IX.10 Life-Cycle Analysis of Air Pollutants Emission for Refinery and Hydrogen Production from SMR

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Project Start Date: October 2016
Project End Date: Project continuation and direction determined annually by DOE

Overall Objectives

- Develop criteria air pollutants emission factors (EF) (in g/MJ) for refinery products to serve as a baseline for comparison with alternative transportation fuels.
- Develop criteria air pollutants emission factors (in g/MJ) for hydrogen production via steam methane reforming (SMR) process.
- Assess the life cycle analysis emissions impact of hydrogen fuel cell vehicles (FCVs), relative to baseline petroleum fuels usage in internal combustion engine (ICE) vehicles.
- Develop emission factors associated with combustion of refinery fuels (e.g., refinery still gas, catalyst petroleum coke).
- Investigate refinery air pollutants emissions variations among regions.
- Incorporate updated criteria air pollutants emission factors in Greenhouse gases, Regulated Emissions, and Energy use in Transportation Model (GREET®) model.

Fiscal Year (FY) 2017 Objectives

- Collect and match U.S. refinery air pollutants emissions data with refinery operation data for individual refineries at facility and sub-facility (unit) levels.
- Develop methodologies to allocate individual refinery air pollutants emissions to individual refinery products (e.g., gasoline, diesel, etc.), and aggregate the results to Petroleum Administration for Defense Districts and national levels.
- Match U.S. standalone SMR air pollutants emissions data with hydrogen production data to derive emission factors, and aggregate the results to national level.
- Conduct life cycle analysis of criteria air pollutants emissions associated with petroleum fuels use in ICE vehicles and hydrogen use in FCVs.

Technical Barriers

This project addresses the following technical barriers from the Systems Analysis section of the Fuel Cell Technologies Office Multi-Year Research, Development, and Demonstration Plan.

(C) Inconsistent Data, Assumptions and Guidelines

(D) Insufficient Suite of Models and Tools

Contribution to Achievement of DOE's Systems Analysis Milestones

This project contributes to achievement of the following DOE milestones from the Systems Analysis section of the Fuel Cell Technologies Office Multi-Year Research, Development, and Demonstration Plan.

- Milestone 2.2: Annual model update and validation. (4Q, 2011 through 4Q, 2020)
- Milestone 3.1: Annual update of Analysis Portfolio. (4Q, 2011 through 4Q, 2020)

FY 2017 Accomplishments

- Collected refinery pollutants emissions data at various levels (unit level, facility level, and national level) and calculated emission factors.
- Conducted life cycle emissions analyses for petroleum fuels (gasoline, diesel, liquefied petroleum gas [LPG]) for updating the default values in GREET. Compared to previous GREET emissions estimates, the updated results show significant reduction in SOx (-20% to -42%), moderate reduction in NOx (-3% to -6%), and minor change (-2% to +3%) in other pollutant emissions (volatile organic compound [VOC], CO, particulate matters with emissions less than 10 µm [PM10] and less than 2.5 µm [PM2.5]).

- Conducted life cycle emissions analyses for hydrogen production via central SMR. Compared to previous emissions estimates in GREET 2016, the updated results are lower for most criteria pollutants emissions (-24% to -60%), with the exception of SOx which had +3% increase.
- Use of SMR hydrogen in FCVs can significantly reduce most criteria pollutant emissions when compared to gasoline ICE vehicles (Figure 1).

