Determining the amount of “green” coke generated when co-processing lipids commercially by fluid catalytic cracking (FCC)

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Abbreviations: FCC, fluid catalytic cracker; IEA, International Energy Agency; LCFS, low carbon fuel standard; UCO, used cooking oil; HTL, hydrothermal liquefaction; ACE, advanced cracking evaluation; VIF, variance inflation factor; VGO, vacuum gas oil.

Highlights

- Year-long commercial FCC co-processing data was used to evaluate green coke production thus providing a potential method for carbon tax rebate
- The biogenic feedstock generated less coke compared with the fossil feed
- A regression approach, incorporating a bootstrap method, was able to quantify the green coke (thus track the “green molecules”)

Abstract

Co-processing biogenic feedstocks in oil refineries will reduce the greenhouse gas emissions normally associated with fossil derived transportation fuels. The fluid catalytic cracker (FCC) within a refinery is a robust processing unit and will likely be a preferred insertion point if biocrudes, produced by the liquefaction of biomass, are co-processed within a refinery. Fluid catalytic cracking results in a wide range of intermediate products which can be upgraded to gasoline, diesel, heavy fuel oil and liquified petroleum gas fractions. Coke is also produced and provides heating for feedstocks, the endothermic catalytic cracking reactions and the regeneration of the FCC catalyst. However, coke combustion also generates carbon dioxide and is a significant source of refinery greenhouse gas emissions.

The hourly data from one year of commercial operation was assessed using linear and Bayesian ridge regression to quantify the burning coefficient of the coke when co-processing lipids at the FCC. When a bootstrap method was used to reduce the uncertainties of the coefficients, this allowed us to quantify the renewable (green) fraction of the coke component, indicating the reduction in carbon dioxide emissions when commercially co-processing biogenic feedstocks.

Keywords

FCC; co-processing lipids; decarbonising coke combustion; bootstrap; regression analyses

1.Introduction

As indicated by organizations such as the International Energy Agency (IEA), to achieve a "net-zero" future will require the decarbonization of every facet of energy production and consumption [1]. The increased use of electric cars, renewable fuels and emerging technologies such as "green" hydrogen will reduce the emissions resulting from burning fossil fuels, which is a major component of petroleum fuel emissions (~70% of the emissions) [2]. However, in the short-to-mid-term, enabling policies will be needed to incentivize the transition to lower carbon intensive fuels. These include pricing fossil carbon [3], in the form of a carbon tax as has been adopted by several jurisdictions around the world, as well as other policies such as the low carbon fuel standard (LCFS) which is used to reduce the carbon emissions of the fuel production and transportation sectors [4–7]. These types of policies have incentivized fuel suppliers to lower the carbon intensity of their products. They have also acted as a catalyst for some companies to develop new business opportunities such as establishing standalone renewable diesel plants (e.g., Neste, Diamond Green, REG) or revamping existing oil refineries to produce renewable diesel/bio jet (e.g., World Energy, Eni, Marathon) [8–10].

Although the development of standalone facilities has typically involved the adaption of the desulfurization processes that are widely used in the petroleum refining [11,12], another way of reducing the carbon intensity of the fuels produced by a refinery is to co-process biogenic, low-carbon intensity feedstocks with the fossil fuel [13–15].

Recent work has shown that co-processing biogenic feedstocks can result in the production of lower carbon intensity fuels with this approach already carried out at a commercial scale [16]. Currently, oleochemical feedstocks such as used cooking oil (UCO) or tallow, can be co-processed at either the hydrotreater (e.g., BP, Preem, Kern Oil) or the fluid catalytic cracker units (e.g., Parkland) [10,16]. However, as oleochemical feedstocks are expected to have limited availability and higher costs as compared to potential “biocrude” feedstocks, various processes are currently under development that involve the gasification or liquefaction of biomass [17,18]. The hope is that these biocrudes will be more plentiful and cheaper than oleochemical feedstocks [17,19]. As these pyrolysis/hydrothermal liquefaction (HTL) processes produce a liquid intermediate that is more challenging and variable than oleochemical feedstocks, it is very likely that biocrude co-processing will take place at the more robust FCC rather than at the hydrotreater [16,18,20]. This is partly due to the FCC’s reactor design which involves a circulating fluidized bed where the catalyst can be regenerated in situ [21–23]. It is also should be noted that the FCC is designed to run in a heat balance mode where the heat demand for the process (heating the feedstocks and providing the heat for reaction) is supplied by burning the coke generated during the process. The heat is also used for miscellaneous heat sinks such as steam stripping, catalyst cooling and heat losses [24,25].

