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APPLICATION OF ARTIFICIAL NEURAL NETWORKS TO PREDICT NO EMISSIONS INTO BIOMASSES COMBUSTION PROCESSES

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Abstract. Replacing part of the use of fossil fuels in energy generation is an attractive solution to reduce its harmful effects on the Earth's atmosphere. During fossil fuel burning, some air pollutants are generated, strongly aggravating the release of greenhouse gases. On the other hand, the use of biofuels reduces the rates that effectively contribute to the Planet's environmental degradation. Several numerical models are widely used as a promising alternative to predict the pollutants emissions from burning biomasses and are considered reliable, saving time and financial resources. This study applied artificial neural networks (ANNs) for the prediction of nitrogen oxide emissions (NO) under different biomasses combustion, using a database with 40 lignocellulosic materials. For evaluation of numerical prediction models, 6 precision criteria were established: Mean Square Error (MSE), Root Mean Square Error (RMSE), Average Absolute Error (AAE), Average Bias Error (ABE), Mean Absolute Error (MAE), and linear regression coefficient (R). It was observed that the feedforward backpropagation model with 10 hidden neurons on the first layer and 15 neurons on the second (FF10x15 model) obtained a precision of 99.98% to estimated and predicted values for the NO emissions of the different samples evaluated, i.e., 1639.14 and 1639.10 mg Nm⁻³, respectively, showing the best performance for the six criteria established. Results showed that numerical prediction presented an excellent performance, and great accuracy, and can serve as a viable alternative for obtaining the NO emissions from different biomasses combustion when the employ of certain experimental processes becomes more difficult.

Keywords: lignocellulosic materials, air pollutants, numerical models, nitrogen oxides.

1. INTRODUCTION

The using fossil fuels in energy generation in recent years have caused high environmental degradation, as well as unrestrained emissions of greenhouse gases (Lela *et al.*, 2016; EPE, 2021). Technological advances and increase energy demand from fossil fuels (coal, oil, and natural gas) will not be enough to meet the main needs required by modern society (REN21, 2020; Covert *et al.*, 2016). Thus, ensuring the world's energy supply has become a huge challenge due to the instability of the current scenario of these fuels (Bharathiraja *et al.*, 2018).

Due to concerns about environmental protection, climate change, as well as human health, animals, and plants, some world leaders have made strong investments in alternative sources for the electricity generation. Such investments aim to reduce demand for non-renewable sources, which are largely responsible for greenhouse gas emissions, in addition to particulate matter emissions and other components harmful to the environment (Williams *et al.*, 2012; Lela *et al.*, 2016; Lopes, 2016). For example, the effects of nitrogen oxides (NO_x) on plants vary considerably for each species, between 0.8 and 1.0 cm³ m⁻³. A characteristic of these species, when affected by excessive emissions of nitrogen oxides is to suffer chlorosis (insufficient chlorophyll production, causing a change in the original color) and necrosis (cell death) in the leaves or simply, reduction in growth (Garbin and Dillenburg, 2008; Sheng and Zhu, 2019). In addition, nitric oxide (NO) emissions contribute approximately 75% of the total emissions of nitrogen oxides and these cannot exceed 4 cm³ m⁻³, ensuring safety for human health (Roy *et al.*, 2014; Cruz *et al.*, 2019).

An alternative to solve these problems is clean energy production from renewable sources, for instance, solid waste, mainly biomasses, as a possible option to replace even partially the energy sources, using fossil fuels (Rockström *et al.*,

2017). Regarding the conditions of renewable energy production through biomass, specific types of equipment are needed to extract biofuels and quantify the energy potential of these, and the air pollutants emissions, whose measurement instruments can be very expensive. Thus, several methodologies can be used to save time and financial resources, since they provide results as accurate as those obtained experimentally (Mateus *et al.*, 2021; Santos and Bordado, 2018; Fournel *et al.*, 2015).

Given the above, this study investigated the prediction of NO emissions through the predictive model of artificial neural networks (ANNs), which are based on the ultimate analysis – contents of carbon (C), hydrogen (H), nitrogen (N), sulfur (S) and oxygen – and proximate analysis (moisture and ash contents). This research used a database with 40 biomasses around the world for the training, validation, and testing steps of the numerical model employed, applying several evaluative metrics to ensure the performance and accuracy of the predictive model.

