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A Rough Set-based Model for Evaluating the Stability of Biochar Based on Accelerated Aging Data

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Climate change is one of the major environmental problems that humanity is facing. Studies suggest the use of negative emission technologies (NETs) to substantially decrease atmospheric CO_2 concentration. Biochar can be applied to the soil for long-term carbon sequestration and simultaneous increase in soil fertility. However, the change in biochar properties after field application and exposure has not yet been thoroughly investigated, necessitating the need to determine if biochar can maintain its desirable properties with aging. Rough set-based machine learning (RSML) can be used to generate a data-driven model to predict the effect of different attributes on % change in C content of biochar. Four rules were accepted that relate the effect of feedstock type, pyrolysis temperature, aging method, and aging duration to % change in C content of biochar. Rules 1 and 2 cover 27 % and 14 % of the training set with 100 % accuracy, while Rules 3 and 4 cover 14 % and 10 % of the training set with 66.67 % accuracy. The performance of the rules was assessed via a k-fold (k=10) cross validation method and checked for mechanistic plausibility. The findings of the study can help maximize the potential benefit of biochar in climate mitigation.

1. Introduction

A large-scale greenhouse gas (GHG) emissions mitigation program is required to limit average atmospheric temperature rise at below 2 °C by 2100. In order to meet this target, there is a need for all countries to commit to net zero carbon emissions by 2050. Negative emission technologies (NETs) are techniques to remove carbon from the atmosphere and sequester it in alternative form (UK Parliament, 2018). Biochar application to soil is an example of a NET (Woolf et al., 2010). The use of biochar technology also has the potential to reduce waste, enhance soil quality, and even generate energy as a byproduct. Biochar is produced during the pyrolysis of biomass. Biochar soil amendment results in long-term storage of stabilized carbon,and can also contribute to reduction of soil GHG emissions, increased cation exchange capacity (CEC), plant nutrient availability, and enhanced soil's water-holding capacity (Leng et al., 2019).

The proper evaluation of the biochar's carbon sequestration potential is essential to achieve its full potential in climate change mitigation (Belmonte et al., 2018). There is a need to determine the stability of biochar in soil (Spears, 2018) since it determines the permanence of carbon sequestration (Wang et al., 2020). Biochar's stability refers to the relative stability of its chemical and physical properties and the length of the life cycle of biochar (biochar's mean residence time) (Wang et al., 2020). Subjecting biochar to aging can alter its physical and chemical properties and effectiveness for soil amendment (Wang et al., 2022). However, it is impossible to directly measure the stability of biochar over the span of hundreds of years needed to give negative emissions (Leng et al., 2019). Artificial accelerated aging methods could be used to simulate the natural biochar aging process (Wang et al., 2020).

The existing biochar stability assessment methods include qualitative evaluation of the physical and chemical properties of biochar and utilization of the kinetic mineralization models (Wang et al., 2020). However, these methods are still unable to accurately estimate the stability of biochar in soil since they do not accurately reflect

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the impact of the interaction between biochar and the influential factors (Wang et al., 2020). Existing data from biochar literature can be used to elucidate the effect of the different factors on the stability of biochar in soil with the aid of machine learning (ML) (Ardabili et al., 2019). It also allows the generation of useful data-driven models (Sen, 2021). Several applications of ML on mitigating climate change cover carbon sequestration, estimating carbon dioxide emissions, predicting GHG concentrations, and identifying climate-vulnerable regions (Kumar, 2022). ML has also been applied to city-level data that characterizes waste management to generate a rule-based model and gain useful insights towards circular city economies (Gue et al., 2021). This study is the first to develop a rule-based model using rough set-based machine learning (RSML) using Rosetta software to evaluate the stability of biochar in soil based on accelerated aging data. The Rosetta software includes additional features (i.e., model validation) and can be implemented with a graphical interface that is user-friendly, hence, it is being widely used in the scientific community (Hvidsten, 2013). The model developed here consists of interpretable if-then rules that can be easily applied in the actual setting. The rule-based model can help determine the effects of the different factors on the stability of biochar after subjecting to different artificial biochar aging methods. In addition, the rule-based model can also guide the proper use of biochar for soil amendment and facilitate the preliminary evaluation of the carbon sequestration potential of biochar. This paper is written as follows. Section 2 details the methodology for the development of the rule-based model. Section 3 discusses the results of this study. Section 4 provides the conclusion and prospects for future work.