Past work has shown that simulating an industrial set-up at a lab/pilot-scale is challenging, primarily due to reactor configuration. Similarly, although lab scale fixed bed and fixed fluidized bed reactors (Advanced cracking evaluation - ACE) are available and simple to operate, they have been shown to provide less accurate results compared to commercial scale circulating fluidized bed reactors [22,23,26]. This difference is particularly apparent by the higher coke yield obtained during advanced cracking evaluation, primarily due to the long residence (reaction) times that are used compared to commercial-scale units [26].

Past work has also shown that the greenhouse gas emissions derived from burning FCC coke can represent 20-35% of the total refinery emissions, even though the coke yield is only around 5 wt% [24,27]. The coke burn value is typically obtained via the flue gas and air blower with an oxygen mass balance performed to derive the mass of carbon, hydrogen and the coke burn value. Although not fully understood, it is likely that the coke originates from multiple sources such as catalytic coke (conversion, catalyst type, reaction time), contaminant coke (metals or impurities in the feedstocks), feed residue coke (heavy fraction of the feed) and catalyst circulation coke (catalyst stripping efficiency and pore size of the catalyst) [24].

Most previous work has observed that the coke yield increased when co-processing biogenic feedstocks [28–30]. This was probably due to the reactor configuration used during lab-scale experiments. In contrast, pilot-scale co-processing work Indicated an opposite trend with the coke yield decreasing when biogenic feedstocks were processed [23,31].

As reported here, when oleochemical feedstocks were co-processed at a commercial scale, the amount of “green” coke generated during FCC co-processing could be determined by collecting hourly data and applying regression models (linear and Bayesian ridge regression) and a bootstrap method to reduce the uncertainty of the coefficients used to quantify coke burning. This method was successfully used to track the “green molecules” derived by co-processing lipid feedstocks and quantify the coke derived from fossil and biogenic sources.

2.Materials and Methods

2.1 Data resource

The Parkland Burnaby refinery (British Columbia, Canada) routinely carries out commercial-scale co-processing of oleochemical feedstocks at their FCC unit [16]. Routine, commercial data from the FCC unit was retrieved using TIBCO Spotfire® which was connected to the refinery database [32]. Several tags were selected based on process knowledge that related to coke generation during FCC operation, with hourly data collected to monitor coke deposition and regeneration. This data was filtered using the FCC feed rate to remove any instances where the unit operated unusually (i.e., during power loss).

2.2 Heat balance and feature selection

The heat balance is key to determining the coke yield with the amount of coke generated equal to the supply of heat needed during heating the feedstocks, supplying the endothermic catalytic cracking energy, heating the air, etc. Consequently, a correlation value was derived, based on selected variables using Minitab® [33], with the first screening process providing an overview of the potential variables that impact the coke yield and their importance in deriving the correlation.

2.3 Multiple linear regression based on least squares

A multiple linear regression model was built using Minitab® [33]. With the commercial unit adjusting itself by changing variables to maximize the feed rate via the advanced control system, a multiple linear regression that normalizes other changing variables was developed when quantifying specific parameter (e.g., the impact of co-processing). A so-called "step change method" (or mass balance based on observed yield) which assumes adding biogenic feedstocks as the only changing variable has been shown problematic when operating commercial units [16]. The P-value and T-value were used to evaluate the significance of various selected parameters [34] and the R2 adjusted value was used to evaluate the model derived from the multiple linear regression. A Variance Inflation Factor (VIF) was also used to prevent multicollinearity [35]. Samples were split into training and test groups to evaluate the model and prevent overfitting. The use of multiple linear regression combined with process knowledge provided a simple method for the first data run.