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INTRODUCTION

Under the Clean Air Act, criteria pollutants, nitrogen dioxide (NO_2), ozone (O_3), sulfur dioxide (SO_2), particulate matter (PM), carbon monoxide (CO), and lead (Pb) are regulated by the Environmental Protection Agency for protecting public health and welfare. In addition to these pollutants, emissions reduction of VOCs is also of interest as they react with NOx to form ozone under sunlight. Among the emission sources, transportation section is a major contributor, responsible for over 50% of NOx, over 30% of VOCs and over 20% of PM emissions of the total emissions inventory in the U.S. This stimulates efforts from both federal and local governments to promote low or zero emission vehicles to reduce air pollutions. The present study focuses on evaluating air pollutants, VOC, CO, nitrogen oxides (NOx), sulfur oxides (SOx), PM10, and PM2.5, associated with the production transportation fuels, to update the GREET model. This study evaluates pollutants emissions from a major industrial sector (petroleum refining and hydrogen production via SMR) using up-to-date emissions inventory data. Allocation methods are used to estimate emissions associated with each refinery product using refinery operation optimization linear programming model for individual refineries. This is particularly important since petroleum fuels (such as gasoline, diesel, and jet) serve as baseline fuels, against which the environmental impacts of alternative transportation fuels and vehicle technologies are compared.

APPROACH

The refinery and SMR facility emission information are collected from National Emission Inventory (NEI) database [1]. The NEI database is updated every three years. Thus, the most recent available datasets are from years 2011 and 2014, both of which are used in the present study. The refinery capacity information is obtained from Energy Information Administration RefCap database [2,3], while hydrogen capacity information is obtained from Pacific Northwest National Laboratory report [4]. The actual production of refinery facilities is estimated by applying Energy Information Administration reported Petroleum Administration for Defense Districts level utilization rate to RefCap capacity [5]. The hydrogen plant production is calculated by multiplying plant capacity with an assumed 80% utilization rate, as provided by industrial partners. Only standalone SMR facilities were investigated, while captive hydrogen production within refineries was excluded since SMR facilities within refineries often do not have clear boundaries in terms of material and energy flows, thus complicating the emissions allocation between refinery products and hydrogen. While the process of calculating emission factors (EFs) (in g/mmbtu hydrogen) for standalone SMRs is straightforward, the allocation of refinery facility emission to various refinery products requires detailed

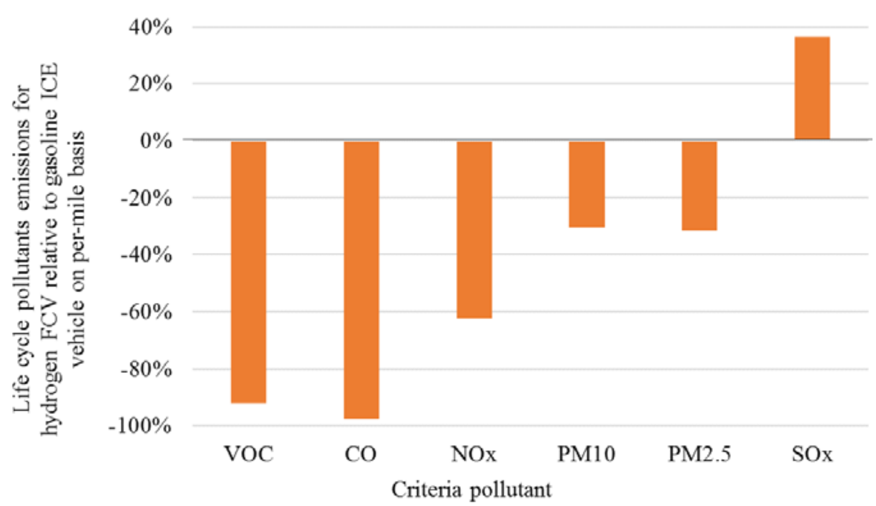

FIGURE 1. Life cycle pollutants emissions of hydrogen FCV relative to spark ignition gasoline vehicle

analysis of refinery energy and material flows at the process unit level, which is different for each refinery.

The allocation of refinery facility emissions to refinery products was guided by refinery operation simulations using a linear programming model. Each refinery has specific configuration, crude quality and products portfolio, and thus requires unique simulation of each refinery operation. Argonne developed a database of material and energy flows for a large number of U.S. refineries, representing 70% of total U.S. refining capacity [6]. Guided by the material and energy flow information in the database, the facility emissions are allocated to intermediate products from each process unit. The energy and emissions burden allocated to intermediate products are carried to the subsequent process units until the facility energy and emissions burdens are cascaded into the final refinery products [6]. The present study includes 14 refineries with 2011 emissions data and 21 refineries with 2014 emissions data. The number of these facilities were constrained by the proper matching of NEI emissions database and the Argonne refinery database.

