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# Identification and Selection of Major Carbon Dioxide Stream Compositions

GV Last  
MT Schmick

June 2011



**Pacific Northwest**  
NATIONAL LABORATORY

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Richland, Washington 99352



## Abstract

A critical component in the assessment of long-term risk from geologic sequestration of carbon dioxide (CO<sub>2</sub>) is the ability to predict mineralogical and geochemical changes within storage reservoirs as a result of rock-brine-CO<sub>2</sub> reactions. Impurities and/or other constituents in CO<sub>2</sub> source streams selected for sequestration can affect both the chemical and physical (e.g., density, viscosity, interfacial tension) properties of CO<sub>2</sub> in the deep subsurface. The nature and concentrations of these impurities are a function of both the industrial source(s) of CO<sub>2</sub>, as well as the carbon capture technology used to extract the CO<sub>2</sub> and produce a concentrated stream for subsurface injection and geologic sequestration.

This report summarizes the relative concentrations of CO<sub>2</sub> and other constituents in exhaust gases from major non-energy-related industrial sources of CO<sub>2</sub>. Assuming that carbon capture technology would remove most of the incondensable gases N<sub>2</sub>, O<sub>2</sub>, and Ar, leaving SO<sub>2</sub> and NO<sub>x</sub> as the main impurities, the authors of this report selected four test fluid compositions for use in geochemical experiments. These included the following: 1) a pure CO<sub>2</sub> stream representative of food-grade CO<sub>2</sub> used in most enhanced oil recovery projects; 2) a test fluid composition containing low concentrations (0.5 mole %) SO<sub>2</sub> and NO<sub>x</sub> (representative of that generated from cement production); 3) a test fluid composition with higher concentrations (2.5 mole %) of SO<sub>2</sub>; and 4) test fluid composition containing 3 mole % H<sub>2</sub>S.



## Acronyms and Abbreviations

ARRA	<i>American Recovery and Reinvestment Act</i>
CaO	calcium oxide
CCS	carbon capture and sequestration
CH <sub>4</sub>	methane
CO	carbon monoxide
CO <sub>2</sub>	carbon dioxide
EOR	enhanced oil recovery
EPA	U.S. Environmental Protection Agency
GHG R&D	IEA's Greenhouse Gas Research and Development (program)
H <sub>2</sub>	hydrogen
H <sub>2</sub> O	water
H <sub>2</sub> S	hydrogen sulfide
HCl	hydrogen chloride
IEA	International Energy Agency
IGCC	Integrated Gasification Cycle
MEA	monoethanolamine
N <sub>2</sub>	nitrogen
NH <sub>3</sub>	ammonia
NO	nitric oxide
NO <sub>2</sub>	nitrogen dioxide
NO <sub>x</sub>	nitrogen oxide
O <sub>2</sub>	oxygen
ppm	parts per million
ppmv	parts per million by volume
scf	standard cubic feet
SCR	selective catalytic reduction
SO <sub>2</sub>	sulfur dioxide
SO <sub>3</sub>	sulfur tri-oxide
SO <sub>x</sub>	sulfur oxide
TDF	tire-derived fuels
VOC	volatile organic compound





## Glossary

**Calcination:** A thermal treatment process typically used for decomposition of calcium carbonate (limestone) to calcium oxide (lime) and carbon dioxide.

**Carbon black:** A fine black amorphous form of carbon, principally used as a reinforcing agent in rubber, and as a black pigment in inks, surface coatings, paper, and plastics.

**Co-contaminants:** Chemical impurities found in CO<sub>2</sub> source streams.

**Coke:** A solid carbonaceous material, derived from destructive distillation of low-ash, low-sulfur bituminous coal, and typically used as a fuel and reducing agent in smelting iron ore in a blast furnace.

**Critical temperature:** The temperature above which a gas cannot be liquefied, regardless of the pressure applied.

**Desulfurization:** The removal of sulfur or sulfur compounds (e.g., SO<sub>2</sub> from exhaust flue gases).

**Feedstock:** Bulk raw material supplied (or fed) to an industrial process.

**Flue gas:** Exhaust gas (typically combustion exhaust gas from power plants) released to the atmosphere via a flue (e.g., pipe).

**Interfacial tension:** The surface tension at the interface between two nonmiscible liquids.

**Mercaptans:** A group of sulfur-containing organic compounds.

**Oxy-fuel combustion:** The process of burning a fuel using pure oxygen instead of air as the primary oxidant.

**Polymers:** A class of large organic molecules (including natural and synthetic materials) composed of repeating chains or networks linked by carbon-carbon bonds and exhibiting a wide variety of properties.

**Saturation pressure:** The pressure at a corresponding boiling point where a liquid boils into its vapor phase.

**Sinter:** A coarse-grained product produced by heating fine grain raw material (powder) below its melting point until its particles adhere to each other. Iron ore sinter is used for charging a blast furnace.

**Supercritical CO<sub>2</sub>:** A fluid state of carbon dioxide where it is held at or above its critical temperature and critical pressure, and where distinct liquid and gas phases do not exist.

**Wettability:** The affinity of a fluid to spread out on a solid substrate. This is dependent on the surface tension (and contact angle) between the solid surface and a drop of fluid.



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## 1.0 Introduction

This report summarizes the chemical impurities typically found in major carbon-dioxide (CO<sub>2</sub>) source streams, and identifies some prototypical source stream compositions for use in geochemical experiments. This work supports the *American Recovery and Reinvestment Act* National Risk Assessment Partnership, aimed at providing scientific basis for risk assessments with respect to the long-term storage of CO<sub>2</sub>.

The primary objectives of this report are to achieve the following:

- Identify major CO<sub>2</sub> source streams from industrial sources outside the power (energy) production industry that contain co-contaminants
- Identify most industrially important co-contaminants to consider in assessing long-term geologic storage of CO<sub>2</sub>
- Select a few prototypical CO<sub>2</sub> waste stream compositions for use in experimental studies.

## 2.0 Background

Geologic sequestration of CO<sub>2</sub> is a promising technology for stabilizing atmospheric greenhouse gas concentrations by separating and capturing CO<sub>2</sub> from industrial or energy-related sources, transporting it to a storage location, and injecting it deep underground for long-term isolation from the atmosphere. A critical component in the assessment of long-term risk from these activities is the ability to predict mineralogical and geochemical changes within storage reservoirs due to rock-brine-CO<sub>2</sub> reactions. Impurities and/or other constituents selected for co-sequestration can affect both the chemical and physical (e.g., density, viscosity, interfacial tension) properties of CO<sub>2</sub> in the deep subsurface. The generic CO<sub>2</sub> Features, Events and Processes database (Savage et al. 2004) lists the composition of the CO<sub>2</sub> source stream(s) as a key feature to be considered for the assessment of long-term performance and safety of geologic storage of CO<sub>2</sub>.

Numerous studies have been performed using pure supercritical CO<sub>2</sub>, and some efforts have been expended to examine co-contaminants relevant to power (energy) production. However, few studies have focused on co-contaminants that are unique to major non-energy-related industrial sources. The intent of the *American Recovery and Reinvestment Act* National Risk Assessment Partnership is to use site-specific materials and CO<sub>2</sub> stream compositions (including co-contaminants) from existing or planned large-scale industrial carbon capture and sequestration (CCS) projects to experimentally identify geochemical mechanisms and their impact on long-term risk profiles.

