Mapping the Source Space for Carbon Capture and Utilization

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1 Key research question

Slowing down and reversing global warming represents one of the greatest challenges of our time. The average global temperature has risen by up to 0.54°F per decade in the past 40 years, and 2016 was the hottest year on record in the U.S.[1]. Out of the greenhouse gases causing global warming, CO_2 is the largest threat owing to the significant amounts that are released to the atmosphere each year. The bulk of CO_2 emissions are due to the power generation sector, which accounted for about 1,500 million metric tons (Mmt) CO_2 equivalent (CO_2e). Other significant contributors are the industrial and manufacturing sectors, e.g., oil refining (160 Mmt/yr) and chemical processing (180 Mmt/yr), with products such as cement and ammonia being particularly CO_2 intensive[2]. Reducing CO_2 emissions is therefore a multi-pronged effort, with key thrusts being the transition of the power generation sector towards renewable sources, and decarbonizing industry.

Progress in renewable generation has been remarkable, with wind power making up a significant portion of the power generation portfolio in several U.S. markets, and solar photovoltaic generation also growing steadily. Nevertheless, renewable sources are inherently variable and power generation rates fluctuate during the day and in between seasons. Grid operators need to balance this fluctuating electricity supply and fluctuating demand at all times. The current fleet of conventional (fossil-fueled) power plants is used to meet peak electricity demand, and is a key asset in compensating for fluctuations and gaps in renewable generation, at least until grid-level electricity storage becomes a practical and economic option. Thus, it is to be expected that fossil fuel-based power generation will be phased out gradually, and carbon capture and sequestration (CCS) will play an essential role in supporting a transition towards sustainable energy.

The decarbonization of the manufacturing sector, however, lags behind. Industrial CO_2 emissions have two sources: endogenous (where the CO_2 is emitted by materials being processed, as is the case, e.g, with a lime kiln that produces quicklime used for making cement) and exogenous (where CO_2 is generated to support processing, such as burning fuel to run a high temperature furnace). Reducing endogenous CO_2 emissions requires new processing routes and technologies, while exogenous emissions can be cut by improving heat use to lower the energy intensity of manufacturing, and by replacing fossil fuels with renewable electricity as the source of process heat and motive power. These are also long-term efforts and, as in the case of power generation, CCS is expected to have in important role while new technologies are implemented and old ones are phased out.

The above reveal that carbon capture (CC) technologies are expected to meet the decarbonization goals of a diverse group of CO_2 sources. Intuitively, the flue gas streams of these sources have different characteristics (flow rate, CO_2 concentration). In order to be economically efficient, a CCS plant should be optimized to match a specific emitter, and no single technology or CC plant configuration can meet the needs of all emitters. Nevertheless, the exact characteristics of the flue gas streams of the major candidates for decarbonization by CCS are not described to the needed level of detail in the literature.

In light of the above, the **key research question that we will pursue is to develop a compre-**

hensive mapping of the types of CO_2 emitters, and their respective flue gas characteristics. The CO_2 sources will be classified by industry, with mean values and upper and lower bounds for important flue gas parameters (flow rate, composition including CO_2 concentration) will be collected in a freely available database and reported in a journal article.

2 Anticipated contributions to the existing literature

Absorbtion in amine-based solvents is currently the most widely studied and used technology for capturing CO_2 from point sources of flue gas (such as the powerplants and manufacturing facilities mentioned earlier). Equivalent processes using ionic liquids as the solvent are being investigated. A schematic of such a solvent-based absorption CC process is shown in Fig. 1. The process is a chemical plant that comprises two main units, the absorber and the stripper. In the absorber, a gas stream (e.g., flue gas from a power plant) containing CO_2 comes into contact with the solvent, which retains the CO_2 via chemical and physical absorption. The treated gas leaving at the top of the absorber can be released to the atmosphere, but may still contain a small fraction of CO_2 (typically, the process is designed to remove 90% of the CO_2 present in the feed stream). The solvent loaded with CO_2 (the rich solvent) is heated in the stripper, releasing CO_2. The CO_2 stream at the top of the stripper is compressed for sequestration in geological formations or for other uses (e.g., enhanced oil recovery, beverage industry). After releasing the CO_2, the lean solvent is cooled by exchanging heat with the rich solvent in a heat exchanger, and returned to the absorber, whence the cycle repeats. Other CO_2 separation technologies are based on physical or chemical adsorption, membrane separation, and cryogenic treatment to condense CO_2 [3].