RESULTS

Emission Factors for Central SMR Hydrogen Plant

The national weighted average pollutant emissions factors for SMR facilities are shown in Figure 2. The dataset includes 36 facilities from 2014 NEI database; the number of considered facilities were constrained by availability of standalone facility capacity/production data, which are often business confidential.

Comparing to previous estimates in GREET 2016, the updated results show much lower pollutants emission factors, except for SOx. The first and third quartiles of the emissions factors from various SMR facilities are displayed in Figure 2, indicating a large variation of emissions among these facilities.

Emission Factors for Refinery Products

Guided by the energy and material flows from individual refinery linear programming simulations, the refinery facility criteria pollutants emissions are allocated to refinery products, resulting in EFs per refinery products. Two sets of EFs, derived from emissions and productions in 2011 [1,2] and 2014 [1,3], respectively, were developed. The EFs from 2014 refinery emissions data are plotted in Figure 3 (the EFs from 2011 data were omitted for legibility of figure).

Figure 3 shows that among the many refinery products, gasoline and LPG carry higher emission burdens relative to the other refinery products, such as diesel and jet. This is consistent with the greenhouse gas emission and energy intensity trends for these products [6]. Gasoline and LPG fuels are produced via more energy intensive process units than diesel and jet. For each pollutant, the wide spread between first and third quartiles in Figure 3 indicate the large variations in criteria pollutants emissions among refineries. This is not surprising given the unique configuration and operation of each individual refinery. Table 1 shows the EFs of refinery products and hydrogen using 2011 and 2014 emissions data, and those from previous GREET 2016 estimates.

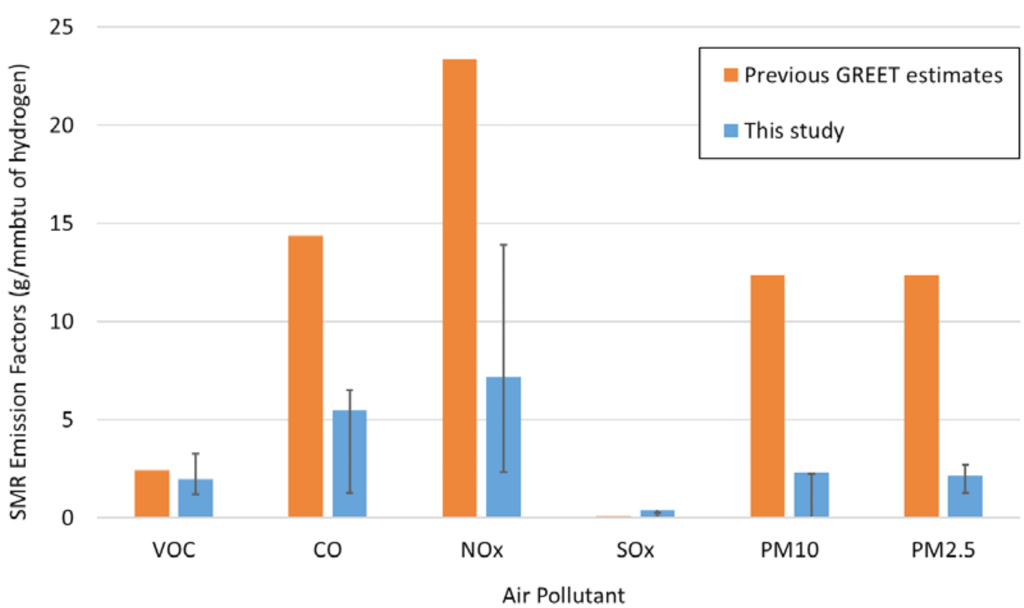

FIGURE 2. National average criteria pollutant emissions factors for SMR facilities

