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Deciphering the origin of organo-sulphur hazes: an exploration of the actual limits of sulphur chemistry with potential implications for prebiotic chemistry, the Early-Earth, and habitability of sulphur exoplanets

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Deciphering the origin of organo-sulphur hazes: an exploration of the actual limits of sulphur chemistry with potential implications for prebiotic chemistry, the Early-Earth, and habitability of sulphur exoplanets

Introduction

Sulphur volatiles are components currently observed in the atmospheres of planetary bodies. This is true for the solar system with the examples of Venus and Io, but also for extrasolar systems as suggested by the recent findings of the James Webb Space Telescope (JWST). Infrared Transit spectroscopy performed with this instrument revealed SO₂ signatures in several different types of exoplanets such as hot-Jupiter, warm-Neptunes, temperate Sub-Neptune, or Super-Earth. This highlights the extended diversity of objects in terms of atmospheric conditions which host sulphur species. In many of these very diversified contexts, photochemical processes involving sulphur volatiles are known to play a key role in the chemistry of the global atmosphere. However, despite the recurrence of sulphur in planetary/exoplanetary atmospheres and the importance of the photochemistry induced by such species, sulphur atmospheric reactivity remains in some respects poorly understood. One of the most relevant examples to illustrate this lack of knowledge concerns sulphur aerosol formation processes. Several laboratory experiments revealed the existence of organo-sulphur hazes which are currently not predicted by atmospheric simulation models. To overcome this discrepancy between experimental observations and model, the chemical mechanisms leading to the formation of such aerosols need to be identified. The understanding of such mechanism could notably have great implications for the Early-Earth at the Archean era. During this period, the atmospheric composition, containing significant amount of CO₂, and CH₄, coupled with emission of sulphur volatiles from volcanic activity could have favoured the formation of such hazes. In this context, organo-sulphur hazes represent a new sink in the sulphur atmospheric budget which could considerably change the description of sulphur chemistry. This unexplored reactivity could change our current understanding of the origin of sulphur isotopic fractionation (Sulphur-Mass Independent Fractionation S-MIF) observed in ancient rocks which is one of the most relevant paleoclimate indicator.

Methods

In this work we adopt an experimental approach to better constrain the mechanisms of formation of organo-sulphur aerosols. The objectives were more precisely to characterise the molecular signatures of the organo-sulphur molecules contained in such hazes with their chemical functions, and to identify potential precursors of these aerosols in gas phase. To do so, we synthesise organo-sulphur aerosols in a N₂/CH₄/S (where S represents either SO₂ either H₂S) plasma mixtures. We

deliberately considered more concentrated conditions in sulphur volatiles than the previous experiments (from 0.1% up to 10%) to enhance the sulphur signatures in the synthesised hazes. Such high sulphur proportions are also necessary to identify the sulphur volatiles precursors in the gas phase, which constrain the first elementary steps leading to the formation of hazes.

Results

The analyses have revealed several sulphur volatiles (such as CS_2 , H_2S , OCS , CH_3SH) but none of them have been observed specifically in presence of organo-sulphur hazes. This suggests that the incorporation of sulphur into the organic matter is mainly done by direct addition of sulphur radicals/intermediates on existing organic chains and not by an organic growth implying organo-sulphur volatile precursors. Analyses were also performed on the solids produced. In the figure below, spectra of two of the organo-sulphur aerosol samples show a strong organo-sulphur band around 2060cm^{-1} attributed to isothiocyanate function ($-\text{N}=\text{C}=\text{S}$). This could result from an addition of sulphur into an isonitrile group of the organic chain. This remark is consistent with the anti-correlation observed between the nitrile/isonitrile band and the isothiocyanate one. Finally, GC-MS measurements performed on the solid particles produced allow to identify the corresponding isothiocyanate organics with the detection of methyl-isothiocyanate, ethyl-isothiocyanate and butyl-isothiocyanate (see Fig.2). Thiocyanide ethers (R-SCN) were also observed. It is the first time that such functions are observed in organo-sulphur hazes matrix. This finding provides new elements in the understanding of the formation of such hazes by highlighting the strong coupling existing between nitriles/isonitriles and sulphur chemistry. These results prompt us to consider more deeply the heterogenous reactivity of sulphur on organic matrix which appears to be the dominant source of organo-sulphur hazes in the conditions studied.

