Artificial Neural Networks To Distinguish Charcoal from Eucalyptus and Native Forests Based on Their Mineral Components

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Abstract

Charcoal has been used as a renewable energy source in many countries. However, the indiscriminate use of wood from native forests is detrimental to sustainability. The development of rapid and efficient methodologies for distinguishing charcoal produced from native Forest or *Eucalyptus* plantations is essential to curb illegal coal transport and trade. The aim of this study was to distinguish charcoal from native and Eucalyptus woods by Artificial Neural Network (ANN) based on their mineral composition. Specimens from native woods (*Apuleia* sp., *Cedrela* sp., *Aspidosperma* sp., *Jacaranda* sp., *Peltogyne* sp., *Dipteryx* sp. and *Gochnatia* sp.) and from *Eucalyptus* sp. hybrid woods from commercial forest plantations were pyrolysed at temperatures from 300 to 700°C in order to simulate the actual pyrolysis conditions and species widely used illegally in southeastern Brazil. The composition and proportion of the mineral elements of charcoal were determined by X-ray fluorescence (XRF). ANNs were trained based on the elemental composition of the charcoal specimens to classify the species and origin of the charcoals (native forest and *Eucalyptus*). ANNs based on mineral element content yielded high percentage of correct classification for charcoal specimens by species (72% accuracy) or origin (97% accuracy) from an independent validation sample set.

Keywords: X-ray fluorescence, artificial intelligence, charcoal distinction, machine learning.

1. INTRODUCTION

Charcoal is a major source of energy in many countries. According to FAOSTAT (2018), Brazil occupies the first position among the main world producers of this product and its consumption is concentrated in the steel industry. Extensive areas of *Eucalyptus* are cultivated to meet the demand of the steel industry in Brazil (IBA 2017). However, wood from native forests has been used illegally.

According to Stange et al. (2018), charcoal producers have used native species from deforestation regions in tropical forests of world. The use of native wood for charcoal production is prohibited in many regions as it increases the deforestation rate in the country. According to Brazil (2013) the Brazilian government has made a national commitment to make 40% of the annual rates of deforestation in Cerrado biome. In 2016 charcoal manufacture from native forest reduced 31.7% (IBGE, 2016). However, enforcement actions to stop the production, transport and trade of illegally produced charcoal are insufficient because there is no official information about ilegal operations. The Brazilian cerrado is one of the most threatened biomes in the country while it is a conservation priority hotspot (Gonçalves et al. 2018).

Fraud is difficult to identify because of the similarity between the coals when observed with the naked eye (Ramalho et al. 2017). Identification of charcoal by anatomical analysis (Gonçalves et al. 2018) is time consuming and requires highly trained technicians. Alternative techniques for charcoal classification have been investigated, such as image analysis (Nisgoski et al. 2014, Maruyama et al. 2018), where some wood characteristics are extracted and analyzed for discriminating the precursory species. Moreover, some studies have shown promising results applying spectrum-based processing systems for classifying charcoal (Devrieux et al. 2010, Ramalho et al. 2017, Costa et al. 2018) but many limitations need to be overcome to apply these models in real situations where pyrolysis temperature and species are unknown.

The possibility of differentiating charcoals produced from planted or native wood from the mineral composition of charcoal was examined in the present study. X-ray fluorescence (XRF) is a technique used in analytical routines for identifying and measuring mineral elements in solid or liquid samples (Weindorf 2014). It is a versatile analytical technique that does not require exhaustive preparation of the material to be analyzed (Wobrauschek 2007).

XRF spectroscopy has been successfully applied in various fields of science that require rapid analytical routines such as agriculture (Freitas et al. 2019), soil science (Pelegrino et al. 2019), mining (Penido et al. 2019), environmental sciences (Muthukalum et al. 2020) and chemical (Szczepanik et al., 2015) and archeological studies (Attaelmanan and Mouton 2014).

Faced with the challenge of differentiating charcoal produced from planted or native wood, the hypothesis of this study is that the mineral composition of charcoal varies trees have grown in native and planted forest. The soil of forest plantations soils are prepared for production of wood for pulp or bioenergy industries and mineral contents are adjusted before planting. Some studies support our hypothesis, although they are not designed to evaluate this issue. For example, Brewer et al. (2009) have studied the ash composition of Switchgrass (grass), maize straw and hardwood (unspecified) samples by XRF spectroscopy. The results show that hardwood presented very different levels of CaO, Fe_2O_3, K_2O, MgO, MnO_2 and SiO_2 from Switchgrass (grass) and maize straw samples. Kim et al. (2013) have evaluated inorganic metals in oak, *Eucalyptus*, Pinus and Japanese cedar biochars by means of XRF spectrometry. They reported the presence of Si, K, Ca, Al, Mg, Na, P and Fe in all studied materials, but in different concentrations: Oak, Pitch pine and Japonese Ceder present much more Si, Ca, K, Al and Na (in g per kg) than *Eucalyptus* charcoals.

The above results clearly show that *Eucalyptus* wood has a very different composition from other biomasses. However, Brewer et al. (2009) and Kim et al. (2013) did not designed their studies to evaluate the potential of this technique to detect the origin of biochar precursor raw material. In this study, artificial neural networks (ANNs) were developed to evaluate the complex information on the mineral composition of charcoal specimens. ANNs are computational techniques based on mathematical models capable of classifying and predicting material properties (Basheer and Hajmeer 2000). ANN approach has been successfully applied in different fields of forest sciences, such as wood defect detection (Wenshu et al., 2015), wood veneer classification (Castellani and Rowlands, 2009) and wood species classification (Nisgoski et al. al., 2017; Cui et al., 2019).