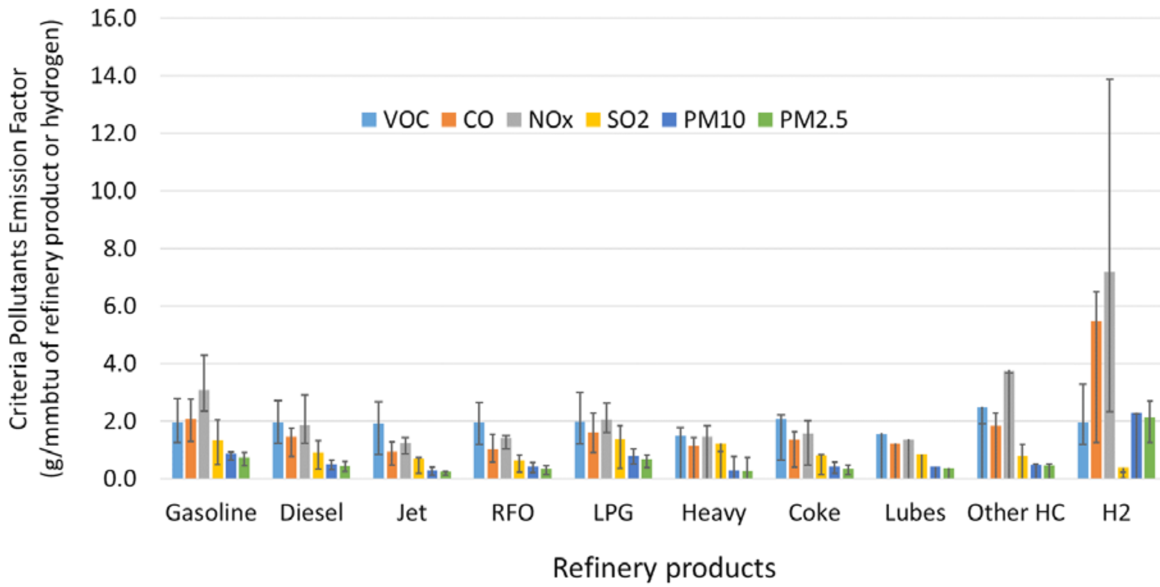

FIGURE 3. Refinery product-specific emission factors derived from NEI 2014 refinery emissions (error bars for each pollutant represent first and third quartiles)

TABLE 1. U.S. Refinery Criteria Pollutants Emission Factors for Refinery Products and Hydrogen (g/mmbtu)

	Data Source	Gasoline	Diesel	Jet	RFO	LPG	Heavy	Coke	Lubes	Other HC	H_2
VOC	GREET 2016	2.55	1.95	0.96	1.08	1.99	--	1.59	--	--	2.44
	2011	2.10	2.09	1.96	1.76	2.13	1.68	2.20	1.26	2.74	2.67
	2014	1.98	1.96	1.92	1.97	1.98	1.48	2.07	1.56	2.48	1.96
CO	GREET 2016	3.75	2.40	1.20	1.56	2.94	--	2.16	--	--	14.37
	2011	1.76	1.14	0.91	0.85	1.36	1.74	1.11	0.77	1.62	9.02
	2014	2.07	1.47	0.93	1.04	1.63	1.13	1.36	1.23	1.83	5.48
NOx	GREET 2016	7.47	4.24	1.99	3.18	6.73	--	3.57	--	--	23.34
	2011	2.76	1.65	0.97	1.20	1.85	1.38	1.41	0.99	3.21	7.97
	2014	3.08	1.86	1.26	1.42	2.04	1.47	1.58	1.35	3.74	7.18
SO2	GREET 2016	16.68	8.21	3.15	7.46	18.85	--	5.19	--	--	0.06
	2011	1.35	0.93	0.56	0.62	0.95	0.87	0.68	0.54	1.35	0.42
	2014	1.34	0.92	0.70	0.63	1.37	1.23	0.81	0.85	0.79	0.39
PM10	GREET 2016	0.70	0.48	0.24	0.29	0.54	--	0.42	--	--	12.38
	2011	1.10	0.65	0.41	0.47	0.94	0.81	0.53	0.46	0.71	2.85
	2014	0.87	0.50	0.30	0.42	0.79	0.29	0.42	0.41	0.48	2.30
PM2.5	GREET 2016	0.54	0.35	0.18	0.22	0.41	--	0.32	--	--	12.38
	2011	0.95	0.58	0.38	0.41	0.82	0.68	0.46	0.43	0.69	2.62
	2014	0.75	0.43	0.24	0.36	0.68	0.26	0.36	0.38	0.46	2.14