2. Methodology

Figure 1 shows the flowchart employed to develop the rule-based model. The data set used in this study is from Wang et al. (2020) and Liu and Chen (2022). The carbon in biochar can be used to assess the stability of biochar in soil (Leng et al., 2019). The effects of the different factors such as feedstock type, aging method, pyrolysis temperature, and aging duration on the stability of C in biochar (expressed in terms of % change in carbon content) are investigated. The higher the value of the % change in C content, the greater the capability of biochar in storing carbon; positive and negative values represent the gain (+) and loss (-) of C in biochar. As can be seen in Table 1, the total acquired data amounting to 164 samples were divided into two sets: 115 samples (70 %) were used for building the information system for the generation of rules (training set), and 49 samples (30 %) were used for the validation of rules (validation set).

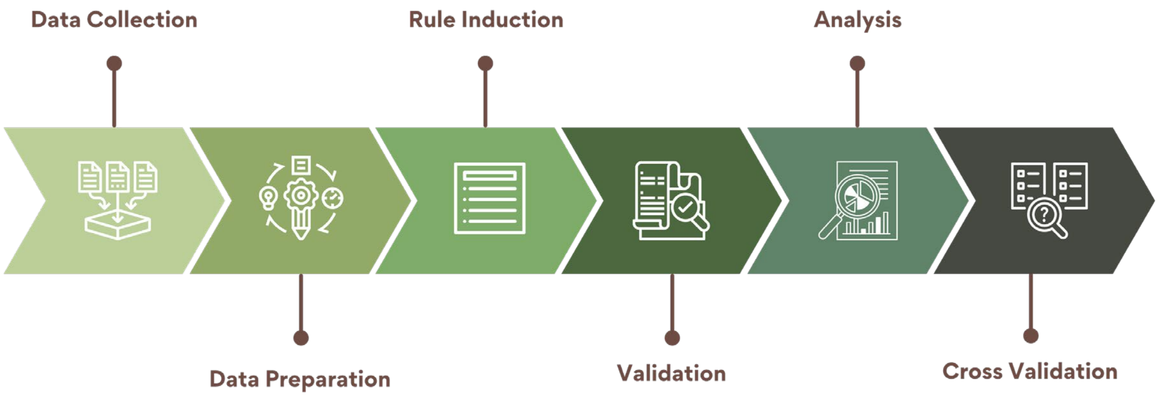

Figure 1: Flowchart of research methodology

Table 1: Data set sample distribution

Total Number of Samples in the Data Set	No. of Samples Used as Training Set (70 %)	No. of Samples Used as Validation Set (30 %)
164	115	49

RSML is implemented using Rosetta software (Hvidsten, 2013). The collected data was preprocessed to discretize the information into number-coded values. The information system includes the values of the attributes in columns (feedstock type, pyrolysis temperature, aging duration, and aging method) that are believed to affect the values of the % change in C content of the biochar (decision column). The missing data were resolved using the mean fill tool of the software. Table 2 shows the discretized data along with its corresponding number-coded value.

Rule induction was done with Johnson's algorithm which generated the rules (Hvidsten, 2013). The rule-based model was generated by employing the training data set. The top-performing if-then rules were accepted based on % coverage, $cov_S(\Phi, \Psi)$, represented by Eq(1) and % accuracy, $acc_S(\Phi, \Psi)$, represented by Eq(2). The coverage factor, $cov_S(\Phi, \Psi)$, finds the contribution of rule, Φ, to the decision, Ψ. The accuracy factor, $acc_S(\Phi, \Psi)$, on the other hand, relates to the probability that an object which meets condition, Φ, will result in decision, Ψ. The plausibility of the accepted rules was further supported by the different studies in biochar literature. The performance of these if-then rules was evaluated using the validation set. Furthermore, a k-fold cross validation was done to avoid overfitting and to further assess the acceptability of the rule-based model.

$$cov_S(\Phi, \Psi) = \frac{card(\| \Phi \ \Lambda \Psi \|)}{card(\Psi)} \tag{1}$$

$$acc_S(\Phi, \Psi) = \frac{card(\| \Phi \ \Lambda \Psi \|)}{card(\Phi)} \tag{2}$$