Most studies that applied RNA to wood and its co-products have reported promising results for classification or estimation of properties. However, to our knowledge there is no study involving ANN for charcoal classification by origin, nor for identification of the precursor wood species. Thus, the aim of this study was to develop artificial neural networks to classify the origin of charcoal (native or planted forest) and the precursor species based on their mineral composition.

2. MATERIAL AND METHODS

2.1 Plant Material

Native tropical wood species from the Cerrado and Amazon biomes and reforestation were used in this study. The native species were *Cedrela* sp. (Cedar, labeled as “C”), *Aspidosperma* sp. (Peroba labeled as “P”), *Jacaranda* sp. (Rosewood, labeled as “J”), *Apuleia* sp. (Garapa labeled as “A”), *Peltogyne* sp. (Pau-roxo, labeled as “R”), *Dipteryx* sp. (Cumaru, labeled as “U”) e *Gochnatia* sp. (Camabará, labeled as “B”).

As for reforestation, two genetic materials from two forest companies were used. One company produces charcoal (6.5 year old Eucalyptus grandis × E. urophylla hybrid clones labeled “Ev”) and the other paper and pulp (6 year old Eucalyptus grandis × E. urophylla hybrid clones). labeled “Ec”) (Ramalho et al. 2017). The seven native species occur in the two largest Brazilian biomes, while Eucayptus hybrids were selected to represent the genetic variation that exists between the clonal materials used in reforestation by forestry companies in the country. Table 1 lists the species, furnaces and temperatures used to generada the dataset of this study.

Table 1 - Pyrolysis plan as a function of biological material, temperature and number of samples.

Vegetal Material	Code	Furnace		Number of specimens by temperature				
		ATG	Muffle	300°C	400°C	500°C	600°C	700°C
Apuleia sp.	A	X		5		6		6
Cedrela sp.	C	X		4		6		6
Aspidosperma sp.	P	X		5		6		6
Jacaranda sp.	J	X		5		6		6
Eucalyptus sp. (1)	Ec	X		5		6		6
Eucalyptus sp. (2)	Ev	X		5		6		6
Peltogyne sp.	R		x		2	2	2	2
Dipteryx sp.	U		x		2	2	2	2
Gochnatia sp.	B		x		2	2	2	2
Eucalyptus sp. (1)	Ec		x		2	2	2	2
Eucalyptus sp. (2)	Ev		x		2	2	2	2

Eucalyptus sp. (1): reforestation hybrids managed for charcoal production while *Eucalyptus* sp. (2): reforestation hybrids managed for pulp and paper industry.

2.2 Specimen Preparation

Central planks were removed from trees. 141 specimens (defect free) were obtained from native and Eucalyptus trees. From the native species, 91 specimens presenting the dimensions of 3.5 cm × 3.5 cm × 10 cm (R × T × L) were produced while 50 specimens (defect free) of Eucalyptus were produced with dimensions of 3.5 cm × 3.5 cm × 10 cm (R × T × L). Sampling was properly identified using a special pencil (labeling did not disappear after pyrolysis). Before pyrolysis, Wood specimens were kept in an acclimatized room and until reaching 12% moisture.

2.3 Pyrolysis Process

Wood specimens were pyrolysed in two laboratory ovens: Muffle furnace and Macro ATG oven (developed by the Center of International Cooperation in Agronomic Research for Development (CIRAD, France) and Universidade Federal de Lavras (UFLA, Brazil) as shown in Figure 2.

Macro ATG furnace

The Macro ATG prototype is equipped with an oven that can reach 1,000°C, a pyrolysis reactor pressure controller, a condensable gas condenser, a load cell, a gas chromatography flowmeter, a control panel and a software. Experiments can be developed using various gases simulating various conditions of partial or complete combustion in the presence of an inert atmosphere (Jesus et al., 2015, Ramalho et al., 2017).

Wood specimens were added in a crucible for pyrolysis in the ATG Macro. The temperature inside the system was monitored by means of four thermocouples and the gases resulting from the pyrolysis process were consensed by means of a condenser attached to the oven. After the prototype cooling period, the charcoals were removed and brought to moisture stabilization in a climate room (Ramalho et al. 2017).

The pyrolysis of the specimens was performed at an initial temperature of 40°C, a heating rate of 5°C. min^{-1} and remained for 1 hour at the final temperatures of 300, 500 and 700°C. After the process of converting wood to charcoal, the material remained inside the oven for cooling for fifteen hours (Ramalho et al. 2017).

The biological materials carbonized in the Macro ATG oven were *Apuleia* sp., *Cedrela* sp., *Aspidosperma* sp. (Peroba), *Jacaranda* sp. (Jacarandá) e *Eucalyptus,* resulting in resulting in hundred one (101) specimens divided into three pyrolysis temperatures.

Muffle furnace

The specimens were pyrolyzed in a muffle furnace (electric; model Q318M; Quimis, São Paulo, Brazil) according to the procedure described in Costa et al. (2018). Pyrolysis conditions were: 100°C initial temperature, 1.67°C.min^{-1} heating rate, 30 minutes at final temperatures 400°C, 500°C, 600°C and 700°C and 16 hours after completion of the conversion process.

The wood specimens were carbonized within a pyrolysis capsule placed inside the muffle furnace. The pyrolysis capsule was connected to a water-cooled condenser coupled to a receiver flask of condensable gases. The charcoal specimens were produced at 400, 500, 600 and 700°C to simulate the temperature range adopted in real situations in most Brazilian industries.

