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Carbon Disulfide (CS₂) Mechanisms in Formation of Atmospheric Carbon Dioxide (CO₂) Formation from Unconventional Shale Gas Extraction and Processing Operations and Global Climate Change

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ABSTRACT: Carbon disulfide (CS₂) has been historically associated with the production of rayon, cellophane, and carbon tetrachloride. This study identifies multiple mechanisms by which CS₂ contributes to the formation of CO₂ in the atmosphere. CS₂ and other associated sulfide compounds were found by this study to be present in emissions from unconventional shale gas extraction and processing (E&P) operations. The breakdown products of CS₂; carbonyl sulfide (COS), carbon monoxide (CO), and sulfur dioxide (SO₂) are indirect greenhouse gases (GHGs) that contribute to CO₂ levels in the atmosphere. The heat-trapping nature of CO₂ has been found to increase the surface temperature, resulting in regional and global climate change. The purpose of this study is to identify five mechanisms by which CS₂ and the breakdown products of CS₂ contribute to atmospheric concentrations of CO₂. The five mechanisms of CO₂ formation are as follows:

1. Chemical Interaction of CS₂ and hydrogen sulfide (H₂S) present in natural gas at high temperatures, resulting in CO₂ formation;
2. Combustion of CS₂ in the presence of oxygen producing SO₂ and CO₂;
3. Photolysis of CS₂ leading to the formation of COS, CO, and SO₂, which are indirect contributors to CO₂ formation;
4. One-step hydrolysis of CS₂, producing reactive intermediates and ultimately forming H₂S and CO₂;
5. Two-step hydrolysis of CS₂ forming the reactive COS intermediate that reacts with an additional water molecule, ultimately forming H₂S and CO₂. CS₂ and COS additionally are implicated in the formation of SO₂ in the stratosphere and/or troposphere. SO₂ is an indirect contributor to CO₂ formation and is implicated in global climate change.

KEYWORDS: global climate change, carbon disulfide, GHG, carbon dioxide

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Introduction

Carbon disulfide (CS₂) is a chemical intermediate best known for its historical use in the production of rayon, cellophane, and carbon tetrachloride.^{1–3} Industrial emissions of CS₂ to the atmosphere have declined as textile manufacturing has shifted from the US to Asia, and the use of CS₂ in carbon tetrachloride production has been phased out.⁴ With energy extraction expanding across the US, emission of CS₂ may be on the rise and its contribution in formation of greenhouse gas (GHGs) is underreported.

CS₂ is a component of sour natural gas and occurs naturally in the environment from the degradation of organic material, including geologic deposits of oil and natural gas; it is a waste gas emitted from the processing of sour natural gas.^{5,6} In natural gas extraction, CS₂ is used as a paraffin solvent for sulfur, phosphorous, selenium, bromine, resins, and rubber.⁷

It is also a component of mercaptan, an odorant added to natural gas.⁸ Mercaptans may also be removed from a gas stream by oxidation to disulfides, which are then easily separated from the gas stream by absorption.⁹ It can be produced from the interaction of natural gas with hydrogen sulfide at high temperatures. CS₂ is also known to be released in fossil fuel combustion, including natural gas combustion. During the extraction of natural gas, venting and flow-back operations release large amounts of gases, including CS₂, directly to the atmosphere.^{10,11}

Recent ambient air monitoring studies have identified CS₂ and other associated sulfide compounds present in emissions emanating from natural gas exploration and production (E&P).^{12–15} As CS₂ is present in many geologic formations and aspects of energy extraction, residential communities experiencing the trend of “urban drilling” – ie, extraction and



processing of natural gas in populated urban communities – may be exposed to higher concentrations of CS₂ in ambient air when compared to other residential areas not experiencing extraction and processing.¹⁶

CS₂ is categorized as a non-methane volatile organic compound (NMVOC) and classified, along with carbonyl sulfide, as a hazardous air pollutant (HAP) according to the Clean Air Act Amendment of 1990 (CAAA). NMVOCs are a category of chemicals commonly used as solvents in industrial processes with the ability to vaporize at room temperature.¹⁷ NMVOCs may be classified as a direct or indirect GHG, and can be involved in indirect radiative reactions that form CO₂.¹⁴ NMVOCs indirectly contribute to global climate change by producing GHGs, such as CO₂, through reactions with other compounds, through their own chemical transformation influencing atmospheric lifetime of other GHGs, and by affecting the absorptive characteristics of the atmosphere such as cloud formation.^{18,19} In an indirect radiative reaction, a chemical breaks down in the atmosphere producing a GHG, or interacts with other chemicals in the atmosphere changing atmospheric concentrations of GHGs. The size of the indirect effect is dependent upon when and where the gas is emitted.^{20,21} CO₂ is a GHG that is transparent to incoming solar radiation but with the capacity to easily absorb and trap infrared radiation. This allows heat to be retained at the Earth's surface, contributing to ground-level ozone and global climate change.²² Ground-level ozone can damage crops and human health, and can lead to reduced crop production.^{23,24}

