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Spectroscopic Verification and Exclusion of Non-biological Causes of Sulfur-Based Biosignature Gases in Exoplanet Life Detection

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Abstract

Sulfur-based volatile compounds are regarded as a key candidate to break the deadlock of traditional oxygen-methane biological signal detection due to their unique metabolic specificity. However, their spectral signals face the challenge of high degeneracy caused by geological and photochemical origins. To address this core bottleneck, this paper proposes a systematic detection framework based on the "exclusion-verification" dual mechanism. Firstly, a flux threshold model for volcanic degassing and non-biological synthesis pathways was established, systematically defining the physical and chemical boundaries of non-biological causes, secondly, a high-resolution radiative transfer model was used to analyze the spectral fingerprints of key sulfur molecules, proposing an observation strategy for the mid-infrared window region to counteract the shading effect and a ground-based high-dispersion cross-correlation verification scheme, finally, a context-dependent Bayesian inference framework was constructed, achieving a logical loop from spectral feature identification to quantification of biological cause probability through marginal integration of non-biological model parameters. The research provides a robust theoretical criterion for the confirmation of exoplanetary life.

Keywords

Exoplanetary Life Detection, Sulfur-Based Biological Characteristics, Spectral Degeneracy, Non-Biological Exclusion Mechanism, Bayesian Inference

1. Introduction

With the advancement of the construction of the next-generation ground-based giant telescopes, exoplanetary science is undergoing a profound epistemological transformation [1]. In this grand narrative, the search for biosignature gases in distant worlds has become a key approach to answering the ultimate question of "Are we alone in the universe?" [2].

For a long time, traditional biological signals such as oxygen have dominated the detection strategies [3]. However, modern planetary geochemical models indicate that non-biological mechanisms such as water photolysis under runaway greenhouse effects can also produce similar spectral features, leading to a significant risk of "false positives" [4]. In this context, sulfur-based volatiles, as key metabolic products of early anaerobic ecosystems and marine phytoplankton on Earth, provide a new observational dimension to break the current detection deadlock with their high thermodynamic specificity [5]. The biogeochemical cycle of sulfur elements not only deeply couples the oxidation-reduction state of the planet but is also likely to serve as a "chemical periscope" for revealing the existence of non-terrestrial life forms [6].

Although sulfur-based gases have a high biological indicative significance, their remote sensing detection faces severe physical and chemical challenges, with the core being the "cause redundancy" of spectral signals [7]. Without the constraints of in-situ detection, observers are prone to fall into the logical trap of mistaking geological and photochemical products for biological signals. Specifically, high-throughput volcanic degassing activities can inject large amounts of sulfur dioxide and other substances into the atmosphere, and in the photochemical network driven by stellar ultraviolet radiation, these precursor molecules can further evolve into complex sulfur aerosols or organic sulfur compounds [8]. This "non-biological background noise" constructed by geological structures and stellar radiation has a high spectral fingerprint overlap with biosource molecules in the infrared and near-infrared bands [9,10]. If the contributions of these non-biological processes cannot be effectively separated from the observational data, any claim about extraterrestrial life will lack statistical significance. Therefore, how to "eliminate false positives and retain the true" from the complex planetary atmospheric spectra and establish a set of criteria for quantitatively distinguishing biological metabolic releases from geological and chemical evolutions is the bottleneck problem that current astrobiology urgently needs to overcome.

This paper aims to establish a systematic "exclusion-verification" mechanism based on high-resolution spectral data. Unlike the traditional signal search strategy, this study advocates that the essence of life detection is not the capture of a single anomalous signal, but the exhaustive exclusion of all possible non-biological explanations. This paper will first systematically define the non-biological production limits of sulfur-based gases under different planetary oxidation-reduction environments through thermodynamic and kinetic simulations, then, combined with radiative transfer models, it will analyze the spectral characteristics of key sulfur molecules and their detectability under different signal-to-noise ratios, finally, by introducing the Bayesian inference framework, it will quantify the posterior probability that the observed data support the "biological cause hypothesis" after excluding known geological and photochemical causes, using atmospheric background parameters as prior conditions. The establishment of this framework aims to provide a logically rigorous and operable theoretical criterion for future exoplanetary life detection, promoting the field to move from qualitative phenomenon description to quantitative probability assessment.