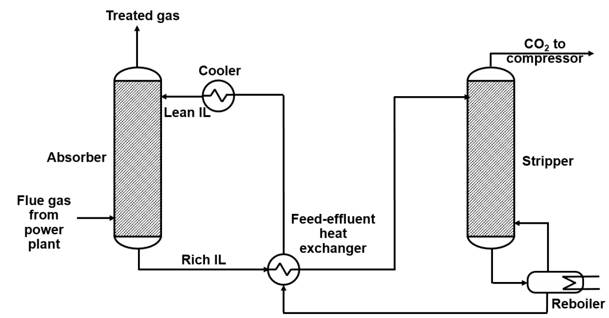

Figure 1: Schematic of solvent-based absorption carbon capture process (ionic liquid – IL – solvent used as an example). Figure is reproduced from Seo et al.[8]

Carbon capture is thus a separation process aiming to remove/retain CO_2 from a gas mixture. At the fundamental level, the choice of separation technology and the design of a separation process (the choice of equipment size and operating parameters such as temperatures, pressures, flow rates and compositions, all off which significantly influence cost) for a given mixture are driven by three key factors:

1. the *composition* of the mixture (for CC, the key factor is the concentration of CO_2 in the stream that needs to be processed, but the presence of water, nitrogen oxides, sulfur, etc. can considerably impact the choice of separation technology)
2. the *flow rate* of the stream to be processed
3. the desired *purity of the products* (in the case of CC, this refers to the percent of CO_2 that must be captured from the feed stream before it is released to the atmosphere. As mentioned above, a typical target is 90% removal, but more stringent environmental mandates may impose higher target values).

This point is illustrated in Fig. 2, which presents the results of a sensitivity study carried out in the PI's group[8] that considered the impact of some these factors on the key equipment design and operating parameters of a solvent-based absorption process as shown in Fig. 1. Fig. 2 a) shows that the height of the absorber increases significantly (by about 50%) as the concentration of the CO_2 in the flue gas decreases. This can be explained by considering the fact that separating a component from a *dilute* mixture (i.e., containing less CO_2) is more difficult than removing it from a concentrated one. Fig. 2 d) shows that lower stripper pressures are optimal when the CO_2 concentration is low, as lower pressures facilitate the release of CO_2 from the solvent during the solvent regeneration step. However, lower stripper operating pressures eventually lead to higher compression costs, as the compressor must "work harder" to increase the pressure of the CO_2 stream for sequestration. Finally, Figs. 2 b) and c) are connected. Fig. 2 b) shows the results of a sensitivity study regarding the enthalpy of chemical absorption for the solvent. Higher absolute values (lower negative numbers) of this parameter indicate that the solvent binds the CO_2 molecules more strongly, which is necessary when the concentration of the CO_2 in the feed gas is lower. Conversely, releasing the CO_2 from the solvent will require more energy, which is reflected in the higher reboiler temperatures (the reboiler is the bottom part of the stripper column) shown in Fig. 2 d).

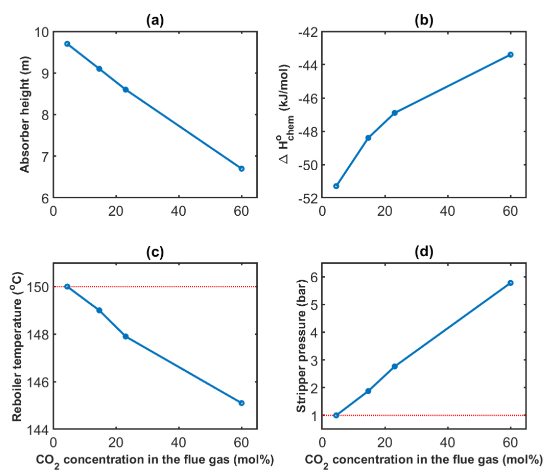

Figure 2: Sensitivity of CC equipment size and operating parameters to the CO_2 content of the flue gas stream. A solvent-based absorption CC process using ionic liquid solvent is considered. Reproduced from Seo et al.[8]

The sensitivity study considered a range of compositions for the flue gas stream to be treated that was based on a few well-known power plant benchmarks: natural gas combined cycle plants, and air- and oxygen- combustion coal-fired power plants[5,6], with CO_2 compositions ranging from 4.5 to 23 mol% (for the sake of completeness, a case with 60 mol% concentration was considered based on the results of a bench scale oxy-combustion study reported by Vega et al.[10]).

Evidently, the design space for CC plants is quite vast; the use cases concerning power plants are relatively well documented in the literature. However, **the detailed parameters of the flue gas streams released by industrial / manufacturing facilities are largely unknown**. A literature review carried out by the author, along with a minimum number of informal inquiries, resulted in the preliminary data shown in Fig. 3.

The **main contribution of this work to the literature** will be to **fill the blank areas of the map** in Fig. 3. The **expected benefits** of this work will be twofold. First, it will provide *strategic guidance*, informing researchers and policy makers on which industries should be targeted and prioritized for CCS, technology development and replacement, etc. Second, it will serve as a *tactical* guide for CCS technology selection and process development for both academic researchers and industry developers. The paper is expected to **complement** existing works (e.g., Waxman et al.[11]) that provide a *global* perspective on the evolution of CO_2 emissions. The paper will also complement the comprehensive survey of Bains et al.[4], which reviews of *process-level* CO_2 emission characteristics based on information reported by emitters to the EPA Greenhouse Gas Reporting Program[1] (rather than industry surveys) and does not quantify the concentration of other components of the flue gas (water vapor, nitrogen oxides, sulfur, etc.). The latter have a significant

[1] `https://www.epa.gov/ghgreporting`

influence on the choice of CC technology and the design of the CC process, and the proposed work will **provide the information granularity** needed to address these decisions. We note here that the aforementioned EPA database largely fails to provide this graular information.