This study is the first to identify five direct and/or indirect mechanisms by which CS₂ contributes CO₂ to the atmosphere and exhibits its capacity as a GHG and contributor to global climate change. This study is also one of the first to identify the presence of CS₂ and other sulfide compounds in emissions from unconventional shale gas E&P operations.

Materials and Methods

A literature review was performed examining previous work related to occupational exposure to CS₂ and any mechanism by which CS₂ contributes to CO₂ atmospheric concentrations and to CS₂ use or presence in emissions from natural gas extraction or processing operations.

Key words searched included CS₂, CS₂ and CO₂, CS₂ occupational exposure, natural gas E&P emissions, and CS₂ mechanism of CO₂ formation. Databases searched included MEDLINE, TOXLINE, Scopus, Science Direct, TOXNET, and PubMed. Although many articles were found identifying CS₂ exposure in occupational settings, they were mostly published prior to 1980. Recent publications on ambient air monitoring in regions experiencing energy extraction analyzed air samples collected for CS₂, but failed to identify the potential CS₂ has as a GHG and contributor to global climate change. No article to date was found identifying all five potential mechanisms by which CS₂ might form CO₂ in the

atmosphere or the potential for COS and SO₂ to be contributors to CO₂ formation.

Results

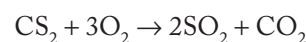
The five potential mechanisms by which CS₂ may form CO₂ either directly or indirectly and contribute to GHG atmospheric concentrations are described in this paper (Fig. 1).

Mechanism 1 – Chemical interaction. The interaction of CS₂ and natural gas with hydrogen sulfide (H₂S) at high temperatures can form CO₂.²⁵



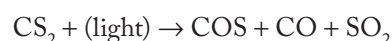
In the document *Report to Congress on Hydrogen Sulfide Air Emissions Associated with the Extraction of Oil and Gas*, gas processing operations were identified as potential sources of H₂S releases. Impurities present in natural gas (produced water, H₂S, and CO₂) are removed prior to the gas being compressed and shipped in pipelines. This removal process is often performed at pad sites located in residential areas where extraction occurs and may be a source of H₂S.²⁶

Mechanism 2 – Combustion. Combustion of CS₂ in the presence of oxygen produces CO₂ and SO₂.



CS₂ is highly flammable and can ignite easily. When exposed to spark or friction, it is known to easily combust. Contact with steam pipes or even light bulbs have been known to initiate the combustion of CS₂.^{27–31}

Mechanism 3 – Photolysis. Photolysis of CS₂ leads to the formation of carbonyl sulfide (COS), CO, and SO₂.



CS₂ may have an indirect effect on global climate change through the main transformation product COS. CO is only a weak direct GHG but has important indirect effects on global climate change. CO reacts with hydroxyl (OH) radicals in the atmosphere, reducing their abundance. As OH radicals help the atmospheric lifetimes of strong GHGs such as methane, CO may indirectly increase the global climate change potential of these gases from OH radical scourging.^{32–34}

The half-life of CS₂ in the atmosphere from photolysis is estimated to be 1 week, whereas that of COS is estimated at 2 years, which provides the opportunity for long-range chemical transport and atmospheric conversions.^{35,36}

Chin reported an additional indirect source of CS₂ present in sulfur recovery operations in the oil industry, which is oxidized COS in the atmosphere.⁶

Mechanism 4 – One-step hydrolysis. Shangguan identified two mechanisms of hydrolysis of CS₂. In the one-step hydrolysis mechanism, CS₂ directly reacts with the hydrogen atoms in two molecules of water, producing CO₂ and H₂S.^{37–39}