2.4 Bootstrap method

Approximately 8,900 lines of data were collected with the typical method used to evaluate the correlation involved splitting the data into training and test groups. However, the uncertainties that occurred over the selection of samples as training and testing groups resulted in uncertainties of the coefficients. Therefore, a bootstrap method was used to randomly select training and test groups while running the models through as many iterations as possible. The bootstrap method provided a useful statistical estimation given the information available [36]. Essentially, the method resampled the data, consequently making statistical inferences on the distribution characteristics of the data. As increased numbers of iterations required longer periods of time initially 10,000 iterations were compared to 100,000 iterations. However, as the results were very similar (although more evenly distributed when using 100,000 iterations) 10,000 iterations were routinely used.

2.5 Bayesian ridge regression

An additional algorithm was used to further validate the regression coefficients. Unlike the linear regression method, which assumes the response is a linear combination of weights multiplied by a set of predictor variables, Bayesian linear regression assumes the response is generated from a normal distribution.

The rationale followed is that we need a distribution of parameters to obtain more information rather than a simple point estimation, as the simple point estimation only tells us the most likely situation. In contrast, the distribution describes what is occurring in the entire space and, thus, as the number of data points increases, there is less uncertainty in the model parameters. As mentioned earlier, as there are uncertainties related to what samples were selected, a bootstrap method was also applied in Bayesian regression. This randomly selected samples and ran the model as many times as possible. The linear regression and Bayesian ridge regression, both with bootstrap, were run using the scikit-learn package in Python [37].

2.6 Statistical analysis

One-way ANOVA was used in Minitab® to compare the means of various groups at different co-processing levels [38]. The amount of coke burned per hour was normalized with the feed rate used to remove the impact of different rates of feed. As observed previously [16], the "signal to noise ratio" derived from co-processing biogenic feedstocks was low when the yields were not significantly different. In this case, with more process data available from the commercial units, we were able to identify the point where the signal was strong enough such that a difference in coke yield could be shown between baseline petroleum processing and co-processing.

3.Results and discussions

3.1 FCC heat balance and feature selections

As mentioned earlier, the overall refinery heat balance is a major component that can be used to better elucidate the coke yield obtained in the catalytic cracker. Key parameters that were initially used for model development were first screened using multiple potential process variables (Figure 1). Although the causal relationships can not be displayed, plus-or-minus signs were used to indicate the directional changes observed when one of the variables changed. For example, increasing the feed quantity increased the coke burn while an increase in riser temperature facilitated the endothermic reaction, resulting in more coke deposition in the unit. Alternatively, reducing the preheat temperature increased the coke yield with more heat (via burning more coke) needed to heat the feedstocks so that they reach the pre-set riser temperature. More heat will also increase the regenerator temperature and produce more steam when a catalyst cooler is used.

The term “delta coke” is used in some literature and company newsletters and it represents the ratio between the coke yield and catalyst-to-oil ratio [25,40,41]. The coke yield can be considered to stay constant as is mainly influenced by the air blower capacity and the availability of the excess oxygen, not by the catalyst coke selectivity or feed coking tendency [24,39]. Although, typically, enough coke is burned to satisfy the heat demand of the FCC unit, if the operating conditions are changed or the feedstock quality changes, the delta coke will also change. In the work reported here, the delta coke value was not used directly but, rather, we monitored changes in the temperature of the regenerator, as it is directly proportional to the delta coke value. As the delta coke value directly corresponds to the temperature difference between the regenerator and riser, the riser temperature is typically set as a fixed value and can therefore be considered as constant [24,39].