The first step in this effort is to identify the most industrially important co-contaminants and their potential concentrations, and to identify a few prototypical CO<sub>2</sub> waste stream compositions for use in these experimental studies. Section 3.0 provides a summary of the major sources of CO<sub>2</sub> and an overview of carbon capture technologies. Section 4.0 provides a description of potential impurities/co-contaminants typically found in stack emissions from the largest non-energy industrial sources. Section 5.0 summarizes some potential post-carbon capture CO<sub>2</sub> sequestration source stream concentrations produced from large non-energy industrial sources for possible use in experimental studies.

### 3.0 Major CO<sub>2</sub> Source Streams

CO<sub>2</sub> is emitted to the atmosphere both naturally through the carbon cycle and through human activities. The focus of this review is on anthropogenic CO<sub>2</sub> emissions (particularly from non-energy-related industrial sources) and their co-contaminants and concentrations that could be targeted for sequestration. These co-contaminants are a function of both the industrial source(s) of the CO<sub>2</sub>, as well as the carbon capture technology used to extract the CO<sub>2</sub> and produce a concentrated stream that can be compressed into liquid form and readily transported to a geologic sequestration site.

#### 3.1 Major Sources of Anthropogenic CO<sub>2</sub> Emissions

The predominant source of anthropogenic CO<sub>2</sub> emissions is the combustion of fossil fuels (coal, oil and natural gas) in power plants, automobiles, industrial facilities, and other sources (EPA 2010, p. 1-3). Non-energy production processes, such as cement production, metal production, and the use of petroleum-based products also emit notable quantities of CO<sub>2</sub>. Forest clearing activities and other biomass burning also contribute to CO<sub>2</sub> emissions. Table 1 summarizes the major sources of CO<sub>2</sub> emission in the United States in 2008.

**Table 1.** Major Sources of U.S. CO<sub>2</sub> Emissions in 2008 (from EPA 2010, Table 2.1)

Source/End-Use Sector	Percent of CO <sub>2</sub> Emissions in the U.S. in 2008
Fossil fuel combustion	94.1
• Electricity generation	39.9
– Coal combustion (e.g., flue gas from coal-fired power plants)	34 <sup>(a)</sup>
• Transportation	30.2
• Industrial (primarily associated with producing steam or heat for industrial processes)	13.8
• Residential	5.79
• Commercial (primarily associated with electricity consumption for lighting, heating, cooling, and operating appliances)	3.71
• U.S. territories <sup>(b)</sup>	0.72
Non-energy use of fuels (e.g., scrap tires, carbon black, and synthetic rubber carbon emissions)	2.27
Iron and steel, and metallurgical coke production	1.17
Cement production	0.69
Natural gas systems	0.51
Lime production (e.g., from lime kilns)	0.24
Incineration of waste	0.22

**Table 1. (contd)**

Source/End-Use Sector	Percent of CO <sub>2</sub> Emissions in the U.S. in 2008
Ammonia production and urea consumption	0.20
Cropland remaining cropland	0.13
Limestone and dolomite use	0.11
Aluminum production	0.08
Soda ash production and consumption	0.07
Petrochemical production	0.06
Titanium dioxide production	0.03
Carbon dioxide consumption	0.03
Ferroalloy production	0.03
Phosphoric acid production	0.02
Wetlands remaining wetlands <sup>(c)</sup>	0.02
Petroleum systems	0.01
Zinc production	0.01
Lead production	0.01
Silicon carbide production and consumption	0.00

Note that shaded rows identify the focus of this study.

(a) Lee et al. (2009).

(b) Fuel consumption by U.S. territories (i.e., American Samoa, Guam, Puerto Rico, U.S. Virgin Islands, Wake Island, and other U.S. Pacific Islands).

(c) CO<sub>2</sub> emissions from the removal of biomass and the decay of drained peat.

### 3.2 Carbon Dioxide Recovery/Capture

The first step in geologic sequestration of CO<sub>2</sub> is to separate the CO<sub>2</sub> from other exhaust gases and produce a concentrated stream of CO<sub>2</sub> that can be compressed into liquid form and readily transported to a geologic sequestration site. CO<sub>2</sub> is routinely separated and captured as a by-product from industrial processes such as synthetic ammonia production, hydrogen (H<sub>2</sub>) production, and limestone calcination, and represents the 19th largest commodity chemical in the United States, based on mass.<sup>1</sup> However, existing capture technologies are not very effective in producing concentrated CO<sub>2</sub> from large air-fired combustion sources—such as power plants—that dilute the CO<sub>2</sub> with nitrogen. Flue gas from coal-fired power plants contains 10–12% CO<sub>2</sub> by volume, while flue gas from natural gas combined cycle plants contains only 3–6% CO<sub>2</sub>.<sup>1</sup>

For effective carbon sequestration, the CO<sub>2</sub> in these exhaust gases must be separated and concentrated. There are a number of technologies available for capturing CO<sub>2</sub>. When discussed in the context of fossil fuel combustion, they are commonly classified as either pre-combustion or post-combustion systems, depending on whether the CO<sub>2</sub> is removed before or after a fuel is burned. The use of carbon capture technologies depends on the specific circumstances under which the capture system is deployed (e.g., the type of plant [pulverized coal or Integrated Gasification Combined Cycle (IGCC) power plant], the vintage and efficiency of the plant, or whether sulfur dioxide (SO<sub>2</sub>), nitrogen oxide

<sup>1</sup> <http://fossil.energy.gov/programs/sequestration/capture/>.

(NO<sub>x</sub>), and other emissions controls are in place). The most commonly used technology for low concentration CO<sub>2</sub> capture is absorption with amine-based chemical solvents (e.g., monoethanolamine [MEA]), adapted from the gas processing industry (GCEP 2005). MEA was developed over 60 years ago as a general, nonselective solvent to remove acid gases, such as CO<sub>2</sub> and hydrogen sulfide (H<sub>2</sub>S), from natural gas streams (Herzog 1999). The process was modified to incorporate inhibitors to resist solvent degradation and equipment corrosion when applied to CO<sub>2</sub> capture from flue gas. If NO<sub>x</sub>, sulfur oxides (SO<sub>x</sub>), or other reactive impurities are present, they are first removed (such as by low NO<sub>x</sub> burners and selective catalytic reduction [SCR]). Otherwise, they can react preferentially with the amines, reducing the capacity for CO<sub>2</sub>, or irreversibly poisoning the solvent. Rao and Rubin (2002) reported that when SO<sub>2</sub> or nitrogen dioxide (NO<sub>2</sub>) react with MEA, they form heat-stable salts that reduce the MEA solvent's capacity for absorbing CO<sub>2</sub>. SO<sub>2</sub> is more of a concern than NO<sub>x</sub> because in flue gas, the most common form of NO<sub>x</sub> is nitric oxide (NO), which is not reactive with MEA.

There are specific advantages and disadvantages to using any CO<sub>2</sub> capture technology (Figuerola et al. 2008). For example, post-combustion technologies are relatively high in cost because CO<sub>2</sub> in the treatment stream has been diluted by combustion air. Pre-combustion capture systems are generally aimed at waste streams that contain higher concentrations of CO<sub>2</sub>. However, precombustion technologies also have high costs. The oxy-fuel combustion capture process involves burning fossil fuels in pure oxygen rather than air, which produces almost pure CO<sub>2</sub> with some water vapor. The main problem with this technology is separating the oxygen from the air, which usually requires a lot of energy.