2. MATERIALS AND METHODS

2.1 Database preparation

Table 1 summarizes a database with 40 (forty) biomasses, which were used in the elaboration of predictive models, and also obtained from the studies developed by García *et al.* (2012), Cruz *et al.* (2019), Malat'ak *et al.* (2020), and Silva *et al.* (2022). All samples contained input data, which are represented by the ultimate analysis of the selected biomasses – carbon (C), hydrogen (H), nitrogen (N), sulfur (S), and oxygen (O) contents – and moisture (W) and ash contents, as well as the respective output data, *i.e.*, NO gaseous emissions. All databases were composed of a numerical range for the minimum and maximum values.

Table 1. Numerical data of inputs and output for all selected biomasses.

| Input data | Samples number | Numerical ranges | | References |
|---------------------------|----------------|------------------|---------|---|
| | | Minimum | Maximum | |
| C (%) | 40 | 20.32 | 54.74 | García <i>et al.</i> (2012) Cruz <i>et al.</i> (2019) Malat'ak <i>et al.</i> (2020) Silva <i>et al.</i> (2022) |
| H (%) | 40 | 0.03 | 6.71 | |
| N (%) | 40 | 0.07 | 6.31 | |
| S (%) | 40 | 0.00 | 0.84 | |
| O (%) | 40 | 32.22 | 69.06 | |
| W (%) | 40 | 5.30 | 24.40 | |
| Ash (%) | 40 | 0.40 | 45.91 | |
| Output data | Samples number | Numerical ranges | | |
| | | Minimum | Maximum | |
| NO [mg Nm ⁻³] | 40 | 10.45 | 3938.94 | |

2.2 Creation of ANNs to predict the NO emissions

The model was created in the software Matlab® version 2020b, through the toolbox *nntool*, in which all networks presented two layers of hidden neurons. According to Silva *et al.* (2019), this setting allows for greater mapping of information from the inputs to the outputs. The first hidden layer ranged from 5 to 10 neurons, and the second layer ranged from 5, 10, and 15 neurons, as can be verified in Table 2.

Table 2. Established typologies for the ANNs created in *nntool* (Matlab®).

| Designation of ANNs | First layer | Second layer |
|---------------------|-------------|--------------|
| FF5x5 | 5 neurons | 5 neurons |
| FF5x10 | 5 neurons | 10 neurons |
| FF5x15 | 5 neurons | 15 neurons |
| FF10x5 | 10 neurons | 5 neurons |
| FF10x10 | 10 neurons | 10 neurons |
| FF10x15 | 10 neurons | 15 neurons |

The model for the prediction of NO emissions consists of 6 ANNs, with 7 inputs, as can be observed in Figure 1.

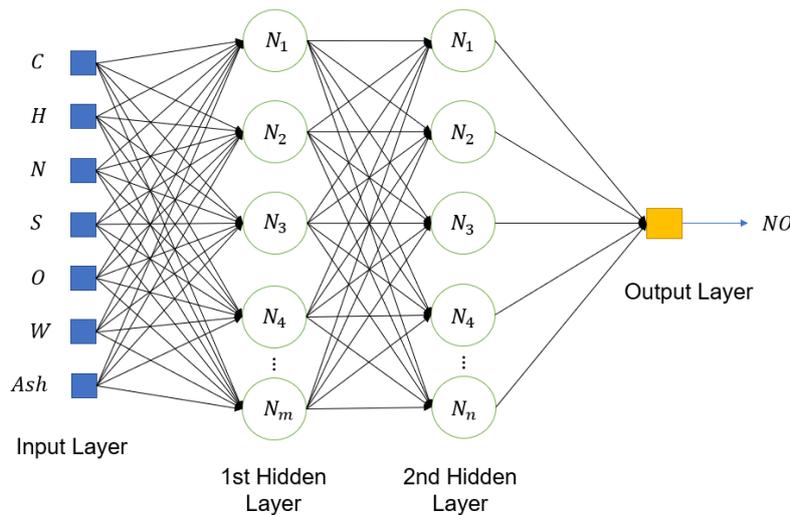


Figure 1. Structure representation of ANNs, with m varying from 5 to 10 neurons, and n between 5, 10 and 15 neurons.

For the learning algorithm, the Levenberg-Marquardt method was designated, which is commonly used in studies that obtained high accuracy of precision (Sharma and Venugopalan, 2014), as well as the gradient descent method used in backpropagation networks. The Mean Square Error (MSE) was chosen as a performance function for the evaluation of the parameters set of training and tests in the prediction of NO emissions. Finally, the activation function established was the hyperbolic tangent, which kept the output results normalized between -1 (one negative) and +1 (one positive). In addition, 1000 (one thousand) times were established as a limit to the training process. Table 3 shows the hyperparameters selected for the training step of the ANNs.