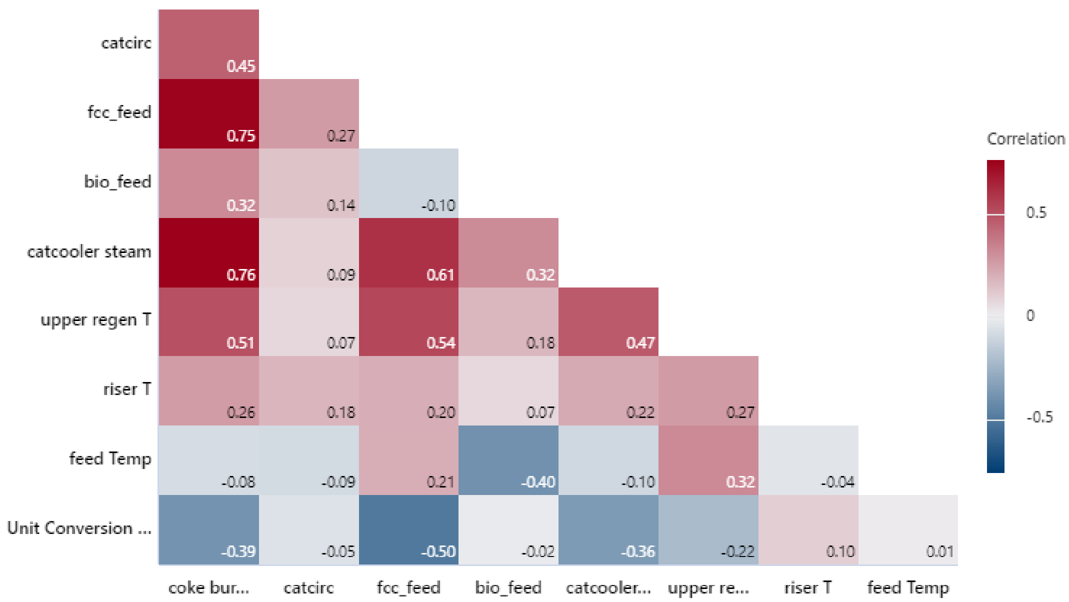

Figure 1 Correlation between the various factors that impact coke yield during commercial FCC operations

3.2 Statistical analysis – did co-processing biogenic feedstocks generate more coke?

The ANOVA test results indicated that a minimum of a 12% ratio of lipid-to-fossil feedstock, was needed to detect a statistically relevant difference in coke burn compared to the petroleum baseline (Figure 2) as, at lower co-processing ratios (low “signal-to-noise” ratio), the coke generated on an hourly basis was not significantly different. Thus, without normalizing the other variables, any the changes resulting from the addition of the biogenic feedstocks were not readily detected.