Scholes et al. (2009) states that certain chemical impurities such as SO<sub>x</sub> and NO<sub>x</sub> have a tendency to degrade polymeric membranes used for CO<sub>2</sub> capture if they come in contact with water. The amount of impurities found in flue gas and carbon streams need to be considered before carbon capture to ensure the membranes are capable of separating both CO<sub>2</sub> and the impurities.

While SO<sub>2</sub> can be destructive to the capture process if MEA solvents are used, it has been shown to have little effect on the geologic sequestration process because it has chemical properties very similar to those of CO<sub>2</sub> (Scholes et al. 2009). According to Nogueira and Mamora (2008), the injection of CO<sub>2</sub> that contains less than one mole % of chemical impurities would result in practically the same volume of CO<sub>2</sub> being sequestered as a stream of pure CO<sub>2</sub>. Not needing to completely purify emissions would reduce the costs of carbon capture, making carbon capture a more feasible option. Carbon capture for pure CO<sub>2</sub> streams, such as those produced from ammonia or ethanol plants, is half the price of CO<sub>2</sub> streams that requires the removal of impurities (Herzog and Drake 1996). If a slightly contaminated stream could be injected into the ground for storage with similar results as CO<sub>2</sub>, the costs of carbon capture for these streams could be potentially reduced by 50%.

### **3.2.1 Recovery of CO<sub>2</sub> from Relatively Pure Sources**

In a few instances, industrial processes emit relatively pure CO<sub>2</sub>, which can be separated and captured relatively cheaply. Such processes generally produce CO<sub>2</sub> as a commercial byproduct for use in a number of products/industries, such as fire extinguishing systems; soft drink carbonation; freezing or cooling of food products; enhanced oil recovery (EOR); and alkaline water treatment.<sup>1</sup>

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<sup>1</sup> <http://www.uiqi.com/carbondioxide.html>.



Capturing and producing CO<sub>2</sub> as a commercial product generally depends on the availability of a relatively high-volume, CO<sub>2</sub>-rich gas stream. These streams primarily come from a large-scale chemical production process or biological process.<sup>1</sup> Typical gas streams used as feedstock for commercially viable liquid CO<sub>2</sub> plants come from the manufacture of some fertilizers, natural gas processing, and cement manufacturing. Large quantities of CO<sub>2</sub> (at concentrations up to 50%<sup>2</sup>) are produced by lime kilns, which calcine the limestone to produce calcium oxide (lime), and magnesium oxide from dolomite (calcium magnesium carbonate). Other industrial activities that produce large amounts of relatively concentrated CO<sub>2</sub> are ammonia production and hydrogen production from natural gas or other hydrocarbon raw materials, corn-to-ethanol plants, and breweries.<sup>3</sup>

CO<sub>2</sub> can also be manufactured directly by burning carbonaceous fuels.<sup>4</sup> However, the concentration of CO<sub>2</sub> in stack exhaust gases from simple combustion sources (heaters, boilers, furnaces) is usually not high enough to make CO<sub>2</sub> recovery commercially feasible. The economic viability of recovery and purification of CO<sub>2</sub> is highly dependent on the source of the CO<sub>2</sub> (i.e., whether the CO<sub>2</sub> is obtained from a natural source, a fermentation source, or a chemical processing source), the specific impurities found within the raw gas stream, and the end-use of the CO<sub>2</sub>.

### **3.2.2 Separation and Capture of CO<sub>2</sub> from Dilute Exhaust Gases**

Most anthropogenic emissions of CO<sub>2</sub> come from coal-fired power stations, which can have flue-gas CO<sub>2</sub> concentrations as low as 10 to 15%.<sup>5</sup> Separating and capturing the CO<sub>2</sub> from such exhaust stack gases is complex due to the low concentration of CO<sub>2</sub> and the presence of undesirable compounds.

There are several CO<sub>2</sub> capture technologies available that can be applied either before (pre-combustion) or after (post-combustion) a fuel is burned (Folger 2010; Sass et al. 2005). The separation/capture technology (e.g., chemical absorption [amines], physical absorption, chemical absorption/flue gas recycling) and its application depend on the desired purity of the CO<sub>2</sub> and on the conditions of the source stream being treated (e.g., CO<sub>2</sub> and impurity concentrations).

Another approach, oxyfuel or oxy-combustion, burns the fuel using pure oxygen instead of air as the primary oxidant. This process produces less flue gas than air-fueled combustion and flue gas consists primarily of CO<sub>2</sub> and water vapor. Thus, this process does not require complicated CO<sub>2</sub> capture technology.

## **4.0 Impurities/Co-Contaminants in CO<sub>2</sub> Captured for Geologic Sequestration**

The composition of CO<sub>2</sub> source stream(s) destined for geologic sequestration is a key feature to be considered for the assessment of long-term performance and safety of geologic storage of CO<sub>2</sub> (Savage et al. 2004). Different combinations of CO<sub>2</sub> feedstock and capture/purification technologies result in

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<sup>1</sup> <http://www.uigi.com/co2recovery.html>.

<sup>2</sup> <http://www.co2crc.com.au/aboutccs/capture.html>.

<sup>3</sup> <http://www.uigi.com>.

<sup>4</sup> <http://www.infoplease.com/ce6/sci/A0810371.html#ixzz1HX2WeYz6>.

<sup>5</sup> <http://www.co2crc.com.au/aboutccs/capture.html>.

different sets of impurities and ranges in impurity concentrations in these source streams. Savage et al. (2004) infers that impurities such as H<sub>2</sub>S, methane (CH<sub>4</sub>), nitrogen (N<sub>2</sub>), NO<sub>x</sub>, SO<sub>2</sub> and mercaptans may be present in CO<sub>2</sub> source streams either intentionally or because it could be particularly difficult to separate them from the CO<sub>2</sub> feedstock. Savage et al. (2004) further indicates that NO<sub>x</sub> and SO<sub>2</sub> might be of particular interest because they are polluting gases that are generated by the same power plants that generate large amounts of CO<sub>2</sub> and attract emission taxes in certain countries (e.g., Italy). Their co-injection with CO<sub>2</sub>, even in small amounts, could help the economics of geologic storage.

Savage et al. (2004) indicated the presence of even small amounts of other gases may have a strong effect on the phase behavior of CO<sub>2</sub>-dominated gases; these gases must be considered in high-pressure equations of state for CO<sub>2</sub>-dominated gas mixtures to account for changes in critical pressures and temperatures. Impurities can reduce the critical temperature, which in turn, has effects on interfacial tension. In addition to changes in interfacial tension, impurities may also change the wettability of the rock, which could lead to the rock needing different sealing capacities to contain the CO<sub>2</sub> and the chemical impurities (Li et al. 2005). Impurities may also affect pore water chemistry (e.g., pH and redox conditions), depending on the impurities involved.

Sass et al. (2005) identified a large variety of potential impurities in the CO<sub>2</sub> source streams for a number of typical CO<sub>2</sub> sources (Table 2). Wang et al. (2010) indicates that oxygen (O<sub>2</sub>) (in addition to N<sub>2</sub>, SO<sub>2</sub>, and H<sub>2</sub>S) can have negative effects on transport, injection, and storage of CO<sub>2</sub>. Wang et al. (2010) found that O<sub>2</sub> and N<sub>2</sub> had the greatest effect on increasing saturation pressure of the liquid and decreasing critical temperatures, but that SO<sub>2</sub> decreases the saturation pressure and increases the critical temperature. In addition, O<sub>2</sub>, N<sub>2</sub>, and H<sub>2</sub> significantly reduce the density of supercritical CO<sub>2</sub> and the solubility of CO<sub>2</sub> in brine by reducing the partial pressure of CO<sub>2</sub>; both of these effects reduce the overall capacity of the storage reservoir.