The biological materials carbonized in the muffle furnace were *Peltogyne* sp., *Dipteryx* sp., *Gochnatia* sp. and again *Eucalyptus*, resulting in resulting in forty (40) specimens divided into four pyrolysis temperatures.

The different furnaces and temperatures were used to verify the influence of the conversion process on the material distinction and to simulate the thermal variation that occurs in an industrial and conventional furnace. After the furnaces were cooled, the charcoals produced were removed and taken to a climate room until moisture stabilization.

2.4 X-ray fluorescence spectrometer

The detection of mineral elements was performed using two X-ray fluorescence spectrometers: M4 Tornado and S8 Tiger spectrometer.

M4 Tornado

The quantity of each mineral element present in the different charcoal samples was determined using an Energy Dispersive X Ray Fluorescence (EDXRF) spectrometer provided from Bruker Nano GmbH (M4 Tornado, Germany). On this typical commercial spectrometer the X-ray tube is a Rh micro-focus side window powered by a low power HV-generator and cooled by air. The spot size of 25 μm is obtained using a poly-capillary lens in a Mo-Kα mode. The X-ray generator was operated at 50 kV and 600 μA and different filters were used to reduce the background (100 μm Al/ 50 μm Ti/ 25 μm Cu). The energy resolution of a detector (thermoelectrically cooled silicon-drift-detector) was of 142 eV for 5.9 keV (Mn-Kα). Measurements were carried out under 20 mbar vacuum conditions [Silva et al. 2017]. According to Dias et al. (2015), the vacuum system avoids back diffusion and improves detection limits.

An inbuilt camera allows visualizing the operating area and permits the analysis in a fully automated mode. According to the required resolution the counting time and the scanning spatial resolution could be freely selected.. The sample is placed directly on a sample holder (360mm × 260 mm), which was attached to a stage translatable along XY. The scanning step size used was 25 μm and the time per analyzed point was 0.5ms × 3 cycles. Each selected area was analyzed over a period to accumulate sufficient data points for high-resolution mapping. Data output was obtained through the X-ray intensity of specific X-ray peaks corresponding to the element signals measured in each point defined by its X and Y coordinate (μm). The data were converted using the software's function into a data matrix, from which XY contour maps (2-dimensional maps) of the data were generated for each element [Silva et al. 2017].

The analysis was performed on five (5) specimens of each charcoal produced at different temperatures in the Macro ATG furnace. Each charcoal specimen was placed inside the equipment and a rectangular area was selected for irradiation during the analysis. In this area 100 points were analyzed and the resulting spectrum was the average of all these points.

The treatment of the X-ray spectra, analyze of the peaks and determination of which mineral elements are present in each sample and in what quantity were performed using the software M4 Tornado.

S8 Tiger

An Wavelength-dispersive X-ray fluorescence spectrometer (WDXRF) spectrometer, model S8 Tiger (Bruker Nano GmbH, Berlin, Germany) was also used to determine and quantify the mineral elements present in the different charcoal samples. This spectrometer is equipped with a Rh anode X-ray tube and 4 kW excitation power. Measurements of the characteristic Br Kα line were performed under vacuum at 20 to 60 kV and 170 mA tube setting and using 8 mm mask, LiF(200) crystal, 0.46° collimator, and scintillation counter. The adjusted peak position of Br Kα1 was set to a 2θ value of 29.97°, the background positions were set at 29.42° and 30.78° [Galina et al. 2016].

Samples were pressed using a semi-automatic hydraulic HERZOG HTP-40 press (Germany) in a 40 mm press tool. Analytical grade crystalline boric acid was used as a backing and rim material. A sample holder fitted with stainless steel masks having openings of 8 mm in diameter was applied for XRF measurements. [Galina et al. 2016]

The spectrometer is equipped with analyzing crystals XS-55, PET, LiF (200), LiF (220), XS-PET-C, XS-C, Al and Cu filters of different thickness, collimators (0.17, 0.23, 0.46, and 1°), and the box for automatic loading of 60 samples. [Suvorova et al., 2017].

The analysis was performed on two specimens of each charcoal sample. Each specimen was ground and sieved through a 150 micrometer nylon sieve. Pressed pellets were made using 4.5 g of ground charcoal and 3.5 g of Hoechst C wax ($C_{38}H_{76}N_2O_2$) of Merck. After homogenization of each sample with the wax, the material was compacted using a Vaneox (Fluxana) hydraulic press with a final pressure of 25 tons. Until reading, the tablets were kept in a desiccator. Soon after, the pellets were placed in specific specimen holders with a diameter of 34 mm and then placed inside the equipment. The analysis was performed by scanning the full length of the sample surface.

The spectrometer is equipped with SPECTRA plus software that allows selecting conditions, measurement parameters, optimal calibration equations, measurement of the calibration set, and mathematical data processing for the calibration set of CRMs [Suvorova et al. 2017]. The treatment of the X-ray spectra, analyze of the peaks and determination of which mineral elements are present in each sample and in what quantity were performed using the software Spectra 2.2.3.2.

2.5 Artificial Neural Network

Artificial Neural Network (ANN) of feedforward multilayer perceptron (MLP) type was developed using the mineral contents of charcoal specimens as input variables and the wood species or charcoal origin as output variables. The ANNs developed in the present study were performed using SPSS statistical software (v. 20).