Chemical characterisation of organo-sulphur aerosol as well as their formation route have potentially great impacts for the habitability of sulphur exoplanets. Indeed, their organic composition may include complex molecules composed of C,H,N,O,S which are five of the six indispensable elements to life on Earth: the CHNOPS. These hazes strongly differ from the inorganic $\text{S}_x/\text{H}_2\text{SO}_4$ clouds which have been the main sulphur condensed forms considered so far for this type of atmosphere. These remarks also highlight the prebiotic potential of such material which can give clues about the origin of sulphur in the organic matter of life.

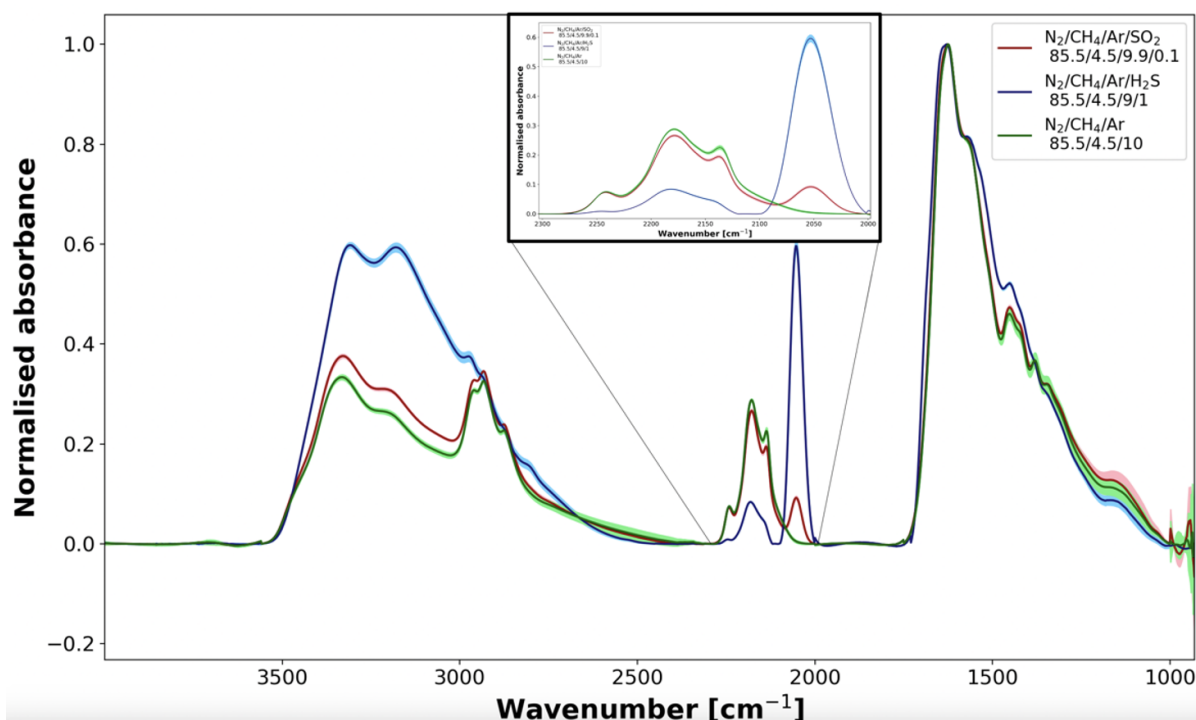


Fig. 1. Comparison between the Infrared spectrums of an organic aerosol produced without sulphur in green, and two organo-sulphur samples produced with an input of H₂S in blue and an input of SO₂ in red. The details of the composition used to form these deposits are indicated in the legend. An intense band at 2060cm⁻¹ is specific to sulphur samples. This suggests the presence of isothiocyanate -N=C=S which gives indication about the sulphur functions present in the sample. Moreover, the blue spectrum (produced with H₂S) present strong differences with the two others. The analysis of its differences gives clues to the mechanism associated in the formation of these organo-sulphur samples.