From a geochemical perspective, Koenen and Tambach (2011) found that O<sub>2</sub>, H<sub>2</sub>S, SO<sub>2</sub>, and NO<sub>x</sub> were probably the most important impurities. Even at low concentrations, these impurities can influence the pH of the formation water, and disturb the geochemical and geomechanical properties of the reservoir rock, cap rock, and well bore material. These impurities were observed to only have significant effects if they accumulated near the injection well. It was further stressed the presence or absence of O<sub>2</sub> is of great importance, particularly in the oxidation of H<sub>2</sub>S.

DOE (2007) indicated that impurities in the captured CO<sub>2</sub> streams (e.g., argon, water [H<sub>2</sub>O], N<sub>2</sub>, and O<sub>2</sub>) may also affect the compressor and pipeline operations.

**Table 2.** Possible Trace Level Impurities in CO<sub>2</sub> Source Streams by Source Type (Excluding Air Gases and Water)

Component	Combustion	Wells/ Geothermal	Fermentation	Hydrogen or Ammonia	Phosphate Rock	Coal Gasification	Ethylene Oxide	Acid Neutralization
Aldehydes	X	X	X	X		X	X	
Amines	X			X				
Benzene (C <sub>6</sub> H <sub>6</sub> )	X	X	X	X		X	X	X
Carbon monoxide (CO)	X	X	X	X	X	X	X	X
Carbonyl sulfide (COS)		X	X	X	X	X		X
Cycloaliphatic hydrocarbons	X	X		X		X	X	
Dimethyl sulfide ((CH <sub>3</sub> ) <sub>2</sub> S)		X	X		X	X		X
Ethanol (C <sub>2</sub> H <sub>5</sub> OH)	X	X	X	X		X	X	
Ether		X	X	X		X	X	
Ethyl acetate (CH <sub>3</sub> COOCH <sub>2</sub> CH <sub>3</sub> )		X	X			X	X	
Ethyl benzene (C <sub>6</sub> H <sub>5</sub> CH <sub>2</sub> CH <sub>3</sub> )		X		X		X	X	
Ethylene oxide (C <sub>2</sub> H <sub>4</sub> O)						X	X	
Halocarbons	X					X	X	
Hydrogen cyanide (HCN)	X					X		
Hydrogen sulfide (H <sub>2</sub> S)	X	X	X	X	X	X	X	X
Ketones	X	X	X	X		X	X	
Mercaptans	X	X	X	X	X	X	X	
Mercury (Hg)	X					X		
Nitrogen oxide (NO <sub>x</sub> )	X		X	X		X	X	X
Phosphine (PH <sub>3</sub> )					X			
Radon (Rn)		X			X			X
Sulfur dioxide (SO <sub>2</sub> )	X	X	X	X	X	X		X
Toluene (C <sub>7</sub> H <sub>8</sub> )		X	X	X		X	X	
Vinyl chloride (C <sub>2</sub> H <sub>3</sub> Cl)	X					X	X	
Volatile hydrocarbons	X	X	X	X		X	X	
Xylene (C <sub>6</sub> H <sub>4</sub> C <sub>2</sub> H <sub>6</sub> )		X	X	X		X	X	

Source: Sass et. al. (2005).

Note: The source types are generic sources and there are variations in individual processes.

EPA (2011) states the composition of the injectate will be reflected in several chemical and physical parameters assigned to the CO<sub>2</sub> fluid in the model simulations. EPA (2011) further indicates that studies by Knauss et al. (2005) and Xu et al. (2007) showed the addition of H<sub>2</sub>S had little impact on geochemical reactions and mineral trapping (formation of carbonate minerals), but the addition of SO<sub>2</sub> resulted in a lower pH in the injection zone, less carbon-bearing mineral precipitation, and more formation-mineral dissolution.

A review of the major CO<sub>2</sub> sources and their specific associated impurities and relative concentrations follows. This review relies mostly on a compilation of air pollutant emission factors (AP-42) taken from the U.S. Environmental Protection Agency's (EPA) Clearinghouse for Inventories & Emissions Factors.<sup>1</sup> For the purposes of this study, it is assumed that carbon capture technology applied to the exhaust gas emissions from these facilities removes most of the N<sub>2</sub>, O<sub>2</sub>, and Ar to produce a concentrated CO<sub>2</sub> stream<sup>2</sup> with the same approximate ratio of other impurities (e.g., NO<sub>x</sub>, SO<sub>x</sub>) to CO<sub>2</sub> as the original exhaust emissions.

The focus of this report is on non-energy-related industrial sources of CO<sub>2</sub> emissions because few studies have focused on these sources. Many non-energy-related industries use fossil fuels as carbon feedstocks for the manufacture of synthetic material and chemical products, such as plastics, fibers, synthetic rubber, paints, solvents, fertilizers, lubricants, and surfactants. Following a brief overview of the impurities associated with CO<sub>2</sub> streams separated from flue gas generated by coal, oil, and natural gas power plants, the authors of this report concentrated on the top five non-energy-related industrial sources of CO<sub>2</sub>: non-energy use of fuels (e.g., scrap tires, carbon black, and synthetic rubber carbon emissions); iron, steel, and metallurgical coke production, cement production, natural gas systems, and lime production (e.g., from lime kilns).

## 4.1 Flue Gas from Coal-Fired Power Plants

Sass et al. (2005) found that flue gas derived from combustion of carbon-rich fuel (e.g., coal) may contain SO<sub>x</sub>, NO<sub>x</sub>, several different low molecular weight hydrocarbons, carbon monoxide (CO), and mercury, and that concentrations of these impurities may vary greatly.

Lee et al. (2009) estimated concentration of impurities in CO<sub>2</sub> streams separated from flue gases of different compositions produced from pulverized coal combustion power plants. Lee et al. (2009) found that flue gas from these plants could range between 10 and 3000 parts per million by volume (ppmv) SO<sub>2</sub>. Wet flue gas desulfurization (e.g., scrubbed using MEA-based absorption process) could reduce the SO<sub>2</sub> concentrations down to 135 ppmv. Heat-stable salt formation could reduce the SO<sub>2</sub> concentration to as low as 34 ppmv. Other impurities included sulfur tri-oxide (SO<sub>3</sub>), NO<sub>2</sub>, hydrogen chloride (HCl), and oxidized mercury.

Table 3 provides an estimate of the relative concentrations of impurities in a CO<sub>2</sub> stream separated from flue gas, as modified from Lee et al. (2009) and Sass et al. (2005). Although this study is focused

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<sup>1</sup> <http://www.epa.gov/ttn/chief/ap42/index.html>.

<sup>2</sup> *Impacts of Impurities on CO<sub>2</sub> Capture, Transport and Storage*. Accessed at <http://www.nrac.wvu.edu/projects/sheia/publications/CarbonSequestration/DOE/ImpactImpuritiesonCO2CaptureTransportandStorage.pdf>.

on the non-energy industrial sources of CO<sub>2</sub> for possible sequestration, information on flue gas sources is provided as a means of comparison for how carbon capture technologies might alter relative concentrations of impurities.