Table 3. Hyperparameters used in the training process of the ANNs predictive models

| Parameters | Specification |
|-----------------------------------|-------------------------------|
| Type of artificial neural network | Feed-forward backpropagation |
| Training algorithm | Levenberg-Marquardt (TRAINLM) |
| Adaptation function | Gradient descent (LEARNGD) |
| Performance function | Mean Square Error (MSE) |
| Activation function | Hyperbolic Tangent (TANSIG) |
| Epochs | 1000 iterations |
| Learning rate | 1% |
| Early stop | 10 epochs |

2.3 Evaluation metrics

For evaluation of the performance of artificial neural networks in the prediction of the NO emissions and accuracy of the results obtained after the application of the test set, some errors types were applied: Average Absolute Error (AAE), Average Bias Error (ABE) and Mean Absolute Error (MAE), which were used for the same purpose by several authors (García *et al.*, 2014; Nhuchhen and Afzal, 2017; Yin, 2011; Han *et al.*, 2017). AAE assessed the accuracy of the results and proximity to the estimated value, *i.e.*, low values of AAE indicates a good accuracy. On the other hand, positive values for ABE indicate overestimated values and, likewise, negative values suggest underestimated values. The MAE represents the amount of error between the estimated and predicted values in the same unit (Monteiro *et al.*, 2021).

It is interesting to note that values close to zero for AAE and ABE show that the model is considered reliable (less than 5%), and low values of MAE indicate a better accuracy with estimated results (up to 7 in the same unit – for the case of the NO emissions [mg Nm^{-3}]) (Chen *et al.*, 2011; Choi *et al.*, 2014; Bousdira *et al.*, 2014; Setyawati *et al.*, 2015).

The accuracy of the models was evaluated using the Mean Square Error (MSE) and Root Mean Square Error (RMSE). The MSE and RMSE represent aggregations of the error magnitude, *i.e.*, the greater the difference between the predicted and actual values, the greater these errors will be.

The statistical errors: AAE, ABE, MAE, MSE, and RMSE are defined using Equations 1 to 5, respectively.

$$AAE(\%) = \frac{1}{n} \sum_{i=1}^n \left| \frac{NO_{(p)i} - NO_{(e)i}}{NO_{(e)i}} \right| \times 100 \quad (1)$$

$$ABE(\%) = \frac{1}{n} \sum_{i=1}^n \left(\frac{NO_{(p)i} - NO_{(e)i}}{NO_{(e)i}} \right) \times 100 \quad (2)$$

$$MAE(mg Nm^{-3}) = \frac{1}{n} \sum_{i=1}^n |NO_{(p)i} - NO_{(e)i}| \quad (3)$$

$$MSE(mg Nm^{-3}) = \frac{1}{n} \sum_{i=1}^n |(NO_{(p)i} - NO_{(e)i})^2| \quad (4)$$

$$RMSE(mg Nm^{-3}) = \sqrt{\frac{\sum_{i=1}^n (NO_{(p)i} - NO_{(e)i})^2}{n}} \quad (5)$$

Where n represents the sample number, and indices p and e were used for predicted and experimental values, respectively.

3. RESULTS AND DISCUSSION

Table 4 summarizes the better results obtained by artificial neural networks in the prediction of NO emissions based on the combustion process, containing the correlation coefficients (R) between estimated and predicted values for the NO emissions, and the MSE of the models for each stage of the learning process extracted from the *nntool*.

Table 4. Performance of the ANNs predictive models for the NO emissions.