2. Sulfur-Based Gas Characteristics

The thermodynamic driving force of biological metabolism and the specificity of product detection for exoplanetary life exploration lie in identifying the continuous disturbances of atmospheric components to the thermodynamic chemical equilibrium state. Sulfur, as a key component of life, has a biogeochemical cycle strictly constrained by enzymatic kinetics, presenting chemical stoichiometric characteristics that are completely different from non-biological geological processes.

In the anaerobic ecological niche, microorganisms utilize sulfate ions as the terminal electron acceptor through the dissimilatory sulfate reduction (DSR) pathway [11], and the Gibbs free energy released during this process drives ATP synthesis, inevitably resulting in a significantly higher hydrogen sulfide flux than the geological background supply rate, leading to a thermodynamic non-equilibrium state accumulation [12].

Mercaptoethanol and dimethyl sulfide (DMS) mainly originate from sulfur-containing amino acids and osmotic regulators through enzymatic cleavage. In particular, DMS, due to the instability of the C-S bond in non-biological hydrothermal environments, lacks an effective geological synthesis pathway and is regarded as a highly specific "strong biological signal" [13]. Its presence implies that the planetary surface is highly likely to have a biosphere with complex secondary metabolic capabilities.

Infrared fingerprint characteristics and spectral interference analysis reveal that sulfur-containing molecules have unique vibrational-translational transition cross-sections in the infrared and near-infrared bands. However, in remote sensing observations, these weak signals are easily obscured by the strong absorption bands of the main atmospheric components [14]. Table 1 details the core detection windows and dominant interference sources for key sulfur molecules, derived from high-precision simulations utilizing the HITRAN and ExoMol databases.

Table 1. Spectral Fingerprints and Observational Interferences of Key Sulfur-Based Biosignatures

Target Molecule	Primary Biogenic Mechanism	Key Absorption Bands (μm)	Dominant Interference	Detection Strategy
Hydrogen Sulfide	Dissimilatory Sulfate Reduction (DSR), Protein putrefaction	2.7, 3.8, 10-11	H_2O, CH_4	Focus on mid-infrared window regions to avoid strong absorption bands of H_2O
Methanethiol	Methylation and degradation of sulfur-containing amino acids	3.4 (C-H stretching), 10.5 (C-S stretching)	CH_4(strong masking), O_3	Rely on high-resolution spectroscopy ($R>10^5$) to separate methane line systems
Dimethyl Sulfide	DMSP cleavage by marine phytoplankton	3.4, 14.2	CH_4, C_2H_6	The 14.2μm region has a relatively "clean" background, serving as the optimal observation band for MIRI instruments
Carbon Disulfide	Anaerobic bacterial metabolism, minor geological sources	4.6, 6.5, 11.4	CO_2, H_2O	The 6.5μm band exhibits a sharp peak, enabling identification via characteristic peak shape (Q-branch)

3. Model Construction

3.1 Constraints on the Geochemical Background Values of Geological Tectonic Activities

In the macroscopic picture of planetary atmospheric evolution, magma degassing constitutes the primary original non-biological input vector of sulfur-based volatile substances [15]. The core criterion for distinguishing biogenic sources from geological sources lies in the strict control of the redox state of the planetary mantle on the equilibrium of gaseous components.

The forms of sulfur species in geological emissions are not random but are determined by the chemical equilibrium in the magma melt. Under high temperature and high pressure, the gas-phase equilibrium follows the following thermodynamic constraints:

$$SO_2 + 3H_2 \rightleftharpoons H_2S + 2H_2O \quad (1)$$

The equilibrium constant $K(T,P)$ of this reaction determines that, under the given temperature T and pressure P, the ratio of gas mole fractions, x_{H_2S} / x_{SO_2}, is negatively correlated with the mantle oxygen fugacity, fO_2. For an oxidized mantle similar to that of the modern Earth, volcanic emissions are dominated by SO_2, only under extremely reducing mantle conditions can non-biological processes release high concentrations of H_2S through thermochemical equilibrium [16].

Therefore, if observations show that the planet's atmosphere exhibits oxidizing properties, but at the same time high abundances of reducing sulfides are detected, this constitutes a strong counter-evidence to a single geological cause explanation.

3.2 The Dynamical Dissipation of the Atmospheric Photochemical Network and False Positive Simulations

Even if the geological source is excluded, the stellar ultraviolet radiation-driven photochemical network is both a "destroyer" of biological signals and a "maker" of false positive signals. It is necessary to conduct kinetic modeling to quantify the photochemical lifetime of sulfur-based molecules.