**Table 3.** Relative Concentrations of Flue Gas Impurities in a Separated CO<sub>2</sub> Stream (modified from Lee et al. 2009 and Sass et al. 2005)

Component	Relative Proportions in Flue Gas (%[v])	Relative Proportions in Separated CO <sub>2</sub> Stream Without Wet Flue Gas Desulfurization Scrubber (%[w]) <sup>(a)</sup>	Relative Proportions in Separated CO <sub>2</sub> Stream with Wet Flue Gas Desulfurization Scrubber (%[w]) <sup>(a)</sup>	Relative Proportions in Separated CO <sub>2</sub> Stream with Low NO <sub>x</sub> Burners, Selective Catalytic Reduction, and Wet Flue Gas Desulfurization Scrubber (%[w]) <sup>(a)</sup>	Estimated Concentrations in Separated CO <sub>2</sub> Stream, Assuming Amine Adsorption (%[v]) <sup>(b)</sup>
CO <sub>2</sub>	13.5	97.45	99.8	99.8	93.2
SO <sub>2</sub>	0.016	2.3	0.12575	0.12575	Trace
SO <sub>3</sub>	0.00325	0.0295	0.01535	0.01535	Trace
N <sub>2</sub>	74.7				0.17
NO <sub>2</sub>	0.0025	0.00585	0.0046	0.00185	
NO <sub>x</sub>	0.06				Trace
HCl	0.00525	0.0422	0.000575	0.000575	
O <sub>2</sub>	4				0.01
H <sub>2</sub> O	7.7				6.5
Hydrocarbons	Trace <sup>(b)</sup>				Trace <sup>(b)</sup>
Metals	Trace <sup>(b)</sup>				Trace <sup>(b)</sup>
Hg(2+)	Trace	0.0000142	0.00000145	0.00000145	

(a) Estimated values (except mercury) include both with and without salt formation.

(b) After Sass et al. (2005).

## 4.2 Non-energy Use of Fuels

About 5–10% of fossil fuels are used for chemical conversion processes (of non-energy use).<sup>1</sup> Non-energy use of fossil fuels for the production of chemicals and certain refinery products results in CO<sub>2</sub> emissions throughout the life cycle in the industrial production, and during product use and subsequent waste treatment. This includes the first use of fossil fuels to create products such as lubricants, paraffin waxes, bitumen/asphalt, and solvents, and secondary uses or disposal of these products after first use (i.e., the combustion of waste oils such as used lubricants). For purposes of this study, the research was focused on three main industries: scrap tire, carbon black, and synthetic rubber.

### 4.2.1 Scrap Tires

Two to three billion ( $2\text{--}3 \times 10^9$ ) scrap tires are in landfills and stockpiles across the United States, with approximately one scrap tire per person generated every year (Reisman 1997). The synthetic rubber in scrap tires consists of about 90% carbon (Freed et al. 2005). CO<sub>2</sub> emissions primarily come from

<sup>1</sup> <http://nws.chem.uu.nl/nenergy/>.

uncontrolled open burning and or controlled burning of the scrap tires as a source of fuel. Tire-derived fuels (TDF) are used in energy-intensive industries (such as cement kilns) as a source of renewable energy. Only the latter provides an opportunity for CO<sub>2</sub> capture and its impurities.

An analysis performed on the rubber portion of passenger car tires indicated they are generally made of carbon, hydrogen, ash, oxygen, sulfur, and nitrogen (EPA 1992). Emissions from burning of scrap tires include a variety of organic and inorganic compounds, many of which may pose health risks. There is a limited amount of emission data available with which to estimate emission factors (EPA 1997). However, use of TDF is similar to the use of coal. TDF has a higher heating value than coal, less moisture content, more carbon, about as much sulfur as medium-sulfur coal, and much less fuel-bound nitrogen.

Because the most likely source of CO<sub>2</sub> emissions suitable for capture and sequestration comes from using TDF as an energy source via a dilute exhaust stream. Refer to flue gas emissions (Section 4.1) for an approximation of exhaust gas concentrations.

#### **4.2.2 Carbon Black**

Carbon black is a fine black amorphous form of carbon, generally 10 to 500 nm in diameter. It is principally used as a reinforcing agent in rubber compounds such as tires, and as a black pigment in inks, surface coatings, paper, and plastics. The tire industry consumes around 80% of the total carbon black (Hisazumi 2006). About 90% of the carbon black manufactured in the United States is produced using an oil furnace process (EPA 1983). Here, an aromatic liquid hydrocarbon feedstock is heated and injected continuously into the combustion zone of a natural gas-fired furnace, where it decomposes to elemental carbon in the form of carbon black. However, typical emissions from this process include particulate matter, CO, organics, NO<sub>x</sub>, sulfur compounds, polycyclic organic matter, and trace elements. The principal source of emissions is from the main process vent, and emissions may vary considerably according to the grade of the carbon black being manufactured, and the chemical makeup of the feedstock. Typical emission factors for carbon black manufacturing, using an oil furnace process, are shown in Table 4.

Hisazumi (2006) indicated the imperfect combustion of carbon black oil (or feedstock) converts half of the hydrocarbons into carbon black while the other half goes into the tail gas. Hisazumi (2006) also indicated the typical composition of this tail gas as shown in Table 5.

Carbon capture technologies for the dilute exhaust gas from carbon black manufacturing would probably resemble those used with flue gas (from coal, oil, or gas-fired power plants) and would likely result in similar levels of impurities.

**Table 4.** Typical Emission Factors for Carbon Black Manufacturing

Component	Emissions, kg/kg
CO <sub>2</sub>	1.9 <sup>(a)</sup> to 3.75 <sup>(b)</sup>
H <sub>2</sub> S	30
SO <sub>x</sub>	25 (from flare)
NO <sub>x</sub>	0.28
CH <sub>4</sub>	25
Non-methane VOC	50

From EPA (1983), Table 6.1-3.

(a) Neelis et al. (2005).

(b) From National Grid and Atlantic Hydrogen, Inc. (2009)<sup>1</sup>.

**Table 5.** Typical Composition of Tail Gas from Carbon Black Manufacturing (from Hisazumi 2006)

Component	Emissions, vol. %
CO <sub>2</sub>	2.4–4.9 <sup>(a)</sup>
CO	10.2–11 <sup>(a)</sup>
N <sub>2</sub>	36.2
H <sub>2</sub>	8.0
CH <sub>4</sub>	0.2
H <sub>2</sub> O	43.0

(a) TRW Systems Group (1970).

### 4.2.3 Synthetic Rubber

Synthetic rubber is an artificial elastomer with the mechanical (or material) property that allows it to undergo much more elastic deformation than most materials and still return to its previous size without permanent deformation. Synthetic rubber is generally made from the polymerization of a variety of monomers including styrene and butadiene. These and other monomers can be mixed in various proportions with other impurities or additives to achieve a wide range of physical, mechanical, and chemical properties.

There are two types of polymerization reactions used to produce styrene-butadiene copolymers – the emulsion type and the solution type (EPA 1982). During these processes, the condenser tail gases (non-condensables and volatile organic compound [VOC] vapors [mostly styrene and butadiene]) are vented to the atmosphere. The estimated emission factor for VOCs from the emulsion latex process is 8.45 g/kg of copolymer produced (EPA 1982). An estimated 6 kg of CO<sub>2</sub> is released to the air for every 1 kg from plastic produced.<sup>2</sup> A similar level of CO<sub>2</sub> release is assumed for the manufacture of

<sup>1</sup> National Grid and Atlantic Hydrogen, Inc. 2009. *Hydrogen-Enriched Natural Gas, Bridge to an Ultra-Low Carbon World*. Accessed at <http://www.ffydd.org/documents/congresspapers/225.pdf>.