Network architectures

The optimal network architectures were established by trying different combinations of number of hidden layers (1 or 2) and neurons (1 to 9). ANN 1 has six (6) neurons in the hidden layer and nine (9) output layer neurons, which represent the nine wooden species converted in charcoal specimens (*Eucalyptus*, *Peltogyne* sp., *Gochnatia* sp., Dipteryx sp., *Apuleia* sp., *Jacaranda* sp., *Aspidosperma* sp., *Cedrela* sp.) while ANN 2 presented two (2) hidden layer neurons and two (2) output layer neurons, which represent the origin of the charcoal (native forest or *Eucalyptus*). The maximum number of epochs of each ANN was 100. The diagrams of the designed ANN for species and for origin are shown in Figures 1 and 2, respectively.

Every neuron in hidden layer and output layer represents an activation function. In this study, a hyperbolic tangent sigmoid function was used as the activation function in the hidden layers while the output layer activation function was softmax. General information on the artificial neural network for classifying wood species or charcoal origin based on mineral composition are listed in Table 2.

Table 2: Information from artificial neural networks to classify the origin of charcoals based on their mineral components.

Layer	Variable	Information	
		ANN 1	ANN 2
Input	Covariate 1	Ca	Ca
	Covariate 2	K	K
	Covariate 3	Mn	Mn
	Covariate 4	Fe	Fe
	Covariate 5	Si	Si
	Covariate 6	S	S
	Covariate 7	Mg	Mg
	Covariate 8	Al	Al
	Covariate 9	Cu	Cu
	Covariate 10	Zn	Zn
	Covariate 11	Sr	Sr
	N of Units	11	11
	Rescaling Method for Covariates	Standardized	Standardized
Hidden	N of Hidden Layers	1	1
	N of Units in Hidden Layer 1 st	6	2
	Activation Function	Hyperbolic tangent	Hyperbolic tangent
Output	Dependent Variables	Wood species	Native or Eucalyptus
	N of Units	9	2
	Activation Function	Softmax	Softmax
	Error Function	Cross-entropy	Cross-entropy

Covariate sets for ANN

The model inputs (covariables) were the concentration values of the mineral components present in the charcoal and the output of the model were species (ANN1) or origin (ANN2). For ANNs, eleven (11) explanatory variables (Ca, K, Mn, Fe, Si, S, Mg, Al, Cu, Zn e Sr, hereafter called covariates) were considered for training the ANN to classify the species (ANN1) or origin (ANN2) of charcoals (Table 2). As the activation function does not generally map into the real numbers, the data set was standardized to a mean of 0 and a variance of 1.

Network training and validations

ANN models were validated by independent test set. To guarantee homogeneity between training and validation sets, the selection of the samples of each subset was made manually.

The sample set (142 observations) was ranked by species, temperature and origin and the data set was split into two uniformly distributed subsets. This procedure allowed higher control of the variability within each subset: the calibration set was composed of 95 specimens while test set had 47 samples with mineral composition information. The selection of ANN models was based on the percentage of correct classifications regarding the charcoal origin of the different species (ANN1) or native and Eucalyptus classes (ANN2).

3. RESULTS AND DISCUSSION

3.1 Mineral composition variation of charcoal

The mineral elements present in the charcoals produced from different species and under different pyrolysis temperatures were detected by X-ray fluorescence analysis. Table 3 presents the mean values as a percentage of the elemental composition of the native and planted wood charcoal samples.

The results show that minerals such as Calcium (Ca) and Iron (Fe) present higher proportion in relation to the others. In addition to varying by species, the percentage of minerals also varies as pyrolysis temperature increases, however, a trend was not detected. These variations are importatn for training the artificial networks to classify the charcoal by its origin. Although all data do not have a clear tendency detectable by visual analysis, the artificial neural network can recognize nonlinear data patterns.

Table 3 - Averaged mineral composition of charcoal by wooden species and temperature of pyrolysis.