| ANNs Typologies | Validated epochs | Stages | R | MSE |
|-----------------|------------------|------------|--------|-----------------------|
| FF5x5 | 12 | Training | 0.9970 | 2.28×10^{-3} |
| | | Validation | 0.9931 | 6.47×10^{-3} |
| | | Test | 0.9944 | 1.08×10^{-2} |
| FF5x10 | 12 | Training | 0.9891 | 7.26×10^{-3} |
| | | Validation | 0.9975 | 4.13×10^{-3} |
| | | Test | 0.9991 | 1.09×10^{-3} |
| FF5x15 | 11 | Training | 0.9985 | 1.24×10^{-3} |
| | | Validation | 0.9999 | 1.93×10^{-4} |
| | | Test | 0.9990 | 1.06×10^{-3} |
| FF10x5 | 15 | Training | 0.9997 | 2.32×10^{-2} |
| | | Validation | 0.9947 | 9.73×10^{-3} |
| | | Test | 0.9924 | 2.39×10^{-4} |
| FF10x10 | 11 | Training | 0.9993 | 7.69×10^{-4} |
| | | Validation | 0.9992 | 9.31×10^{-4} |
| | | Test | 0.9997 | 1.45×10^{-3} |
| FF10x15 | 13 | Training | 0.9999 | 1.11×10^{-4} |
| | | Validation | 0.9999 | 3.21×10^{-5} |
| | | Test | 0.9994 | 5.92×10^{-4} |

The ANN with typology FF5x5 obtained the worst performances in the validation and test stages among all selected models, with the lowest MSE and correlation coefficients (R) in the training stage (2.28×10^{-3} and 0.9970, respectively). Three models presented satisfactory performances: FF5x10, FF5x15, and FF10x15 ANNs. The FF5x10 and FF5x15 models presented both MSE errors and R-coefficient in the validation and test stages better than those of the training stage. However, the smallest MSE errors of the FF5x10 ANN occurred in the test stage (1.09×10^{-3}), while for the FF5x15 model such an effect was observed in the validation stage (1.93×10^{-4}). On the other hand, the FF10x15 ANN presented the lowest errors among all steps about the mentioned networks, with the lowest MSE error and better R-coefficient in the validation stage (3.21×10^{-5} and 0.9999, respectively). The FF5x10, FF5x15, and FF10x15 models for the prediction of the NO emissions operated until 12, 11, and 13 epochs, respectively, in which there were 10 iterations and MSE error increased in the test stage, after this situation, the training step is interrupted by early stop.

Table 5 shows the performance of the main three selected models. These models were chosen considering that the MSE errors in the test and validation stage were smaller than in the training stage, *i.e.*, they have not characterized

overfitting (MSE errors in the training stage lower than those in the test and validation stages, harming network learning). Based on the statistical errors analysis, the FF10x15 model presented a better performance, with the lowest errors AAE and ABE (6.50% for both), indicating an optimal accuracy, with the predicted values for the NO emissions slightly higher than estimated values, besides presenting an optimal accuracy based on MAE error (0.04 mg Nm⁻³). On the other hand, the largest AAE and ABE errors evaluated were for the F5x15 model with nitric oxide prediction of order 12.25% and -12.25%, respectively, which indicated a regular accuracy, with the predicted values not much lower than the estimated values. The AAE and ABE values found were better compared to other predictive models, using biomasses ultimate analysis in the range from 10.92 to 29.19% for AAE, and from -29.19 to 10.97% for ABE (Boumanchar *et al.*, 2019; Demirbas A. and Demirbas A.H., 2004; Yin, 2011; Sheng and Azevedo, 2005; García *et al.*, 2014; Toscano *et al.*, 2013; Elneel *et al.*, 2013; Maksimuk *et al.*, 2016; Qian *et al.*, 2016).

Table 5. Statistic errors analysis for the better selected ANN models.

| Models | Errors | | | | |
|---------|---------|---------|----------------------------|-----------------------------|--------|
| | AAE (%) | ABE (%) | MAE (mg Nm ⁻³) | RMSE (mg Nm ⁻³) | R |
| FF5x10 | 7.77 | -7.77 | 6.24 | 129.00 | 0.9950 |
| FF5x15 | 12.25 | 12.25 | 3.93 | 63.75 | 0.9988 |
| FF10x15 | 6.50 | 6.50 | 0.04 | 25.74 | 0.9998 |

The lowest errors for the NO prediction were found for MAE and RMSE, when using the FF10x15 ANN model, with 0.04 and 25.74 mg Nm⁻³, respectively, indicating an excellent accuracy and minimal variances between predicted and estimated values for NO emissions. On the other hand, the highest MAE and RMSE errors belonged to the FF5x10 model, for the NO prediction, at values of 6.24 and 129.00 mg Nm⁻³, indicating low accuracy and high variances between predicted and estimated values, respectively. In addition, the RMSE value for FF5x10 suggested the presence of outliers, *i.e.*, point differences between predicted and estimated values for this predictive model, whose variance is so discrepant to the point of impairing (or increasing) the RMSE calculation (RMSE > MAE). It is noteworthy that the FF10x15 model also presented high values for the RMSE to the MAE errors, which indicates the presence of outliers in the predictive model. However, the variances pointed out by RMSE not affected the prediction of ANN for the NO emissions. The MAE values were better when compared to other predictive models, using biomasses ultimate and proximate analyses, ranging from 3.80 to 8.75% for MAE (Sheng and Azevedo, 2005; Huang *et al.*, 2008; Elneel *et al.*, 2013; Cellejon-Ferre *et al.*, 2011). The RMSE values for the [mg Nm⁻³] units were not found in the literature for comparative purposes.