<sup>2</sup> <http://www.powerplantccs.com/ccs/cap/fut/c2p/plastics.html>.

synthetic rubber. The San Joaquin Valley Air Pollution Control District found only VOCs in area and point source emissions for rubber and rubber products manufacturing.<sup>1</sup> They reported zero emissions for NO<sub>x</sub>, CO, and SO<sub>x</sub>.

The carbon content of synthetic rubber is estimated at 90% for tire synthetic rubber and 85% for nontire synthetic rubber.<sup>2</sup> Approximately 2.5 lb per passenger tire and 10 lb per commercial tire are assumed to be abraded from the tires during use and considered to be 100% emitted (presumably as CO<sub>2</sub>). Other than the abraded rubber, no other emissions from the use of synthetic rubber were identified.<sup>3</sup>

Based on this study, the authors of this report have been unable to find relative proportions of impurities in a potential CO<sub>2</sub> source stream for carbon sequestration from the manufacture or use of synthetic rubber other than as a TDF (see Section 4.2.1).

### 4.3 Iron and Steel, and Metallurgical Coke Production

Iron and steel production is an energy-intensive activity that also generates process-related emissions of CO<sub>2</sub>, CH<sub>4</sub>, and other gasses. Process emissions occur at each step of the production process. Metallurgical coke is an important component of this process. Coke is used to produce iron or pig iron from raw iron ore, and is produced both onsite at “integrated” iron and steel plants and offsite at “merchant” coke plants.

#### 4.3.1 Iron and Steel Production

Steel production at an integrated iron and steel plant is accomplished using several interrelated processes. The major operations are as follows: 1) coke production; 2) sinter production; 3) iron production; 4) iron preparation; 5) steel production; 6) semi-finished product preparation; 7) finished product preparation; 8) heat and electricity supply; and 9) handling and transport of raw, intermediate, and waste materials.

EPA (2010, p. 50) indicated the vast majority of greenhouse gases (i.e., CO<sub>2</sub>) from steel production is emitted from blast furnace stove stacks where the combustion gases from the stoves are discharged. A small amount of emissions may also occur from flares, leaks in the ductwork for conveying the gas, and from blast furnace emergency venting. Emissions of CO<sub>2</sub> are also generated from the combustion of natural gas using flame suppression to reduce emissions of particulate matter. It was estimated in EPA (2010) that the relative composition of blast furnace gas contains about 60% nitrogen, 28% CO, and 12% CO<sub>2</sub>. Carbon capture technology applied to this dilute CO<sub>2</sub> exhaust stream would likely produce similar impurity estimates to those of flue gas (Section 4.1).

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<sup>1</sup> 2008 Area Source Emissions Inventory Methodology; 410 – Rubber, Fiberglass and Plastics Manufacturing, San Joaquin Valley Air Pollution Control District.

<sup>2</sup> EPA (2011). ANNEX 2, “Methodology and Data for Estimating CO<sub>2</sub> Emissions from Fossil Fuel Combustion,” p. A-93–A-94. Accessed at <http://www.epa.gov/climatechange/emissions/downloads11/US-GHG-Inventory-2011-Annex-2.pdf>.

<sup>3</sup> EPA (2011). ANNEX 2, “Methodology and Data for Estimating CO<sub>2</sub> Emissions from Fossil Fuel Combustion,” p. A-93–A-94. Accessed at <http://www.epa.gov/climatechange/emissions/downloads11/US-GHG-Inventory-2011-Annex-2.pdf>.



### 4.3.2 Metallurgical Coke Production

Metallurgical coke is used in iron and steel industry processes (primarily in blast furnaces) to reduce iron ore to iron. Metallurgical coke is produced by destructive distillation of coal in coke ovens, in an oxygen-free atmosphere (–coked–) until most volatile components are removed. Most coke plants are co-located with iron and steel production facilities, and the demand for coke generally corresponds with the production of iron and steel.<sup>1</sup> An estimate of the relative proportions of concentrated (post carbon capture) combustion stack gases from coke production is provided in Table 6 (from EPA 2008).

**Table 6.** Emission Factors and Relative Proportions of Combustion Stack Emissions from Coke Production

Component	Emissions, kg/Mg	Relative Proportion
Extractable organic matter	0.012	0.00247%
CO	0.34	0.07010%
CO <sub>2</sub> (BFG)	482	99.37313%
NO <sub>x</sub>	0.82	0.16906%
SO <sub>x</sub> (DCOG)	1.47	0.30307%
HCl (DCOG)	0.013	0.00268%
Total organic compounds	0.19	0.03917%
Methane (CH <sub>4</sub> )	0.1	0.02062%
Ethane	0.005	0.00103%
Acetone	0.0295	0.00608%
VOC	0.047	0.00969%
Benzene	0.0075	0.00155%
Toluene	0.0033	0.00068%
Chloromethane	0.0032	0.00066%
Benzoic acid	4.14E-05	0.00001%
Bis(2-ethylhexyl)phthalate	3.40E-06	0.00000%
Diethyl phtalate	9.90E-06	0.00000%
2,4-Dimethylphenol	4.17E-06	0.00000%
Phenol	2.56E-06	0.00000%

BFG = Blast furnace gas.  
DCOG = Desulfurized coke oven gas.  
VOC = Volatile organic compound.

### 4.4 Cement Production

Portland cement accounts for 95% of the hydraulic cement production in the United States (EPA 1995a). Portland cement consists of a mixture of calcium silicates, aluminates,

<sup>1</sup> EPA (U.S. Environmental Protection Agency). “Fact Sheet Final Rule to Reduce Toxic Air Emissions From Lime Manufacturing Plants.” Accessed at [www.epa.gov/ttn/atw/lime/limefs8-19-03.pdf](http://www.epa.gov/ttn/atw/lime/limefs8-19-03.pdf).

aluminoferrites. More than 30 raw materials are known to be used in the manufacture of Portland cement, the most notable of which is limestone. These materials are chemically combined through pyroprocessing and subjected to subsequent mechanical processing operations.

NO<sub>x</sub>, SO<sub>2</sub>, CO, and CO<sub>2</sub> are the primary gaseous emissions in the manufacture of Portland cement. Small quantities of VOC, ammonia (NH<sub>3</sub>), chlorine, and HCl may also be emitted. Emissions may also include products of incomplete combustion that are considered to be hazardous. Because some facilities burn waste fuels, particularly spent solvents in the kiln, these systems may also emit small quantities of additional hazardous organic pollutants. Also, raw material feeds and fuels typically contain trace amounts of heavy metals that may be emitted as a particulate or vapor (EPA 1995a).

In addition, calcium oxide (CaO) is produced from concrete plants and has been shown capable of being stored with sequestered CO<sub>2</sub> (Stolaroff et al. 2005). CaO reacts with CO<sub>2</sub> to create CaCO<sub>3</sub>, which has proven to be a stable compound that can be stored safely underground. In the presence of water, CaO also reacts relatively quickly with CO<sub>2</sub> so it is not difficult to create this compound. This is also an option for steel plants because high levels of CaO are present in steel slag.

Emission factors from Portland cement kilns and their relative proportions expected in a concentrated (post-carbon capture) CO<sub>2</sub> source stream are summarized in Table 7.