Species	Temp	Percentage (%)										
		Ca	K	Mn	Fe	Si	S	Mg	Al	Cu	Zn	Sr
EV	300	21.08	3.59	2.77	42.07	3.16	0.28	1.92	4.12	1.21	2.60	1.27
	400	65.96	13.84	5.00	3.06	3.32	1.65	1.17	1.46	0.97	1.27	2.91
	500	31.33	13.58	4.64	36.39	1.95	0.69	0.76	1.30	1.29	1.19	1.32
	600	23.06	2.87	3.29	2.69	17.21	1.56	1.39	5.94	0.88	0.93	3.10
	700	27.37	19.41	3.26	23.75	9.85	1.37	0.52	4.55	1.18	1.52	1.28
EC	300	19.37	5.27	2.36	42.40	4.39	0.11	1.79	6.75	1.23	3.03	1.52
	400	53.81	24.36	2.72	2.15	3.59	2.29	2.09	1.32	1.20	1.46	3.10
	500	30.34	22.92	1.78	22.83	2.61	0.64	0.71	1.31	0.64	0.77	1.30
	600	22.78	32.93	1.49	2.32	5.68	1.51	1.05	3.76	1.00	1.00	2.54
	700	38.06	17.31	1.80	25.14	4.29	0.97	0.47	2.41	0.91	1.22	1.51
R	400	74.42	6.36	3.04	0.83	2.08	1.66	5.37	0.49	1.15	0.68	3.99
	500	78.75	4.55	2.89	0.57	0.89	1.47	5.23	0.32	0.87	0.49	4.34
	600	74.54	5.04	5.07	0.59	1.14	1.38	6.83	0.14	1.52	0.52	4.37
	700	79.34	4.24	3.24	0.46	0.74	1.32	5.38	0.99	0.99	0.28	4.41
B	400	4.59	4.38	0.57	0.50	12.07	1.49	0.51	75.09	0.30	0.45	0.57
	500	4.96	1.16	0.28	0.54	14.85	0.97	0.73	74.62	0.39	0.44	0.50
	600	5.46	1.64	0.31	0.68	12.42	0.94	1.07	66.31	0.41	0.38	0.66
	700	5.56	2.47	0.35	0.61	21.92	1.27	1.62	64.76	0.39	0.43	0.55
U	400	81.66	1.38	2.33	0.83	6.38	0.66	0.80	2.24	0.40	0.18	2.91
	500	76.72	2.56	1.68	1.18	9.08	0.88	0.93	3.22	0.65	0.36	2.31
	600	64.70	3.26	2.91	1.72	15.63	0.87	1.17	5.83	0.63	0.32	2.51
	700	67.89	2.57	2.53	1.40	14.66	0.70	0.98	5.35	0.39	0.23	2.60
A	300	53.60	13.90	1.62	6.22	0.22	1.56	1.73	0.90	0.34	0.87	1.08
	500	57.98	21.54	1.27	9.53	0.15	1.47	1.51	0.52	0.40	0.84	0.81
	700	62.96	23.28	1.31	6.26	0.17	0.85	0.91	0.43	0.28	0.58	0.77
J	300	48.09	1.31	2.53	21.00	1.98	0.35	4.24	3.09	0.63	0.65	1.24
	500	66.62	5.42	3.91	11.98	0.50	0.53	2.25	0.45	1.09	0.93	0.98
	700	62.91	1.14	3.38	14.80	1.26	1.01	3.91	1.20	0.96	0.96	0.68
C	300	57.98	6.16	0.21	5.27	1.08	0.17	3.60	2.11	0.33	1.01	1.87
	500	61.42	10.18	0.34	14.45	1.25	0.78	2.30	0.99	0.32	0.79	1.18
	700	69.02	8.87	0.49	10.56	0.33	0.35	2.66	0.58	0.34	0.86	1.38
P	300	46.07	21.81	7.89	1.38	3.16	0.19	1.79	11.30	0.17	1.25	1.50
	500	38.31	27.11	7.37	11.71	1.22	0.21	1.99	6.48	0.13	0.73	1.04
	700	47.95	13.31	6.48	8.13	2.13	0.31	4.16	11.98	0.28	0.66	1.08

There are few studies that have evaluated the composition and proportion of mineral elements in charcoal or forest biomass. Kim et al. (2013) have evaluated inorganic metals in oak, eucalyptus, pine and Japanese cedar biochars by X-ray fluorescence spectrometry and found Si, K, Ca, Al, Mg, Na, P and Fe in all studied materials. The elements that stood out in Eucalyptus were Si, K and Ca. In the present study the last two elements are present in high percentage. Brewer et al. (2009) studied the ash composition of Switchgrass (grass), maize straw and hardwood (unspecified) samples by X-ray fluorescence spectroscopy by pressed tablet method and found Al_2O_3, CaO, Cl, Fe_2O_3, K_2O, MgO, MnO_2, Na_2O, P_2O_5, SiO_2 and SO_3 in all varieties studied, and CaO presented the highest percentage (22.37%) for wood. Bouraoui et al. (2015) have analyzed the mineral content of faveira and found significant amounts of silicon (4430 mg / kg), calcium (1260 mg / kg) and potassium (990 mg / kg) while magnesium was detected in smaller amounts (550 mg / kg). All minerals reported by Bouraoui et al. (2015) were found in the charcoal samples analyzed in the present study.

3.2 Neural Network Architecture

The artificial neural networks architectures developed in this study are presented, respectively, in Figures 1 and 2 with their respective input layers, hidden layers, neurons, output layers and synaptic weights. Both ANNs to estimate the charcoal origin as a function of wooden species (Figure 1) as well as native Forest or Eucalyptus plantation category (Figure 2) were obtained using eleven (11) input neurons and one (1) hidden layer, with six (6) and two (2) neurons, respectively.