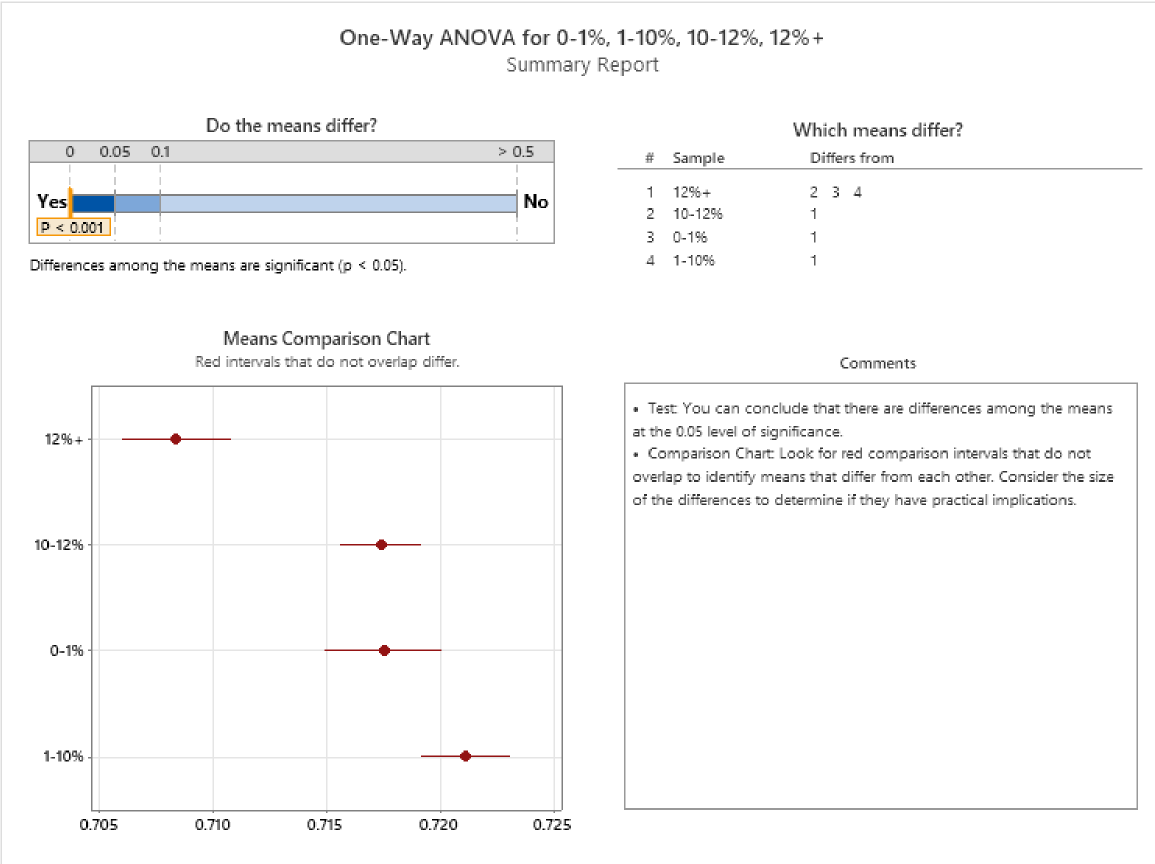

Figure 2 Statistical results obtained when comparing the average coke generation at various co-processing levels

3.3 Regression analysis

To develop a method of detecting any changes due to biogenic feedstock addition, a multiple linear regression approach, based on least-squares, was built to quantify the relative impact of the previously selected variables (Figure 3). Statistical values (p-, t-value) were used to quantify the significance of the variable while removing any variables that were not statistically significant. The variance inflation factor (VIF) provided by Minitab also ensured there was no multi-collinearity (less than 10) [35]. The standard error and R2-adjusted values indicated that the model performed well, particularly considering the limited filtering that was performed on the “real-world” data-set. The Pareto chart derived from the t-values confirmed that the biogenic feed was not the only changing variable and it was also not the most important variable impacting coke yield.

The Minitab calculated the 95%-confidence interval of the coefficient based on its standard error, indicating the likely range for the various coefficients. Although the analysis indicated that the coke generation rate per unit of biogenic feedstocks was less than fossil feedstocks, the exact value or ratio between the two was not clearly defined. This was likely due to the uncertainty range of the two coefficients. To try to reduce the uncertainty and derive a ratio that could quantify the renewable fraction in the coke (green coke) a bootstrap method, which generated a distribution of the coefficients after running the model 10,000 times (bootstrap with linear regression and Bayesian ridge regression), was applied. It was apparent that the average of the coefficients of the distribution confirmed early regression results and further reduced the uncertainties when quantifying the changes (Figure 4). These results indicated that the biogenic feedstock generated 76% of the coke, on a per unit of feed basis, compared with the fossil fuels (on average, for the annualised data). Thus, when incorporating the coefficient, on a per-barrel basis of feed, the co-processing of oleochemical feedstocks at the FCC resulted in the generation of less coke.

As the source of the coke is derived from either the fossil or bio feed, the equation below was used to quantify the amount of fossil and green coke generated:

$$Total\ coke\ =\ A * fossil\ feed\ +\ B * bio - feed \qquad \textit{Equation 1}$$

$$B = 0.76 * A \qquad \textit{Equation 2}$$

$$Bio - feed\ =\ fossil\ feed\ * co - processing\ percent. \qquad \textit{Equation 3}$$

$$A\ =\ total\ coke/(fossil\ feed * (1 +\ 0.76 *\ co - processing\ percent) \qquad \textit{Equation 4}$$