**Table 7.** Summary of Emission Factors and Relative Proportions for Portland Cement Kilns (from EPA 1995a)

Component	Max. Emissions, kg/Mg	Relative Proportion
Total organic carbon	0.09	0.00810%
CO	1.8	0.16208%
CO <sub>2</sub>	1100	99.04792%
NO <sub>x</sub>	3.7	0.33316%
SO <sub>2</sub>	4.9	0.44121%
HCl	0.073	0.00657%
Acetone	0.00019	0.00002%
Benzene	0.008	0.00072%
Toluene	0.0001	0.00001%
Chloromethane	0.00019	0.00002%
Benzoic acid	1.80E-03	0.00016%
Bis(2-ethylhexyl)phthalate	4.80E-05	0.00000%
Phenol	5.50E-05	0.00000%
Hg	1.10E-04	0.00001%

## 4.5 Natural Gas Processing

Raw natural gas is usually passed through field separators at the wellhead to remove hydrocarbon condensate and water. Natural gas contains a number of impurities, principally CO<sub>2</sub> and H<sub>2</sub>S, that must be removed before a number of separable commodities can be utilized. This is called “sweetening” the gas. The typical mole percent of CO<sub>2</sub> remaining in the processed natural gas is reportedly about

0.6 mol %.<sup>1</sup> Major emission sources in natural gas processing come from compressor engines, acid gas wastes, fugitive emissions from leaking equipment, and glycol dehydrator vent streams (if present). Most plants employ elevated smokeless flares or tail gas incinerators for complete combustion of all waste gas constituents, including virtually 100% conversion of the H<sub>2</sub>S to SO<sub>2</sub>. Thus, the major pollutant is SO<sub>2</sub>. The emission factor for SO<sub>2</sub> from gas sweetening plants is 26.98 kg/10<sup>3</sup> m<sup>3</sup> gas produced, while those for CO and NO<sub>x</sub> are negligible (EPA 1995b). Due to the high level of impurities and low levels of CO<sub>2</sub>, it is unlikely that natural gas processing plants would be targeted for carbon capture and sequestration, at least in the near future.

## 4.6 Natural Gas Combustion

Natural gas is one of the major fuels used to generate electric power, produce industrial process steam and heat, and heat commercial and residential buildings. Natural gas contains a high percentage (generally <85%) of CH<sub>4</sub> and varying amounts of ethane, propane, butane, and inerts (typically N<sub>2</sub>, CO<sub>2</sub>, and helium) (EPA 1998a). There are three major types of boilers used for natural gas combustion for utility and industrial purposes: watertube, firetube, and cast iron. Residential boilers and furnaces are generally similar to firetube boilers. The emissions from natural gas-fired boilers and furnaces include NO<sub>x</sub>, CO, CO<sub>2</sub>, CH<sub>4</sub>, N<sub>2</sub>O, VOCs, trace amounts of SO<sub>2</sub>, and particulate matter (EPA 1998a). A number of control techniques (both during and after combustion) are used to reduce these emissions (particularly NO<sub>x</sub>). For the purposes of developing emission factors, EPA organized natural gas combustion processes into three categories: large wall-fired boilers, boilers and residential furnaces, and tangential-fired boilers. Emission factors, in lb/million standard cubic feet (scf) of natural gas fired, are summarized in Table 8.

**Table 8.** Emission Factors and Relative Proportions from Natural Gas Combustion (from EPA 1998a)

Component	Maximum Emissions, lbs/10 <sup>6</sup> scf	Relative Proportion
CO <sub>2</sub>	120,000	99.7
CO	98	0.0814
N <sub>2</sub> O	2.2	0.00183
SO <sub>2</sub>	0.6	0.0005
NO <sub>x</sub>	280	0.233
CH <sub>4</sub>	2.3	0.00191
VOC	5.5	0.00457
TOC	11	0.00914
Lead	0.0005	0.00000

Taken from EPA (1998a), Table 1.4-2.

<sup>1</sup> Picard D. “Fugitive Emissions from Oil and Natural Gas Activities.” *Good Practice Guidance and Uncertainty Management in National Greenhouse Gas Inventories*. Accessed at [www.ipcc-nggip.iges.or.jp/public/gp/bgp/2\\_6\\_Fugitive\\_Emissions\\_from\\_Oil\\_and\\_Natural\\_Gas.pdf](http://www.ipcc-nggip.iges.or.jp/public/gp/bgp/2_6_Fugitive_Emissions_from_Oil_and_Natural_Gas.pdf).

## 4.7 Lime Production

Lime is produced through calcination of limestone, dolomite or other natural materials. The center of lime production is the lime kiln, and the most prevalent type of kiln is the rotary kiln, accounting for about 90% of all lime production in the United States (EPA 1998b). CO<sub>2</sub>, CO, SO<sub>2</sub>, and NO<sub>x</sub> are all produced in lime kilns. Emissions are influenced by the content of the fuel used to heat the kiln, content and mineralogic form of the feed material, quality of the lime produced, type of kiln used, and type of pollution control equipment used. An estimate of the relative proportions of these gases expected in a concentrated (post-carbon capture) CO<sub>2</sub> source stream is provided in Table 9 (from EPA 1998b).

**Table 9.** Emission Factors and Relative Proportions of Gases from Lime Manufacturing

Component	Emission Factor (kg/Mg)	Relative Percent
SO <sub>2</sub>	2.7	0.17%
SO <sub>3</sub>	0.11	0.01%
NO <sub>x</sub>	1.7	0.11%
CO	3.2	0.20%
CO <sub>2</sub>	1600	99.52%

The primary air toxics present in the exhaust gases from lime kilns are metals such as arsenic, cadmium, chromium, and nickel, and HCl.<sup>1</sup>

An example of lime kiln exhaust gas emission concentrations comes from EPA testing of a lime kiln in Alabaster, Alabama, in 1998 (EPA 2000), Table 10. Note these data would be concentrations/relative proportions prior to carbon capture.

**Table 10.** Concentrations and Relative Proportions of Gases from a Lime Kiln in Alabaster, Alabama

Component	Maximum Concentration	Minimum Concentration	Average Concentration	Units	Relative Proportion
O <sub>2</sub>			10	% by volume (assumed)	23.5379%
CO <sub>2</sub>			20	% by volume (assumed)	47.0758%
Moisture	21.1	20	20.467	% by volume	29.3447%
Total TCDD			ND	ng/dscm	
Total TCDF			ND	ng/dscm	
HCl	1.33	0.968	1.11	ppmvd	0.0261%
Ammonia (as NH <sub>4</sub> )	0.433	0.257	0.326	ppmvd	0.0077%
Aluminum, Al			ND	ppmvd	
Calcium, Ca			0.13	ppmvd	0.0031%
Magnesium, Mg			0.042	ppmvd	0.0010%
Potassium, K			0.045	ppmvd	0.0011%
Sodium, Na			0.115	ppmvd	0.0027%

<sup>1</sup> EPA. “Fact Sheet Final Rule to Reduce Toxic Air Emissions From Lime Manufacturing Plants.” Accessed at [www.epa.gov/ttn/atw/lime/limefs8-19-03.pdf](http://www.epa.gov/ttn/atw/lime/limefs8-19-03.pdf).