RFO – residual fuel oil; HC - hydrocarbon

Table 1 shows that the EFs derived from 2011 and 2014 refinery emission datasets are consistent. For most pollutants, the EFs from the present study are less than previous GREET 2016 estimates. In particular, the current emission factors for SOx are significantly lower compared to previous GREET 2016 estimates across all refinery products.

Comparison between Life Cycle Emissions of Refinery Fuels and Hydrogen on a Per-Mile Basis

A life cycle analysis was conducted for SMR-hydrogen use in FCV, gasoline use in spark ignition vehicle, diesel use in compression ignition direct injection vehicle-diesel, and spark ignition LPG vehicle. Fuel economy estimates for these four vehicle technologies are shown in Table 2.

TABLE 2. Fuel Economy of Various Fuel–Vehicle Technologies

	Fuel economy (mile/gge)
Gasoline ICE	26
Diesel ICE	31
LPG ICE	26
Hydrogen FCV	55

With the fuel economy estimates in Table 2, the life cycle criteria pollutants emissions can be estimated on per-mile. The comparison of life cycle criteria pollutants emissions for various fuel–vehicle technologies is provided in Table 3, which shows that hydrogen FCV has significantly lower per-mile WTW emissions for most pollutants except for SOx. The FCV does not have tailpipe emissions. The WTW SOx emissions is attributed to the emissions associated with electricity generation for hydrogen compression, which is required for both hydrogen delivery and refueling into FCVs. As the future grid electricity generation mix is projected to have reduced share of coal-based generation, the WTW SOx emissions for FCVs is expected to be proportionally reduced.

TABLE 3. Life Cycle Emissions for Various Fuel–Vehicle Technologies on Per-Energy Basis and Per-Mile Basis (Using 2014 Emissions Dataset)

(g/mmbtu basis)	VOC	CO	NOx	SOx	PM10	PM2.5
Gasoline ICE	84.7	645	65.3	21.3	8.89	4.58
Diesel ICE	42.3	796	67.0	14.4	8.58	4.42
LPG ICE	59.2	645	56.4	21.5	6.82	3.36
Hydrogen FCV	13.7	33.6	51.8	61.0	13.2	6.35
(g/mile basis)						
Gasoline ICE	0.364	2.78	0.281	0.091	0.038	0.020
Diesel ICE	0.152	2.86	0.240	0.051	0.031	0.016
LPG ICE	0.255	2.77	0.243	0.092	0.029	0.014
Hydrogen FCV	0.028	0.069	0.106	0.125	0.027	0.013

CONCLUSIONS AND UPCOMING ACTIVITIES

The national criteria pollutants emissions from refinery and SMR facilities have been investigated to derive emission factors for the refining of petroleum fuels and SMR-hydrogen. A methodology is developed to allocate refinery facility emissions to individual refinery products. Relative to previous GREET 2016 model estimates, the results from current study demonstrate significant reduction in most pollutants for these fuels. The future work will update the GREET 2017 model with emission factors from this analysis.

FY 2017 PUBLICATIONS/PRESENTATIONS

1. Pingping Sun, Amgad Elgowainy, Zifeng Lu, Jeongwoo Han, Michael Wang, Troy Hawkins, Ben Morelli, and Ben Young, "A Life Cycle Analysis of Refinery Fuel Products Air Emissions Using Refinery Emissions Inventory Data," International Emissions Inventory Conference, Baltimore, MD, August 14–18, 2017.

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