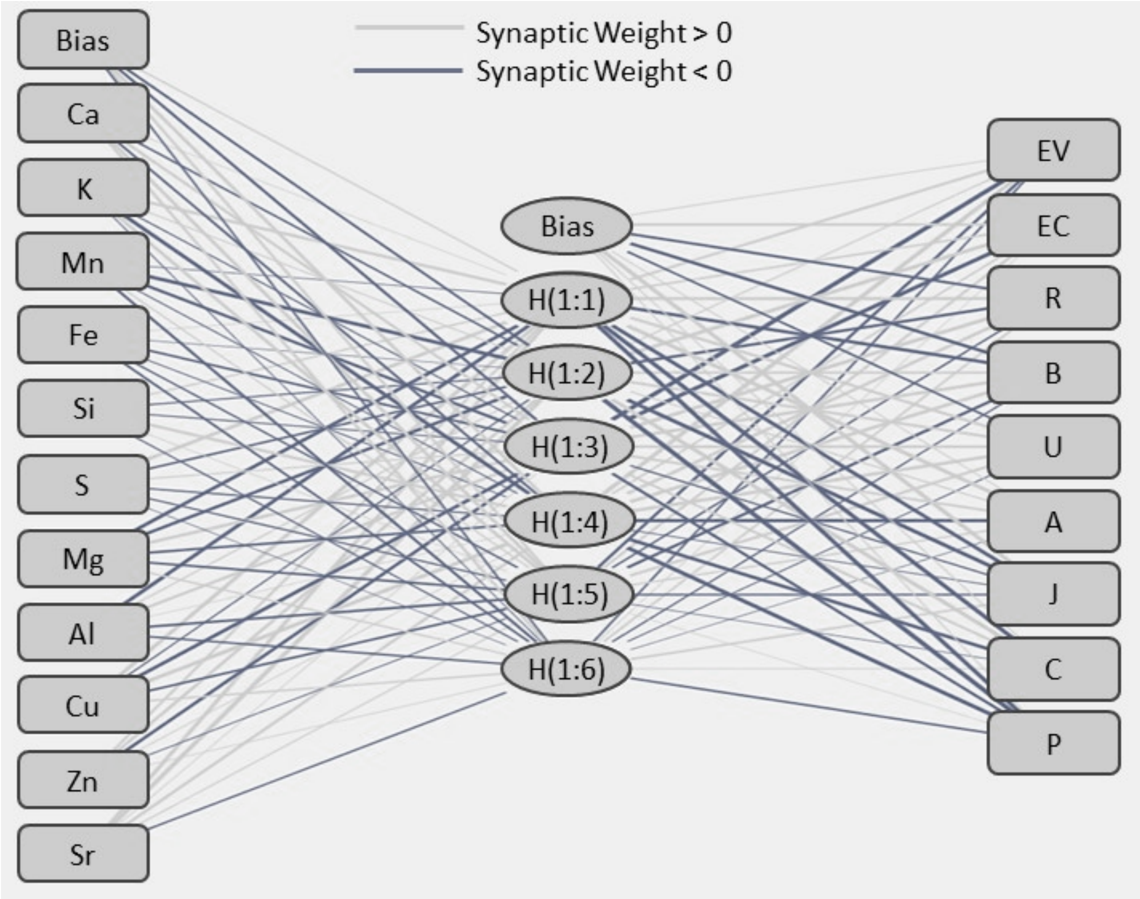

Figure 1 - Network Diagram to estimate the wooden species of charcoal based on the mineral composition.

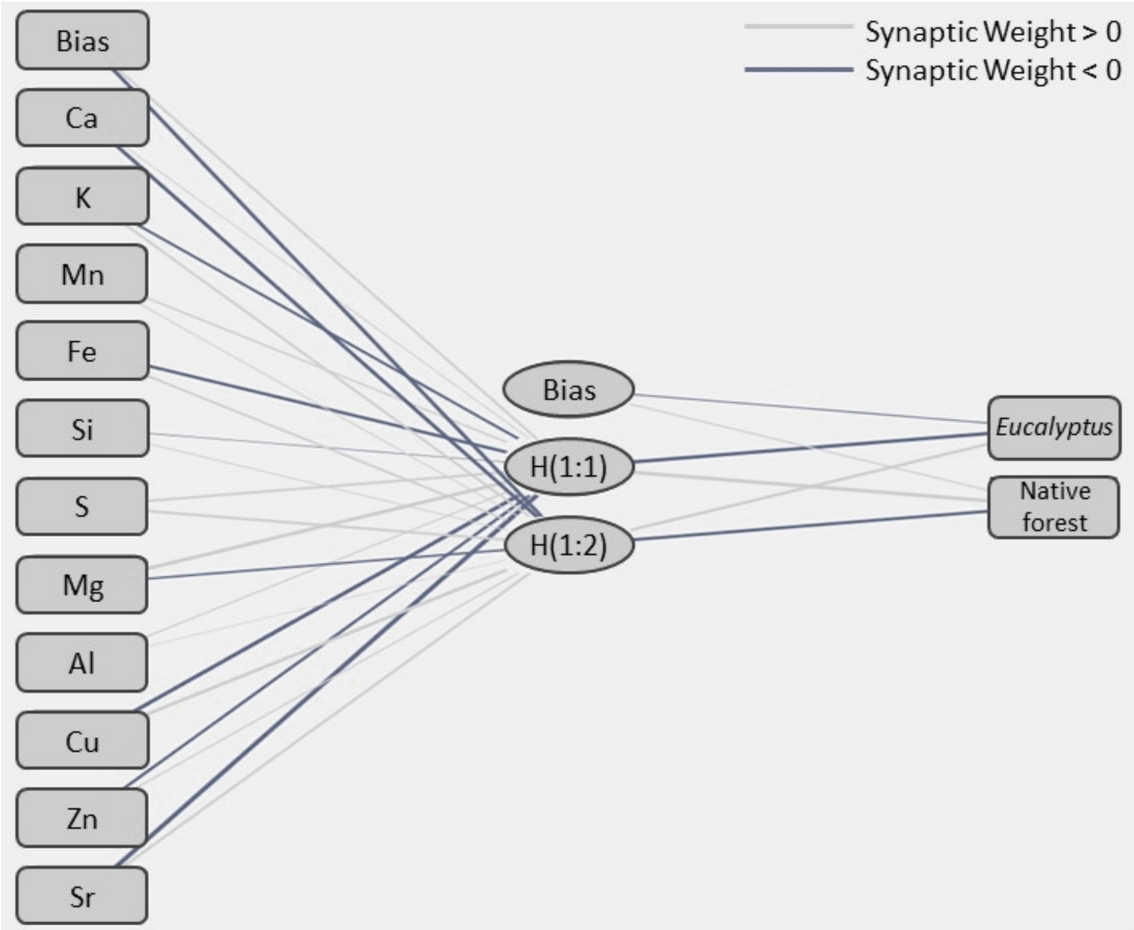

Figure 2 - Network Diagram to estimate the wooden origin (native Forest or Eucalypt plantation) of charcoal based on the mineral composition.