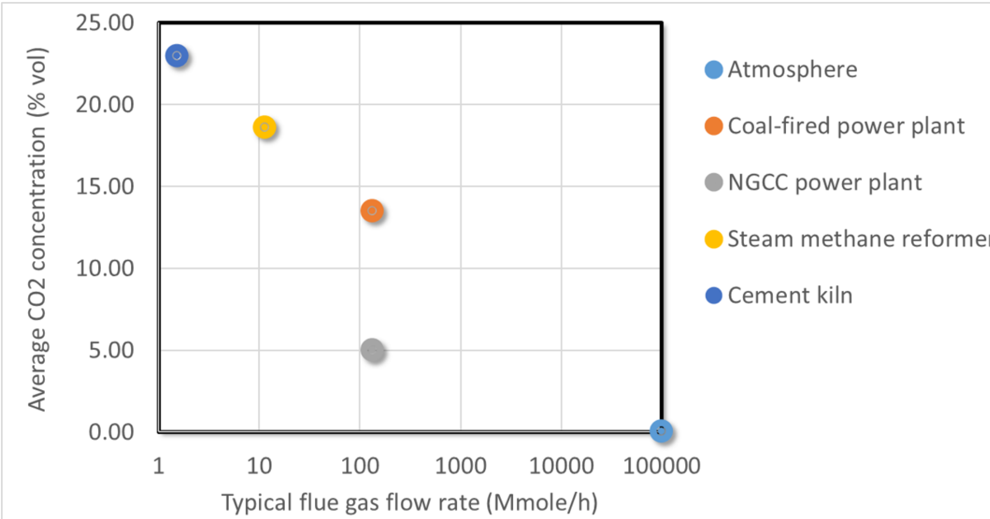

Figure 3: An incipient mapping of the flue gas characteristics of industrial CO_2 emitters. Data sources: power plants: James et al.[6], cement: Worrell et al.[12], Olsen et al.[7], steam methane reforming: personal communication with subject matter expert.

3 Research design and approach

The project will be centered around an industry survey focusing on the chemical and manufacturing industries. Examples of industries to be targeted include:

1. Cement
2. Glass
3. Ethanol production
4. Ethylene production
5. Hydrogen production
6. Crude oil processing
7. Coal and biomass gasification
8. Electronics manufacturing
9. Oil and gas extraction

Specifically, we will seek a precise and comprehensive characterization of industrial CO_2 emitters, including detailed flue gas composition and flow rate. In cases where multiple technologies are available for making the same product, a break-down will be estimated. These results will be universal (since by and large the same technologies are used around the world to make a specific product). Estimates of the *total* emissions of a specific industry in the U.S. will also be computed. Further information will be obtained from the EPA Greenhouse Gas Reporting Program in order to quantify uncertainty and variability in the survey data, and complete missing information if necessary (and possible). Table 1 shows a sample data entry.

Table 1: Sample table for collecting survey results. Confidence intervals (c.i.) and missing information to be determined from survey data

Industry	Technology	Flue gas flow rate Mmole/h	Flue gas CO_2 content mol %	Other flue gas components	Further info
Hydrogen	Steam reforming	11.33 $\pm$c.i.	18.60$\pm$c.i.	N_2, O_2, NO_x	

4 Data collection plan

Industry researchers and decision-makers will be contacted and interviewed. The PI has extensive experience carrying out such surveys, collecting and analyzing the data, and publishing the results. An example recent work is the paper by Tsay, Pattison, Piana and Baldea[9], which reports the results of a survey comprising 110 interviews with industry experts, concentrating on process design practices and capabilities in diverse chemical industries. The PI also has extensive industry contacts (having worked in industry for five years prior to joining UT Austin) and will leverage the contacts and membership of the two industry-academia consortia where he is deeply engaged: the Texas Wisconsin California Control Consortium (involving UT, University of California Santa Barbara and University of Wisconsin - Madison along with about 12 companies), and the Process Science and Technology Center at UT (involving about 30 industry sponsors).

The PI will involve the graduate research assistants in his group, as well as undergraduate researchers in the interview process. All interviews will be conducted by the PIs, with the students in attendance. Students will help with taking notes, organizing the responses and drafting the proposed manuscript. The **target** for publishing the manuscript is the AIChE Journal (`https://aiche.onlinelibrary.wiley.com/journal/15475905`), the flagship journal of the American Institute of Chemical Engineers (AIChE), where the PI has published extensively.

Conflict of interest statement

The author declares that he has no conflict of interest impacting the proposed work.

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