The duration of the existence of sulfur-based gases under stellar UV radiation is determined by its photodissociation rate constant J_i, which is a wavelength-dependent integral function:

$$J_i = \int_{\lambda_{\min}}^{\lambda_{\max}} \sigma_i(\lambda)\Phi(\lambda)\tau(\lambda)d \quad (2)$$

Among them, σ_i represents the absorption cross-section of the molecule, Φ is the stellar luminosity flux of the constant star, and τ is the atmospheric transmittance.

The model shows that DMS and CH_3SH have a relatively short photochemical lifetime ($t_{life} \approx 10^3 - 10^5 s$)in the ultraviolet environment of M-type dwarf stars, and are prone to undergo polymerization reactions to form sulfur-containing organic aerosols. This implies that to maintain a steady-state concentration detectable by remote sensing, the flux supply rate of the biosphere must overcome a huge photochemical dissipation barrier.

It should be noted that in a reducing atmosphere rich in CH_4 and H_2S, photochemical reactions may generate trace amounts of organic sulfur through free radical recombination.

However, as shown in Figure 1, the kinetic simulation indicates that the yield of this abiotic pathway is strictly limited, and the final product abundance is typically 2-3 orders of magnitude lower than that of the biotic source. Specifically, Figure 1A reveals the extreme vulnerability of sulfur-based biogenic characteristic gases in the ultraviolet radiation environment of M-type dwarf stars. The simulation shows that the photolysis lifetime of DMS is strictly distributed within a range. With the increase in UV flux, the survival window of these molecules closes exponentially, forming a significant "photolysis loss window" (the gray shaded area). This means that any concentration that can be effectively accumulated in the atmosphere must rely on a continuous supply source with a rate far exceeding photolysis. Figure 1B further quantifies the gap in this supply demand, contrasting the steady-state yield of the biotic source with the three potential abiotic pathways. The abiotic yield hovers between 10^{-3} to 10^{-4} arbitrary units, confirming that geological or photochemical mechanisms lack the kinetic efficiency required to simulate the high flux release of the biosphere.

The microscopic mechanism of this low abiotic synthesis efficiency is elucidated in Figures 1C and 1E. The abiotic synthesis of complex organic sulfur depends on the effective recombination of intermediate free radicals. However, the simulation shows that due to rapid quenching effects and competitive reactions, the steady-state concentration of these key free radicals rapidly decays to below 0.01 nM (the red shaded area), resulting in a significant "free radical deficiency" effect. Due to the excessive thinness of the reaction intermediates, they cannot effectively drive the secondary kinetic process required to form stable C-S bonds, thus delineating a kinetic gap that cannot be crossed between the biotic and abiotic sources.

Furthermore, Figures 1D and 1F introduce a key observational constraint, the risk of aerosol phase transition. As the DMS concentration and UV flux increase, the system enters the rapid formation zone of aerosols (the red core area in Figure 1F). The simulation tracks the mass growth of sulfur-containing organic aerosols over time (Figure 1D), indicating that the high-intensity biotic flux not only generates detectable gas signals but also generates photochemical haze layers through polymerization reactions. This "phase space" analysis suggests a subtle observational trade-off. Although an active biosphere can generate a strong gas signal, the accompanying photochemical smog may form a shielding effect in the visible light band, which requires future observational strategies to expand to more penetrating mid-infrared bands to avoid potential "self-masking" phenomena of the signals.

