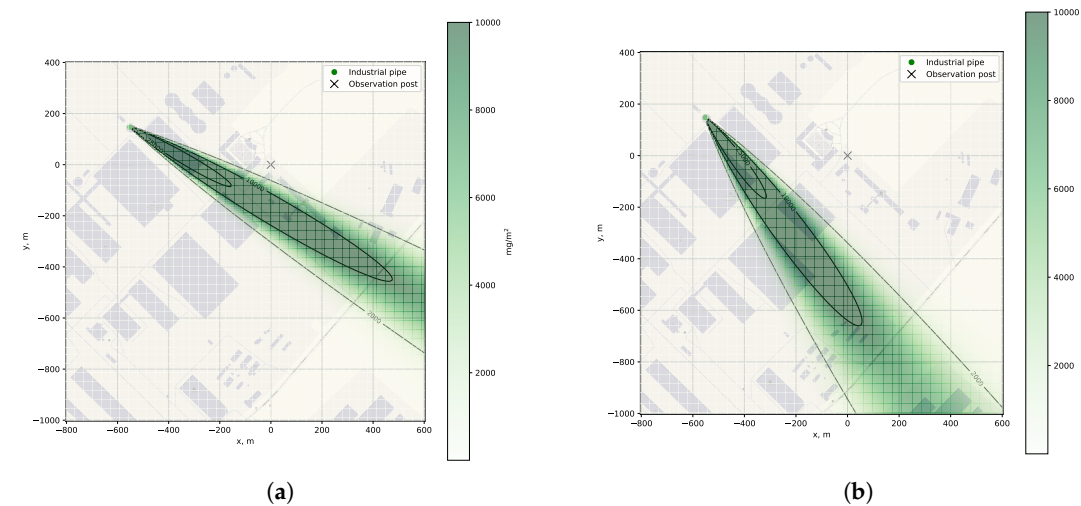


Figure 7. Exhaust gas plume: (a) – Series 1, (b) – Series. 2. The black cross is an IR Fourier spectrometer, the green dot is a chimney.

The analysis of the concentration isolines demonstrates a characteristic expansion in the zone of maximum elevation, followed by a stabilized spread at altitudes of about 350-400 m. Data on the height of the plume rise in a specific location allow the spectrometer to be optimally positioned for the accurate targeting of the area of maximum concentration of pollutants. Knowledge of the vertical distribution of the plume ensures the correct selection of elevation and azimuth angles during scanning, which increases the reliability of measurements of path-integrated gas concentration and minimizes errors associated with the incomplete capture of the cross-section of the plume.

To choose the optimal location for the observation point, it is necessary to combine the terrain map with data on the density and direction of winds, as well as information on the distribution of pollution sources and possible obstacles. Preliminary data on wind and emission rates allow us to estimate the pollution density in advance and determine the most suitable location for an observation point.

When calculating the emission rate of n sources (chimneys), a linear superposition of the emissions from each individual source is applied simultaneously, taking into account the position of the sources on the terrain map $c(x, y, z) = \sum_{i=1}^n c(x - x_i, y - y_i, z)$.

During series No. 1, 15 experiments were performed to register the IR spectra of SO₂ while scanning the FTIR spectrometer along the plume. During series No. 2, 25 measurements were performed. The results of the analysis of the two series are shown in Table 4.

Table 4. Results of emission rate calculation.

Parameter	Value	Series 1	Series 2
The average value	kg/s	15.0	22.0
Average square deviation	kg/s	7.4	7.3
Coefficient of variation	%	45.2	32.8

Accurate emission data are not available for remote measuring, as it is difficult to obtain local monitoring by simultaneously measuring gas flows in all pipes entering the chimney. Another way to estimate the emission rate may be through PEMS (Predictive Emissions Monitoring Systems) models. However, the PEMS data are indirect and also require validation. The above errors in the emission rate are determined to be random errors, such as errors in registering IR spectra and in determining wind speed (especially the vertical component, where the error of anemometers reaches $\pm 5\%$), as well as methodological errors related to solving the multilayer problem of atmospheric optics for determining concentrations while considering atmospheric turbulence, etc.

5. Conclusions

This paper presents a methodology for the remote quantification of chimney emission rates from metallurgical plants. Remote optical techniques enable the acquisition of path-integrated gas concentration data across a plume’s cross-section, a parameter not accessible through conventional in-situ point sampling. The integration of such line-integrated measurements with atmospheric dispersion modeling allows for the comprehensive reconstruction of total substance flow. Specifically, the method employs ground-based Fourier transform infrared spectrometry to measure the path-integrated concentration within the gas flux. The FTIR spectrometer, operating in the mid-infrared range (7–14 μm) at a spectral resolution of 4 cm^{−1} and a scanning frequency of 1 Hz, was deployed at a distance of 570 m from the emission source.

The emission rates of sulfur dioxide were calculated using the Pasquill-Briggs Gaussian dispersion model. Model inputs included source parameters (chimney height, nozzle diameter, and gas exit velocity) and real-time meteorological data (wind speed and atmospheric stability class). Analysis of two distinct 30-minute measurement series, conducted on separate days, yielded average emission rates of 15.0 kg/s and 22.0 kg/s, with corresponding coefficients of variation of 45.2% and 32.8%. This level of uncertainty is consistent with established dispersion modeling approaches, which report inter-model discrepancies on the order of 35% for emission rate estimations [33].

The presented remote sensing approach provides a practical tool for estimating industrial emission fluxes. Furthermore, the derived data can serve as critical input for modeling the dispersion and transport of gaseous emissions in complex topographic environments.

Author Contributions: V.G.: conceptualization, investigation, writing (original draft), numerical modeling; I.G.: data processing, writing, editing; I.F.: writing, review and editing; I.V. and R.G.: experiments and data preparation; A.M.: project management. All authors have read and agreed to the published version of the manuscript.

Data Availability Statement: The data presented in this study are available on request from the corresponding author. The data are not publicly available because of privacy restrictions.

Conflicts of Interest: The authors declare no conflicts of interest.

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