Synaptic weights represent the connecting forces between neurons and are used to store acquired knowledge (Haykin, 2001). Weight is considered excitatory when it is positive (> 0) and inhibitory when it is negative (<0). High synaptic weights are indicated by thick lines while low weights are represented by thin connections. Synaptic weight greater than zero is indicated in light gray color while synaptic weight below zero is indicated in dark gray (Figures 1 and 2). Since very negative or very positive weights can generate thicker connections, the more positive or the more negative a weight, the thicker the connection. Input variables can be evaluated by considering the connections between the hidden or the output layer.

The two ANN models were developed based on the values of the proportion of mineral components present in the material to estimate the origin of the charcoals as a function of woonden species and as a function of origin classes: native forest or Eucalypt plantation. For ANN 1 (Figure 1) the thick connections with very negative synaptic weights occurred at the

Ca, K, Si, S, Mg, Al, Cu, Zn and Sr inputs and with very positive weights occurred at Ca, K, Mn, Fe, S, Mg, Al, Cu and Zn (Table 4). For ANN 2 (Figure 2) the very negative weights were highlighted in the K, Fe, Si, Cu, Zn and Sr inputs and very positive in the Ca, Mn, S, Mg and Al inputs (Table 5). The thicker, very positive and negative connections indicate that the input variable is important to define the output variable, most of the mineral elements used in the input layer had such connections.

Table 4 - Training parameters of artificial neural networks 1 (ANN 1) used to estimate the origin of charcoal based on mineral components.

Predictor		Predicted								
		Hidden Layer 1								
		H(1:1)	H(1:2)	H(1:3)	H(1:4)	H(1:5)	H(1:6)			
Input Layer	(Bias)	0.293	-0.499	-0.544	0.753	0.461	-0.284			
	Ca	0.013	-0.392	0.610	-0.438	0.786	0.471			
	K	1.485	0.215	-0.169	-1.404	0.787	-0.022			
	Mn	-0.073	-2.817	-1.104	-0.122	0.198	-0.530			
	Fe	0.064	-0.008	-0.654	-0.134	-0.345	-0.430			
	Si	0.409	-0.338	-0.271	1.225	0.053	-0.219			
	S	1.658	-0.535	0.382	-0.378	-0.364	-0.002			
	Mg	-1.414	-1.397	1.567	-0.824	-0.736	0.433			
	Al	-1.390	0.289	1.046	0.029	-0.919	-0.503			
	Cu	1.317	-0.170	-1.491	2.003	-0.665	0.501			
	Zn	0.450	0.721	-0.996	0.074	-0.109	0.091			
	Sr	1.062	1.577	0.010	0.677	0.211	-0.397			
		Output Layer								
		[Ev]	[Ec]	[R]	[B]	[U]	[A]	[J]	[C]	[P]
Hidden Layer 1	(Bias)	0.217	1.180	-1.027	-1.556	-0.467	0.102	1.570	0.111	0.082
	H(1:1)	0.877	0.564	1.804	-1.832	1.194	2.084	-1.545	-1.673	-1.519
	H(1:2)	0.287	2.227	-1.288	0.818	1.213	0.295	-1.926	1.690	-2.486
	H(1:3)	-3.054	-2.728	2.124	1.777	1.177	0.858	-0.354	0.653	-0.714
	H(1:4)	1.726	0.398	1.305	1.671	1.838	-2.785	-0.152	-1.796	-2.561
	H(1:5)	-0.622	0.534	-0.336	-0.879	1.041	0.444	-0.483	-0.061	0.009
	H(1:6)	-0.306	0.004	0.385	-0.226	-0.130	-0.007	0.659	0.185	-0.619

Table 5 - Training parameters of artificial neural networks (ANN 2) used to estimate the origin of charcoal based on mineral components.

Predictor		Predicted			
		Hidden Layer 1		Output Layer	
		H(1:1)	H(1:2)	[Tipo=1]	[Tipo=2]
Input Layer	(Bias)	0.383	-0.605		
	Ca	0.103	-0.663		
	K	-0.467	0.403		
	Mn	0.374	0.145		
	Fe	-0.882	0.429		
	Si	-0.052	0.224		
	S	0.542	0.553		
	Mg	1.559	-0.455		
	Al	0.265	0.098		
	Cu	-0.922	0.840		
	Zn	-0.486	0.258		
	Sr	-1.038	0.450		
Hidden Layer 1	(Bias)			-0.256	0.370
	H(1:1)			-1.807	1.933
	H(1:2)			0.813	-0.911

3.3 Identificating the charcoal origin

The model for classifying species (ANN 1, Table 6) was able to correctly predict 88.3% of the specimens of the independent test set and 74.5% of specimens belonging to the training set. Of the erroneous classifications in the test set, only two (2) specimens of the native genus (Jacaranda) were confused with Eucalypt specimens and only one (1) specimen from plantation (Eucalypt) was classified as Peroba (native). Most incorrect predictions were of the genera of native specimens among themselves or Eucalyptus specimens among themselves. This type of error within each category is positive for classification purposes, as it is possible to identify illegal native charcoals independent of the tree genus.