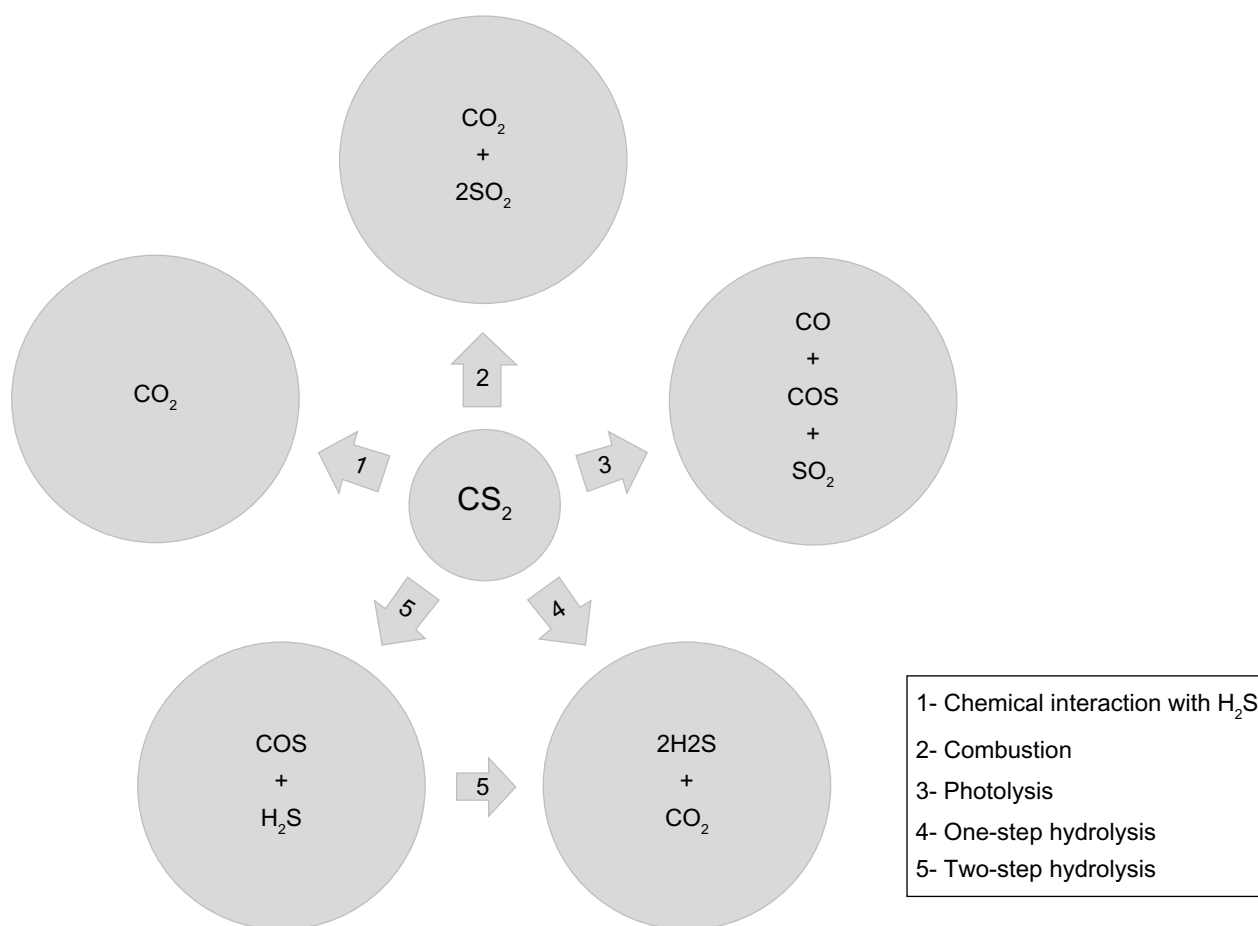
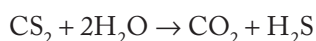
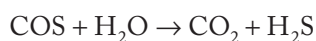
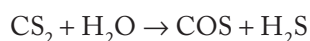


Figure 1. Five mechanisms of CO₂ formation from CS₂.



Mechanism 5 – Two-step hydrolysis. In a two-step hydrolysis mechanism, CS₂ reacts with a hydrogen atom in a water molecule, leading to reactive intermediate COS. COS then reacts with an additional water molecule to form CO₂ and H₂S.



Research has shown that hydrolysis of CS₂ can occur at room temperature at a slow rate.⁴⁰

A previous study by the authors identified CS₂ and 12 other sulfide compounds present in ambient air in residential communities in the Barnett Shale geologic formation where natural gas E&P operations were being carried out.⁴¹ CS₂ concentrations and the detection frequency were found to vary with distance from the natural gas emission source. Values ranged from 0.3 to 200 parts per billion by volume (ppbv) based on a 24-hour air monitoring period, and from 1.5 to 980 ppbv based on a 1-hour monitoring period. The results presented in Table 1.