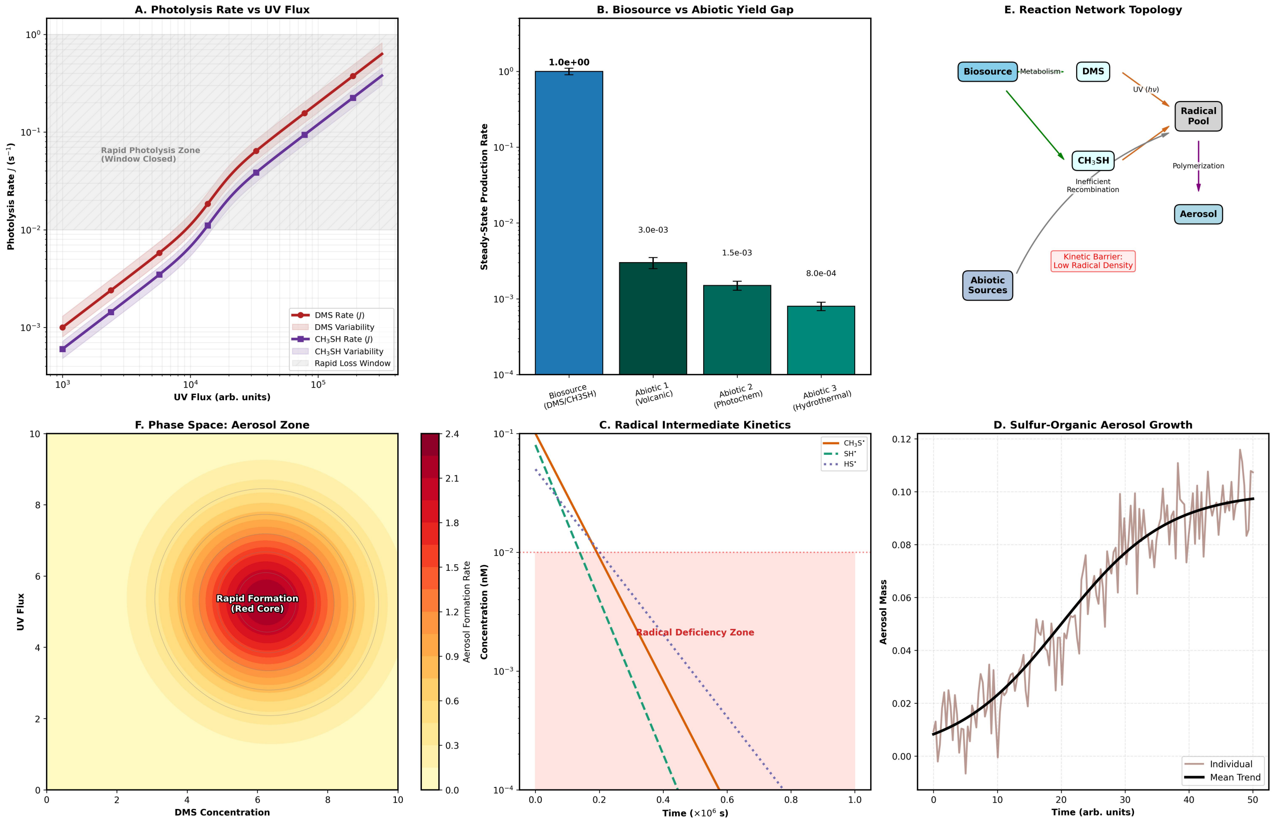

Figure 1. Dynamics Simulation

3.3 Construction of the "Elimination-Confirmation" Decision Matrix

Based on the above thermodynamic and dynamic analyses, we have established a multi-dimensional abiotic cause exclusion matrix. Specifically as shown in Table 2, this matrix couples the observed gas signals with the background of the planetary environment, aiming to eliminate "false positives" and identify biologically candidate targets with high confidence.

Table 2. Abiotic Exclusion and Confidence Assessment Matrix for Sulfur-Based Biosignatures

Observational Context	Predominant Sulfur Species	Abiotic Interpretation	Biosignature Confidence
Oxidized Atmosphere (High CO_2)	SO_2	High (Volcanic)	Null
Oxidized Atmosphere (High CO_2)	H_2S/DMS	Very Low	High
Reduced Atmosphere (High H_2, CH_4)	H_2S	Moderate	Ambiguous
Reduced Atmosphere (High H_2, CH_4)	DMS/CH_3SH	Low	Moderate to High
Hyper-active Star (High UV)	Any VOS (Volatile Organic Sulfur)	N/A (Rapid Photolysis)	Very High

4. Observation Strategy

Although a single sulfur-based signal has indicative significance, isolated evidence always has "uncertainty in interpretation" in statistics. This study advocates the establishment of a "multivariate collaborative verification" system, with the core strategy being to capture the "redox binary opposition" among atmospheric components. Specifically, when reducing sulfur species coexist with highly oxidizing atmospheric background, this strong chemical potential gradient violates the relaxation law of non-biological chemical equilibrium. This "contextual correlation" has higher biological diagnostic value than single gas detection.

Furthermore, it is suggested to introduce associated biogases as auxiliary verification. If DMS and CH_4 or N_2O show synchronous abnormal abundance in the spectrum, this cross-metabolic-path signal coupling will greatly reduce the posterior probability of non-biological causes.