Table 6 - ANN classification of charcoal by wooden species (Ev, Ec, R, B, U, A, J, C and P) using the mineral composition of the charcoals produced at temperatures from 300 to 700°C

Observed	Predicted by ANN									Correct classifications (%)
	EV	EC	R	B	U	A	J	C	P	
Training set										
EV	12	5								70.6
EC	3	13						1		76.5
R			5							100.0
B				5						100.0
U					6					100.0
A						11				100.0
J							10		1	90.9
C								11		100.0
P							1		10	90.9
Overall Percent (%)	16.0	19.1	5.3	5.3	6.4	11.7	11.7	12.8	11.7	88.3
Test set										
EV	5	2							1	62.5
EC	1	7								87.5
R			3							100.0
B				3						100.0
U					2					100.0
A						2	1	2	1	33.3
J	1	1					4			66.7
C						1		3	1	60.0
P									6	100.0
Overall Percent (%)	14.9	21.3	6.4	6.4	4.3	6.4	10.6	10.6	19.1	74.5

EV: *Eucalyptus* EC: *Eucalyptus* R: *Peltogyne* sp. G: *Gochnatia* sp. D: *Dipteryx* sp. A: *Apuleia* sp. J: *Jacaranda* sp. P: *Aspidosperma* sp. C: *Cedrela* sp.

The model to classify the origin (native or Eucalypt) of charcoal (ANN 2, Table 7) was able to estimate the classes correctly with 97.9% success in the test set. Only one native specimen was misclassified as *Eucalyptus* and the entire remaining set was correctly predicted. This finding indicates that the use of artificial neural networks can be an efficient tool classifying charcoal samplings based on the proportion of mineral elements as input data. In addition to the high percentage of correct classifications, the only mistake that occurred should not lead to an accusation of false fraud, which would be serious if the mistake is to classify Eucalyptus charcoal (legal) as native charcoal (mostly illegal).

Table 7 - ANN classification of charcoal by source (Eucalypt, E or Native, N) using the mineral composition of the charcoals produced at temperatures from 300 to 700°C

Sample	Observed	Predicted by NIR		Correct classifications (%)
		E	N	
Training	E	33	1	97.1
	N	0	60	100
			Overall Percent	98.9
Testing	E	16	0	100
	N	1	30	96.8
			Overall Percent	97.9

Artificial neural networks have proven to be a powerful machine learning tool for function approximation and pattern recognition. ANN has been applied as a modeling tool to overcome various challenges in a number of timber forestry sectors. Some studies have developed ANN models to estimate the wood density (Leite et al., 2016; Demertzis et al., 2017), wood stiffness (García-Iruela et al., 2016), wood strenght (Zanuncio et al., 2017), to assess the surface quality of wood (Hazir and Koc 2018) and to predict the moisture content of wood during drying (Chai et al., 2018; Zanuncio et al., 2016).

In regard to the application of ANN approach in classifications, most studies have shown promissing findings. For insctance, Cui et al. (2019) have used laser-induced breakdown spectroscopy (LIBS) combined with ANN to classify four wooden species and reported a rate of correct classification of specimens of 100% in test set using a model with multilayer perceptron network and Broyden-Fletcher-Goldfarb-Shanno iterative algorithm. Nisgoski et al. (2017) have compared an ANN and SIMCA classifications to identify some Brazilian wood species based on near infrared spectra. Their neural network resulted in no misidentification for a ± 2% margin using a spectral range of 10,000 to 4,000 cm^{-1} while SIMCA produced over 60% misidentification using the raw spectra. Esteban et al. (2017) have developed ANNs to differentiate wood from *Pinus sylvestris* and *Pinus nigra* and their network achieved 90.4% accuracy for the training set and 81.2% for the validation in test set. Wenshu et al. (2015) have studied the detection of defects in wood board based on ANN with an identification rate of 86.67% of success. Castellani and Rowlands (2009) have built na evolutionary artificial neural networks for classifying wood veneers from statistical characteristics of wood sub-images. Experimental evidence from this study showed that their algorithm builds highly compact multilayer perceptron structures capable of accurate and robust learning.

The studies reported above show that artificial neural networks are robust techniques capable of analyzing complex data. To our knowledge, no study has applied neural networks for charcoal classifications, especially to evaluate the mineral composition of charcoal.

The data used as input variables in ANN for evaluating wood can be physical and mechanical characteristics (Nasir et al., 2018), heat treatment temperature (Van Nguyen et al., 2018, Zanuncio et al., 2017), tree age (Leite et al., 2016), wood species (Van Nguyen et al., 2018), basic density (Zanuncio et al., 2016), basal area (in m^2 / ha), annual average increment (in m^3/ ha / year), total height and diameter at 1.3 m from the ground (Leite et al., 2016). This study is pioneering in using mineral elements contained in charcoals as predictive variable in ANN modeling.

3.4 Limitations of this study

The rapid identification of charcoal origin can be carried out through the artificial neural networks developed on this exploratory study. The approach used in this study shows that it is possible to create an automated process to determine the legality of the charcoal load and then reduce fraudulent trade of charcoal. However, robust models may be further developed taking into account more wooden species and pirolysis processes. Complementary studies are necessary to build a robust data of mineral composition of charcoal, including samples of several wooden species, pyrolysis kilns, temperatures, dimensions, moisture, etc.

4. CONCLUDING REMARKS

The findings reported in this study show the great potential for the use of artificial neural networks as systems to identify the charcoal origin when traditional qualitative or quantitative methods cannot be used.

Classification of charcoal specimens by origin (native or Eucalyptus) by ANN 1 reached 97.9% of correct classification in validations from independent test set while the ANN 2 correctely predicted 74.5% of charcoal specimens by wooden species in test set.

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