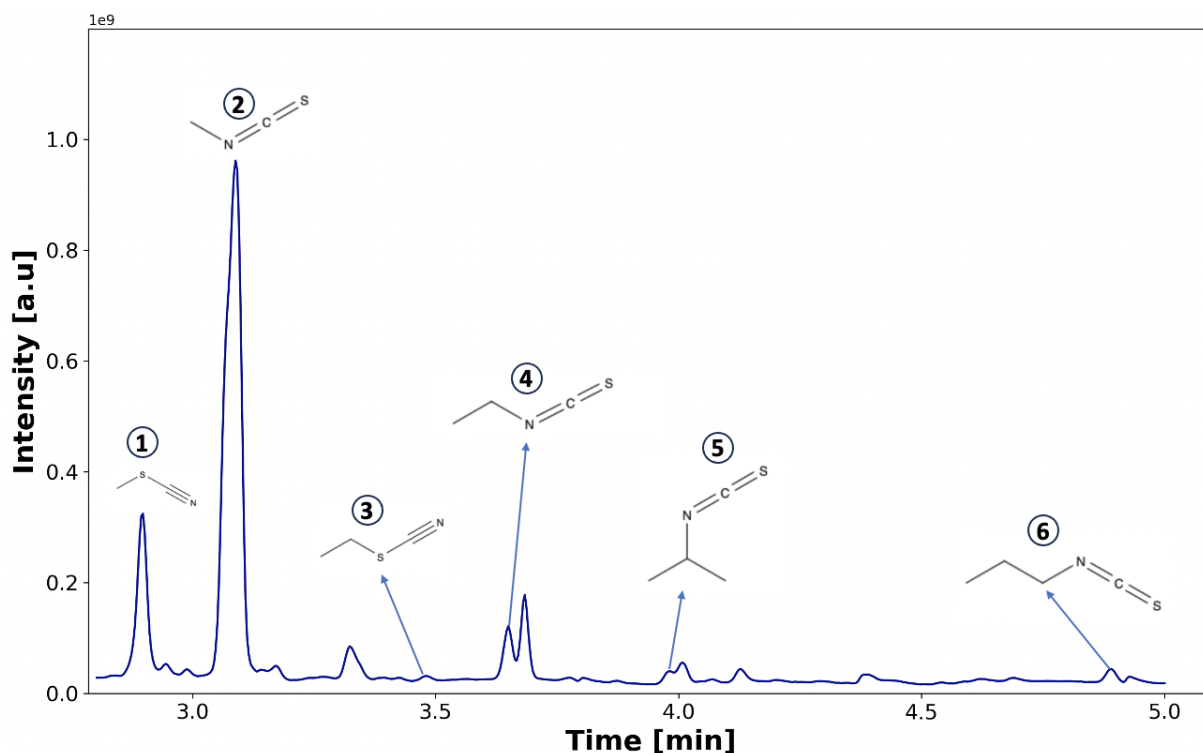


Fig. 2. Example of chromatogram of one organo-sulphur aerosol sample produced (N₂/CH₄/Ar/H₂S 85.5/4.5/9/1) got after a pyrolysis between 200 and 400°C with a ramp of 35°C/min. Several organic isothiocyanate (2=Methyl, 4=Ethyle, 5=Isopropyle, 6=Butyle) and thiocyanide ethers (1=Methyl, 3=Ethyl) are observed in this chromatogram. The different peaks are associated to their corresponding species with the arrows.