Method

Test set fraction 25.0%

Regression Equation

coke burn hourly = -5.61 + 0.22540 catcirc + 0.000375 fcc_feed + 0.000296 bio_feed + 0.05215 catcooler steam + 0.004171 upper regen T + 0.00494 riser T - 0.005037 feed Temp

Coefficients

Term	Coef	SE Coef	95% CI	T-Value	P-Value	VIF
Constant	-5.61	1.60	(-8.74, -2.48)	-3.51	0.000	
catcirc	0.22540	0.00484	(0.21591, 0.23489)	46.55	0.000	1.20
fcc_feed	0.000375	0.000006	(0.000363, 0.000388)	58.95	0.000	2.53
bio_feed	0.000296	0.000010	(0.000276, 0.000316)	28.68	0.000	1.73
catcooler steam	0.05215	0.00106	(0.05007, 0.05422)	49.23	0.000	2.27
upper regen T	0.004171	0.000614	(0.002968, 0.005375)	6.79	0.000	2.02
riser T	0.00494	0.00168	(0.00165, 0.00824)	2.94	0.003	1.16
feed Temp	-0.005037	0.000507	(-0.006031, -0.004043)	-9.93	0.000	1.58

Model Summary

S	R-sq	R-sq(adj)	PRESS	R-sq(pred)	AICc	BIC	Test S	Test R-sq
0.577595	83.46%	83.44%	2279.13	83.15%	11696.61	11757.89	0.540182	85.35%

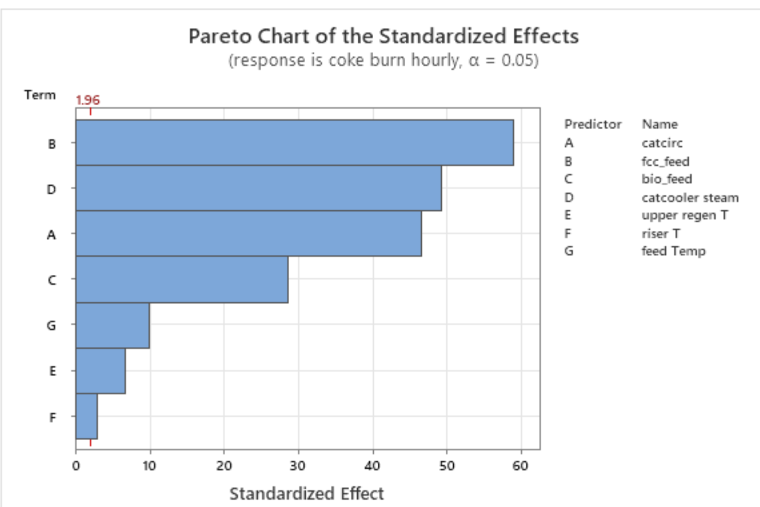

Figure 3 Multiple linear regression results from the Minitab used to quantify the impact of co-processing biogenic and fossil feedstocks. Note, factor importance based on t value from multiple linear regression