Figure 2 presents a linear regression for the FF10x15 model, reaffirming that was observed employing statistical errors analysis (excellent prediction and minimal variances between predicted and estimated values for the NO emissions), and also illustrating the determination coefficient (R²).

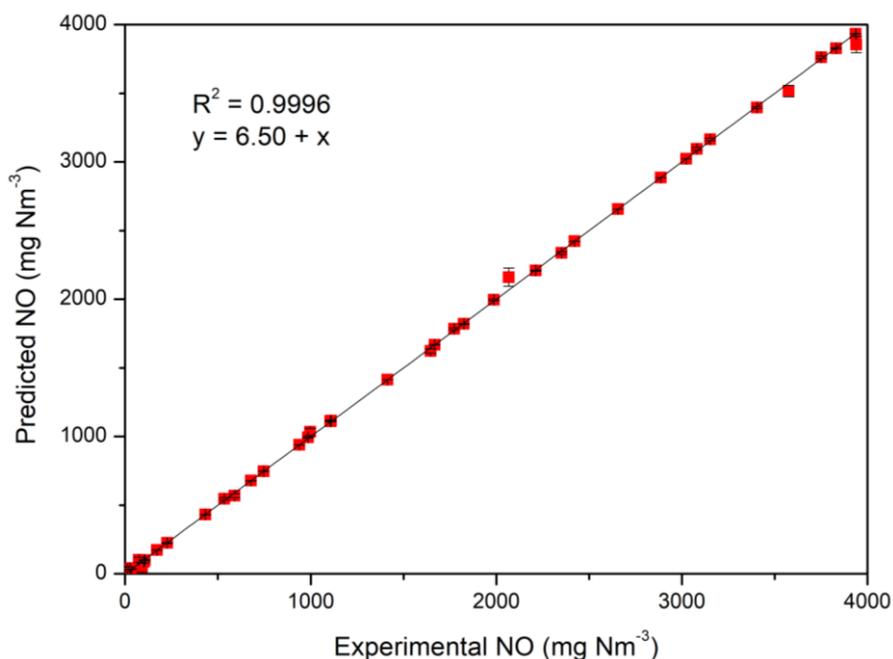


Figure 2. Experimental *versus* predicted NO values for the biomasses selected by FF10x15 model.

For the samples used in the database, which were employed for the training step of the ANNs created, 6 (six) biomasses were selected to be evaluated for the predicted values for the nitric oxide (NO) emissions as illustrated in Figure 3. All biomasses presented the NO concentrations values measured experimentally through the studies carried out by our research group members (Cruz *et al.*, 2019; Silva *et al.*, 2022).