Table 2: Discretization of the attributes

	Feedstock Type	Aging Method	Pyrolysis Temperature (°C)	Aging Duration (y)	% Change in C Content
1	Mixed Crop Straws	Field Experiment, Freeze Thaw Cycles	[200, 225)	[0.0005, 0.0009)	[-40.7, -30)
2	Corn Stover, Wheat Straw	20 % Acidification, Dry Wet Cycles	[225, 375)	[0.0009, 0.0062)	[-30, -20)
3	Elephant Grass, Peanut Straw	40 % Acidification	[375, 490)	[0.0062, 0.0076)	[-20, -10)
4	Corn Straw	60 % Acidification, HCl+HF, $NaOH+H_2O_2$	[490, 575)	[0.0076, 0.0137)	[-10, 0)
5	Apple Tree Branches, Maize Stalks, Rice Straw, Sunflower Seed Shells	Ethanoic Acids	[575, 700)	[0.0137, 0.1793)	[0, 10)
6	Alfafa Meal	Malic Acids		[0.1793, 6.94)	[10, 17.1)
7	Cellulose	5 % H_2O_2, Citric Acids			
8	Corn Cob, Eucalyptus Wood, Furfural Production Residues, Maize Stover, Reed	10 % H_2O_2			
9	Maize Straw	15 % H_2O_2, 3 % H_2O_2, $NaOH+H_2O_2$			
10	Corn Stalk	0.01 mol H_2O_2, 20 % H_2O_2, 25 % Acidification, Microbial Cultivation, Plant Cultivation, Tartaric Acids			
11	Bamboo	Citric Acids, Co-composting, $HCl+H_2O_2$			
12	Alternanthera Philoxerides, Pig Manure, Pinewood Chips, Wheat Straw Pellets, Wood	6 % NaClO, $CaCl_2$, $HNO_3+H_2SO_4$, $HNO_3+H_2SO_5$, Microbial Inoculation, Wet Dry Cycles			
13	Alfafa, Biosolid + Grass, Cotton Straw, Dairy Manure, Grass, Peanut Shell, Pig Manure, Rice Hull	Natural Clay			
14	Elephant Grass, Peanut Straw	Kaolinite			
15	Sawdust	25 % HNO_3, Montmorillonite			

3. Results and Discussion

This section presents the discussion on the generation and evaluation of the rules in determining the stability of biochar in soil in terms of % change in C content after subjecting to artificial accelerated aging. It also presents the related studies on biochar literature that further support the plausibility of the top performing rules.

Table 3 shows the confusion matrix. It consists of columns that contain the values of the % change in C content (see Table 2 for the actual values) predicted by the rule-based model and rows which contain the actual values. Overall, the generated rules have correctly classified 96.64 % of the training set. It is important to note that out of 115 objects in the training set, only 112 were captured, and out of 112, 108 objects were correctly classified which are the highlighted values in green.

The top performing rules generated in the training set in terms of % coverage and % accuracy are accepted and shown in Table 4. Rule 1 states that if the feedstock type is corn cob, eucalyptus wood, furfural production residues, maize stover, or reed, and the aging duration ranges from 5 d (0.0137 y) to 65 d (0.1793 y), the % change (increase) in carbon content ranges from 0 to 10 %. This result shows that plant-based and wood-based biomass feedstock can increase the carbon content after being accelerated to aging. The increase in carbon content of aged biochar can be explained by the deashing treatment using HCl and HF solution and the loss of organic matter with low carbon content during aging (Liu and Chen, 2022). The deashing treatment facilitated the dissolution of minerals which in turn exposed the hidden carbon surface and pores blocked with minerals (Liu et al., 2017). Rule 1 can also be supported by other studies on biochar stability. Since more lignin and cellulose are contained in plant-derived biochar, there is a higher concentration of aromatic carbon, which gives the biochar higher stability and resistance to microbial decomposition (Tomczyk et al., 2020). Similar findings were reported for biochar produced from other biomass feedstock types. Ali et al. (2021) observed higher carbon content in the characterization of corn cob biochar samples, while Purakayastha et al. (2015) presented the potential of maize stover for long-term C sequestration in soil because it was found to be the most stable among other biochar samples. Biochar derived from eucalyptus wood has high stability because of its increased carbon concentration, producing a more condensed carbon structure (Domingues et al., 2017), which can occur regardless of the pyrolysis mode (Inkoua et al., 2022). No significant finding was obtained from the literature for the artificial aging duration in relation to Rule 1 but this may suggest that shorter aging duration can have a favorable interaction effect with feedstock type in stabilizing the C in biochar. The coverage of Rule 1 in the training set is 27 % at 100 % accuracy (see Table 5).

Table 3: Confusion matrix of the generated rules from the training set

		Predicted			
		3	4	5	
Actual	3	14	0	0	
	4	2	64	2	Overall Accuracy
	5	0	0	30	96.43 %

Table 4: Accepted rules for biochar carbon stability

Top Performing Rules	
Rule 1	IF Feedstock Type = 8 AND Aging Duration (y) = 5 THEN % Change in C Content = 5
Rule 2	IF Aging Type = 4 AND Aging Duration (y) = 1 THEN % Change in C Content = 3
Rule 3	IF Feedstock Type = 12 AND Pyrolysis Temperature (°C) = 4 AND Aging Duration (y) = 6 THEN % Change in C Content = 3
Rule 4	IF Feedstock Type = 12 AND Pyrolysis Temperature (°C) = 5 AND Aging Duration (y) = 6 THEN % Change in C Content = 3