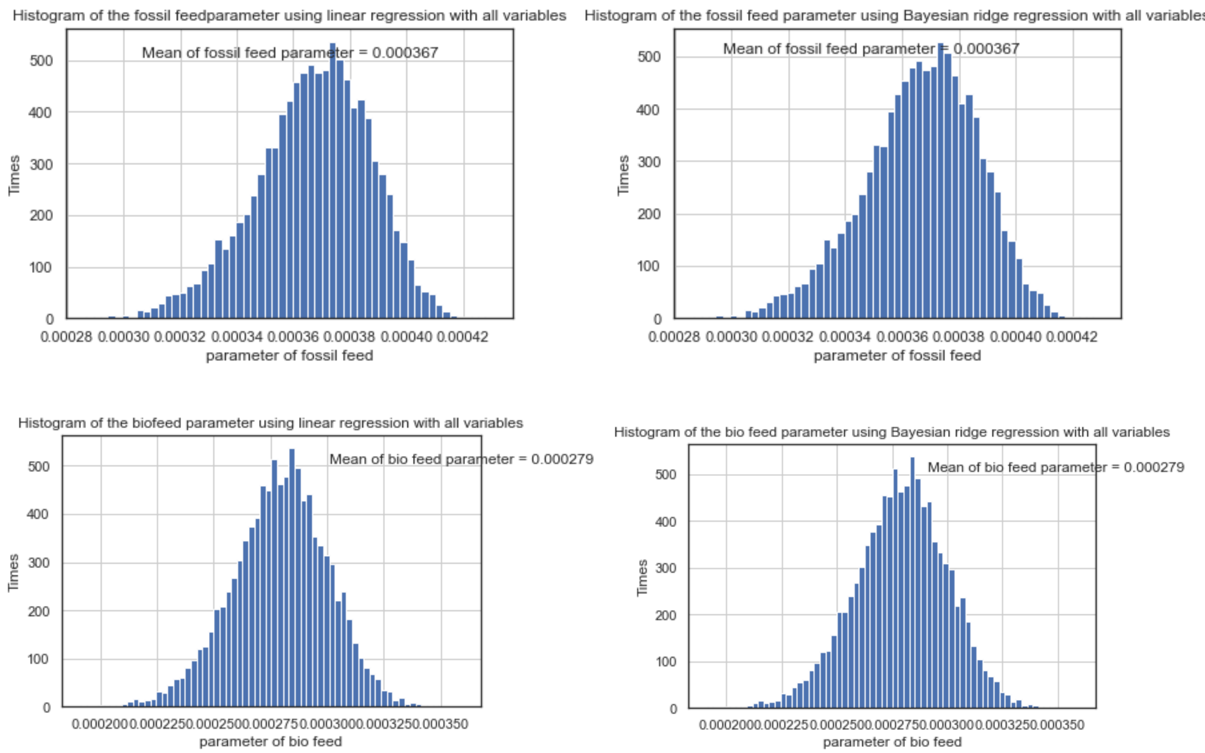

Figure 4 Coefficients of fossil and bio feed estimated from linear regression, Bayesian ridge regression using the bootstrap method

It was apparent that several variables impacted the coke produced by a commercial FCC besides the addition of biogenic feedstocks. As indicted here, at a low co-processing level (below 12% here), the coke yield was not statistically different. Thus, it is probable that the exact boundary will vary with other commercial units, depending on their operational co-processing conditions. As mentioned earlier, contradictory results have been reported in the literature regrading whether co-processing bio-feeds generate more-or-less coke [14,22,29,41]. When earlier pilot work compared the cracking of 100% soybean oil with 100% fossil vacuum gas oil (VGO) these researchers found that the heat of cracking was only 15% of the selected VGO (the temperature variance from the top of the riser to the bottom is less for soybean oil cracking [31]). This was likely due to the deoxygenation reaction where the major product is water (exothermic reaction) [10]. Thus, the heat requirement was lowered and, in theory, the coke yield should be lower when processing biogenic feedstocks. This agrees with the work reported here, when assessing the operation of a commercial FCC unit, where the coke yield decreased when the co-processing level was high. By further normalizing the other variables, by using a combination of regression and bootstrap, the coefficients for the bio feed and fossil feed could be determined, further indicating the lower coke yield generated after adding the biogenic feedstock.

4.Conclusions

Typically, when co-processing biogenic feedstocks at existing oil refineries some “green coke molecules” are produced, displacing fossil derived coke. During FCC co-processing, unlike the liquid streams where the flow can be directly metered, the coke yield has to be inferred from the air blower and the flue gas derived from the FCC regenerator. When a year’s worth of commercial-scale FCC co-processing data (hourly data) was assessed it was apparent that, at low co-processing ratios, the coke yield changes were not significant when compared to 100% petroleum processing. However, after normalizing other critical factors via multiple linear regression, the coke generating coefficients of the fossil and bio feed could be determined. When a bootstrap method and Bayesian ridge regression were also incorporated, it was apparent that, when co-processing biogenic feedstocks, less coke was generated. It also provided a reproducible way to quantify the amount of green coke produced when co-processing biogenic feedstocks at the FCC.

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