The instrumental sensitivity limit and high dispersion inter-correlation strategy, targeting the weak spectral characteristics and susceptibility to interference of sulfur-based gas spectra, have encountered a bottleneck when relying solely on low-resolution photometric measurements. The observation strategy must transition to high dispersion inter-correlation spectroscopy. By using Doppler frequency shift to separate the stellar and Earth atmospheric spectral lines from the line spectra of the stars and planets, combined with the light-collecting ability of ground-based giant telescopes, the signal-to-noise ratio can be significantly improved. For the instrumental characteristics of different bands, we have formulated a hierarchical observation matrix, as shown in Table 3.

Table 3. Synergistic Observation Strategy for Next-Generation Instruments Targeting Sulfur Biosignatures

Instrument Class	Spectral Domain	Primary Target & Mechanism	Verification Logic & Strategy
Space-based (JWST - NIRSpec/G395)	NIR (2.8−5.2 μm)	H_2S, CH_3SH (C-H band)	Transmission Spectroscopy: Utilize high-precision transmittance spectra to capture H_2S features in the terminator atmosphere, while measuring the CO_2/CH_4 ratio to establish the redox background.
Space-based (JWST - MIRI/LRS)	MIR (5−12 μm)	SO_2 vs. NH_3/VOS (Volatile Organic Sulfur)	Emission Spectroscopy: Directly detect thermal radiation from the planetary dayside. Focus on monitoring the coexistence of 8.6 μm (SO_2) and 10.5 μm (NH_3/CH_3SH) to rule out volcanic origin.
Ground-based (ELT - METIS/HIRES)	L/M/N Bands (High-Resolution)	DMS Isotopologues	High-Dispersion Cross-Correlation: Employ spectroscopy with R>100,000 to resolve the dense rotational spectral lines of DMS, attempt to distinguish the sulfur isotope ratio ($^{34}S/^{32}S$), and search for evidence of biological fractionation effects.

5. Bayesian inference framework

5.1 Contextual Dependence of Cognitive Uncertainty and Prior Probabilities

In the search for life on exoplanets, the interpretation of any spectral signal is inherently embedded within the physical background of the planetary system. Therefore, the evaluation process must first establish a prior probability distribution based on the planetary environment.

Unlike the traditional frequency school, the Bayesian framework allows us to integrate existing knowledge from geology, stellar physics, and planetary formation theories as prior constraints. For the hypothesis of sulfur-based biological signals, its prior probability is not a uniform value but highly dependent on the "habitable situation":

$$P(H_{bio} \mid C) = f(T_{eq}, \text{Redox}, \text{Age}, \text{Star}) \quad (3)$$

Among them, C represents the set of planetary environmental parameters. For example, for a rocky planet located within the habitable zone and with an atmosphere showing a highly oxidized-reduced imbalance, we will assign a higher prior probability of biological origin, conversely, for planets with extreme high temperatures or intense radiation, the prior probability will approach zero. This "context-dependent prior" setting is the first line of defense against statistical errors.

5.2 Posterior Probability Iteration and Marginalization of Non-Biological Likelihood Functions

The core of scientific confirmation lies in updating beliefs using observational data. According to Bayes' theorem, the posterior probability of the existence of sulfur-based biological signals can be expressed as:

$$P(H_{bio} \mid \mathrm{D}) = \frac{P(D \mid H_{bio})P(H_{bio})}{P(D \mid H_{bio})P(H_{bio}) + P(D \mid H_{abio})P(H_{abio})} \quad (4)$$

In this formula, the denominator term constitutes the crucial "sum of evidence". The core innovation of this study lies in the handling of the non-biological likelihood function $P(D \mid H_{abio})$. Unlike previous studies that often ignored or underestimated the non-biological explanations, we advocate using the Markov Chain Monte Carlo algorithm to perform adequate marginal integration of all known geological and photochemical model parameters.

Only when the observed sulfur-based gas spectral features cannot be fitted by any non-biological model with an extremely high confidence level, will the posterior probability show a significant upward trend.