## 5.0 Selection of CO<sub>2</sub> Waste-Stream Compositions for Use in Experimental Studies

The co-contaminants and concentrations in CO<sub>2</sub> source streams that could be targeted for geologic sequestration are a function of both the industrial source(s) of the CO<sub>2</sub>, as well as the carbon capture technology used to extract the CO<sub>2</sub>. Previous sections addressed relative concentrations of CO<sub>2</sub> and other constituents in exhaust gases from major non-energy-related industrial sources of CO<sub>2</sub>, based on reviews of available information from both published and unpublished literature. Consistent information on stack emissions was difficult to find for most industries, so the authors of this report relied mostly on a compilation of air pollutant emission factors (AP-42) taken from EPA's Clearinghouse for Inventories & Emissions Factors.<sup>1</sup> It was also assumed that carbon capture technology applied to these industrial exhaust gases streams would remove most of the air (N<sub>2</sub>, O<sub>2</sub>, and Ar) to produce a concentrated CO<sub>2</sub> stream with the same approximate ratio of other impurities (e.g., NO<sub>x</sub>, SO<sub>x</sub>) to CO<sub>2</sub> as the original exhaust emissions. This is a similar approach to that used by the International Energy Agency's (IEA) Greenhouse Gas Research and Development (IEA GHG R&D) program.<sup>2</sup> Table 11 summarizes the relative proportions of the major impurities assumed to be present in post-carbon capture CO<sub>2</sub> source streams from major non-energy-related industries contributing to CO<sub>2</sub> emissions that could be targeted for geologic sequestration.

**Table 11.** Summary of Relative Proportions of Major Impurities in Post Carbon Capture CO<sub>2</sub> Source Streams from Major Non-energy Emitters of CO<sub>2</sub>

Component	Flue Gas with Flue Gas Desulfurization	Combustion Stack from Coke Production	Portland Cement Kilns	Natural Gas Combustion	Lime Production
CO <sub>2</sub>	97.50000%	99.40000%	99.00000%	99.70000%	99.52000%
CO		0.07010%	0.16200%	0.08140%	0.20000%
N <sub>2</sub> O				0.00183%	
NO <sub>2</sub>	0.00585%				
NO <sub>x</sub>		0.16900%	0.33300%	0.23300%	0.11000%
HCl	0.04220%	0.00268%	0.00657%		
SO <sub>2</sub>	2.30000%		0.44100%	0.00050%	0.17000%
SO <sub>3</sub>	0.02950%				0.01000%
SO <sub>x</sub>		0.30300%			
CH <sub>4</sub>		0.02060%		0.00191%	
VOC		0.00969%		0.00457%	
TOC			0.00810%	0.00914%	
Lead				0.00000%	
Mercury (Hg[2+])	0.00001%		0.00001%		

<sup>1</sup> <http://www.epa.gov/ttn/chief/ap42/index.html>.

<sup>2</sup> *Impacts of Impurities on CO<sub>2</sub> Capture, Transport and Storage*. Accessed at <http://www.nrac.wvu.edu/projects/sheia/publications/CarbonSequestration/DOE/ImpactImpuritiesonCO2CaptureTransportandStorage.pdf>.

Also reviewed were CO<sub>2</sub> sequestration source stream concentrations evaluated by other researchers; it was found that Koenen et al. (2010) had selected two possible CO<sub>2</sub> streams for investigating the effects of impurities on geologic sequestration (see Table 12).

**Table 12.** CO<sub>2</sub> Streams Selected for Investigation (by Koenen et al. 2010)

Component	Pre-Combustion	Semipurified Oxyfuel
CO <sub>2</sub>	99.64000%	98.00000%
CO	0.00300%	0.00500%
O <sub>2</sub>	0.04500%	0.70000%
N <sub>2</sub>	0.07700%	0.70000%
NO	-	0.01000%
H <sub>2</sub>	0.14000%	-
H <sub>2</sub> S	0.00014%	-
SO <sub>2</sub>	-	0.00700%
Ar	-	0.60000%

Yang et al. (2007) examined the effect of three different injection gases (pure CO<sub>2</sub>, impure CO<sub>2</sub> containing 5.19 mol % O<sub>2</sub>, and impure CO<sub>2</sub> containing 9.99 mol % O<sub>2</sub>) on miscible CO<sub>2</sub> flooding. Yang et al. (2007) found the minimum miscibility pressures for the tested oils increase unfavorably as the O<sub>2</sub> concentration in the CO<sub>2</sub> stream increased.

In selecting possible fluid concentrations for use in geochemical experiments, the authors of this report chose to include a test fluid representative of food-grade CO<sub>2</sub> as a control. Nearly pure, food-grade CO<sub>2</sub> is used for most EOR projects (Bryant and Lake 2005), and is often used as the base case for evaluation of carbon sequestration. Battelle (2002)<sup>1</sup> found commercial food-grade CO<sub>2</sub> has a minimum purity of 99.90%. Nobles and Swenson (1984) found the final product of food-grade CO<sub>2</sub> must not have more than 35 ppm CH<sub>4</sub> (preferably no more than 5–10 ppm), 10 ppm CO, 5 ppm SO<sub>2</sub>, preferably no more than 0.1 ppm H<sub>2</sub>S, 0.5 ppm carbonyl sulfide, and 1 ppm total sulfur content. The first test fluid composition was selected to equal 100% CO<sub>2</sub>, to be representative of a food-grade CO<sub>2</sub> carbon sequestration source stream (Table 12).

The IEA GHG R&D program<sup>2</sup> found the most important impurities expected in co-captured CO<sub>2</sub> were H<sub>2</sub>S and SO<sub>2</sub>, with NO<sub>x</sub> and CO also listed as other significant impurities. The authors of this report used a similar approach, assuming the incondensable gases in air (O<sub>2</sub>, N<sub>2</sub>, and Ar) are removed from the CO<sub>2</sub> waste streams, leaving SO<sub>2</sub> and NO<sub>x</sub> as the main impurities. Drawing from a summary of relative proportions of various nonenergy-related industrial sources, the authors found carbon capture from most sources (except from cement production) would produce similar concentrations, not too different from that of food-grade CO<sub>2</sub> with greater than 99% CO<sub>2</sub>. Thus, the authors selected their second test fluid composition to be representative of the cement production industry (Table 13).

<sup>1</sup> Battelle. 2002. *Purity Specifications for Commodity Uses of Carbon Dioxide in the United States*. Battelle, Columbus, Ohio.

<sup>2</sup> *Impacts of Impurities on CO<sub>2</sub> Capture, Transport and Storage*. Accessed at [www.nrac.wvu.edu/projects/sheia/publications/CarbonSequestration/DOE/ImpactImpuritiesonCO2CaptureTransportandStorage.pdf](http://www.nrac.wvu.edu/projects/sheia/publications/CarbonSequestration/DOE/ImpactImpuritiesonCO2CaptureTransportandStorage.pdf).

To provide some diversity for testing purposes, the authors of this report also selected a test fluid composition that might be representative of concentrations coming from flue gas (Test Fluid 3, Table 13), and a test concentration that might be representative of a combined CO<sub>2</sub>/H<sub>2</sub>S stream for co-sequestration (Test Fluid 4, Table 13). Although H<sub>2</sub>S concentrations as high as 21 mole % have been used for EOR (Bryant and Lake 2005), most co-capture source streams (such as those from IGCC) plants are expected to be about 3 mole % (Haines et al. 2004); thus, a similar concentration for Test Fluid 4 was selected (Table 13).

**Table 13.** Recommended Test Concentrations in Mole %

Component	Test Fluid 1 – Food Grade CO <sub>2</sub>	Test Fluid 2 – Cement Production	Test Fluid 3 – Flue Gas	Test Fluid 4 – Co-Captured CO <sub>2</sub> and H <sub>2</sub> S
CO <sub>2</sub>	100.0%	99.0%	97.5%	97.0%
NO <sub>x</sub> (NO <sub>2</sub> )		0.5%		
SO <sub>2</sub>		0.5%	2.5%	
H <sub>2</sub> S				3.0%

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