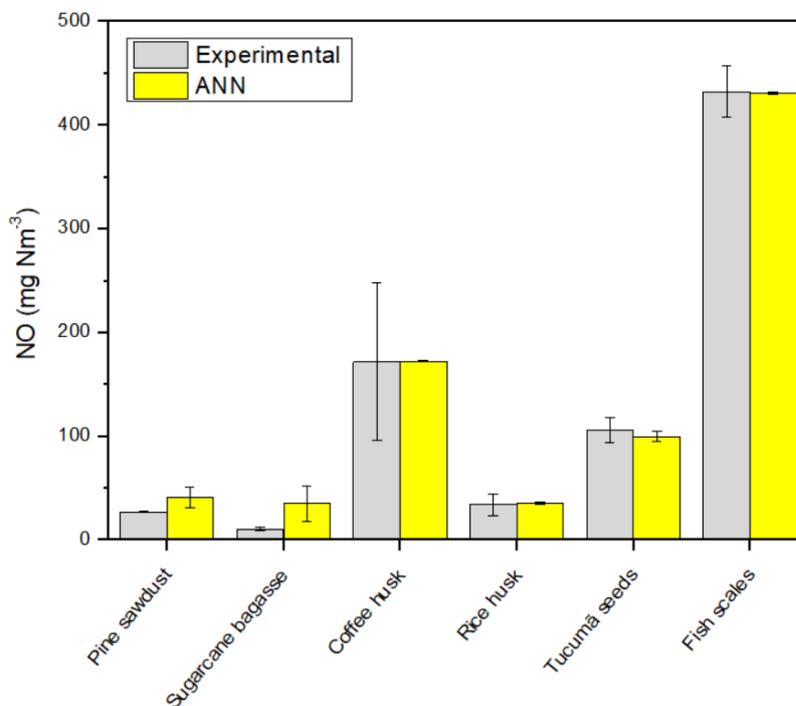


Figure 3. Experimental values for the NO emissions from some biomasses compared to the values predicted by ANN FF10x15 model.

It was noted that the greater divergences were between the real and predicted values for the NO emissions from the samples of pine sawdust, sugarcane bagasse, and *tucumã* seeds, which presented outliers in the input data, possibly due to the sulfur contents obtained by ultimate analysis, which were not experimentally detected, as noted by Mateus *et al.* (2021), and can be observed in Table 6. According to Rousseeuw and Leroy (1987), Barnett and Lewis (1995), Azme and Mokhtar (2004), Khamis *et al.* (2005), Araújo Júnior *et al.* (2021), and Nguyen *et al.* (2021), an outlier is a set of data to be an observation or subset of data which appears to be inconsistent with the remainder of that data set, which will influence the modeling accuracy as well as the estimated parameters, especially, in statistical analysis. It is interesting to highlight, that sulfur contents for the samples mentioned were defined as null values for the creation of predictive models in Matlab® software.

Table 6. Input data of the selected biomasses with some not detected values for the sulfur contents.

| Biomasses | C (%) | H (%) | N (%) | S (%) | O (%) | W (%) | Ash (%) | References |
|---------------------|-------|-------|-------|-------|-------|-------|---------|-------------------------------|
| Pine sawdust | 46.60 | 6.17 | 0.40 | n.d. | 38.43 | 7.20 | 1.20 | Cruz <i>et al.</i> (2019) |
| Sugarcane bagasse | 45.05 | 5.57 | 0.25 | n.d. | 37.93 | 6.40 | 4.80 | |
| Coffee husks | 43.13 | 5.93 | 1.55 | 0.67 | 32.22 | 8.20 | 8.30 | |
| Rice husks | 39.11 | 4.91 | 0.31 | 0.59 | 36.78 | 7.10 | 11.20 | |
| <i>Tucumã</i> seeds | 48.83 | 6.71 | 0.88 | n.d. | 32.98 | 5.30 | 5.30 | |
| Fish scales | 20.32 | 3.52 | 6.31 | 0.79 | 69.06 | 11.34 | 45.91 | Silva <i>et al.</i> (2022) |

n.d.: not detected or below the equipment detection limit.

Even so, although the presence of outliers affected some biomasses, in general, they do not impair the performance and accuracy of the FF10x15 model, selected as the best ANN in the prediction of NO emissions, reaffirming the higher confiability of the prediction model's performances for the selected database.

4. CONCLUSION

This study presented the investigation of nitric oxide (NO) emitted from various biomasses under combustion processes for using as biofuels for the renewable energy generation, from predictive numerical models as an alternative to experimental work, in the lack of financial resources and operational time reduction. Numerical modeling was developed, taking into account the ultimate analysis of the samples as input data, and moisture and ash contents of the proximate analysis. Statistical errors analysis demonstrated an optimal accuracy of artificial neural network (ANN), with low values for the AAE and ABE (6.50%), MAE (0.04 mg Nm⁻³), and an excellent linear regression coefficient (R), with an accuracy of 99.98% concerning predicted and experimental values.

In general, the FF10x15 model (10 hidden neurons in the first layer and 15 neurons in the second layer) presented better performance and excellent reliability for estimation of the values for the NO emissions, with a determination coefficient (R) of the order of 99.96%. Therefore, it was justified the use of numerical models for the prediction of biomasses NO emissions, when these are submitted to combustion processes, like a viable alternative to the application of experimental and theoretical methods, whose the prediction of this pollutant air and other gaseous emissions, which are extremely essentials for using of these biofuels in real thermal processes, considering that artificial neural network proved results reliable, confiability and great precision.

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