Table 5: Performance of accepted rules

Rules	Training Set		Validation Set		10-Fold Cross Validation		
	% Coverage	% Accuracy	% Coverage	% Accuracy	Number of Appearances	Highest % Coverage	Highest % Accuracy
1	27	100	50	100	10	75 %	100 %
2	14	100	18	100	8	50 %	100 %
3	14	66.67	0	0	-	-	-
3’	-	-	-	-	7	100 %	100 %
4	10	66.67	0	0	-	-	-

Rule 2 states that if the aging type is 60 % Acidification, HCl+HF, or $NaOH+H_2O_2$ and the aging duration is less than 8 h (0.0009 y), then the change in carbon content is -10 % to -20 %. A study by Fan et al. (2018) shows that the carbon content of biochar subjected to 60 % HNO_3/H_2SO_4 solution decreased by 18.07 %. In their findings, oxidized biochar was found to have a higher percentage loss in carbon content than the original biochar because more carbon would dissolve if subjected to oxidation. This finding is consistent with Rule 2. No significant finding was found in the literature for the artificial aging duration in relation to Rule 2 but it may also appear that shorter aging duration can have a negligible interaction effect with the mentioned accelerated aging methods in decreasing the carbon content of biochar in soil. This hypothesis is made since the aging duration is less than 8 h (0.0009 y) only which may suggest that it is not enough to induce a significant effect on the C content of biochar in soil. Rule 2 covers 14 % of the training set at 100 % accuracy.

Rule 3 states that if the feedstock type is *Alternanthera philoxeroides*, pig manure, pinewood chips, wheat straw pellets, or wood, the pyrolysis temperature is 490 °C to 575 °C, and the aging duration is greater than 65 d (0.1793 y), then the change in carbon content is -10 % to -20 %. Similarly, Rule 4 states that if the feedstock

type is *Alternanthera philoxeroides*, pig manure, pinewood chips, wheat straw pellets, or wood, the pyrolysis temperature is greater than 575 °C, and the aging duration is greater than 65 d (0.1793 y), then the change in carbon content is -10 % to -20 %. Wei et al. (2019) studied the stability of biochar samples using rice straw, pine wood, pig manure, and sewage sludge at varying pyrolysis temperatures of 300 °C, 400 °C, 500 °C, 600 °C, and 700 °C. Their results show that biochar samples have higher carbon content and stability at temperatures greater than 500 °C. The study of Wei et al. (2019) can be extended to reveal new insights by considering different accelerated aging methods and aging duration as additional parameters during the investigation to know their influence in the stability of biochar in soil. Due to this scientific finding by Wei et al. (2019), it can be hypothesized that the dominating attribute in Rules 3 and 4 is the aging duration. This may suggest that longer aging duration can significantly affect the C content of biochar in soil. Since the effect of aging on biochar properties, composition, and C sequestration is still under discourse (de la Rosa et al., 2018), there is a need to investigate further the effect of aging duration on the C content in biochar as well as its interaction with other attributes not included in this study. Rules 3 and 4 cover 14 % and 10 % of the training set at 66.67 % accuracy.
The accepted if-then rules for biochar carbon stability were subjected to validation to assess their performance further. Table 5 shows the % coverage and % accuracy of the rules in the validation set. It is evident that Rules 1 and 2 perform well in the validation set. Overall, Rules 1 and 2 have correctly classified 12.5 % of the validation set.
A k-fold (k=10) cross validation (see Table 5) was performed to avoid overfitting and to further evaluate the performance of the rules. This was done by randomly splitting the dataset into training set and validation set ten times. Out of the 160 data points, 90 % (144 objects) was identified as the training set to train the model, and the 10 % (16 objects) served as the cross validation set.
The recurring top performing rules from the cross validation, the number of trials they appeared in the method, the highest % coverage, and the highest % accuracy are shown in Table 5. Rule 1 appeared in all 10 trials, Rule 2 showed up in 8 trials, and new Rule (Rule 3') was generated and validated in 7 trials. The results show that the first two top-performing rules in the first validation method also exhibit good performance during the cross validation. This implies that the methodology done in the Rosetta software provides satisfactory results.

4. Conclusion

A rule-based model was developed using RSML to determine the stability of biochar in soil based on accelerated aging data. The model is comprised of component if-then rules that relate the condition attributes (feedstock type, pyrolysis temperature, aging method, and aging duration) with the decision attribute (% change in C content of biochar). The results show that these attributes can significantly affect the stability of biochar in soil. It is evident that Rules 1 and 2 performed well in the training, validation and cross validation datasets. Rules 1 and 2 cover 27 % and 14 % of the training set with 100 % accuracy. They also cover 50 % and 18 % of the validation set with 100 % accuracy. The rule-based models were also examined for mechanistic plausibility. The cross validation method was implemented as part of the methodology to avoid overfitting. The rule-based model can provide useful insight to maximize the benefits of biochar in climate change mitigation. Future work can use larger datasets and other attributes not included in this study to further improve the prediction of the rules.

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