The U.S. Environmental Protection Agency's Urban Air Toxics Monitoring Program (NMP) comprising 52 sites around the US reported only CS₂ as a monitored sulfide compound comparable to the values in this study. CS₂ atmospheric concentrations based on a 24-hour air monitoring period showed a minimum level for CS₂ of 0.005 ppbv and a maximum of 0.193 ppbv. Sampling period was for up to 12 consecutive months.⁴² CS₂ is denser than air (2.62 vapor density when compared to air = 1) and can settle close to the ground in ground-level breathing zones.⁴³

Discussion

The potential contribution of CS₂ to atmospheric CO₂ and GHG levels warrant further examination due to its unique qualities as a NMVOC and HAP. COS, CO, SO₂, breakdown products of CS₂, and indirect greenhouse gases additionally contribute to CO₂ formation in the atmosphere. The transparency of CO₂ to incoming solar radiation allows it to trap and absorb infrared radiation at the Earth's surface. This, in turn, gives rise to ground-level ozone formation, which is a major factor in climate change.

Many urban cities experiencing natural gas E&P are designated as nonattainment areas with air quality below National Ambient Air Quality Standards (NAAQS). Iden-



Table 1. CS₂ and associated sulfide compounds in natural gas emissions 24- and 1-hour minimum and maximum concentrations in parts per billion by volume (ppbv).

CHEMICAL (PPBV)	MIN. 24 HOURS	MAX. 24 HOURS	MIN. 1 HOUR	MAX. 1 HOUR
Carbon disulfide	0.7	103.0	3.4	504.6
Carbonyl sulfide	0.3	36.7	1.5	180.0
Dimethyl disulfide	0.3	200.0	1.5	980.0
Methyl ethyl disulfide	0.3	145.0	1.5	710.0
Methyl propyl disulfide	0.3	41.6	1.5	204.0
Diethyl disulfide	0.3	32.7	1.5	160.2
Ethyl, methylethyl disulfide	0.3	46.7	1.5	228.8
Dimethyl trisulfide	1.2	46.3	5.9	226.8
Ethyl <i>n</i> -propyl disulfide	0.3	25.2	1.5	123.5
Diethyl trisulfide	0.3	8.2	1.5	40.3
Methyl <i>n</i> -butyl disulfide	0.3	15.5	1.5	76.0
Propyl <i>n</i> -butyl disulfide	0.3	14.6	1.5	71.5
Dipropyl disulfide	0.3	23.1	1.5	113.2

tification of airborne chemicals contributing to CO₂ levels as well as the sources of those emissions is critical in order to manage air quality and achieve NAAQS compliance. The quantification of emissions from chemical manufacturing facilities producing CS₂ in the US may be easy due to the major source reporting requirements. Quantification of CS₂ emissions from natural gas E&P operations is more difficult due to the lack of required emission reporting and the vast number of well sites. Additionally, each pad site may have multiple sources of CS₂ emission. As CS₂ escapes into the atmosphere, to quantify secondary atmospheric conversion becomes a challenge. While CS₂ is a direct contributor to atmospheric CO₂ levels, it is also an indirect contributor and is responsible for intermediates that may also produce CO₂ in the atmosphere.

Currently, there is a profound lack of information and quantification of CS₂ emissions from natural gas E&P operations. At facilities where combustion takes place, CS₂ may be counted overall in VOC emissions. Quantification of CS₂ or speciation of sulfide compounds is not currently required. Additionally, these data are self-reported and may be more qualitative rather than quantitative. Although CS₂ and COS are HAPs, capable of adversely impacting health, this chemical has been considered to be primarily an occupational exposure agent. Current peer-reviewed nonoccupational health studies are lacking and outside the scope of this paper.

This study provides additional knowledge to the atmospheric puzzle, but more questions remain. Future studies are needed to examine and quantify the direct and indirect contribution of CS₂ and intermediates of CS₂ to atmospheric CO₂ formation and GHG levels. Quantification of direct source CS₂ emissions from natural gas E&P operations is also required to

better understand how CS₂ originating from energy extraction may contribute to local and regional CO₂, GHG, and HAP levels. With additional knowledge, we can prepare proper mitigation strategies for this previously unidentified and underreported chemical that contributes to CO₂ concentrations in the atmospheric and contributes to global climate change.

Author Contributions

Conceived and designed the experiments: ALR, JTP. Analyzed the data: ALR, JTP. Wrote the first draft of the manuscript: ALR, JTP. Contributed to the writing of the manuscript: ALR, JTP. Agree with manuscript results and conclusions: ALR, JTP. Jointly developed the structure and arguments for the paper: ALR, JTP. Made critical revisions and approved final version: ALR, JTP. Both authors reviewed and approved of the final manuscript.

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