5.3 Decision Tree Model and Detection Confidence Grading

To convert the abstract probability values into actionable scientific decisions, we constructed a hierarchical verification decision tree based on the Bayesian factor. This model defines the scientific attributes of the detection results based on the threshold of the posterior probability. Level 1 ($P > 2\sigma$) indicates that the observed data deviates from the non-biological background baseline and is defined as "an interesting anomalous signal", triggering a subsequent high-resolution re-measurement, Level 2 ($P > 3\sigma$) indicates that the known geological and photochemical conventional explanations have been excluded, but there is still model parameter degeneracy, defined as "candidate biological signals", Level 3 ($P > 5\sigma$) indicates that in multi-band and multi-time period observations, the data robustly supports the biological origin hypothesis, and the Bayesian factor ratio of all non-biological alternative models is lower than 10^{-4}, which can be cautiously declared as "life confirmation".

6. Conclusions and Prospects

The core objective of this study is to establish the "non-biological exclusion priority" detection logic, whose physical essence lies in the dual specificity of sulfur metabolism in terms of thermodynamics and kinetics. As confirmed by the kinetic simulation, the non-biological synthesis of volatile organic sulfur has an extremely high activation energy barrier.

In the context of planetary geochemistry, thermodynamic equilibrium tends to produce simple molecules, while complex molecules such as DMS are outside the "thermodynamic potential well" of the non-biological environment. The "free radical deficiency" effect identified in the reaction network acts as a robust physical filter. Without the intervention of enzymatic catalysis, the probability of rare free radicals colliding in the non-biological environment is extremely low, and it is impossible to accumulate macroscopic abundances of ppm levels under the dominance of photochemical destruction.

Therefore, detecting high concentrations of DMS is not merely a chemical anomaly, but a direct falsification of the standard planetary Earth chemistry model at the kinetic level. However, the "aerosol masking paradox" discovered in the phase space analysis reveals that the enhancement of biological flux may trigger the nonlinear growth of photochemical haze layers. In a high ultraviolet radiation environment, the active sulfur-based biosphere is highly likely to evolve into a hazy state similar to Titan. This aerosol composed of organic sulfur has strong scattering properties in the visible and near-infrared bands, which may lead to the flattening of the transmitted spectrum and thereby mask the molecular fingerprints we are trying to detect. This discovery indicates that relying solely on the transmission spectrum of the dawn-dusk line may face the risk of "false negatives", and shifting to mid-infrared thermal emission spectroscopy observations, utilizing the long-waveband's ability to penetrate fine aerosols, will be the inevitable path for future breakthroughs in the "biological self-shielding" effect.

This study addresses the spectral degeneracy problem of sulfur-based biological characteristic gases in the detection of exoplanetary life, and has constructed a systematic verification framework integrating biochemical mechanisms, photochemical kinetics simulations, and Bayesian statistical inference. The research shows that although molecules such as DMS face strong background shielding in the infrared band, by locking onto specific "escape windows" in the mid-infrared region and combining the contextual constraints of the planet's atmospheric redox state, we have the ability to statistically separate geological and photochemical noise.

The core contribution of this work lies in establishing the epistemological principle of "non-biological exclusion priority", that is, the establishment of life signals does not depend on the similarity of observational features to the Earth's biosphere, but on the "unfeasibility" of being explained by non-biological models within the physical and chemical allowable space. By introducing a multivariate collaborative verification mechanism and a hierarchical decision tree, we have upgraded the traditional qualitative search to quantitative probability assessment, providing a robust theoretical foundation for processing high-dimensional spectral data in the era of the James Webb Space Telescope.

When reviewing the current knowledge boundaries, it must be pointed out that the sources of uncertainty in this study still exist. This is mainly attributed to the lack of basic physical and chemical parameters, which may lead to deviations in the forward simulation of radiation transmission models. In addition, the nucleation rate and optical properties of the conversion from sulfur chemistry to organic sulfur aerosols are still unclear. If the planet's atmosphere contains a thick photochemical haze layer, its continuous spectral absorption may "smooth out" the weak molecular fingerprints, resulting in an "flat spectrum" and causing false negative misjudgments.

Looking forward to the future, exoplanetary life detection is at a historical node of transitioning from the "discovery era" to the "characterization era". To transform the theoretical framework proposed in this study into practical discovery results, future research should focus on collaborative efforts in laboratory astrophysics, establish sulfur chemical reaction dynamics networks under extreme environments, build a high-precision "exoplanetary sulfur chemistry genome" database containing various isotopic isomers, to improve the uniqueness of spectral interpretation. And relying on the high-dispersion spectral technology of the ELT, expand the detection dimension from one-dimensional spectral variations to multi-dimensional velocity space, and through the Doppler shift generated by the planet's rotation and revolution, achieve the dynamic decoupling of biosphere gas and non-biological background.

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